



Title	Subharmonic lock-in detection and its optimization for femtosecond noise correlation spectroscopy
Author(s)	Weiss, M. A.; Herbst, F. S.; Eggert, S. et al.
Citation	Review of Scientific Instruments. 2024, 95(8), p. 083005
Version Type	VoR
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RESEARCH ARTICLE | AUGUST 20 2024

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Rev. Sci. Instrum. 95, 083005 (2024)

<https://doi.org/10.1063/5.0208499>



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Cite as: Rev. Sci. Instrum. 95, 083005 (2024); doi: 10.1063/5.0208499

Submitted: 15 March 2024 • Accepted: 29 July 2024 •

Published Online: 20 August 2024



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ABSTRACT

Although often viewed as detrimental, fluctuations carry valuable information about the physical system from which they emerge. Femtosecond noise correlation spectroscopy (FemNoC) has recently been established to probe the ultrafast fluctuation dynamics of thermally populated magnons by measurement of their amplitude autocorrelation. Subharmonic lock-in detection is the key technique in this method, allowing us to extract the pulse-to-pulse polarization fluctuations of two femtosecond optical pulse trains transmitted through a magnetic sample. Here, we present a thorough technical description of the subharmonic demodulation technique and the FemNoC measurement system. We mathematically model the data acquisition process and identify the essential parameters that critically influence the signal-to-noise ratio of the signals. Comparing the model calculations to real datasets allows validating the predicted parameter dependences and provides a means to optimize FemNoC experiments.

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I. INTRODUCTION

Fluctuations in condensed matter systems contain rich information about their microscopic interactions. In equilibrium, spontaneous fluctuations are connected to susceptibility.^{1,2} Nevertheless, measuring their dynamical properties in the time domain provides access to the fundamental mechanisms of quantum electromagnetic fields^{3,4} as well as the thermodynamical nature of solid-state systems.^{5–10} In spin systems, spin noise spectroscopy (SNS) is a widely established tool to study the paramagnetic spin fluctuations in the megahertz to gigahertz regime.¹¹ In SNS, the spin noise is imprinted onto the polarization state of a continuous-wave laser by the Faraday effect. Using SNS based on ultrashort laser pulses, the investigation of spin fluctuations with long coherence times in paramagnets was reported.^{8–10,12} However, fluctuations of the magnon systems in exchange-coupled solids often exhibit picosec-

ond dephasing times and, in particular in antiferromagnets, natural resonance frequencies up to several terahertz—well beyond the reach of conventional SNS.

In the past decade, experimental approaches granting access to fluctuation dynamics in the terahertz frequency range were developed in the emerging field of subcycle quantum optics.^{3,4,13,14} The common principle of these techniques relies on probing the polarization noise encoded, e.g., by stochastic fluctuations of the vacuum electric field in a high-repetition rate (tens to hundreds of megahertz) femtosecond probe pulse train. In practice, a variety of methodologies exist, which grant access to the encoded information: In one of the early examples of subcycle quantum optics (Ref. 3), the ultrafast stochastic properties of mid-infrared vacuum fluctuations are probed by analyzing the polarization state histogram of a single probe-pulse train detected via electro-optic sampling. While this approach enables femtosecond time resolution,

dynamical information of the vacuum field remains unresolved. To access the femtosecond dynamics, a different method, which we denote as femtosecond noise correlation spectroscopy (FemNoC), was proposed in Ref. 4. Here, the polarization noise imprinted by the vacuum electric field on two femtosecond optical probes separated in time and transmitted through an electro-optic crystal is detected using two individual polarimetric detectors. By real-time multiplication and averaging of the output signals, the autocorrelation function of the vacuum fluctuations is directly revealed at ultrafast time scales. Recently, we combined this FemNoC technology with SNS and applied it to the investigation of magnetization noise in the orthoferrite $\text{Sm}_{0.7}\text{Er}_{0.3}\text{FeO}_3$, thereby successfully resolving the incoherent noise dynamics of antiferromagnetic magnons with femtosecond resolution.¹⁵

The crucial ingredient for the implementation of FemNoC experiments is the extraction of pulse-to-pulse polarization fluctuations that carry information about the stochastic properties of the investigated system. Since a femtosecond laser system operating at high repetition rates is used in these measurements, the detection of pulse-to-pulse fluctuations requires a high-speed electronