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Osaka University
Quadrupole Effects in NMR Spectra on Short-Lived $\beta$-Radioactive Nuclei, $^{12}\text{B}$ and $^{12}\text{N}$

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Abstract

Quadrupole effects in NMR spectra on polarized $^{12}$B($I^r = 1^+$, $T_1 = 20$ ms) and $^{12}$N($I^r = 1^+$, $T_1 = 11$ ms) produced through $^{11}$B(d, p)$^{12}$B and $^{10}$B($^3$He, n)$^{12}$N reactions respectively and implanted in polycrystalline samples of bcc metals, TiB$_2$ and ZrB$_2$ were studied. The nuclear polarization was preserved by use of the preservation magnetic field and monitored by the resultant asymmetric $\beta$-decay. The initial polarization of recoiled nuclei at a reaction angle of 30 degrees were $\sim 16 \%$ at the incident energies $E(d) = 1.43$ MeV and $E(^3$He) = 3.8 MeV for respective reactions.

In order to observe the quadrupole effects in NMR spectra, a frequency modulated rf field of a suitable range had to be employed because the expectable change of the $\beta$-decay asymmetry was only a small fraction of initial one. The spectra on $^{12}$B and $^{12}$N implanted in bcc metals showed clear characteristics of the quadrupole interaction, which indicated the interstitial sites of the implanted elements in the bcc metals of which atoms were heavier compared with $^{12}$B and $^{12}$N. The observed spectra were analyzed by use of the formula of quadrupole effects in high magnetic field. The analyses confirmed the nuclear spins to be $I(^{12}$B) = 1 and $I(^{12}$N) = 1 which were in agreement with the results from the $\beta$-decay data. The results on quadrupole coupling constants are $\mathcal{J}_Q(^{12}$B in Ta) = $3eQ\sqrt{2} = (202 \pm 10)$kHz, $\mathcal{J}_Q(^{12}$B in Nb) = $105 \pm 11$kHz, $\mathcal{J}_Q(^{12}$B in Mo) = $186 \pm 26$kHz, $\mathcal{J}_Q(^{12}$B in W) = $(340 \pm 40)$kHz, $\mathcal{J}_Q(^{12}$N in Ta) = $(0.91 \pm 0.12)$MHz, and $\mathcal{J}_Q(^{12}$N in Nb) = $(0.96 \pm 0.11)$MHz. Large differences among the coupling constants were found, i.e. the field gradient acted on $^{12}$B in the metal of the Vth group in the periodic table was almost twice larger than that in the metal of the Vth group in the same period, and the field gradient acted on $^{12}$N(Vth group) in a metal of Vth group were $5 \sim 10$ times larger than that acted on $^{12}$B(IIIth group).

In order to determine the nuclear quadrupole moment of $^{12}$B, measurements were made on TiB$_2$ and ZrB$_2$ of which quadrupole coupling constants on $^{11}$B have been
known. It was essential that the implanted $^{12}\text{B}$ were substituted for the original $\text{B}$ in the samples so that the known internal field was effective. The substitution was supported by the fact that the ratio of the field gradients which acted on $^{12}\text{B}$ was in agreement with the known ratio on $^{11}\text{B}$. The results are $\mathcal{E}(^{12}\text{B} \text{ in TiB}_2) = (231 \pm 24)\text{kHz}$, and $\mathcal{E}(^{12}\text{B} \text{ in ZrB}_2) = (74 \pm 7)\text{kHz}$. Comparison with known coupling constants on $^{11}\text{B}$ gives the ratio of the quadrupole moments to be $\left| q(^{12}\text{B})/q(^{11}\text{B}) \right| = 0.42 \pm 0.04$. The known value of the quadrupole moment of $^{11}\text{B}$ yields the quadrupole moment to be $\left| q(^{12}\text{B}) \right| = (0.0151 \pm 0.0014) \text{ barns}$. 
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1 Introduction

The nuclear electric quadrupole moment is one of the basic physical quantities. The studies on the quadrupole moments together with the magnetic moments play important roles in the nuclear physics, in the atomic physics, and in the solid state physics.

Historically the experimental study on the nuclear moments of atomic nuclei started in the investigation of the hyperfine structure of atomic spectra. The interaction of the nuclear magnetic moment with the magnetic field produced by the atomic electrons gives rise to hyperfine spectra. Among the data, there were a few spectra in which marked departures from the empirical rule occurred. These could be attributed to the presence of a quadrupole interaction. The radio-frequency-resonance technique which was invented by Rabi et al.\(^1\) and successfully applied to the atomic beam technique greatly improved the situation of the study. This principle was further developed into the nuclear magnetic resonance (NMR) technique by Bloch, Purcell, and Pound.\(^2\) These methods have not only brought much more precise determination of the nuclear moments but also vast increase of the related knowledges.

The applicability of these methods are restricted to the stable and the long-lived nuclear states by the experimental conditions in spite of the vast contributions to the studies on the nuclear moments. The nuclear lifetime has to be longer than a few minutes. Recently a new NMR technique has been developed by K. Sugimoto et al.\(^3,4\) which is applicable to the measurements of the nuclear magnetic moments of the \(\beta\)-radioactive nuclei of which lifetime lie within the range of \(10^{-5}\) sec \(\sim\) 10 sec. The method essentially is based on the following four points: the production of the polarized \(\beta\)-radioactive nuclei through a nuclear reaction, the preservation of the polarization by use of a preservation magnetic field and by means of the implantation of polarized recoil nuclei in a suitable sample, the observation
of the nuclear polarization by the asymmetric $\beta$-decay of the polarized nuclei, and the resonant destruction of the polarization by a radio frequency (rf) magnetic field.

This technique can be extended to the investigation of the interaction of implanted nuclei with the internal electromagnetic fields, if the above mentioned four conditions are satisfied. In the present experiment, the electric quadrupole interactions were investigated. In the course of the study, it was shown that the samples of face centered cubic (fcc) metals were good implantation media for recoiled $^{12}$B and $^{12}$N to observe the NMR effects. When the body-centered cubic polycrystalline metals like tantalum were used, no prominent NMR effect was observed while the polarizations were well preserved. The causes were considered to be due to the quadrupole interaction resulted from the asymmetric distribution of the surrounding host ions at the interstitial sites in the body centered cubic (bcc) metals. The causes of the interaction was well certified to be quadrupole interaction in the present study.

In order to determine the nuclear quadrupole moment, the use of the known internal field gradient was attempted. The usable strength of the interaction was limited by the applicability of the present method, i.e. the quadrupole interaction must be weak so that the externally applied magnetic field can preserve the nuclear polarization. The quadrupole coupling constants on $^{11}$B in TiB$_2$ and ZrB$_2$ are known and these samples were suitable for the present purposes.

In this paper we report the studies on the quadrupole effects in NMR spectra of $^{12}$B and $^{12}$N in bcc metals, and on the determination of the quadrupole moment of $^{12}$B. The NMR technique which is applicable on the short-lived $\beta$-radioactive nuclei is summarized in Chapter 3. The formula on the quadrupole effects in NMR spectra, and the theoretical line shapes related to the present work are summarized in Chapter 4. The implantation and the possible sites of the recoil nuclei in the
samples are explained in Chapter 5. The experimental provisions which were improved in the present study, and the procedures are explained in Chapter 6. The experimental results are described in Chapter 7. The results of the present works are summarized in Chapter 9.
Outline of the Experimental Method

Short-lived $\beta$-radioactive nuclei $^{12}\text{B}$ and $^{12}\text{N}$ were produced through $^{11}\text{B} \ (d,p) \ ^{12}\text{B}$, $^{10}\text{B} \ (^{3}\text{He},n) \ ^{12}\text{N}$ reactions. The nuclei ejected to 30 degrees were implanted in a suitable host medium by using their recoil energies as shown in Fig. 2-1. The mean recoil energies were $\sim 400 \text{ keV}$ and $\sim 500 \text{ keV}$ respectively. A static high magnetic field was employed to preserve the initial polarization. The polarization was detected by the asymmetric $\beta$-decay of the nuclei. The polarizations of $\sim 16\%$ were obtained at $E(d) = 1.43 \text{ MeV}$ and $E(^{3}\text{He}) = 3.8 \text{ MeV}$ for respective reactions. A radio frequency (rf) magnetic field was applied perpendicular to the external magnetic field to destroy the polarization resonantly (NMR). Thus the dependence of the $\beta$-decay-asymmetry change on the frequency of the rf magnetic field and on the static magnetic field gives rise to the nuclear magnetic resonance spectrum.

In order to observe the quadrupole effects in NMR spectra, special provisions were necessary because the expected change of the $\beta$-decay asymmetry was only a small fraction of the initial one. A frequency modulated rf-field of a suitable range was employed to increase the observable effects. Still one had to observe a few $\%$ of asymmetry change. To reject the inevitable long term instability of the $\beta$-particle-detecting system, an automatic-data-normalization system was employed which consisted of the periodic rf-excitation in the rf coil and of the signal distribution to the related $\beta$-particle counters in accordance with each period.

The reliability of the system and the conditions which were necessary to the experiment were checked by use of the fcc metals which were good implantation media to observe NMR spectra. The ranges of the rf modulation were examined by analyzing the NMR spectra observed by means of the modulated rf field. The rf field intensity necessary to the experiment was determined by observing the intensity dependence of the $\beta$-decay asymmetry.

The NMR spectra on $^{12}\text{B}$ and $^{12}\text{N}$ in bcc metals such as Nb, Mo, Ta, and W were
Fig. 2-1  Experimental arrangement for the nuclear-magnetic-resonance observation.
examined by means of the modulated rf field to clarify the causes of the wide spread of the NMR spectra, and to justify the considerations on the final sites of the captured elements in the heavy host elements.

To determine a nuclear quadrupole moment, a known field gradient must be used. TiB₂ and ZrB₂ were suitable for the present purposes. The implanted ¹²B had to be substituted for the original B in the samples so that the known field gradient might be used. The substitution of ¹²B was supported by the agreement of the ratio of the field gradients acted on the ¹²B in the samples to that acted on ¹¹B, which means the majority of the ¹²B were substituted. The known coupling constants on ¹¹B give rise to the ratio of the quadrupole moment, and the known quadrupole moment of ¹¹B gives rise to the quadrupole moment of ¹²B.
a) Principles of the Method

The method consists of the four processes as stated in the previous chapter: The production of the polarized nuclei through a nuclear reaction; The recoil implantation and the preservation of the polarization in a suitable host medium; The detection of the asymmetric β-decay of the polarized nuclei and the resonant destruction of the polarization by an rf magnetic field.

The region of the nuclear lifetime to which this method is applicable is limited by the observability of NMR effects and by the actual experimental conditions. A spin I with the magnetic moment $\mu$ precesses in the magnetic field with the frequency of $\frac{1}{\hbar}$, where $H$ is the magnetic field and $\hbar$ is the Larmor frequency. When the frequency of the magnetic field $H \cos \omega t$ which is applied to the implanted nuclei and is perpendicular to the static magnetic field, equal to the Larmor frequency $\hbar$, the rf transitions among the Zeeman levels are induced and the nuclear polarization can be destructed. Thus the knowledge of the strength of the static magnetic field and the frequency of the rf magnetic field at the resonant condition give rise to a nuclear $g$ factor $-\mu/I$ of the nucleus.

According to the uncertainty principle, there is a condition between the energy and life time,$$
\hbar \tau > \hbar.
$$
A nucleus with a magnetic moment $\mu$ has an energy of $E = -\mu H \cdot Iz/I$ in a magnetic field. If $I = 1$, $H = 10^4$ and the uncertainty about the moment to be observed is smaller than $10^{-3}$, the life time must be longer than $2 \cdot 10^{-5}$ sec. The boundary from the actual experimental conditions is that the transitions among the Zeeman levels can be caused sufficiently during the life time of the nucleus. This relation can be written as $\frac{\mu Hz}{\hbar} > 1$.\[3-2\]
If \( \mu = 1 \text{ (nm)} \), \( I = 1 \) and \( H_\parallel \text{ < 20G} \), the life time must be,

\[ T > 10^{-5} \text{ sec} \]

The upper limit of the life time of a nucleus to which the method is applicable, is determined from the view point of the preservation of the polarization. From the various experimental data, a suitable host medium can be selected and the polarization may be preserved for a few minutes.

1) Production of polarized nuclei

The short lived \( \beta \)-radioactive nuclei are produced through a nuclear reaction. The nuclei ejected to a confined recoil angle are expected to be polarized in the direction normal to the reaction plane.

The nuclear polarization depends on many causes involved in the reaction; the direct process or compound process through which the reaction comes, the spin-orbit interaction, and the interference term of the direct and the compound processes. It is considered that the processes are very complicated and that quantitative predictions on the polarization is not so good in spite of the success in the qualitative discussions. Neume and Ref. treated \((\delta, p), (\delta, n)\) reactions in the direct process. They explained the polarization mechanism by the degree of distortion of the incident and outgoing particles. The orbital angular momenta taking part in the reaction are perpendicular to the reaction plane. Thus if there exist some differences between the mean free paths of the incident and the outgoing particles in the nucleus, then the nuclei recoiled to a confined angle are probably polarized. The polarization direction which is parallel or anti-parallel to the normal of the reaction plane will be determined by the relative differences between the mean free paths. In this case the \( L - s \) interaction and the compound process are not included. A. Simon et. al. take the \( L - s \) interaction and the compound process into account, but the theory is difficult in quantitative predictions
in respective cases.

The polarization of $^{12}$B through $^{11}$B(d, p) reaction is experimentally examined by K. Sugimoto$^4$ and L. Pfeuffer$^7$ independently. They observed 45% and 8% polarization respectively in different conditions. The positive sign is defined by the wave vectors $\vec{k}_1$ and $\vec{k}_2$ of the incident and outgoing particles as \( \frac{(\vec{k}_1 \times \vec{k}_2)}{|\vec{k}_1 \times \vec{k}_2|} \).

In this case the rate of polarization larger than 5% is expectable by the improvement of the experimental conditions because L. Pfeuffer did not decouple the hyperfine interaction during the flight of $^{12}$B in vacuum. In the present work polarization of 16% at $E(d) = 1.43$ MeV was observed. On $^{12}$N, K. Sugimoto et. al. observed polarization of 16% through $^{10}$B(3He, n)$^{12}$N reaction.

ii) Preservation of the polarization

When the recoil atoms are ejected from the target into vacuum with their recoil energies, they are in various ionized states of atom. During their flight in the vacuum the states are not filled up by the electrons, and the nuclei are under the influence of a strong hyperfine field. If this interaction is not decoupled, the nuclear polarization will be smeared out. The atomic spin $J$ with the atomic moment $\mu_a$ precesses around the external magnetic field $H$ with the frequency of $(\mu_a H)/(\hbar J)$. If this frequency is far larger than the hyperfine frequency $\omega_H$ of the nuclear spin, the interaction between the atomic spin and the nuclear spin will be decoupled. A high magnetic field applied parallel to the nuclear polarization axis may preserve the nuclear polarization during its flight. In the actual experiment on B or N case, 2kG is sufficient to decouple the interaction.

The recoil nuclei that come into a confined angle which is defined by a collimator are captured in a suitable recoil stopper. Their probable sites in the implantation media and the recoil-range distribution are described in Chapter 5.

In the samples the nuclei are exposed to the several depolarization mechanisms
caused by the collision processes during their flight in the sample and by the surrounding electric charges and nuclear magnetic moments at their final sites. The depolarization mechanisms in a host media are summarized as follows. The recoil nucleus takes less than $10^{-12}$ sec to pass through its range in a solid medium. Although some electromagnetic interactions which arise in the collision processes may be active on the recoil nuclei, the spins of the nuclei will hardly flip their directions during the short period in such atomic processes. When they are trapped in final sites, the lack of the electronic shells have to be quenched. From this view point, metallic stoppers are preferable as implantation media in which the free electrons quickly fill up the electronic shells. Non metallic samples which contain the elements of the same atomic number of the implanted element are also preferable, because the implanted element is expected to be captured in the substitutional. (See Chapter 5.5).

There are long term relaxation mechanisms, the spin-lattice relaxation caused by the thermal motions of the surrounding nuclear magnetic moments and of the surrounding electric charges. N. Bloembergen intensively examined the magnetic relaxation mechanisms of $^{19}$F in CaF$_2$ crystal. He found that the magnetic spin-lattice relaxation time $T_1$ in the pure crystal is longer than $10^2$ seconds in a room temperature. And he also pointed out that the main cause that shortens the relaxation time $T_1$ is the paramagnetic impurity in the crystal. Thus the samples of high purity have to be selected as suitable implantation media. In the case of the pure metal the relaxation time $T_1$ caused by the magnetic interaction is mainly governed by the interaction with the conduction electrons. This is mainly due to the contact interaction of the unpaired $s$ electrons in the conduction band with the nucleus. The relaxation time $T_1$ can be approximately written as follows:

$$ T_1 = \frac{\hbar}{4\pi kT} \frac{(\gamma e)^2}{(\Delta H/H)^2} $$

$$ (\Delta H/H)^2 = \frac{3}{8\pi} < |\psi_{K\ell}(0)|^2 > \chi_p M. $$

- 10 -
where \( T \) is the temperature of the sample, \( \gamma_e \) and \( \gamma_n \) are gyromagnetic ratios of the electrons and the nucleus respectively, \( \chi_p \) is the magnetic susceptibility of conduction electrons per unit mass, and \( M \) is the atomic mass weight. Presuming the \( (\Delta H/\Delta T) \) from various data, \( T_1 \) can be estimated to be \( \sim 0.1 \) sec at a room temperature for the light elements like B and N. J. Wells observed \( T_1 \) to be as large as 257 sec⋅K by the case of \( ^{12}\text{B} \) in Pt \(^{10}\).

By the case the nuclear spin is larger than \( \frac{1}{2} \), the nucleus may have an electric quadrupole moment. Then there arises an electric relaxation mechanism due to the interaction between the moment and an electric field gradient produced at the nucleus by the surrounding electric charge distribution. The static component, however, of the field gradient has no effect on the nuclear depolarization mechanism even when the poly-crystals are used if the strength of the interaction is far smaller than the static magnetic interaction. The relaxation time \( T_1 \) which is governed by the time dependent part of the electric field gradient is approximately

\[
\left( \frac{1}{T_1} \right) \approx \frac{81 \pi}{10} \left( \frac{\alpha^2}{x^2} \right)^2 \left( \frac{\gamma_e \Theta}{\hbar \nu} \right)^2 \left( \frac{T}{\Theta} \right)^2 \Omega
\]

where \( \Theta \) is the Deby characteristic temperature, \( \Omega \) is connected with Boltzmann constant \( k \) and \( \Theta \) by the relation of \( k \Theta = \hbar \Omega \), \( T \) is the temperature of the sample and \( x^2 \) is the strength of the quadrupole interaction which is approximately \( (e/\ell^3)(eQ) \) where \( r \) is the distance to the ionic elements. Assuming that \( Q \) is \( \sim 10^{-25} \) barns, \( \ell \) and \( r \) is \( \sim 2\ell \), \( T_1 \) becomes approximately \( \sim 5 \cdot 10^2 \) sec. In the actual experiment a few seconds of \( T_1 \) was observed \(^9\). This is 100 times smaller than the estimated value. But the relaxation time is far longer compared with the nuclear life time of \(^{12}\text{B} \) and \(^{12}\text{N} \).

Due to the above discussions it is known that by using metallic media: \(^{12}\text{B} \) and \(^{12}\text{N} \), \( ^{7}\text{N} \) and \( ^{11}\text{B} \), \( ^{12}\text{B} \) and \( ^{11}\text{B} \), the nuclear polarization can be preserved.
in polycrystalline samples sufficiently longer than the nuclear life times of $^{12}\text{B}$, $^{12}\text{N}$ whose life times are 20 ms and 11 ms respectively.

iii) Detection of the polarization

The nuclear polarization is observed by detecting the asymmetric $\beta$-decay of the polarized nuclei. The angular distribution of the $\beta$ rays from the polarized nuclei are asymmetric because of the parity nonconservation in weak interactions. The angular distribution of the $\beta$ particle is expressed by

$$W(\theta) = 1 + A \frac{\cos \theta}{C} P \cos \theta,$$

where $\cos \theta$ is the direction cosine between the polarization axis and the momentum of the outgoing $\beta$ particle, $A$ is the asymmetry constant which is determined by the transition matrix element, $\nu$ is the ratio of the velocity of the $\beta$ particle to the light velocity, and $P$ is the nuclear polarization which is connected to the population in the magnetic sublevels,

$$P = \frac{<I_z>/I}{\sum m_p P_m},$$

where $\sum P_m = 1$.

In the case the kinetic energy of the $\beta$ particle is larger than 500 keV, the ratio $\nu$ is nearly equal to one. As the mean energies of the $\beta$ particle emitted from $^{12}\text{B}$ and $^{12}\text{N}$ are about 8 MeV, the ratio $\nu$ is nearly equal to 1. The asymmetry parameter $A$ is written

$$A = \frac{\pm |C_x|^2 |<0>|^2 \Delta I_f^2 - 2 C_x^* <l> C_A <o> \sqrt{I_f/(I_f+1)}}{|C_x|^2 |<l>|^2 + |C_A|^2 |<o>|^2},$$

where the coefficient $\Delta I_f$ is

$$\Delta I_f = \begin{cases} 1 & I_f = I_i - 1 \\ \frac{1}{(I_i+1)} & I_f = I_i \\ -\frac{1}{I_i(I_i+1)} & I_f = I_i + 1 \end{cases}$$

The upper sign in $A$ is applied to the positron emission, $<l>$ is the Fermi-matrix element, $C_x$ is the Fermi-coupling constant, $<0>$ is the Gamow-Teller matrix element, and $C_A$ is the Gamow-Teller coupling constant. According to the experimental data
on its values as shown in Table 3-1, the \( \beta \) decays of \( ^{12}_8 \text{B} \) and \( ^{12}_N \) have been assigned to be allowed and the type of the transitions are assigned to be possibly pure Gamow-Teller. Then the asymmetry parameter \( A \) is reduced to be

\[
A = \pm 1 \quad \text{where} \quad \begin{cases} + & \text{for } ^{12}_N \\ - & \text{for } ^{12}_B \end{cases}
\]

In the actual measurement the counting rate-ratio \( R \) is obtained from the counting rates of \( \beta \) particles emitted parallel to the polarization axis (Ny) and antiparallel \( (N_D) \),

\[
R = \frac{N_U}{N_D} = (1 - p) \frac{G_U}{G_D} \left(1 + p^2 \right)
\]

where \( G_U \) and \( G_D \) contain both geometrical and instrumental asymmetries. When an rf frequency is applied, the polarization is diminished to \( (p - \Delta p) \) depending on the rf intensity and resonance conditions. And the ratio \( R_{on} \) is

\[
R_{on} = \frac{1 - \frac{(p - \Delta p)}{(1 + (p - \Delta p))} \cdot \frac{G_U}{G_D}}{1}
\]

This ratio is normalized by a ratio for the rf off period. Then the asymmetry changes to be observed are

\[
\Delta R = \left\{ \frac{(R_{on})}{(R_{off})} \right\} - 1 \equiv 2\Delta p/(1 - p^2)
\]

where \( \Delta p \), \( p < 1 \).

iv) Resonant destruction of polarization

The magnetization \( M_z \) of the nuclei with a magnetic moment \( \mu \) and the polarization \( P_0 \) is written \( M_z = n \cdot \mu \cdot P_0 \), where \( n \) is the concentration of the nuclei per unit volume. The time dependence of the magnetization is written

\[
\frac{d}{dt} (M_z) = q \mu P_0 - (\Lambda + \lambda + \gamma) H_1 \chi f(\omega) M_z
\]

where \( q \) is the rate of the production of the nuclei per unit volume and unit time, \( \lambda \) is the reciprocal of life time, \( \Lambda \) is the reciprocal of the spin lattice relaxation time \( T_1 \), \( \gamma \) is the gyro-magnetic ratio of the nuclei, \( H_1 \) is the amplitude
of the rf magnetic field, and \( f(\omega) \) is the line shape function; either Lorentzian or Gaussian will do. The number of the nuclei per unit volume is expressed

\[
\frac{dN}{dV}(n) = q - \lambda n
\]

Above equations can be solved by taking the experimental conditions into consideration. The period of nuclear production and the \( p \) countings are alternatively repeated as shown in Fig 3-1.

\[
\left( \frac{P/P_0}{P/P_0} \right) = \frac{\lambda^2}{\mathcal{L}^2} \cdot \frac{1 - \varepsilon - \lambda}{1 - \varepsilon - \lambda - \lambda^2} \cdot \frac{1 - \varepsilon - \lambda - \lambda^2}{1 - \varepsilon - \lambda - \lambda^2} \cdot \frac{1 - \varepsilon - \lambda - \lambda^2}{1 - \varepsilon - \lambda - \lambda^2}
\]

where \( \mathcal{L} = \lambda + \gamma + \mathcal{R} \gamma^2 \mathcal{H}_1^{-2} f(\omega) \)

b) Causes of the Line Broadening

In this section the NMR line width due to various causes other than quadrupole interaction is discussed. The experimental NMR line width is determined by the nuclear dipolar interaction, the finite life-time \( T_1 \) caused by the relaxation mechanism, the finite nuclear life-time, the intensity of the rf magnetic field and the instrumental ones. The hamiltonian of the dipolar interaction between two nuclei is

\[
\mathcal{H}_{12} = \frac{r_{12}^3}{\mathcal{R}_{12}^3} \cdot \left\{ \mathbb{I}_1 \cdot \mathbb{I}_2 - \frac{3(\mathbb{I}_1 \cdot \mathbb{I}_2)(\mathbb{I}_1 \cdot \mathbb{I}_2)}{r_{12}^{-2}} \right\}
\]

where \( r_{12} \) is the distance between the implanted and the host nuclei, \( \gamma_i \) and \( \gamma_k \) are their gyromagnetic ratios. The second moment of the NMR line due to the dipolar interaction with different element in a polycrystalline sample is given by

\[
\left( \frac{1}{T_2} \right)^2 = \frac{4}{15} \gamma_i^2 \gamma_k^2 I_1(I_1+1) \mathcal{H}_{12}^2 \sum_{K} \frac{1}{r_{12}^4}
\]

where the summation \( K \) extends over the nuclei in the crystal and \( I_2 \) is the spin of the surrounding nuclei. By the case the face centered cubic crystal and the body centered cubic crystal in which interstitial sites \( ^{12}_N \) and \( ^{12}_N \) are implanted, the square roots of the second moments calculated by using the equation 3-13 are listed in Table 7-2 together with the experimental widths. The probable sites of the
Fig. 3-1

Period of the nuclear production and the $\beta$-particle counting are alternatively repeated.
recoil nuclei will be discussed in the next section.

When a metal is used for an implantation medium, there may be a broadening due to the anisotropy \(^{11}\) of the Knight-shift. The anisotropy is caused by the interaction of the nuclear spin with the conduction electrons of which distribution in the space is asymmetric. Thus the interaction exists only when the sample is noncubic symmetry about the site under the consideration. This interaction produces a shift of resonance line, which depends on the orientation of the crystallographic axes with respect to the external magnetic field. Then the resonance lines are distributed around the isotropic Knight-shift for a poly crystalline sample. Thus in the actual case of \(^{12}\)B or \(^{12}\)N implanted in the fcc crystal, such an interaction is not expected because of the asymmetric distribution of the neighbouring elements. On the other hand such an interaction may occur in the cases of the bcc crystals. The broadening, however, is estimable to be under the order of the Knight-shift. From the various data, the Knight-shift of the light elements like boron and nitrogen are smaller than \(10^{-3}\). The width of B and N in the metal due to the anisotropy is at most about a few kHz which is comparable with the dipolar width.

Above discussions means that any width which is wider than the dipolar broadening has an important meaning; the existence of the quadrupole interaction.

c) Frequency Modulation

The resonance condition of an unknown nuclear moment in a static magnetic field is searched by use of a frequency modulated rf field of a suitable range. If the following conditions are satisfied, (\(L\)) in equation 3-11 can be approximated to be

\[
L = \Lambda + \lambda + \frac{1}{4\eta}(\pi \gamma^2 H_i^2)\]

The conditions are: \(\delta \ll \Delta \) and \(\lambda \gg \frac{1}{f_m}\), where \(\delta\) is the resonance width and \(f_m\) is the period of the frequency modulation. Then the rf field intensity necessary for the complete polarization-destruction in the experiment by means of
a modulated rf can be estimated.

d) Application of the Present Method

The NMR method which has been successfully applied to the determination of
nuclear magnetic moments of short lived nuclei, can also be applied to various
other fields of studies. The possibilities are summarized.

1) The measurement of the nuclear magnetic moments of short lived \( ^{\beta} \)-radioactive
nuclei.

There are still many short lived \( ^{\beta} \)-radioactive nuclei of which magnetic moments
are not yet measured. Systematic studies of the moment of isospin multiplet are
usefull for the study of the nuclear structure and the mesonic effect in nuclei.

2) The measurement of the nuclear quadrupole moments of short lived \( ^{\beta} \)-radioactive
nuclei.

The method is extended to observe the nuclear quadrupole interaction. In this
method, the strength of the quadrupole interaction is observed as an NMR line width.
And a knowledge of the field gradient determines the quadrupole moments.

3) The research of the solid state physics and the atomic structure.

The recoil implantation technique is employed in the present method. This tech-
nique will be a useful means to observe the interactions between the host material
and the implanted nuclei such as Knight-shift, dipolar interaction, magnetic
interactions and quadrupole interactions with the internal field. The implanted
nuclei can be used as a microscopic probes to explore the internal magnetic field
and the electric field gradient.

4) The study on the isomeric states of the nuclei.

There are still many short lived isomeric nuclei (\( MS \sim 10^5 \)) which decays
by emitting \( \gamma \) rays. It is an interesting problem to study on these nuclei.
4 Electric Quadrupole Interaction

Theoretical treatments of quadrupole interactions together with magnetic interactions are reviewed by many authors\(^2, 13, 14, 15\). In this section, the quadrupole effects in NMR spectra in high magnetic field related to the present study are summarized. One of the conditions on the strength of the quadrupole interaction in the present method by use of the polycrystalline implantation media is

\[
\text{eq}Q \ll \mu H \tag{4-1}
\]

where \(\text{eq}Q\) is the electric-quadrupole coupling constant and \(\mu H\) is the magnetic coupling constant. The condition is necessary for the preservation of the initial polarization in the polycrystal sample in which field gradient is active. The 1st and 2nd order solutions are given in section a) and b). The polycrystal patterns are given section c).

a) Symmetric Electric Field Gradient : 1st Order Perturbation

A space fixed cartesian coordinate \((x, y, z)\) is defined as follows to describe the interaction hamiltonian. The \(z\) axis is taken in the space parallel to the magnetic field as a quantization axis. The origin of the coordinate is indicated by \(O\).

According to the electrodynamics the electric field gradient can be expressed by two parameters, defined by \(\eta = V_{zz}\) and \(\xi = (V_{xx} - V_{yy})/\xi\), where \(|V_{zz}| \leq |V_{yy}| \leq |V_{xx}|\). Then the interaction hamiltonian can be expressed when the electric field gradient is uniaxially symmetric by putting \(\varphi\) in the \(xOz\) plane as shown in Fig. 4-1.

\[
H = H_M + H_a \tag{4-2}
\]

\[
H_M = -\hbar^2 \frac{\eta}{\xi} H \cdot I_z \equiv -\hbar^2 \eta H \cdot I_z \tag{4-3}
\]

\[
H_a = \frac{e_0 Q}{4 I (2 I - 1)} \left[ \frac{1}{2} (3 \cos^2 \theta - 1) (3 I_z^2 - I_z) \right. \\
\left. + \frac{3}{4} \sin^2 \theta (I_z^2 (I_z + I_z) + (I_z + I_z) I_z) \right] \tag{4-4}
\]
The $(x, y, z)$ is the space fixed cartesian coordinate, and $(x', y', z')$ is fixed to the field gradient.

Fig. 4-1

The quadrupole shifts are exaggerated compared with the magnetic shifts.

Fig. 4-2

$\Theta = 0, \pi \pm \sin \frac{1}{\sqrt{3}}$

Fig. 4-3

$\Theta \Theta Q = 0$

Fig. 4-4
where \( H_M \), \( H_Q \) are the magnetic and quadrupole interaction hamiltonians respectively, and the \( H_Q \) is regarded as a perturbation of \( H_M \), where \( \gamma \) is the gyromagnetic ratio which is connected to the Larmor frequency \( \Omega_L \) by \( \Omega_L = \gamma H_0 \). \( I \) is the nuclear spin and \( H_0 \) is the uniform magnetic field. By the 1st order perturbation calculation the energies of the magnetic sublevels are obtained.

\[
E_{0m} = -m \hbar \Omega_L \\
E_{1m} = \frac{\gamma \hbar}{12} \left\{ 3m^2 - I(I+1) \right\} \left( 3 \cos^2 \theta - 1 \right) \hbar
\]

where \( E_{0m} \) is the energy of magnetic sublevel \( m \) of 0th order perturbation. When the nuclear spin is \( I = 1 \), two transitions among the sublevels are possible. Each transition frequency can be written as \( \Omega_L \pm \Omega_\perp \), corresponding to the transitions between the magnetic sublevels \( m = 1 \) and \( m = 0 \), and between \( m = 0 \) and \( m = -1 \) respectively.

\[
\Omega_\perp = \frac{\gamma \hbar}{4} \left( 3 \cos^2 \theta - 1 \right).
\]

As seen in the equation (4-7) the \( \Omega_\perp \) depend on the direction cosine between the electric field gradient and the magnetic field. They take extrem values when the two axes are parallel or perpendicular. This trends are shown in Fig. 4-2.

b) Asymmetric Electric Field Gradient

The hamiltonian of the quadrupole interaction by the case of an asymmetric electric field gradient, depends not only on the angle between \( q \) and \( H_0 \) but also on the relative relations of \( V_{xx} \) and \( V_{yy} \) as to \( H_0 \). Thus the principal axes in Fig. 4-1 is rotated around the \( q \) to represent this orientation in general. This relation is drawn in Fig. 4-3. The hamiltonian \( \hat{H}_Q \) in the equation (4-2) is rewritten

\[
\hat{H}_Q = \frac{e^2 q Q}{4I(2I-1)} \left[ \frac{1}{2} (3 \cos^2 \theta - 1) \left\{ 3I_z^2 - I(I+1) \right\} + \frac{I}{2} \sin \theta \cos \theta \left\{ I_x (I+1)_x + (I+1)_x I_z \right\} \right. \\
\left. + \frac{3}{4} \sin^2 \theta (I^2 + I_z^2) + \frac{1}{4} \cos 2\psi (I^2 + I_z^2 \cos \theta) (I^2 + I_z^2 \sin \theta). \right]
\]

-20-
where \( I_\pm = I_\chi \pm i I_y \)

By using the 1st and 2nd order perturbation theory, the energies of the magnetic sublevels can be calculated.

\[
E_{\text{om}} = -m\lambda_L h
\]

\[
E_{1m} = \left( \frac{\lambda_L}{12} \right) \{ 3m^2 - 7(I+1) \} \{ (3\cos^2 \theta - 1) + h \sin^2 \theta \cos 2\psi \} \]

the 2nd order solutions by the case the nuclear spin is one,

\[
E_{20} = 0
\]

\[
E_{2 \pm 1} = \pm \frac{1}{2\lambda_L} \left( \frac{\lambda_L}{12} \right)^2 \left\{ 9(\sin^2 \theta)(1 + 3\cos^2 \theta) + 6h \cos 2\psi \sin \theta \right. \\
+ h^2 (4 - 3 \cos^2 2\psi \sin^2 \theta) \left\} \right.
\]

Transition frequencies \( \omega_L \pm \omega_\pm \) are also defined as in chapter 4, a). Then \( \omega_\pm \) are written by

\[
\omega_\pm = \frac{1}{4} \lambda_L \left\{ 3 \cos^2 \theta - 1 + h \sin^2 \theta \cos 2\psi \right. \\
+ \left. \frac{1}{2\lambda_L} \left( \frac{\lambda_L}{12} \right)^2 \{ 9 \sin^2 \theta (1 + 3 \cos^2 \theta) \right. \\
+ 6h \sin^2 \theta \cos 2\psi \right. \\
+ h^2 (4 - 3 \sin^2 \theta \cos^2 2\psi) \left. \right\}
\]

The order of the contribution of the second order solutions in equation 4-12 is estimated. The contributions were

\[
\frac{\text{2nd order}}{\text{1st order}} = \left\{ \frac{9(\lambda_L/12)^2}{(3/4)(\lambda_L)} \right\} = \frac{1}{4} \frac{\lambda_L^2}{\lambda_L} = \left\{ \begin{array}{ll}
0.2 \% & \text{for } ^{12}\text{B} \\
2.3 \% & \text{for } ^{13}\text{B}
\end{array} \right.
\]

where the quadrupole coupling constants in the present experiment were \( \sim 0.2 \text{ MHz} \), \( \sim 1 \text{ MHz} \) respectively for \(^{12}\text{B}\) and \(^{13}\text{B}\) in metal sample, and \( \omega_L \equiv 6.753 \text{ MHz} \left(^{12}\text{B}\right) \), and \( 3.079 \text{ MHz} \left(^{13}\text{B}\right) \) respectively.

c) NMR Spectra

i) Single crystal pattern (I = 1)

when the nuclei of which spins are one sit at the equivalent sites in a single crystal, two transition frequencies are possible as shown in equation 4-7. The splitting of the two lines depend on the direction cosine of the electric field gradient against the magnetic field direction as shown in Fig. 4-4.

The intensity distribution in the NMR spectrum are considered as follows. The
polarization is expressed by the populations in the magnetic sublevels. When there is no quadrupole interaction, the polarization can be completely destructed by an unique resonance condition and the maximum intensity is proportional to the initial polarization \( P = P_1 - P_4 \). If there exists quadrupole interaction, the levels are only partially equalized by an unique resonance condition, and the respective intensities are proportional to \( \frac{1}{2}(P_1 - P_5) \) and \( \frac{1}{2}(P_6 - P_4) \) corresponding to each transition. A simple sum of these independent intensities becomes only one half of the initial polarization \( P = P_1 - P_4 \). This is essentially different from the case when the two transitions are induced simultaneously and the initial polarization is completely destructible.

ii) Polycrystal pattern
A) The random orientation of the field gradient in a polycrystalline sample gives rise to a continuous distribution of the transition frequencies. We assume in this section that \( \lambda QP \) is unique and field gradient are uniaxially symmetric. For the sake of simplicity, the sign of \( \lambda QP \) is assumed to be positive. The intensity of a line shape is proportional to the number of the nuclei of which transition frequencies belong to a unit frequency range. By using equation 4-7 the line shape function \( f_4(\lambda \lambda) \) is reduced for \( \lambda \lambda \):

\[
f_4(\lambda \lambda) \propto \frac{d(\cos \theta)}{d\lambda \lambda} \propto \left| \sqrt{\lambda \lambda} \right|
\]

As for \( \lambda \lambda \), the shape function is a mirror image of \( \lambda \lambda \) to the Larmor frequency \( \lambda L \). Thus the intensity \( \frac{1}{2}(P_1 - P_5) \) or \( \frac{1}{2}(P_6 - P_4) \) spread over the range of \( \frac{1}{2} \lambda \lambda \). By superposing the two line shape functions, an NMR spectrum of quadrupole interaction can be obtained. The over all width is \( \lambda \lambda \). If \( \lambda \lambda \) is a few hundred kHz, the polarization destruction by an rf field at unique condition can be estimated by considering the calculated dipolar broadening (\( \sim \) a few kHz) to be only a fraction
of a %. Thus the effect is very difficult to detect by a usual rf field. The rf field must be modulated in frequency to integrate the NMR effects within the modulation range to make the effects observable. The calculated spectra calculated by assuming the frequency modulated rf field are shown in Fig. 4-5 in which their modulation dependences are clearly seen. The asymmetry of a spectrum is the result of the asymmetric distribution of the level population which is simulated to the $^{12}$B case. The center peak shown in the figure is the effect of the rf modulation. As seen in Fig. 4-2, $\mathcal{L}_+$ and $\mathcal{L}_-$ of a nucleus are nearly equal to zero when $\theta \sim \omega^* \frac{1}{\Omega^*}$. If the range of the frequency modulation cover the frequency $\mathcal{L}_L$, the two transitions may be induced at the same time, and the polarization of the nuclei may be destructed completely.

The line shape shown in Fig. 4-5 can be expressed by using the line shape function 4-13 in which the dipolar broadening is neglected. The intensity of the rf magnetic field is assumed to be sufficiently strong, which means that the equalization of the populations related to the transition is complete. The polarization change is given by

$$
\Delta P = \int_{-\infty}^{\infty} \left[ \frac{1}{2} (\mathcal{L}_+ - \mathcal{L}_-) f_0(u) + \frac{1}{2} (\mathcal{L}_+ + \mathcal{L}_-) f_0(u') \right] du + \int_{-\infty}^{\infty} \left[ \frac{1}{2} (\mathcal{L}_+ + \mathcal{L}_-) f_0(u') \right] du',
$$

where $\Delta P$ is the polarization changes by the case the center frequency of the modulation is $\mathcal{L}_L$, the modulation is $\Delta \mathcal{L}$,

1) $\mathcal{L} \leq \mathcal{L}_L - \frac{1}{2} \Delta \mathcal{L}$

$$
A = \mathcal{L} - \frac{1}{2} \Delta \mathcal{L}, \quad B = \mathcal{L} + \frac{1}{2} \Delta \mathcal{L}, \quad C = D = 0
$$

2) $\mathcal{L}_L > \mathcal{L} > \mathcal{L}_L - \frac{1}{2} \Delta \mathcal{L}$

$$
A = \mathcal{L} - \frac{1}{2} \Delta \mathcal{L}, \quad B = 2 \mathcal{L}_L - \mathcal{L} - \frac{1}{2} \Delta \mathcal{L}, \quad B = C, \quad D = \mathcal{L} + \frac{1}{2} \Delta \mathcal{L}
$$

3) $\mathcal{L}_L + \frac{1}{2} \Delta \mathcal{L} > \mathcal{L} > \mathcal{L}_L$

$$
A = 2 \mathcal{L}_L - \mathcal{L} + \frac{1}{2} \Delta \mathcal{L}, \quad B = \mathcal{L} + \frac{1}{2} \Delta \mathcal{L}, \quad C = \mathcal{L} - \frac{1}{2} \Delta \mathcal{L}, \quad D = 2 \mathcal{L}_L - \mathcal{L} + \frac{1}{2} \Delta \mathcal{L}.
$$
Fig. 4-5 Poly crystal pattern

The first order solutions were used. The dipolar broadening was neglected. The ratio of the related population differences were assumed to be 2.9 which was simulated to the $^{12}$B in Ta case. The asymmetric heights in the spectra are the results of the asymmetric populations. The center peak is the result of the frequency modulation.
B) $\eta \neq 0$, dipolar width $\neq 0$.

The transition frequencies of more general cases are given in equation 4-12. It is complicated to solve the line shape functions analytically by taking the 1st and the 2nd order solutions and the asymmetric electric field gradient into consideration. The modulation in frequency and the dipolar broadening make the case more difficult. Still the spectra can be calculated numerically. In order to make the calculation easier, the rate of the polarization destruction of the nuclei is expressed by simple equations by assuming averaged transition probabilities.

The probabilities of the two transitions are generally different according to the frequency differences from the applied rf frequency. As the rf frequency is modulated, they are also time dependent due to the periodical sweep of the frequency. The repetition of the modulation is comparably fast compared with the nuclear lifetime, and the time averaged probabilities may be used. These quantities can get experimentally in the modulation-check run, in which $^{12}$B nuclei are implanted in a platinum stopper.

The polarization change is estimated by summing up all the changes of the nuclei whose transition frequencies which correspond to an electric field gradient of an orientation defined by $\theta$ and $\psi$, belong to the modulation range.

$$\Delta P(\omega) = \int_{0}^{2\pi} \int_{0}^{\pi} \left\{ 1 - T(k_+, k_-) \right\} P \frac{\sin \theta}{4\pi} d\theta d\psi$$  \hspace{1cm} 4-15

where $k_+$ and $k_-$ are the averaged transition probabilities for the two transitions.
\( \lambda_{\parallel} \) and \( \lambda_{\perp} \) of a nucleus respectively, which are the function of the differences between the transition frequencies and the center frequency of the modulation, and the \( \{ 1 - T(k_+, k_-) \} \) is the rate of the polarization destruction of the nucleus. If the averaged probabilities are used, the time dependences of the level populations of the nuclei are expressed by the following simple equations,

\[
\begin{align*}
\frac{dP_i}{dt} &= P_i^0 - \frac{1}{2} k_+ (P_j - P_i) - \lambda P_i \\
\frac{dP_j}{dt} &= P_j^0 + \frac{1}{2} k_+ (P_i - P_j) - \frac{1}{2} k_- (P_0 - P_j) - \lambda P_j \\
\frac{dP_0}{dt} &= P_0^0 + \frac{1}{2} k_- (P_0 - P_1) - \lambda P_0
\end{align*}
\]

where \( P_1, P_0 \) and \( P_{-1} \) are the populations in the magnetic sublevels which are not normalized, the \( P_i^0, P_j^0 \) and \( P_0^0 \) are the production rate of each magnetic sublevel, the \( \lambda \) is the reciprocal of the nuclear life time and \( k_\perp \) include the effect of rf transitions. The spin lattice relaxation time is assumed to be fairly long compared with the nuclear life time. The result of the equation is shown in Appendix C.

If there is no quadrupole interaction, \( k_+ \) and \( k_- \) take a same value, and equation 4-16 gives equation 3-11. Then the analysis of a sharp resonance obtained by applying modulated rf field may give the transition probabilities as a function of the distance between the transition frequency under the consideration, and the applied rf frequency.

In the actual experiment, the rf field was selected so that the populations related to the transition may be completely equalized, and the condition \( \{ 1 - T(k_+, k_-) \} \approx 1 \) is obtained. Because of the dipolar broadening, the polarization destructions at the outside the modulation range are also induced. In this case the effects should be computed by using the equations written above. As the dipolar broadening, however, is very narrow compared with the modulation range, the following approximation can be made in the numerical calculation. Instead of obtaining the transition probabilities, experimental spectra of polarization destruction observed in the experiment of the sharp resonance such as \( ^{12} \text{B} \) in Pt case are used in the approxima-
tion. Thus \( \{1 - T(k_x, k_y)\} \) is reduced from the experimental spectra on \(^{12}\text{B}\) in Pt.

A rate of the population equalization among the related two magnetic sublevels depend on the experimental conditions; the rf intensity, the modulation width and the dipolar width. The resultant line shape of the NMR spectra on fcc metals were well assigned to be Lorenzian-type from the analyses of the experimental data as shown in Chapter 7 a), Table 7-1. Then the extent of the NMR intensity to be observed at \( \mathcal{J} \) is proportional to

\[
f(\mathcal{J}/-\mathcal{J}^*) = \sqrt{\frac{4}{\pi}} \delta^2 + (\mathcal{J}'-\mathcal{J}^*)^2 \]

where an rf is applied at \( \mathcal{J} \) and the \( \delta \) is the half width at half maximum (HWHM) of the experimental line broadening which was comparable to the calculated dipolar broadening. When the rf was modulated in frequency an rf of sufficient intensity which can induce a complete polarization destruction within the modulation range is experimentally selected, and the NMR intensity to be observed is proportional to

\[
\int_{\mathcal{J}-\frac{1}{2}\Delta \mathcal{J}}^{\mathcal{J}+\frac{1}{2}\Delta \mathcal{J}} f(\mathcal{J}/-\mathcal{J}^*) d\mathcal{J}^* \]

where the range of the modulation is \( \mathcal{J} - \frac{1}{2}\Delta \mathcal{J} \sim \mathcal{J} + \frac{1}{2}\Delta \mathcal{J} \). This can be rearranged by taking the experimental conditions, and the maximum value of the polarization into consideration.

\[
T'(\mathcal{J}, \mathcal{J}') = \begin{cases} \tan^{-1}(\mathcal{J}/\mathcal{J} + \frac{1}{2}\Delta \mathcal{J})/\delta - \tan^{-1}(\mathcal{J}/\mathcal{J} - \frac{1}{2}\Delta \mathcal{J})/\delta, & \text{or } |\mathcal{J} - \mathcal{J}'| < \frac{1}{2}\Delta \mathcal{J} \\ 1, & \text{otherwise} \end{cases} \]

4-19

4-20

The related populations whose transition frequencies are contained within the modulation range are assumed to be completely equalized. The \( T'(\mathcal{J}, \mathcal{J}') \) can be directly connected to the differences of the resultant populations of the related two magnetic sublevels. The populations \( \mathcal{J} \) in the levels are \( P_1, P_2, P_4 \), and \( P'_1, P'_0, P'_4 \) corresponding to the before and the after the application of an rf field.

\[
T'(\mathcal{J}, \mathcal{J} + \mathcal{J}_c) = 1 - \frac{(P'_1 - P'_0)}{(P_1 - P_0)} \]

4-21

\[
T'(\mathcal{J}, \mathcal{J} + \mathcal{J}_c) = 1 - \frac{(P'_4 - P'_1)}{(P_4 - P_1)} \]

4-22
where the level populations are normalized to satisfy the following conditions:
\[ P_r + P_u + P_l = P'_r + P'_u + P'_l = 1 \]
The \( \{1 - T(k_+, k_-)\} \) can be approximated to be
\[
{\left\{ T'(\omega, \omega + \omega_1) - T'(\omega, \omega + \omega_2) + \{ T'(\omega, \omega + \omega_1) - T'(\omega, \omega + \omega_2) \right\} \frac{1}{2}} (P_r - P_u)
\]
where \( T'(\omega, \omega + \omega_1, \omega_2) = T'(\omega, \omega + \omega_1) - T'(\omega, \omega + \omega_2) \), \( \omega_1 \) are calculated by equation 4-12. The relation is correct when the only one transition frequency of a nucleus is caused by a unique resonance condition, and when the two transitions of a nucleus are caused by the strong rf field with which the population related to the transitions is completely destructed. By the case when the two transition frequencies are under the influence of the rf field and the populations of the magnetic sublevels related to the transitions are not completely equalized, the relation 4-23 is an approximation. Then the range where the equation is approximation is just the modulation width around the Larmor frequency, \( \omega_L \).

An rf with a sufficient intensity and a wider width of frequency modulation compared with the dipolar width, reduces the difference between the real value and the calculated. For instance, by the case of \(^{12}\)B implanted in tantalum the difference is numerically estimated to be smaller than 7% of the asymmetry at the center peak, by the case the range of the modulation was 13 kHz. The absolute value of the difference is estimated to be smaller than 0.3%, since the asymmetry change at the peak is at most 4%. This is smaller than the statistical error in the present experiment, which is \( \approx 0.5\% \). The relative differences reduce, as the modulation range is widened as described in Appendix D. Typical quadrupole spectra of general cases are shown in Fig. 4-6. These are numerically computed by assuming the quadrupole width, the asymmetry of the electric field gradient, the populations of the magnetic sublevels, and the dipolar broadening.
Fig. 4-6 Poly crystal pattern ($\eta \neq 0$)

The spectra were calculated by assuming $\Delta \mu /\mu_b = 6.5\%$, dipolar broadening = 0.8% of the coupling constant, and the ratio of the population differences to be 2.9.
Fig. 4-7 The 1st order solutions with symmetric field gradient were used to obtain these spectra. The ratio of the population differences were assumed to be 2.9.
Fig. 4-8 Poly-crystal pattern

Finite asymmetry of the field gradient, and the distribution of the coupling constants were assumed.
C) Factors which affect the actual quadrupole spectra

There are several causes which deform the polycrystal spectra observed by use of uniaxially symmetric field gradient. One of the causes as already shown in the previous section is asymmetry of the field gradient caused by the crystalline defects; internal strains, dislocations, lattice defects and impurities. Another is some kinds of distribution of field-gradient values which must be closely related to the asymmetry of the field gradient. There is one another factor which deforms the spectra; the anisotropic Knight shift. (See cf. p. 16 and 41) in this section)

i) Continuous distribution of the field gradient

As an extreme case uniaxially symmetric field gradients are considered. The distribution of the coupling constants is assumed to be square type with half width \( \Delta u_{\text{g}} \). The resultant spectra are obtained by summing up each spectrum which correspond to each coupling constant. The typical spectra calculated by the present method are shown in Fig. 4-7 in which we can not recognize prominent differences from the spectra in Fig. 4-6. To simulate an actual case typical spectra are calculated in which the two causes, i.e. the asymmetry of the field gradient and the continuous distribution of the coupling constant are included. They are shown in Fig. 4-8.

ii) Anisotropic Knight shift

According to N. Bloembergen the anisotropic part of the Knight shift is written as,

\[
\frac{\Delta U_{\text{anis}}}{U} = -\frac{\Delta H_{\text{aniso}}}{H_0} = \beta^2 V_0 N(E_F) \frac{\gamma(3 \cos^2 \theta - 1)}{4} \tag{4-25}
\]

where \( \beta \) is the Bohr magneton \( N(E_F) \) is the density of states at Fermi surface per unit volume and per unit energy interval, \( V_0 \) is one atomic volume, \( \gamma \) is determined
by the radial wave function, and $\theta$ is the angle between the magnetic field and the crystallographic axis. The angle dependence of the anisotropy is very similar to that of quadrupole splitting, and the quadrupole splitting $\alpha$ are expanded or reduced according to the sign of the anisotropy. In the spectra on $^{12}$B in Ta and Mo, clear effects were observed in which the frequency difference of the left peak to the center peak ($\alpha \lambda$) is systematically large compared with that of the right peak.
5 Recoil Implantation

a) Possible Sites of the Recoil Nuclei

The recoil nuclei ejected from the target are implanted in the various host media by using their recoil energies. The depth of the implanted nuclei in the host medium can be estimated by the range-energy relation of a charged particle of low energy. The range spectrum of the implanted particles have a certain width corresponding to the energy spread due to the target thickness and the spread of the recoil angle. For instance the recoil nuclei $^{12}$B which come out from a thin target of which effective width for the recoil nuclei is $\sim 200 \mu g/cm^2$ and captured in the recoil stopper are evenly distributed in the host medium, and the spread of the range is almost equal to the target thickness. (See Fig. 5-1 and Appendix E.)

When there is a large mass difference between the recoil and the host elements, the probable sites of the recoil elements are interstitial. On the other hand if the mass weights are comparable, the probable sites are substitutional.

1) fcc metal

The probable sites of $^{12}$B and $^{12}$N are interstitial.

They sit at the center of an octahedral (A) or a tetrahedral (B) surrounding of the nearest neighbours as shown in Fig. 5-2. Thus the sites are free from an electric field gradient because the symmetry of the nearest neighbours are at least 3 fold symmetric about independent 3 axes. As the NMR width to be observed is mainly due to nuclear dipolar and instrumental width, the careful observation of the width and NMR intensity certify the discussions on the recoil implantation and the causes of the line broadening. By analyzing the spectra on fcc metals, the necessary treatments and conditions are obtained for bcc metals that is to be used in quadrupole interaction experiment. In the experiment, it was found that the recoil sat at the electric field gradient free sites.
Fig. 5-1 The distribution in the sample was calculated by use of the range energy relation of the low energy $^{12}$B particle, by assuming a thin target and a sharp recoil angle.

Erecoil = 0.2 MeV
Target ~ 200 $\mu$g/cm$^2$
Host: Copper
Skindepth ~ 22 mg/cm$^2$
Fig. 5-2 Probable sites of the light elements in the fcc crystal of heavy element are shown by the larger spheres. The sizes of the elements are arbitrary. The implanted elements sit in the center of the octahedral (A) and the tetrahedral (B) surroundings of the nearest neighbours.
Fig. 5-3 The probable sites of the light elements in the bcc crystal of heavy elements are shown by the larger spheres. The sizes are arbitrary. The implanted particle sit in the face center of a unit cell (C), and in the center of the two perpendicular arrays formed by nearest two host elements.
ii) bcc metal

The probable sites of light elements like $^{12}\text{B}$ in the heavy elements like Ta are also interstitial. According to the consideration on the symmetry of the host elements, two probable sites are expected as shown Fig. 5-3. (See Appendix F.) Electric field gradients are expected there due to the uniaxially symmetric distribution of the neighbouring elements. The observation on quadrupole interaction in these bcc metals in the present study certified the expected sites to be true.

iii) TiB$_2$ and ZrB$_2$ cases

The atomic mass of $^{12}\text{B}$ is so close to that of initial B in the sample that the probability of substitution of $^{12}\text{B}$ in the sample may be considered very high in the collision processes. Then the probable sites of $^{12}\text{B}$ is the substitutional in the sample. This process was successfully used by K. Sugimoto et. al. in the experiment of F$^{19}$ and F$^{17}$ $^{19}$. In the present experiment, the ratio of the field gradient acted in these samples agreed to the known ratio of $^{11}\text{B}$. This supports that the majority of $^{12}\text{B}$ sat at the substitutional.

The electric quadrupole interaction of $^{11}\text{B}$ in the samples have been measured by the usual NMR experiment by A. H. Silver. As the electronic state of $^{11}\text{B}$ and $^{12}\text{B}$ is identical, the same electric field gradient will act on them. Thus a measurement of the strength of the quadrupole interaction gives the absolute ratio of quadrupole moments of $^{12}\text{B}$ to $^{11}\text{B}$. A knowledge of the quadrupole moment of $^{11}\text{B}$ determines the quadrupole moment of $^{12}\text{B}$.

According to the crystallographic study about the boron in the sample, it sits at the center of the prism formed by two regular triangle which is composed of three metal ions and which is parallel with each other. As the surroundings of the sites are uniaxially symmetric, the electric field gradient expected there is uniaxially symmetric.
b) Electric Field Gradient

The electric-field gradient at a nucleus is given by

$$\mathbf{q} = 2 \int_{\text{all space}} \mathbf{f}(r) \cdot \frac{1}{3} P_2^0(\cos \theta) d\tau$$

where \( \mathbf{f}(r) \) is the nuclear and electronic charge density at \( r \). According to R. E. Watson, the field gradient in a metal is considered as arising from three separate sources. A sphere about the nuclear site can be drawn. The sphere can be chosen so that spheres touch for near neighboring atomic sites. First gradient contribution is \( \mathbf{q}_{\text{lat}} \) arising from the region outside the sphere, the nuclei and the electrons. Second contribution \( \mathbf{q}_{\text{loc}} \) comes from the conduction electrons inside the sphere. Finally there is a contribution from the electrons in the closed shells at the atomic site in question. They are distorted under the influence of spherical environment and interact with the nucleus. This interaction is well known as Sternheimer antishielding factors \( \gamma_{\infty} \) and \( R_q \). Then electric field gradient at the nucleus is

$$\mathbf{q} = \mathbf{q}_{\text{lat}} (1 - \gamma_{\infty}) + \mathbf{q}_{\text{loc}} (1 - R_q).$$

The \( \mathbf{q}_{\text{loc}} \) arises from the free electrons in the distorted electric potential within the sphere due to the \( \mathbf{q}_{\text{lat}} \) and is strongly related and proportional to \( \mathbf{r} \). Thus the electric field gradient of a site in the crystal is determined by the distribution of the surrounding ions at the lattice.

Naturally the nuclei of an element who sit at the equivalent interstitial of or substitutional sites of a crystal receive an identical electric field gradient. There are several sources that destroy the uniformity of the field. They are dislocations, local strains and the existence of impurities in the crystal.

In the actual experiment the dislocations and the local strains can be removed by the annealing.

The purity of the sample has to be high. The availability of the treatment was certified in the NMR intensity and width of \(^{12}\)N and \(^{12}\)B in a fcc metal. In this case it had narrow line width. The observed line
widths after the annealing of the sample were comparable to that of the calculated dipolar width \textsuperscript{4}). The purity of 99.9 \% of the host medium was sufficient for the purpose of this experiment. It is difficult to calculate the quantitative electric field gradient, in the case \textsuperscript{12}B and \textsuperscript{12}N are implanted in the interstitial of a crystal, because of the ambiguity of the atomic state and of the charge states of surrounding ions. The existence and the symmetry of the electric field gradient can be seen by the distribution of the surrounding ions, since each source of the field gradient strongly depends on their asymmetric distribution.

\textbf{Fig. 5-4}
Experimental Apparatus and Procedure

a) Production of Polarized $^{12}$B and $^{12}$N Nuclei

$^{12}$B and $^{12}$N were produced through the nuclear reactions $^{11}$B(d, p)$^{12}$B and $^{10}$B($^3$He, n)$^{12}$N respectively. The incident particles of deuterons and helium were accelerated by the Osaka University 4MV Van de Graaff, and their energies were 1.43 MeV and 3.8 MeV respectively. The incident particles were analyzed by a magnetic particle analyzer. The beam of particles was chopped to a pulsed beam of 20 ms width and every 60 ms interval by an electric beam pulser to separate the time of the nuclear production and the $\beta$-particle countings. The focused condition of the beam at the target was adjusted by a quadrupole magnet. An electrostatic deflector was employed to correct the beam path wandered slightly by a static strong magnetic field applied at the target position. (see Fig. 6-1)

The target and the target holder were developed to fit for the use of the high beam intensity to increase the yield of nuclear production and higher incident beam energy. A thick tantalum or copper backings were used. The target side of the backings were exposed to the incident beam and the recoil nuclei were ejected from the same surface. Metallic boron was evaporated on the backings by the electron bombardment technique. Natural boron was used for (d, p) reaction and enriched $^{10}$B isotope was used for ($^3$He, n) reaction. The thickness of the target was about 40 $\mu$g/cm$^2$. It was set in the chamber so that the angle between the surface of the target and the incident beam was 10 degree$^\circ$. The collimator for recoil nuclei was set at 30 degrees from the incident beam and its angular width was confined to 10 degrees. The relation between the recoil angle and the recoil energy are shown in Fig. 6-4, 6-5.

The $\beta$ particles were detected by two coincidence couples of Si (Li) detectors (Fig. 6-6) which were set at the upper and at the lower sides of the recoil stopper. The sensitive layers of the solid state detectors were $\sim$1.5 mm X 2 cm$^2$. Only the $\beta$ particles which pass through a pair of detectors were counted. The energy and
time spectrum of the $\beta$ particles observed by the detectors are shown in Fig. 6-7, 6-8, and 6-9. The observed time spectra agreed with the results so far measured; $T_1 = 20 \text{ ms for } ^{12}\text{B}$, and $T_2 = 11 \text{ ms for } ^{12}\text{N}$ respectively. The block diagram of the detection system used in the experiment are shown in Fig. 6-18 and described in the next section. The energy of the incident particle was decided so that the quantity of $(\beta \text{ ray yield}) \times (\text{observed polarization})^2$ may become maximum. Their trends are shown in Fig. 6-10, and 6-11.

The ejected nuclei were implanted in suitable implantation media (recoil stopper). The recoil stopper was attached to a frame of copper line of 1 mm in diameter and was fixed to a holder as shown in Fig. 6-12, and 6-13. The treatments of the stoppers are described in Chapter 7.

b) Data Accumulation and Its Control System

In the course of the experiment the following technical improvements were made. The expected change of $\beta$-decay asymmetry by an rf magnetic field in existence of the quadrupole interaction is only a small fraction of the initial asymmetry as explained in Chapter 4-c). The one way to increase the NMR effect is to use a frequency modulated rf field of a suitable range to integrate the effects within the range. Still one must prepare to detect a few percent effect in the counting rate in order to observe the clear quadrupole pattern. To avoid the inevitable long term instability of the detecting system, we employed the system as shown in Fig. 6-18.

An rf oscillator described in Appendix I was employed. To get an rf modulated in frequency, a varicap (variable capacitance) was attached parallel to the capacitance of LC-type-tank circuit in an rf oscillator, and a suitably attenuated electric saw tooth wave of 500 or 1000 Hz in repetition was applied. The modulation range was able to be selected by changing the amplitude of the wave. The
modulation range was variable from a few kHz to 300 kHz. A typical time spectrum of a frequency modulated rf observed at rf coil in the chamber is shown if Fig. 6-14A. Its linearity was detected roughly by "Carrier deviation meter" and observed by a synchroscope as shown in Fig. 6-14A. The non linearity of the modulation was negligibly small by the case the modulation range is smaller than 50 kHz. The precise measurement of the range was done in the experiment on fcc metals by use of the frequency modulated rf field. (Fig. 6-15,6-16, 6-17). In those cases the observed width of the spectrum was mainly caused by the modulation of the rf field. Taking the dipolar width into account, calculated curves were fitted to the data. The curve has to be predicted by equation 4-20. Thus the modulation ranges were obtained.

An automatic data normalization system (Fig. 6-18) was employed to reject the data fluctuations due to inevitable long term instability of $\beta$ counting system and signal amplifying system. The method essentially consists of the automatic normalization of the data. A counting rate ratio calculated by the data obtained by applying an rf field, was normalized by that obtained without the rf field. The rf field was excited on the rf coil periodically. The signals from the two $\beta$ ray detecting systems were distributed into two pairs of scalars corresponding to the rf on and off periods. A typical repetition period was $\sim 1$ sec. From the $\beta$ -ray counting rates, the ratios $R_{on}$ and $R_{off}$ were calculated and a normalized ratio $R_{on} / R_{off}$ was obtained (see equation 3-8). Likewise the time of the production of nuclei and the time of $\beta$ -ray detection were separated as already described in the previous section. For 20 ms the target was bombarded by the incident beam of particles and the following 40 ms was used for $\beta$ -ray detection alternately and repeatedly. This system was supervised by a programmed timer shown in the system-block diagram. The four scalars were connected to a computer by way of a scalar control. When the scalars were stopped, their contents were perforated out in a
paper tape. The flow chart of this process was shown in Appendix H.

c) Data Analysis

The quadrupole spectra were analyzed by using the 1st and the 2nd order perturbation theory of the quadrupole interaction in a high magnetic field with conditions that the sites where the recoil nuclei were captured were all crystallographically equivalent and that the crystalline internal electric field gradient was time independent and was able to be described by the classical formulations as described in Chapter 4.

A quadrupole spectrum was calculated by using the equation 4-15, by assuming the quadrupole width \( \Delta q \), the finite asymmetry or/and distribution in magnitude of the electric field gradient \( q \) or \( \Delta q \), the populations in the magnetic sublevels, and the dipolar broadening. The theoretical curves were compared with the experimental data. The goodness of fit was evaluated by the following equation,

\[
\chi^2 = \sum_i \frac{(R_i^{exp} - R_i^{calc})^2}{\sigma_i^2}
\]

\[
\Delta \chi^2 = 2 \sqrt{\chi^2}
\]

where \( R_i^{exp} \) and \( \sigma_i^{exp} \) were the observed asymmetry change and its uncertainty due to the counting statistics respectively, \( R_i^{calc} \) was the calculated asymmetry change, \( \Delta (\chi^2) \) was the uncertainty of \( \chi^2 \) due to the statistical uncertainties of \( R_i^{exp} \). The parameters were searched so as to minimize the \( \chi^2 \). A typical \( \chi^2 \)-parameter curve is shown in Fig. 6-19.

In an course of an experiment the conditions about an rf magnetic field was fixed; the center frequency of a modulated rf field, the rf amplitude and the range of the modulation. The quadrupole spectra were obtained by scanning the magnetic field. For the sake of convenience, however, in the data fitting processes, the spectra were converted by the following procedure into the spectra as to the fre-
frequency of the rf field. The intensity of the applied homogeneous magnetic field is given in the proton-resonance frequency. The resonance frequency due to the external magnetic field \( f_p \) (in proton resonance frequency) of \( \text{^{12}B} \) (or \( \text{^{12}N} \)) is

\[
f = \left( -\frac{\mu_s}{2\mu_p} \right) f_p
\]

where \( \mu_s \) and \( \mu_p \) are the magnetic moments of \( \text{^{12}B} \) (or \( \text{^{12}N} \)) and of a proton respectively. The quadrupole width and the dipolar width are independent of the magnetic field intensity. However, even when the field dependent term such as Knight shift exists, it must be comparable order of Knight shift which may be smaller than a thousandth about \( \text{^{12}B} \) and \( \text{^{12}N} \). The magnetic field was scanned only about 10%. Thus the dependence is very small compared with the width due to quadrupole interaction in the present experiment, and the field intensity dependence can be neglected. Then the application of homogeneous magnetic field of \( f_p \) (in proton resonance frequency) corresponds to the application of an rf field of \( (2f_p - \frac{\mu_s}{2\mu_p} f_p) \) in a magnetic field of \( f_p = \frac{2\mu_s}{\mu_p} f_o \), where \( f_o \) is the proton resonance frequency of the employed magnetic field in the experiment.
Fig. 6-1 Beam Transport System.
Fig. 6.2 Target Holder

Incident beam

(Ta or Cu)

Back ing

Target

Recoil Nuclei

Recoil Collimator
Fig. 6-3A Target chamber
Fig. 6-35 Target Chamber

Incident Beam

Pb Block (Shield)

Recoil Stopper
Window for \( \beta \) Particle

rf Coil

Target Holder

Stopper Holder Guide

Recoil Stopper
rf Coil

Recoil Collimator

Window for \( \beta \) Particle
Fig. 6-4

\[ ^{11}\text{B}(d,p)^{12}\text{B} \]

\[ E_d = 1.43 \text{ MeV} \]
Fig. 6-5

$^{10}\text{B} \left( ^3\text{He}, n \right) ^{12}\text{N}$

$E( ^3\text{He}) = 3.8 \text{ MeV}$
Fig. 6-6 Couples of Si(Li) detectors.
Fig. 6-7  Pulse-height distribution of the Si(Li) detector signals in the off-beam periods by the $^{11}\text{B}(d,p)^{12}\text{B}$ reaction.
Fig. 6-8 Typical time spectrum of the β counting by the $^{11}\text{B}(d,p)^{12}\text{B}$ reaction. The normal counting time scheme for the asymmetry measurements are shown by hatched areas.
Fig. 6-9 Typical time spectrum of the $\beta$ counting by the $^{10}_B(\text{He},n)^{12}_N$ reaction.
Fig. 6-10  Energy spectra of the recoil polarization and the $\beta$ particle yield by the case of $^{11}\text{B}(d,p)^{12}\text{B}$ reaction. Recoil angle was $\theta = (30 \pm 5)$ degrees.
Fig. 6.12. Energy spectra of the recoil polarization and the particle yield by the case of $^{10}$B($^3$He,$n$)$^7$N reaction. The recoil angle was $(30 \pm 5)$ degrees.

Asymmetry Change in %

E($^3$He) vs. Recoil Angle

- Asymmetry Change
- Relative $\beta$-ray Yield

counts/µA.sec
Fig. 6-12  Recoil stopper and its holder

Fig. 6-13  rf coil and recoil stopper
Fig. 6-14A
Frequency modulation was detected by a carrier deviation meter.

Fig. 6-14B
Time spectrum of the rf amplitude detected at the head of the rf coil.
Fig. 6-15 An NMR spectrum for a sharp resonance obtained by use of the frequency modulated rf field. The observed width of the spectrum was mainly due to the modulation width $\Delta W$, because the natural width of the resonance by the case of $^{12}$B implanted in Pt was as narrow as $\approx 2.4$ kHz (FWHM).
Fig. 6-16 An NMR spectrum for a sharp resonance by use of the frequency modulated rf field.
Fig. 6-17 An NMR spectrum for a sharp resonance by use of the frequency modulated rf field.
Fig. 6-18 Block diagram of the data accumulation system together with its control. The rf field was periodically excited in the rf coil, and the signals from the \( \beta \) particle detectors were automatically distributed into the related four scalers corresponding to the rf on and off periods. The system was supervised by a programmed timer.
Fig. 6-19
Typical $\chi^2$-parameter curve

$\mathcal{Z}_q = 85.5 \pm 16$ kHz

$^{\text{12}}\text{B in ZrB}_2$
Experimental Result

a) Experiment on fcc Metals

Expectations on the probable sites in the recoil-implantation technique were confirmed on examination of the widths and the line shapes of the NMR spectra observed by using the samples of fcc metals.

The rf intensity dependences of the NMR line width were observed by using Pt foil of \( \sim 50 \mu \text{m} \). The observed typical spectra are shown in Fig. 7-1. The experimental half width at half maximum (HWHM) of each spectrum was obtained by fitting Lorentzian curves to the data. The width was widened as the rf intensity was increased. (See Fig. 7-2). The widths observed at the rf intensity below \( \sim 0.5 \text{ G} \) were almost constant and were \( \sim 1.2 \text{ kHz} \).

The NMR line widths were examined by use of Pt, Cu, Au, and Al metals. Considering the above mentioned experiment on \(^{12}\text{B} \) in Pt, the rf field intensity was selected to be \( H_1 \sim 0.5 \text{ G} \). Among the samples Cu, and Au metals must be annealed to observe sharp line widths comparable to the calculated dipolar broadening due to surrounding nuclear magnetic moments of the host elements. They were annealed by heating in high vacuum \( (\sim 10^{-6} \text{ mmHg}) \). The observed NMR spectra are shown in Fig. 7-3 and 7-4. For comparison of the line shapes and widths, they are drawn as the center frequencies coincide with each other. The spectra should be fitted to the curves calculated by Equation 3-11 by assuming the rf intensity and relaxation time \( T_1 \). For the sake of simplicity, however, the pure Lorentzian or Gaussian curves were fitted to the data. The goodness of fit are shown in Table 7-1. Thus the line shape except for the cases of \(^{12}\text{B} \) in Cu and Al are in favour of the Lorentzian type. They fitted well to the data within the experimental errors. This shows that the resultant line shape to be calculated by equation 3-11 can be well approximated by the Lorentzian curve within the experimental errors. The HWHM of the data are shown in Table 7-2. They are all a few kHz and are comparable to the calculated dipolar broadening due to the sur-
rounding nuclear magnetic moments. Asymmetry changes of $\beta$-decay of more than 20% were observed at resonance conditions in all cases by means of the fixed rf conditions (H$\sim$0.5 G for $^{12}$B, and H$\sim$1 G for $^{12}$N respectively). Among the fcc metals, Al metal was the most suitable implantation medium in which $\sim$30% of asymmetry changes were observed. This is mainly due to the high purity of the sample and its self-removal of dislocations at room temperature. Thus almost maximum asymmetry changes may be expected by the fixed rf conditions. By the other metals, the asymmetry changes comparable to the Al cases were also observed. Above results on the line width and the NMR intensity confirmed the expectations that the the recoil stoppers were not damaged by the recoil implantation and by the radiations which accompanied to the nuclear reaction, that the recoil stoppers of 99.9% were sufficient for the present experiment, and that the recoil elements sat at sites where any electric field gradient did not act. Thus the sites must be the center of the tetrahedral (A) or octahedral (B) surrounding of the nearest neighbours as expected in Chapter 5 a) i).

The rf-field-intensity dependences of the asymmetry changes were observed experimentally by examining $^{12}$B in Pt spectra obtained by using both the modulated and non-modulated rf-field. And the suitable conditions of rf-magnetic-field intensity were obtained. (See Fig.7-5)

b) Experiment on bcc Metals

i) $^{12}$B in bcc metals

The NMR spectra of $^{12}$B implanted in various body-centered-cubic (bcc) metals were observed. The metals used were Ta, Nb, Mo, and W. Their purities were all

* Obtained from Kobe Seiko Company.
above 99.9%. The thickness were a few tens \( \mu m \). They were all annealed by heating in high vacuum \(( \sim 10^{-6} \text{ mmHg} ) \). The conditions of the treatments are shown in Table 7-3. After this annealing their surfaces were etched off a few \( \mu m \) in thickness by suitable etchants.**

The nuclear polarization comparable to that observed by use of the fcc metals were preserved in the bcc metals. The asymmetry changes which were reduced from the analyses of the spectra which were observed by means of the wide rf-modulation were consistent to that observed by use of fcc metals.

The NMR spectra were obtained by scanning the magnetic field. The homogeneous magnetic field intensity was about 8.8 kG. The rf intensity and the tuning conditions of the resonance amplifier for the rf field were fixed during a run of experiment. The center frequency of the modulated rf field was 6.753 MHz. Then the skin depth of each metal could be calculated to be about 60 \( \mu m \) which was far deeper than the recoil range. Thus the variation of the rf magnetic field within the recoil range was very small. The rf field intensity was so determined that the asymmetry change may be saturated but not too strong in NMR experiment on \(^{12}\text{B}\) in Pt stopper (Fig. 7-5).

For each sample, two or three independent NMR spectra were obtained by using the rf field with different ranges of frequency modulation. At first rough spectrum was observed by a wide rf modulation. Following experiments, precise line shapes were examined by narrower rf modulations. Then the spectra were independently analyzed by taking the quantities related to the quadrupole interaction: the coupling constant \( \lambda_q \), the asymmetry of the electric field gradient or the distribution of the coupling constant, (which means the finite distribution of the magnitude of the field gradient), the degree of the polarization, and the dipolar broadening. The results are listed in Table 7-4, and the averaged values

** Ta in HF, W in HF+HNO\(_3\), Mo in HNO\(_3\), and Nb in HF.
were tabulated in Table 7-6.

\[ \text{Ta case} \]

The NMR spectra were obtained by the cases the range of the frequency modulation were 33 kHz and 13 kHz. They are shown in Fig. 7-6. The spectra show clear patterns of quadrupole interaction. A small asymmetry of the electric field gradient or 15% of coupling-constant distribution is necessary to obtain the best fitted curve as shown in Fig. 7-7, Fig. 7-16A, and B. The curves were fitted to respective spectra, and the values used to obtain the best fit are shown in Table 7-4A, and B. The values obtained for each sample are averaged and listed in Table 7-6. Finally the results by the extreme assumptions are

\[
\begin{align*}
\mathcal{L}_a & = (202 \pm 10) \text{ kHz} \\
\mathcal{L}_b & = (204 \pm 13) \text{ kHz} \\
\epsilon_a & = 0.12 + 0.13 \\
\epsilon_b & = (30 \pm 30) \text{ kHz} \\
\epsilon & = 0
\end{align*}
\]

In spite of the independent assumptions i.e., \( \mathcal{L} = 0, \epsilon_a = 0 \), \( \mathcal{L} = 0, \epsilon_b = 0 \), and \( \mathcal{L} = 0, \epsilon_a = 0 \), the coupling constants are insensitive to those assumptions.

\[ \text{Nb case} \]

The calculated dipolar broadening (square root of the second moment) is 5.5 kHz. An rf field which had a relatively narrow modulation range was necessary to observe a clear pattern. The spectra obtained by use of \( \mathcal{L} = 75 \text{ kHz}, 13 \text{ kHz}, \) and 0 kHz are shown in Fig. 7-8, Fig. 7-16A, and B. The dependences of the NMR intensities of the spectra to the modulation ranges are clearly observed. The spectra are relatively smooth due to the wide dipolar width compared with the coupling constant, i.e., \( \sim 100 \text{ kHz} \). The field gradient asymmetry \( \eta \), or \( \epsilon \) have large errors to say something significant, but they are certainly a finite values within the errors. (See Table 7-4A, and B)

\[ \text{No case} \]

The spectra observed by use of the rf fields with \( \mathcal{L} = 25.6 \text{ kHz}, 34.0 \text{ kHz} \) and
90 kHz are shown in Fig. 7-9A, 7-16A, and B. The center peak of the spectrum shown in the bottom of the figure is abnormally high compared with the two peaks at the shoulders of the spectrum. One may understand it as the result of high $\eta$ value or wide spread of the coupling constant. This phenomenon is not yet clearly understood. To obtain the best fitted curves in each assumption, i.e., $\eta = 0$ or $\omega_0 = 0$, fairly large values must be used as shown in Table 7-4A, and B. The peaks at the shoulders are rather well reproduced by a fitting with $\eta = 0$ and $\omega_0 = 0$ (See Fig. 7-9B). But in this treatment the center peak is completely out of fit.

The spectra obtained by using an rf field with $\omega_1 = 94.8$ kHz and 48.5 kHz are shown in Fig. 7-10. In this case, relatively large coupling constants are observed as shown in Table 7-4A, and B. In the course of the data fitting relatively large asymmetry and distribution of the coupling constant are necessary to obtain the best fitted curves in each assumption.

Summary of $^{12}$B experiment

In each data analysis, a set of level populations was obtained. The ratio of the two population differences were in agreement within the experimental errors. The values are tabulated in Table 7-4A, and B. They are averaged to be

$$\left\{ \frac{(P_1 - P_2)}{(P_0 - P_1)} \right\}^{\pm 1} = 2.3 \pm 2.9$$

where the sign $\pm$ comes from the situation that the sign of $\omega_1$ could not be determined in the present method, because the two peaks of the spectrum could not be identified to each transition of the related levels. From the $^{12}$B in Pt experiment the observed polarization was $P = \sim +16\%$ where positive sign is defined by

$$\frac{(R_d \times R_p)}{|R_d \times R_p|}$$
where $k_i, k_f$ are the momentum of the incident and the outgoing particles. Using the normalization condition $(P_1 + P_0 + P_{-1} = 1)$ the populations could be derived. Thus the two independent solutions were possible. They were shown in Fig. 7-18.

Systematic deviations are found between the data and theoretical curves in Ta and Mo cases. (See Fig. 7-9 A, Fig. 7-6, and Fig. 7-7). The separation frequencies of the left peak to the center peak are systematically wider than that of the right peak. This discrepancies could be qualitatively well identified to the anisotropic Knight shift of $\sim 5$ kHz. Present spectra result q' in 4-25 (p. 32) to be positive.

i) $^{12}_N$ in bcc metal

The nuclear polarization comparable to that obtained by use of the fcc metals were also preserved in bcc metals. A quadrupole resonance on $^{12}_N$ in Ta and Nb were performed. The quality of Ta and Nb samples were the same as that used for the $^{12}_B$ experiment. The quadrupole frequencies were $\sim 10^5$ Hz. Thus the modulation ranges must be widened at least to $\sim 300$ kHz. Consequently the amplitude of the rf field must be increased. The necessary amplitude for sufficient rf transitions between the related levels could be estimated by using the equation 3.11. In case the range of the modulation was far wider than the dipolar width, $(L_i)$ in 3-11 could be approximated to be

$$ L = L + \frac{f}{\Delta L} \left( \eta \gamma^2 I_i^2 \right) $$

If the modulation range was increased by n times the rf field amplitude must be increased by $\sqrt{n}$ times of the initial intensity so that the same rate of the transitions at every modulation range may occur.

A uniform amplitude for an rf field is favourable to the present experiment throughout the modulation range. The resonance width of the tank circuit of the rf oscillator and resonance amplifier which included the rf coil has to be widened.
Consequently the power dissipation of the tank circuit increased. The widest modulation range which was obtainable in the present system was 256 kHz that was about 8% of the \( \nu_L \). The non uniformity of the rf amplitude is well seen in the NMR spectrum of \(^{12}\text{N}\) in Al by use of the rf (See Fig. 7-13).

By the cases of \(^{12}\text{N}\) in Mo and W, the quadrupole coupling constants seemed too large compared with the modulation widths and any reliable spectrum could not be obtained.

Ta case

The ranges of the frequency modulation were 116 kHz and 254 kHz. The spectra are shown in Fig. 7-14, Fig. 7-17A, and B. The data were also analyzed by using the 1st and 2nd order perturbation theory by neglecting the non uniformity of the rf magnetic field within the modulation range. Finite values of \( \eta \) and/or \( \gamma_q \) must be used in the data analyses. The results are tabulated in Table 7-5 and 7-6.

Nb case

The ranges of the frequency modulation were 95 kHz and 245 kHz. The spectra are shown in Fig. 7-15, 7-17A, and B.

Summary of bcc experiment

The quadrupole coupling constant of \(^{12}\text{N}\) in Nb and Ta are almost 5~10 times larger than that of \(^{12}\text{B}\) in respective metals. As nuclear quadrupole moment of \(^{12}\text{B}\) and \(^{12}\text{N}\) are expected to be comparable (See Chapter 8), the field gradients are expected to be 5~10 times larger.

Two sets of populations in magnetic sublevels for respective nuclei are obtained and are shown in Fig. 7-18 and Fig. 7-19.

The spectra on \(^{12}\text{B}\) and \(^{12}\text{N}\) are summarized in Fig. 7-16, and Fig. 7-17.

The implanted nuclei in bcc metals were exposed to the crystalline internal field gradient without any exception. The existence of the field gradient in bcc metals shows the captured sites of the recoiled \(^{12}\text{B}\) and \(^{12}\text{N}\) to be interstitial.
If the sites were substitutional, any field gradient would not be expected. This fact together with the sharp NMR spectra obtained by use of fcc metals justify the expectations on the implanted sites of recoiled light elements like B and N in the heavy host elements like Ta, to be interstitial.

c) Quadrupole Moment of $^{12}B$

The quadrupole spectra of $^{12}B$ implanted in TiB$_2$ and ZrB$_2$ were observed. The samples used in the experiment were powders of which grains were $5 \sim 30 \mu m$ in diameter. A recoil stopper was made by depositing a sample ($\sim 1.5 mg/cm^2$) on a thin aluminum foil. A small amount of sample was mixed with alcohol, and was deposited on an aluminum foil by means of a centrifugal separator. This stopper was also attached to a frame made of a copper line. The resonance line of $^{12}B$ in aluminum was so narrow as measured in the section a) that if the sample was peeled off from the surface of the aluminum, a sharp high resonance line would be observed. The experimental results are shown in Fig. 7-20 and Fig. 7-21.

The borons in the sample must receive an uniaxially symmetric electric field gradient at each site, because a boron in the sample sits at the center of the prism formed by two parallel regular triangles of the nearest metal ions. To get, however, the best fitted theoretical curve, finite values of $\zeta$ or $\Delta I/A$ must be taken into account, which were mainly due to the crystalline dislocations and the strains. The samples were not annealed and the surfaces were not etched off. The smearing effects by these causes on the spectral shape showed the same tendencies as shown in Chapter 4-c) (See Figs. 4-6, 7, and 8), and unique assignments were not possible by the present experimental accuracies. The results obtained by assuming each cause are tabulated in Table 7-7. In these cases, fairly large values of $\zeta$ and $\Delta I/A$, resulted i.e. $\zeta \sim 0.5$, $\Delta I/A \sim 70\%$ to obtain the best fitted curve, whereas the coupling

* Obtained from Alfa Inorganics, Inc. USA.
constants of the two cases of a sample are in agreement within the experimental errors. The actual situations are considered to have both the two causes. As stated above the tendencies of each cause are very similar so that the following assumption was made to obtain final results; a half value of that obtained in the extreme cases are effective. The results obtained by assuming \( \frac{d\theta}{d\phi} = 35\% \) and \( \eta = 0.3 \) are tabulated in Table 7-8, and are also in agreement to the coupling constants by the extreme cases. Present results on the two causes are qualitatively consistent with the results from X-ray analyses of the samples.

A ratio of the quadrupole width of the both cases is equal to the ratio of the electric field gradient of the both samples. It reduces to

\[ \frac{\zeta}{\zeta'}(^{12}\text{B in ZrB}_2) \bigg/ \frac{\zeta}{\zeta'}(^{12}\text{B in TiB}_2) = 0.32 \pm 0.05. \]

This must agree with the ratio of the case of \(^{11}\text{B}\). The quadrupole interactions of \(^{11}\text{B}\) in the TiB\(_2\) and the ZrB\(_2\) have been studied by A. H. Silver\(^{16}\). The quadrupole coupling constants which were measured by the usual NMR in a high magnetic field were \( e\xi Q = (360 \pm 20)\) kHz for \(^{11}\text{B}\) in TiB\(_2\) and \( e\xi Q = (118 \pm 10)\) kHz for \(^{11}\text{B}\) in ZrB\(_2\).

The ratio is

\[ \frac{e\xi Q(^{11}\text{B in ZrB}_2)}{e\xi Q(^{11}\text{B in TiB}_2)} = 0.328 \pm 0.033. \]

Their consistency within the errors strongly supports the experimental facts on the substitution of the boron in the sample. Then the known coupling constants can be used

The ratio of the quadrupole moments of \(^{12}\text{B}\) to \(^{11}\text{B}\) are obtained by comparing the known coupling constants of \(^{11}\text{B}\)

\[ \left| \frac{Q(^{12}\text{B})}{Q(^{11}\text{B})} \right| = 0.42 \pm 0.04. \]

The quadrupole moment of \(^{11}\text{B}\) has been determined by the atomic beam method\(^{17}\).

The quadrupole moment of \(^{11}\text{B}\) is known by the atomic beam method as

\[ Q(^{11}\text{B}) = +0.0355 \pm 0.0002 \text{ barns}\]

without correction from the configuration-interaction effects and the Sternheimer polarization. The absolute value of the quadrupole moment of \(^{12}\text{B}\) is determined,

\[ \left| Q(^{12}\text{B}) \right| = 0.0151 \pm 0.0014 \text{ barns} \]

- 73 -
The rf field intensity dependences of the NMR spectra.

The solid lines are the best fitted Lorezian curve to the data.
Fig 7.2: The rf intensity dependence of the NMR width.

- \( \Delta U = 0 \)
- \( \mathcal{U}_0 = 6.753 \text{ MHz} \)

- H_{1} \sim 0.5 \text{ G} 
- \sim 1.0 \text{ G} 
- \sim 2.0 

\( \sqrt{\text{(HWHM)}} \)

rf Intensity (Volts)
Fig. 7-3

$^1\text{H}$ in Metal

Cu

Asymmetry Change in %

Au

Pt

Al

$k$ (kHz)
Fig. 7-3. NMR spectra on $^{12}$B in fcc metals.

The spectra were obtained in a high magnetic field ($\sim 8.8$ kG), with the rf field $B_{0.5}$G. The spectra are so arranged that the center frequencies coincide with each other; the Knight shifts are neglected.

Fig. 7-4. NMR spectra on $^{12}$B in fcc metals.

The spectra were obtained in a high magnetic field ($\sim 8.8$ kG), with the rf field $B_{0.5}$G. The spectra are so arranged that the center frequencies coincide with each other; the Knight shifts are neglected.
Fig 7-4

$^{12}\text{N}$ in Metal

Al

Cu

Pt

Asymmetry Change in %

$\nu (\nu_0 = 3.0785\text{ MHz})$

kHz

-8 -6 -4 -2 0 2 4 6 8
<table>
<thead>
<tr>
<th>Host</th>
<th>( \chi^2 )</th>
<th>( \chi^2 )</th>
<th>( \chi^2 )</th>
<th>( \chi^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>6.4</td>
<td>2.1</td>
<td>0.63</td>
<td>1.42</td>
</tr>
<tr>
<td>Au</td>
<td>0.76</td>
<td>0.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pt</td>
<td>0.46</td>
<td>0.6</td>
<td>0.53</td>
<td>0.9</td>
</tr>
<tr>
<td>Al</td>
<td>4.00</td>
<td></td>
<td>0.51</td>
<td>0.8</td>
</tr>
</tbody>
</table>

\( \chi^2_L \); The minimum value of \( \chi^2 \) with Lorenzian curve.

\( \chi^2_G \); The minimum value of \( \chi^2 \) with Gaussian curve.

The data in Fig. 7-3, and 7-4 are analyzed by the Lorenzian and Gaussian curves.
### Table 7-2  Dipolar width in kHz

<table>
<thead>
<tr>
<th>Host</th>
<th>12B (1^+/)</th>
<th>12N (1^+)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>site A</td>
<td>site B</td>
</tr>
<tr>
<td></td>
<td>Δ calc, Δ calc,</td>
<td>Δ calc, Δ calc,</td>
</tr>
<tr>
<td>fcc</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cu</td>
<td>3.2</td>
<td>2.5</td>
</tr>
<tr>
<td>Pt</td>
<td>0.52</td>
<td>0.41</td>
</tr>
<tr>
<td>Au</td>
<td>0.14</td>
<td>0.11</td>
</tr>
<tr>
<td>Al</td>
<td>3.4</td>
<td>2.7</td>
</tr>
</tbody>
</table>

| bcc  | site C     | site D     | site C     | site D     |
|      | Δ calc, Δ calc | Δ calc, Δ calc | Δ calc, Δ calc | Δ calc, Δ calc |
| Nb   | 5.5        | 5.8        | 5.4 ± 0.7  |
| Mo   | 0.51       | 0.54       | 0.45 ± 1.1 |
| Ta   | 1.9        | 2.0        | 0.81 ± 0.8 |
| W    | 6.4x10^-2  | 6.7x10^-2  | 1.0 ± 2.8  | 3.0x10^-2  | 3.1x10^-2 |

* Square root of the second moment.

** The dipolar widths of 12N in fcc metals written in Jour. Phys. Soc. Japan 25 1258 (1968) should be amended by the present Table.

### Table 7-3  Annealing condition

<table>
<thead>
<tr>
<th>Element (thickness)</th>
<th>Temperature</th>
<th>Duration</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nb(100 m)</td>
<td>~1000°C</td>
<td>2 hours</td>
</tr>
<tr>
<td>Mo(50 m)</td>
<td>~2000</td>
<td>0.5</td>
</tr>
<tr>
<td>Ta(50 m)</td>
<td>~1000</td>
<td>0.5</td>
</tr>
<tr>
<td>W (50 m)</td>
<td>~2000</td>
<td>2</td>
</tr>
</tbody>
</table>
Fig 7-5  rf-field intensity dependences of the NMR effects.
The ranges of the frequency modulation were 33 kHz, and 13 kHz respectively. The solid lines are the best fitted theoretical curves to the data by assuming $\gamma \neq 0$, and $A_0 = 0$. The static magnetic field was ~8.8kG.
Fig 7-7 The spectrum showed at the bottom in Fig. 7-6 is expanded. This spectrum is not relatively smeared out, and can be explained by a symmetric field gradient with no distribution.
The ranges of the modulation were 75 kHz, 100 kHz, and 0 kHz respectively. The solid lines are the best fitted theoretical curves to the data by assuming $\hbar \neq 0, \Delta W = 0$. The magnetic field employed in the experiment was 8.8 kG.
Fig. 7-9A The ranges of the frequency modulation were 95 kHz, 34 kHz, and 25.6 kHz respectively. The solid lines are the best fitted curve obtained by assuming $\varphi_{40}, \lambda_0 = 0$. The magnetic field employed in the experiment was $\sim 8.8$ kG.
Fig 7-3B $^{12}$B in Mo

The solid line is calculated by assuming $\xi=0, \delta\xi=0$. The center peak is completely neglected in these conditions.
The ranges of the frequency modulation were 95 kHz, and 48.5 kHz respectively. The solid lines are the best fitted curves calculated by assuming \( B, Q, \Delta \nu = 0 \). The magnetic field employed in the experiment was \( \approx 8.8 \) kG.
Fig 7-13 $^{12}$N in Al

$\nu_L = 3,079 \text{ MHz}$

$H_0 = 37.632 \text{ MHz}$
The ranges of the frequency modulation were 254 kHz, and 116 kHz respectively. The solid lines are the best fitted curves calculated by assuming $\xi_{\parallel 0}^{10} = 0$. The magnetic field employed in the experiment was $\sim 8.8$ kG.
The ranges of the frequency modulation were 254 kHz, and 95.3 kHz respectively. The solid lines were the best fitted curves calculated by assuming $\eta = 0$ and $\zeta = 0$. The magnetic field employed in the experiment was \( \approx 8.8 \text{ kG} \).
### Quadrupole Resonance of $^{12}$B

| Host | Fitting conditions | $(P_1-P_0)/(P_0-P_1)$ | $h$ | $D$ | $|\Delta|_{\text{in kHz}}$ | $\chi^2$/data No | Data No. |
|------|--------------------|------------------------|-----|-----|-----------------|----------------|----------|
| Ta   | 13.0 \{0 0 \neq 0\} | 2.9±3.4 | 0.04±0.14 | 0.25±0.8 | 200 | 0.74 | 26 |
|      | 34.0 \{0 0 \neq 0\} | 6.8±24 | 0.29±0.33 | 0.18±0.8 | 210±28 | 1.1  | 26 |
| Nb   | 0 \{(0 0) \neq 0\} | 2.9±7.6 | 0.5±0.43 | 5.5±0.7 | 100±14 | 0.57 | (2.3) | 21 |
|      | 13.0 \{0 0 \neq 0\} | 1.5±12 | 0.01±0.92 | 5.1±1.7 | 112±18 | 0.49 | 26 |
| No   | 25.6 \{0 0 \neq 0\} | 2.9 | 0.95±0.31 | 0.5±0.16 | 280 | 2.6  | 28 |
|      | 34.0 \{0 0 \neq 0\} | --- | 0.39±0.22 | 0.4±0.15 | 180±30 | 0.84 | --- |
| W    | 48.5 \{0 0 \neq 0\} | 2.4 | 0.60±0.25 | 0.1±0.3 | 324±55 | 1.40 | 17 |
|      | 94.8 \{0 0 \neq 0\} | 5.2±3.1 | 0.56±0.44 | 2.0±0.55 | 370±60 | 0.62 | 21 |

- $D$: Dipolar broadening.
- $\Delta$: Modulation range.

Table 7-4A The best fitted values were obtained by assuming $\Delta \neq 0$, $\Delta \neq 0$. 
Table 7-4B  Quadrupole resonance of $^{12}$B

The best fitted values were obtained by fitting the theoretical curves which are calculated by assuming $\eta = 0$, $\omega_Q \neq 0$, to the data.

<table>
<thead>
<tr>
<th>Host</th>
<th>Fitting condition</th>
<th>$(P_1 - P_0)/(P_0 - P_{-1})$</th>
<th>$\eta$</th>
<th>$\omega_Q$</th>
<th>$\Delta \lambda_Q$</th>
<th>$\chi^2$/data</th>
<th>$A%$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ta (13)</td>
<td>0 0</td>
<td>2.9</td>
<td>$201 \pm 15$</td>
<td>$20 \pm 35$</td>
<td>0.86</td>
<td>36</td>
<td></td>
</tr>
<tr>
<td>(34)</td>
<td>0 0</td>
<td>5.8</td>
<td>$215 \pm 28$</td>
<td>$80 \pm 80$</td>
<td>1.1</td>
<td>32.5</td>
<td></td>
</tr>
<tr>
<td>Nb (13)</td>
<td>0 0</td>
<td>1.5</td>
<td>$116 \pm 17$</td>
<td>$54 \pm 54$</td>
<td>0.4</td>
<td>23</td>
<td></td>
</tr>
<tr>
<td>(58)</td>
<td>0 0</td>
<td>2.5</td>
<td>$102 \pm 15$</td>
<td>$64 \pm 20$</td>
<td>0.85</td>
<td>31.5</td>
<td></td>
</tr>
<tr>
<td>Mo (26)</td>
<td>0 0</td>
<td>4.6</td>
<td>$202 \pm 45$</td>
<td>$210$</td>
<td>1.7</td>
<td>26</td>
<td></td>
</tr>
<tr>
<td>(38)</td>
<td>0 0</td>
<td>1.0</td>
<td>$180 \pm 21$</td>
<td>$80 \pm 80$</td>
<td>0.8</td>
<td>29.3</td>
<td></td>
</tr>
<tr>
<td>W (49)</td>
<td>0 0</td>
<td>3.5</td>
<td>$333 \pm 75$</td>
<td>$226 \pm 113$</td>
<td>1.0</td>
<td>23.0</td>
<td></td>
</tr>
</tbody>
</table>

$\eta$; asymmetry of the field gradient.
$D$; dipolar broadening.
$A$; initial decay asymmetry obtained in the fitting.
$\omega_Q$; distribution of the quadrupole coupling constant.
The least-squared residuals were obtained by assuming \( \Delta h = 0 \).

<table>
<thead>
<tr>
<th>No.</th>
<th>kHz</th>
<th>( \Delta h )</th>
<th>( \eta )</th>
<th>( \eta )</th>
</tr>
</thead>
<tbody>
<tr>
<td>116</td>
<td>0</td>
<td>0 0 0 0</td>
<td>0.5 0.41 0.27 2.3 1.7 4.7</td>
<td>1.7 ± 9 0.77 ± 5.3 ± 0.3</td>
</tr>
<tr>
<td>254</td>
<td>0</td>
<td>0 0 0 0</td>
<td>0.5 0.41 0.27 2.3 1.7 4.7</td>
<td>1.7 ± 9 0.77 ± 5.3 ± 0.3</td>
</tr>
</tbody>
</table>

Table 1: Residuals for fitting conditions (\( \Delta h = \eta \)).
Table 7-5B  Quadrupole resonance of \(^{12}\text{N}\)

The best fitted values were obtained by fitting the theoretical curves which were calculated by assuming \(\eta = 0 \Delta m = 0\) to the data.

<table>
<thead>
<tr>
<th>Host</th>
<th>Fitting condition</th>
<th>(\frac{P_1-P_0}{P_0-P_{-1}})</th>
<th>(\Delta U_Q) in kHz</th>
<th>(\Delta Z_Q) in kHz</th>
<th>(\frac{\gamma^2}{\text{data n.}})</th>
<th>(A%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ta (116)</td>
<td>0 0</td>
<td>0.33</td>
<td>855 ± 170</td>
<td>300 ± 450</td>
<td>0.9</td>
<td>30</td>
</tr>
<tr>
<td>Nb (96)</td>
<td>0 0</td>
<td>0.38</td>
<td>855 ± 160</td>
<td>340 ± 350</td>
<td>0.53</td>
<td>36.4</td>
</tr>
</tbody>
</table>
Quadrupole Resonance of $^{12}\text{B}$ and $^{12}\text{N}$

<table>
<thead>
<tr>
<th>Host (bcc)</th>
<th>$^{12}\text{B}$ ($I^\pi = 1^+$)</th>
<th>$^{12}\text{N}$ ($I^\pi = 1^+$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\mathcal{U}_q$ in MHz</td>
<td>$\eta$</td>
</tr>
<tr>
<td>Nb (V)</td>
<td>$0.105 \pm 0.011$</td>
<td>$0.45 \pm 0.40$</td>
</tr>
<tr>
<td>Mo (VI)</td>
<td>$0.186 \pm 0.026$</td>
<td>$0.64 \pm 0.18$</td>
</tr>
<tr>
<td>Ta (V)</td>
<td>$0.202 \pm 0.010$</td>
<td>$0.12 \pm 0.13$</td>
</tr>
<tr>
<td>W (VI)</td>
<td>$0.340 \pm 0.041$</td>
<td>$0.59 \pm 0.22$</td>
</tr>
</tbody>
</table>

\[\mathcal{U}_q = \frac{3eqQ}{4\hbar} \quad \frac{Q(12\text{B})}{Q(12\text{N})} = \frac{1 + \beta'}{\beta}\]

Fig. 7-6  The data were analyzed by assuming $\mathcal{U}_q = 0$. The data in Table 7.4-A are averaged.
Q-resonance on $^{12}$B($I^r = I^\pi$)

\[ \Delta \nu = 0 \]

Asymmetry Change in %

Fig. 7-16A The spectra on $^{12}$B in bcc metals are summarized. The solid lines are theoretically calculated by assuming $\chi_0 \approx 0$, and fitted to the data.

$\nu (\nu_0 = 6.753 \text{ MHz})$
Fig 7-16B Summary
\( I = 0, \Delta J_Q = 0 \)

\( \Delta J \) \( (J_0 = 6.753 \text{ MHz}) \)

\( \Delta J_Q \neq 0 \)
$Q$-resonance on $^{12}\text{N}(I^\pi = 1^+)$

![Graph showing asymmetry change in Nb and Ta](image)

$\Delta \nu$ (Hz) vs. kHz

- $\nu = 3.079 \text{ MHz}$

Fig. 7-17 A Summary ($\zeta \neq 0, \chi \neq 0$)
Fig 7-17B Summary

$\Delta U_a \neq 0$

$\Delta U = 3.079 \text{ MHz}$
Fig 9-18 Populations in Magnetic Sublevels.

$^{11}\text{B}(d, p)^{12}\text{B}$

$\theta = 30 \pm 5$

$P = 0.16$

$^{10}\text{B}(^3\text{He}, n)^{12}\text{N}$

$\theta = 30 \pm 5$

$P = 0.16$
Fig. 7-20 The magnetic field employed in the experiment was 37.61 MHz in proton resonance frequency. The solid line is the best fitted curve calculated by assuming $\eta \neq 0, \Delta B = 0$, and the broken line is the best fitted curve calculated by assuming $h = 0, \Delta B = 0$. 
Fig. 7-21  The magnetic field employed in the experiment was 37.60 MHz in proton resonance frequency. The solid line is the best fitted theoretical curve calculated by assuming \( \eta \neq 0, \alpha_0 = 0 \).
Fig. 7-22  The solid lines are calculated by assuming
\[ \eta = 0.3, \quad \Delta \nu / \nu_0 = 35\% \]
Fig. 7-23  The solid lines are calculated by assuming \( \eta = 0.3 \), \( \Delta \omega / \omega_a = 35\% \).
### Table 7-7A  Quadrupole resonance of $^{12}$B in TiB$_2$ and ZrB$_2$

<table>
<thead>
<tr>
<th>Host</th>
<th>Fitting condition</th>
<th>$(\Delta l/l_0)$</th>
<th>$\eta$</th>
<th>D (in kHz)</th>
<th>$\lambda$ (in kHz)</th>
<th>$\Delta\eta$</th>
<th>$A_{99}$</th>
<th>$\chi^2$/data n.</th>
</tr>
</thead>
<tbody>
<tr>
<td>TiB$_2$ (15.9)</td>
<td>0 0 0</td>
<td>0.52</td>
<td>192 ± 26</td>
<td>1.55</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(15.9)</td>
<td>0 0 0</td>
<td>0.43 ± 0.74</td>
<td>0.5 ± 0.3</td>
<td>0.3</td>
<td>193 ± 30</td>
<td>35.2</td>
<td>0.87</td>
<td></td>
</tr>
<tr>
<td>(23.4)</td>
<td>0 0 0</td>
<td>1.43</td>
<td>0.65 ± 0.35</td>
<td>1.7</td>
<td>184 ± 34</td>
<td>35.4</td>
<td>1.45</td>
<td></td>
</tr>
<tr>
<td>ZrB$_2$ (7.5)</td>
<td>0 0 0</td>
<td>1.00</td>
<td>0.42 ± 0.33</td>
<td>1.3</td>
<td>85.5 ± 18</td>
<td>29.3</td>
<td>0.56</td>
<td></td>
</tr>
<tr>
<td>(7.5)</td>
<td>0 0 0</td>
<td>0.75</td>
<td>0.59 ± 0.24</td>
<td>2.0</td>
<td>73.4 ± 7.6</td>
<td>25</td>
<td>0.92</td>
<td></td>
</tr>
</tbody>
</table>

D: dipolar broadening obtained in the present data fitting.
$A$: initial $\beta$-decay asymmetry obtained in the present data fitting.
$\Delta\eta$: distribution of the coupling constant.
Table 7-7B Quadrupole resonance of $^12_B$ in TiB$_2$ and ZrB$_2$

<table>
<thead>
<tr>
<th>Host condition</th>
<th>Fitting $\frac{(p_1-p_0)}{(p_0-p_{-1})}$</th>
<th>$\nu$ (kHz)</th>
<th>$\Delta\nu$ (kHz)</th>
<th>$\Delta\nu_B$ (kHz)</th>
<th>A %</th>
<th>$\chi^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>TiB$_2$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(18.9) 0 40</td>
<td>0.31</td>
<td>215±30</td>
<td>142</td>
<td>39.5</td>
<td>0.68</td>
<td></td>
</tr>
<tr>
<td>(23.4) 0 40</td>
<td>0.51</td>
<td>202±30</td>
<td>152</td>
<td>37</td>
<td>1.1</td>
<td></td>
</tr>
<tr>
<td>ZrB$_2$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(8.0) 0 40</td>
<td>0.84</td>
<td>82±8</td>
<td>60</td>
<td>35</td>
<td>0.98</td>
<td></td>
</tr>
<tr>
<td>(7.5) 0 40</td>
<td>0.9</td>
<td>87±13</td>
<td>50</td>
<td>35</td>
<td>0.52</td>
<td></td>
</tr>
<tr>
<td>Sample</td>
<td>rf modulation in kHz</td>
<td>$\chi_q = (3/2h) e \Omega(^{12}\text{B})$</td>
<td>$\eta$</td>
<td>$\Delta \chi_q$</td>
<td>$\text{eq}(^{11}\text{B})/h$</td>
<td></td>
</tr>
<tr>
<td>--------</td>
<td>----------------------</td>
<td>-----------------------------------</td>
<td>-------</td>
<td>-----------------</td>
<td>-----------------</td>
<td></td>
</tr>
<tr>
<td>TiB$_2$</td>
<td>18.9</td>
<td>232 ± 30</td>
<td>0.3</td>
<td>70</td>
<td>360 ± 20</td>
<td></td>
</tr>
<tr>
<td></td>
<td>23.4</td>
<td>230 ± 40</td>
<td>0.3</td>
<td>70</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>averaged</td>
<td>231 ± 24</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>ZrB$_2$</td>
<td>7.5</td>
<td>86 ± 15</td>
<td>0.3</td>
<td>30</td>
<td>118 ± 10</td>
<td></td>
</tr>
<tr>
<td></td>
<td>8.0</td>
<td>70 ± 8</td>
<td>0.3</td>
<td>30</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>averaged</td>
<td>74 ± 7</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Data taken from ref. 16)

** A half value of the best fit obtained by extreme assumptions $\eta = 0$ $\Delta \chi_q = 0$.

*** A half value of the best fit obtained by extreme assumptions $\eta = 0$ $\Delta \chi_q = 0$. 

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Discussion

The quadrupole effects were observed in the NMR spectra on $^{12}$B and $^{12}$N in various bcc metals. As clearly seen in the spectrum on $^{12}$B in Ta (Fig. 7-6, Fig. 7-7), each quadrupole spectrum observed in the present experiment consists of three peaks. These structure can be attributed to the corresponding transition with the nuclear spin $I = 1$, and to the rf modulation effects by use of polycrystalline samples. Considering the agreements of the theoretical curves calculated by assuming the nuclear spin $I = 1$ to the data, one can conclude that the grand state spins of $^{12}$B and $^{12}$N are $I = 1$ without any ambiguity. These are consistent with the results from the $\beta$-decay data. The $\beta$-decay branching ratios and fit values are known experimentally and are listed in Table 8-1. According to the $\beta$-decay systematics, the decays to the grand state($0^+$) and to the 1st excited state($2^+$) of $^{12}$C are assigned to be allowed transitions and the spins of the grand states of the $^{12}$B and $^{12}$N are deduced to be $I = 1^+$. From the stand point of the shell model, $^{12}$B and $^{12}$N belong to the IP shell nuclei, and the Nordheim rule can predict their spins to be $I = 1$. The present results are consistent with the prediction.

The asymmetric heights of a quadrupole spectrum can be explained by the population differences of the related magnetic sublevels. The sign of the $\nu_Q$, however, cannot be assigned in the present experiment alone since two set of populations are possible to explain the data. The two sets of the populations are shown in Fig. 8-18, and Fig. 8-19.

The known quadrupole coupling constants $^{16}$ of $^{11}$B in TiB$_2$ and ZrB$_2$ were utilized to determine the ratio of the quadrupole moment of $^{12}$B to $^{11}$B. In the present experiment the ratio of the field gradients acted on $^{12}$B in the both samples are obtained to be

$$\frac{\nu_Q (^{11}B \text{ in TiB}_2)}{\nu_Q (^{12}B \text{ in TiB}_2)} = 0.32 \pm 0.05$$

This ratio is in agreement to the ratio on $^{11}$B deduced from the known coupling.
constants,\n\[ \frac{g Q (^{11}B \text{ in } Xr B_2)}{g Q (^{11}B \text{ in } Pb_2)} = 0.328 \pm 0.033. \]

In the present experiment, the NMR effects of ~5 percent were observed. These effects require 30-35 percent of the initial \( \beta \)-decay asymmetry of \(^{12}B\) as shown in Table 7-7A,B. These are in agreement with the observed one in fcc metals. Above two facts support the considerations that the exchange collisions were probable in the final stage of implantation, and the major sites of the implanted \(^{12}B\) in these samples were substitutional. Thus the known coupling constants on \(^{11}B\) can be used to deduce the ratio of the quadrupole moment of \(^{12}B\) to \(^{11}B\) to be

\[ |Q (^{12}B)/Q (^{11}B)| = 0.42 \pm 0.04. \]

G. Wessel obtained the quadrupole moment of \(^{11}B\) by the atomic beam method to be \(Q (^{11}B) = +0.0355 \pm 0.0002\) barns, without corrections from the configuration-interaction and Sternheimer polarization.

The main cause of the ambiguity which must be taken into consideration other than the statistical error in the present result arises from the uncertainty of \(Q (^{11}B)\) due to the above mentioned configuration-interaction and Sternheimer polarization. As to the configuration-interaction, G. Wessel found that the ground configuration of \(^{11}B\) was relatively free from the interaction because the ratio of the magnetic dipole interaction constants \(Q''\) and \(Q'\) of the \(2P_{1/2}\) and \(2P_{3/2}\) states observed in the atomic beam method was \(Q''/Q' = 4.993 \pm 0.001\) which was consistent to the theoretically predicted one of 5.0014 without considering the interaction. The upper limit of the interaction in \(^{11}B\) can be estimated by considering the Al(3p \(^2P_1\)) case. H. Lew and G. Wessel obtained the correction factor for Al of less than 5 percent.

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The amount of the configuration-interaction is considered large for heavier atoms due to the increase in relative importance of the coulomb repulsion between electrons as compared to the attraction of the nuclear charge. Thus the correction from this interaction for lighter element B is less than 5 percent which is comparatively small.

Another correction for \( Q^{(11)B} \) is the Sternheimer polarization. The correction for the nuclear quadrupole moment is calculated by Sternheimer to be \( 1/(1-R) = 1.17 \). The test on the reliability of the correction factors are not sufficient. Some coarse experimental tests have been made, for instance as to p or d electrons of \( \text{Eu}^{153} \), \( \text{La}^{175} \), \( \text{Ta}^{181} \), and \( \text{Re}^{185} \) atoms. In these cases the quadrupole moments obtained by hyperfine structure are systematically \( \sim 18 \) percent larger than that obtained by coulomb excitation. These results are consistent with the calculated shielding factors.

Thus the correction for \( Q^{(11)B} \) is mainly governed by the Sternheimer polarization. And the quadrupole moment of \( ^{12}\text{B} \) can be corrected by neglecting the one from configuration-interaction to be \( Q^{(12)B} \approx 0.018 \) barns.

The quadrupole moment of \( ^{12}\text{B} \) can be predicted by using the simple particle model in which the nuclear properties are attributed to an odd proton and an odd neutron each in its appropriate shell model state. The quadrupole moment of an odd-odd nucleus on the particle model is

\[
Q = \frac{(2J+1)!}{2J_p!} \frac{\sqrt{(2J_r+2)!(2J_r+3)!}}{(2J_r-2)!(2J_r+5)!} W(J_p J_r J_n) (-1)^{J_p+J_r} Q_{j_p}
\]

where \( Q_{j_p} \) is the quadrupole moment of a proton in the \( J_p \) state, \( W(J_p J_r J_n) \) is a Racah coefficient, and \( j_n \) is the neutron state. This reduces to \( \frac{1}{2}Q_{j_p} \) in the case of \( ^{12}\text{B} \). If the nuclear radius is assumed to be \( 1.22 \times 10^{-13} A^{1/3} \) cm, where \( A \) is the mass number, it reduces to \( 0.017 \) barns and this is in good agreement to the corrected result. The comparison of the calculated moments with the observed one of
the boron isotopes are shown in Table 8-2 in which experimental values are not corrected as to the configuration-interaction and the Sternheimer polarization.

On the other hand the quadrupole moment of $^{12}\text{N}$ is zero in the simple particle model. The existence of the quadrupole moment is clearly shown in the present experiment on $^{12}\text{N}$ in bcc metals. The quadrupole moment may be attributed to the "effective charge" of the last neutron in $1F_{3/2}$ state. If the effective charge of the last neutron in $^{12}\text{N}$ is assumed to be the same as that of the last neutron in other even-odd($1S^21P_{3/2}^j$) nuclei, the nuclear quadrupole moment of $^{12}\text{N}$ can be deduced. The effective charge $\beta$ is defined as $\beta = \frac{Q_{\text{el}}}{\epsilon_0}$, where $j = \frac{3}{2}$ in the present cases. By using the data on $^{11}\text{C}$ and $^9\text{Be}$ of which last neutron states are $1P_{3/2}$, the effective charge of $^{12}\text{N}$ may be assumed to be ~1 as shown in Table 8-3. Therefore the quadrupole moment may be 0.017 barns which is the same amount of $^{12}\text{B}$. Further discussions require more refined calculations.

Large differences are found among the internal field gradient acted on $^{12}\text{B}$ and $^{12}\text{N}$ in Ta, Nb, Mo, and W. As stated above, the nuclear quadrupole moment is determined in the present experiment, the absolute values of the electric field gradient with which $^{12}\text{B}$ interacted in the bcc metals can be deduced, and they are shown in Table 8-4. The electric field gradient which acted on $^{12}\text{B}$ in W is ~3 time stronger than that in Nb. The quadrupole moment of $^{12}\text{N}$ is considered to be comparable to that of $^{12}\text{B}$ as stated above. The field gradient acted on $^{12}\text{N}$ in Ta is considered to be 4.5 times larger than that of $^{12}\text{B}$ in Ta. By the case of Nb, it may be 9 times.

In order to interpret the quadrupole spectra on bcc metals, TiB$_2$ and ZrB$_2$, it is necessary to take into account a finite distribution of the quadrupole coupling constant and/or the asymmetry of the field gradient. The smearing effects by these causes on the spectral shape show the same tendencies and unique assignment is not possible by the present accuracies. The causes of the distribution of the coupling constant and the asymmetry of the field gradient are considered mainly due to
the crystalline defects, dislocations and the impurities in the samples.

The experimental half width at half maximum of the spectra on \( ^{12}\text{B} \) in Pt are about three times wider than the estimated value. The half width of the spectra on \( ^{12}\text{B} \) in Au are as narrow as 0.6 kHz. Thus the 1.2 kHz in Pt cases show the existence of other causes than that of the dipolar broadening. In fact Fig. 7-5 in which the rf-field-intensity dependences of the observed asymmetry changes are shown, suggests some comparatively narrow static broadening. If the cause is the dipolar broadening, the rf intensity must be increased to obtain the same amount of NMR effects as the modulation ranges widened. By the case of the Pt, however, an rf field with wider modulation (13 kHz) gets a larger NMR effect than the rf field which is not modulated in frequency with the same rf field intensity.
Fig. 8-1  Energy-level diagram of $A = 12$ triplet. The uncertainties of the quadrupole and magnetic moment values are given in parentheses at the last decimal.
### Table 8-1. $\beta$-Decay Data

<table>
<thead>
<tr>
<th>Element</th>
<th>$^{12}_B$</th>
<th>$^{12}_N$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$T^+_1$</td>
<td>Branching Ratio</td>
</tr>
<tr>
<td>$^{12}_C$ g.s.</td>
<td>$0^+$</td>
<td>97%</td>
</tr>
<tr>
<td>1st</td>
<td>$2^+$</td>
<td>1.4</td>
</tr>
<tr>
<td>2nd</td>
<td>$0^+$</td>
<td>1.3</td>
</tr>
</tbody>
</table>

### Table 8-2

<table>
<thead>
<tr>
<th>Element</th>
<th>Spin</th>
<th>${Q/Q(11B)}_{\text{exp}}^\alpha$</th>
<th>${Q/Q(11B)}_{\text{calc}}^\beta$</th>
<th>$Q_{\text{calc}}^\gamma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{10}_B$</td>
<td>$3$</td>
<td>$2.064 \pm 0.002$</td>
<td>$0.94$</td>
<td>$+0.074$</td>
</tr>
<tr>
<td>$^{11}_B$</td>
<td>$3/2$</td>
<td>$1$</td>
<td>$1$</td>
<td>$+0.03555$</td>
</tr>
<tr>
<td>$^{12}_B$</td>
<td>$1$</td>
<td>$0.42 \pm 0.04$</td>
<td>$0.53$</td>
<td>$0.015$</td>
</tr>
</tbody>
</table>

\(a\); D. Dehmelt, Z. Physik 135, 528 (1952), \(b\); ref. 16), \(c\); present result

*Calculated by \#Particle model.

### Table 8-3

<table>
<thead>
<tr>
<th>Element</th>
<th>Spin</th>
<th>$Q_{\text{exp}}^{\alpha}$</th>
<th>$Q_{\text{calc}}^{\beta}$</th>
<th>$\beta^\gamma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{11}_C$</td>
<td>$3/2$</td>
<td>$0.031$</td>
<td>$0.031$</td>
<td>$1$</td>
</tr>
<tr>
<td>$^9$Be</td>
<td>$3/2$</td>
<td>$0.03$</td>
<td>$0.028$</td>
<td>$1.07$</td>
</tr>
<tr>
<td>$^{12}_N$</td>
<td>$1$</td>
<td>$0.017$</td>
<td>$\sim 1$</td>
<td></td>
</tr>
</tbody>
</table>

** Effective charge of the odd neutron.

** Q\(_{\text{exp}}\); experimentally measured value (known).

### Table 8-4. Electric Field Gradient Acted on $^{12}_B$ in bcc Metals

<table>
<thead>
<tr>
<th>Element</th>
<th>$q$ (esu)*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nb(V)</td>
<td>$(0.64 \pm 0.09) \times 10^{14}$</td>
</tr>
<tr>
<td>Mo(VI)</td>
<td>$(1.13 \pm 0.19) \times 10^{14}$</td>
</tr>
<tr>
<td>Ta(V)</td>
<td>$(1.23 \pm 0.13) \times 10^{14}$</td>
</tr>
<tr>
<td>W (VI)</td>
<td>$(2.1 \pm 0.3) \times 10^{14}$</td>
</tr>
</tbody>
</table>

* Without Sternheimer correction on $Q(11B)$. If the correction is accepted $q$ must be multiplied by $1/1.17$. 
a) The results obtained in the present study are summarized.

A) Nuclear Properties
i) Nuclear spin of $^{12}\text{B} (T_\frac{1}{2} = 20\text{ ms})$ are confirmed to be $I = 1$; the quadrupole spectra on $^{12}\text{B}$ in bcc metals especially in Ta clearly showed the nuclear spin to be $I = 1$. The spectra on $^{12}\text{N}$ are also consistent with the assumption that the nuclear spin is $I = 1$. These results are consistent with the results from $\beta$-decay data.

ii) The ratio of the nuclear quadrupole moment of $^{12}\text{B}$ to $^{11}\text{B}$ are determined to be

\[ \left| \frac{Q(^{12}\text{B})}{Q(^{11}\text{B})} \right| = 0.42 \pm 0.04. \]

The nuclear quadrupole moment of $^{12}\text{B}$ are deduced by using the known value $Q(^{11}\text{B}) = (+0.03555 \pm 0.0002) \text{ barns}$ to be,

\[ Q(^{12}\text{B}) = (0.015 \pm 0.0014) \text{ barns.} \] (See cf p.110)

iii) The nuclear polarizations of $\sim +16\%$ were observed in $^{11}\text{B}(d, p)^{12}\text{B}$ reaction at $E(d) = 1.43\text{ MeV}$ and recoil angle $\theta = 30' \pm 5'$, and in $^{10}\text{B}(^3\text{He}, n)^{12}\text{N}$ reaction at $E(^3\text{He}) = 3.6\text{ MeV}$ and recoil angle $\theta = 30' \pm 5'$ respectively.

The possible two sets of populations in magnetic sublevels are obtained for each polarization (see Fig. 7-18 and Fig. 7-19).

B) Solid-State Properties
i) The observed NMR spectra on $^{12}\text{B}$ and $^{12}\text{N}$ in fcc and bcc metals are consistent with the assumption that the captured sites of the recoiled $^{12}\text{B}$ and $^{12}\text{N}$ in the samples of the heavy host metals (bcc and fcc) are interstitial. The consistency of the ratio of the field gradient acted on $^{12}\text{B}$ in TiB$_2$ and ZrB$_2$ to that of the field gradient acted on $^{11}\text{B}$ in these samples supported the consideration that the exchange collisions were probable in the final stage of implantation and the major

* Without corrections from configuration interaction and Sternheimer polarization.
sites of implanted $^{12}\text{B}$ in these samples were substitutional.

ii) Absolute values of field gradient which acted on $^{12}\text{B}$ in bcc metals are obtained by using the $Q(^{12}\text{B})$ obtained in the present experiment. They are shown in Table 8-4. Large differences among the field gradient are found. The field gradient acted on $^{12}\text{B}$ in the metal of Vth group in the periodic Table is almost twice larger than that in the metal of IVth group in the same period. And the field gradient acted on $^{12}\text{N}(\text{Vth group})$ in a metal of Vth group are considered to be $5 \sim 10$ times larger than that acted on $^{12}\text{B}(\text{IIIth group})$, if the effective charge of the last neutron in $1P_{3/2}$ state is assumed to be $\sim 1$.

The field gradient acted on $^{12}\text{B}$ in bcc metals Ta, Nb, Mo and W are found to be the same sign. On the other hand, the field gradient acted on $^{12}\text{B}$ in TiB$_2$ and ZrB$_2$ are opposite to the metal cases. And the relative signs of the field gradient on $^{12}\text{N}$ in Ta and Nb are same with each other.

iii) Knight shift anisotropy are observed in the quadrupole spectra on $^{12}\text{B}$ in Ta and Mo. The spectra are qualitatively explained by assuming positive anisotropic shift of $\sim 5 \text{ kHz}$ for the crystallographic axes oriented parallel to the external magnetic field.

b) Future Prospect

There are still many short-lived $\beta$-radioactive nuclei of which nuclear properties are expected to be studied. By selecting suitable implantation media of which internal fields are known, nuclear quadrupole moments can be measured by the present method.

The experimental method in which the automatic data normalization method and rf modulation are employed, is also fit for the measurements of the magnetic moments of other nuclei with small asymmetry factor $A$.

Present method can be utilized to the study of the nuclear polarization.
mechanism in the nuclear reaction. The absolute value of the nuclear polarization can be easily obtained if the $\beta$-decay is pure Gamow-Teller transition or mirror transition. The observation of the energy and recoil angle dependences of the polarization will give insight into the polarization mechanism.
Acknowledgement

I would like to express my indebtedness to many people who gave off their time and energy to bring this study to fruition, including the following:

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Mr. Yutaka Takahashi, for his cooperation in operation of Osaka University Van de Graaf and for his technical assistance.

Honami Minamisono, my wife, for her patient encouragement, understanding and cooperation.
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Appendix A. Quadrupole-Interaction Hamiltonian

a) The Hamiltonian of the quadrupole interaction is expressed by using the Cartesian coordinate to which the principal components of the electric field gradient are superposed.

\[ H_q = \frac{eQ}{4I(2I-1)} \left\{ I_x^2 - I_z^2 + \frac{1}{2} \alpha (I_x + I_z) \right\}. \]

In the system shown in Fig. 5-1, this can be rewritten by a transformation of the spin operators.

\[
\begin{align*}
    I_x &\rightarrow \cos \theta \cdot \cos \psi I_x + \sin \psi I_y - \sin \theta \cdot \sin \psi I_z \\
    I_y &\rightarrow \cos \psi \sin \theta I_x + \cos \psi I_y + \sin \theta \sin \psi I_z \\
    I_z &\rightarrow \sin \theta I_x + \cos \theta I_z
\end{align*}
\]

The results are shown in equation 4-8.

b) The expectation values of the spin operators can be calculated by the following formula.

\[
\begin{align*}
    \langle m | I_z^{m'} | m' \rangle &= \sqrt{I(I+1)(I+2)} \cdot \delta_{m', m+2} \\
    \langle m | I_{+}^{m'} | m' \rangle &= \sqrt{I(I+1)(I+2)} \cdot \delta_{m', m+1}
\end{align*}
\]

By the case the spin I=1, the matrix elements of the Hamiltonian are calculated as follows.

\[
\begin{align*}
    H_{11} &= \frac{1}{12} \lambda_0 \left[ 3 \cos^2 \theta - 1 \right] + \frac{1}{12} \lambda_0 \eta \cos 2 \psi \sin^2 \theta \\
    H_{00} &= -\frac{1}{6} \lambda_0 \left[ 3 \cos^2 \theta - 1 \right] - \frac{1}{6} \lambda_0 \eta \cos 2 \psi \sin^2 \theta \\
    H_{10} &= \frac{1}{12} \lambda_0 \left[ \cos^2 \theta - 1 \right] + \frac{1}{12} \lambda_0 \eta \cos 2 \psi \sin^2 \theta \\
    H_{1-1} &= -\frac{1}{12} \lambda_0 \left[ \sin^2 \theta \right] - \frac{1}{12} \lambda_0 \eta \sin 2 \cos 2 \psi + \frac{1}{12} \lambda_0 \eta \sin \theta \sin 2 \psi \\
    H_{01} &= -\frac{1}{12} \lambda_0 \left[ \sin^2 \theta \right] + \frac{1}{12} \lambda_0 \eta \sin 2 \cos 2 \psi - \frac{1}{12} \lambda_0 \eta \sin \theta \sin 2 \psi \\
    H_{0-1} &= -\frac{1}{12} \lambda_0 \left[ \sin^2 \theta \right] - \frac{1}{12} \lambda_0 \eta \sin 2 \cos 2 \psi + \frac{1}{12} \lambda_0 \eta \sin \theta \sin 2 \psi \\
    H_{-1} &= \frac{1}{12} \lambda_0 \left[ \sin^2 \theta \right] + \frac{1}{12} \lambda_0 \eta \left( 1 + \cos^2 \theta \right) \cos 2 \psi - \frac{1}{12} \lambda_0 \eta \cos \theta \sin 2 \psi \\
    H_{-2} &= -\frac{1}{12} \lambda_0 \left[ \sin^2 \theta \right] + \frac{1}{12} \lambda_0 \eta \left( 1 + \cos^2 \theta \right) \cos 2 \psi + \frac{1}{12} \lambda_0 \eta \cos \theta \sin 2 \psi
\end{align*}
\]
Appendix B. Dipolar Broadening Calculation

The numerical value used in the calculation of the dipolar broadening are tabulated below. In the course of the numerical calculations of \( \sum \frac{1}{r^6} \), the nuclei included in a sphere formed by a radius of 2.5a are mainly taken into consideration, where a is the lattice constant of the sample. Because these are the main contributions in the summation, the contribution from the outside of the sphere can be estimated to be

\[
a^6 \sum \left( \frac{S(r)}{r^6} \right) = a^6 \frac{4\pi S N}{M} \sum \frac{1}{r_0^6} \frac{dS}{dr} \\
\approx \left( a^6 \frac{4\pi S N}{M} \right) \int_{2.5a}^{\infty} \frac{dr}{r^4} \\
= \frac{4\pi S N}{3M} \frac{a^6}{(2.5a)^3}
\]

where \( S \) is the density of the sample, \( M \) is the atomic mass weight, \( N \) is the Avogadro number.

By the case of Pt this reduces to 0.18. As for the contribution from the nuclei within the sphere is \( 6.6 \times 10^{-2} \). This shows that the contribution from the outside the sphere is negligibly small. The calculated values are tabulated in the Table B-1.

<table>
<thead>
<tr>
<th>sites</th>
<th>fcc</th>
<th>bcc</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>422</td>
<td>134</td>
</tr>
<tr>
<td>B</td>
<td>659</td>
<td>148</td>
</tr>
</tbody>
</table>

**Table B-1**

<table>
<thead>
<tr>
<th>Table B-2</th>
<th>Numerical values used in the calculation (fcc)</th>
</tr>
</thead>
<tbody>
<tr>
<td>element</td>
<td>abundance</td>
</tr>
<tr>
<td>Cu(^{63} )</td>
<td>69.1%</td>
</tr>
<tr>
<td>Cu(^{65} )</td>
<td>30.9</td>
</tr>
<tr>
<td>Pt(^{195} )</td>
<td>33.7</td>
</tr>
<tr>
<td>Au(^{197} )</td>
<td>100</td>
</tr>
<tr>
<td>Al(^{27} )</td>
<td>100</td>
</tr>
</tbody>
</table>

* Square root of the second moment
Table B-3  Numerical values used in the calculation (bcc)

<table>
<thead>
<tr>
<th>element</th>
<th>abundance</th>
<th>$I^\pi$</th>
<th>$\mu$ (nm)</th>
<th>$a$ (Å)</th>
<th>$\Delta$ calc (site C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nb$^{93}$</td>
<td>92.9%</td>
<td>$9/2$</td>
<td>+6.167</td>
<td>3.29</td>
<td>5.5 kHz</td>
</tr>
<tr>
<td>Mo$^{95}$</td>
<td>15.7</td>
<td>$5/2^+$</td>
<td>-0.9135</td>
<td>3.14</td>
<td>0.51</td>
</tr>
<tr>
<td>Mo$^{97}$</td>
<td>9.5</td>
<td>$5/2^+$</td>
<td>-0.9327</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ta$^{181}$</td>
<td>99.99</td>
<td>$7/2$</td>
<td>+2.36</td>
<td>3.30</td>
<td>1.94</td>
</tr>
<tr>
<td>W$^{183}$</td>
<td>14.4</td>
<td>$1/2$</td>
<td>+0.117</td>
<td>3.16</td>
<td>0.064</td>
</tr>
</tbody>
</table>
Appendix C  Resonant destruction of polarization

A) $M_z(t)$ and $n_1(t)$ can be calculated by using equation 4-9, and 4-10. At the end of beam bombardment, the magnetization is

$$M_{z1} = \frac{\mu P}{L} \left\{ 1 - \exp(-\lambda t_1) \right\} \sum_{m=0}^{\infty} \exp(-\lambda t_m)$$

$$n_1 = \frac{\mu P}{L} \left\{ 1 - \exp(-\lambda t_1) \right\} \sum_{m=0}^{\infty} \exp(-\lambda t_m)$$

and at the end of cooling time $t_2$ they reduce to be

$$M_{z2} = M_{z1} \exp(-\lambda t_2)$$

$$n_2 = n_1 \exp(-\lambda t_2)$$

Finally at $t$ in $t_3$ range

$$M_z(t) = M_{z2} \exp(-\lambda t)$$

$$n(t) = n_2 \exp(-\lambda t).$$

Then the polarization to be observed is

$$P = \frac{1}{\mu} \int_0^{t_3} M_z(t) \, dt / \int_0^{t_3} n(t) \, dt.$$  

A simple calculation gives the equation 3-11.

b) General Case.

$$P = \left[ \frac{1}{2} \left\{ 2(1 + P_0^2 P_1^2) \right\} \exp(\lambda t_2) \left\{ 1 - \exp(-\lambda t_2) \right\} \left\{ 1 - \exp(-2(1 + P_0^2 P_1^2)) \right\} \right]$$

$$+ \left\{ 1 + 2(1 + P_0^2 P_1^2) \right\} \exp(\lambda t_2) \left\{ 1 - \exp(-\lambda t_2) \right\} \left\{ 1 - \exp(-2(1 + P_0^2 P_1^2)) \right\}$$

$$\times \left\{ 1 - \exp(-\lambda t) \right\} \left\{ 1 - \exp(-\lambda t_2) \right\} \left\{ 1 - \exp(-\lambda t_3) \right\} (x + x_0)$$

Where

$$x_t = \frac{1}{2} \left\{ 2(1 + P_0^2 P_1^2) \right\} \exp(\lambda t_2) \left\{ 1 - \exp(-\lambda t_2) \right\} \left\{ 1 - \exp(-2(1 + P_0^2 P_1^2)) \right\}$$

$$A_t = \frac{1}{2} k_1 P + (x + k_1 + \lambda) P$$

$$P_t = 2 \left\{ (x + k_1 + \lambda) \left\{ k_1^2 + (x + k_1 + \lambda) b \right\} \exp(\lambda t_2) + (x + k_1 + \lambda) \left\{ k_1^2 + (x + k_1 + \lambda) b \right\} \right\}$$

$$\times \left\{ 1 - \exp(-\lambda t) \right\} \left\{ 1 - \exp(-\lambda t_2) \right\} \left\{ 1 - \exp(-\lambda t_3) \right\} (x_t + x)$$

$$Q = \left\{ k_1 + (x + k_1 + \lambda) b \right\} \left\{ 1 - \exp(-\lambda t_3) \right\} \left\{ 1 - \exp(-\lambda t_2) \right\} \left\{ 1 - \exp(-\lambda t) \right\}$$

$$A = \left\{ (k_1 + \lambda) (P_0^2 P_1^2) + \frac{1}{2} k_1 (P_0^2 P_1^2) \right\} \left\{ 1 - \exp(-\lambda t_3) \right\} \left\{ 1 - \exp(-\lambda t_2) \right\}$$

$$b = \left\{ \frac{1}{2} k_1 (P_0^2 P_1^2) + (k_1 + \lambda) (P_0^2 P_1^2) \right\} \left\{ 1 - \exp(-\lambda t_3) \right\} \left\{ 1 - \exp(-\lambda t_2) \right\} \left\{ 1 - \exp(-\lambda t) \right\}$$

$t_1$, $t_2$, $t_3$ and $t_0$ are defined in Fig. 3-1. The $T(K_+, K_-)$ in the equation 4-15 is rewritten by, $P/P_0 = T(K_+, K_-)$. 

"b."
Appendix D

Approximate Calculation of the Center Peak

In the calculation of the center peak in the quadrupole spectra an approximation method is employed as already stated in the chapter 4. As far as the following condition is satisfied further approximation other than the equation 4-20 is unnecessary in the calculation of the center peak; the populations related to a transition of the nuclei can be completely equalized. If the suitable intensity of the rf magnetic field is selected this condition is satisfied at least within the modulation range. However, at the outside of the modulation range, a certain rate of population equalization is induced because of the dipolar broadening, and there the equalization is only partial of the initial population difference. To take the effects outside the modulation range simply into the calculation, the following approximation is made for the polarization change instead of analytical reduction of the transition probabilities by using the equation 4-17, 4-21, and 4-22;

\[
\left\{1 - T(k_+ k_-)\right\} = \left[\left\{T'(\omega, \omega_L + \omega_L) - T'(\omega, \omega_L, \omega_L)\right\} \frac{1}{2} (p_0 - p_1) + \frac{1}{2} \left[\frac{1}{2} (p_0 - p_1) + T'(\omega, \omega_L, \omega_L) (p_0 - p_1)\right]\right] \\
\text{where} \quad T'_L = T'(\omega, \omega_L, \omega_L) = T'(\omega, \omega_L + \omega_L) = T'(\omega, \omega_L + \omega_L) \\
\text{This means the approximation is confined only outside the modulation range and the differences of the value with the true value is decreases as the range is increased.}
\]

If the Larmor frequency is not covered by the modulation range, one of the two transitions within the range must be zero and \(T'(\omega, \omega_L, \omega_L)\) is zero.

In this case the calculation becomes correct one.

If the center of the modulation range is coincide to the Larmor frequency the \(k_+\) and \(k_-\) take the same value and the equation of polarization change \(\text{eq.4-15}\) reduces to the eq. 3-11 which is used for the calculation of the line shape of dipolar broadening.

In this case all over the range outside the modulation must be taken into the approximation that the difference of the calculated value from the true must be greatest. By assumption that the quadrupole width is far wide compared with the modulation range and with the dipolar broadening, the goodness of the approximation can be estimated by a numerical calculation. The half line shape in the experiment must be \(\text{eq.4-18}\) as checked in the line fitting. The difference from the approximation is \(\frac{1}{2} (f'(\omega) - f'(\omega)) P_0\).
According to the numerical calculation by using a computer, an example of the calculation is shown in Table D-1. This is simulated to the case of $^{12}$B in Ta. By the case narrowest range was 13 kHz and the absolute difference becomes $\sim 0.3\%$ which is included within the experimental errors. As the center of the modulation is separated from the Larmor frequency the differences may decrease gradually. And the range in which the equation D-1 is approximation is just the modulation width around the Larmor frequency.

<table>
<thead>
<tr>
<th>$\Delta U$ kHz</th>
<th>$\frac{\Delta P_{\text{exp}} - \Delta P_{\text{app}}}{\Delta P_{\text{exp}}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>21 %</td>
</tr>
<tr>
<td>0.2</td>
<td>21 %</td>
</tr>
<tr>
<td>13</td>
<td>7</td>
</tr>
<tr>
<td>20</td>
<td>5</td>
</tr>
<tr>
<td>40</td>
<td>3</td>
</tr>
</tbody>
</table>

*Table D-1*
Appendix E

Recoil Range Spectrum

The rate of the energy loss of a charged particle whose energy is a few hundred keV/nucleon or below, can be approximated as follows by considering effective charges of the particle during its flight:

\[ \frac{dE}{dx} = -k \sqrt{E} \]

where \( k \) is a constant determined by the atomic numbers of the particle and the host medium, and by the mass weights. A particle produced at the depth \( x \) and ejected from the target as shown in Fig. E-1 has energy,

\[ E'(x) = \frac{1}{2} (2\sqrt{E_0} - kx)^2 \]

where \( E_0 \) is the initial energy. Then the energy spectrum of the particles ejected to a direction can be written by assuming a homogeneous production rate through the target depth,

\[ \frac{dE}{dE} \ll \frac{dx}{dE} \ll \frac{1}{k \sqrt{E}} \].

On the other hand the range of a particle with energy \( E \) in the stopper is,

\[ R = \frac{2}{k'} \sqrt{E} \].

Thus the range spectrum is

\[ f(R) 
\approx (f(E) dE)/dR = \frac{1}{k' \sqrt{E}} \cdot (k' \sqrt{E}) = \frac{k'}{k} \].

This result shows that the recoil nuclei spread in the recoil stopper, and the width of the distribution almost to the target thickness.

Extreme Points of Electric Potential in bcc Crystal

When light elements are implanted in a heavy metal, they sit in the interstitial. The captured elements may probably be ionized in the host medium. If they do not bond with the host element, they may sit at the site where no electric field is expected; electric potential takes extreme value. From a brief consideration about the distribution of the surrounding metal ions, C, D points in Fig. 5-3 are expected to be electric field free. The variation of the electric potential can be estimated by considering nearest neighbours. The electric potential at x is

\[ V_c = \frac{4e}{a} \left( \frac{1}{ \sqrt{x^2+1} } + \frac{1}{ \sqrt{(x-y)^2+1} } + \frac{1}{ \sqrt{(x+y)^2+1} } \right) \]

where \( x = \frac{\alpha}{2} \) and \( \frac{dV_c}{dx} = -e \frac{x}{(\alpha/2)} \) for \( |x| \ll \frac{\alpha}{2} \).

About D point the potential is

\[ V_D = \frac{8e}{a} \left( \frac{1}{ \sqrt{(x-\alpha/2)^2+4} } + \frac{1}{ \sqrt{(x+\alpha/2)^2+4} } + \frac{1}{ \sqrt{(x-\alpha/2)^2+4} } + \frac{1}{ \sqrt{(x+\alpha/2)^2+4} } \right) \]

where \( x = \frac{\alpha}{2} \) and \( \frac{dV_D}{dx} = e \frac{x}{(\alpha/2)^2} \) for \( |x| \ll \frac{\alpha}{2} \).

These results show that at the two points electric potential takes extreme values. If the electric potential is maximum at one site, it takes minimum value at the other site.
Appendix G  Flow chart of the data analysis program

Main flow chart

Data in

Data Plot.

m, n in

Parameter in

(A)

χ² Calc.

χ², X Print Out

Rth Curve Plott.

i=m

j=n

k=l

no

go

yes

o<1/n

no

yes

j>n

no

yes

j=n

i=i-1

θ_i, θ_j

\( \theta_i^j \) Calc.

T(\( \mu_j^j \))

Calc.

\( \Delta_{pij} \)

Calc.

j=j-1

\( \sum_{pij} \)

Rth Calc.

χ², X Calc.

k=k-1
Appendix H  Flow chart of the data accumulation program

Program Start

XY Plotter Setting

Start Comm.  no

Pause yes

Scaler Preset

rf Cycle Signal

H. frf Data In

Scaler Start

rf Cycle Signal

Overflow Signal

SC Preset

no

yes

PL? use

no

yes

Start Comm.

Pause no

yes

SC Stop R's Data Out

Start Comm.

Pause yes

Scalar Preset

rf Cycle Signal
Appendix I  Details of the Present Experimental System

1) Programmed Timer
2) rf Oscillator
3) Resonance Amplifier for rf Field
4) Beam Pulser
1) Programmed Timer

- MV (Multivibrator)
- UV (Univibrator)
- FF (Flip Flop)
- Coincidence

- Beam Cycle
- Beam Pulser
- Scalar Control
- rf on, off
- Shaper

Logic gates and connections for various components.
2) rf oscillator

Saw Tooth Wave

Oscillator

Amplifier

2SC373 2SC373 2SC373 2SC373

-12V -12V 10K 0.05K

2SC373 2SC373 2SC373 2SC373

+20V
3) Resonance amplifier for rf field
4) Beam pulser

\[ C_{11} = 4 \mu F \]
\[ C_{21} = 8 \mu F \]
\[ C_{31} = 8 \mu F \]
\[ V_{11} = HV972 \]
\[ V_{31}, V_{2} = 1 \times 4 \]
\[ V_{0} = 5P7 \]

\[ L_{2} \quad \text{VR105} + \text{VR150} \times 3 \]
\[ \text{Defl. Plate} \]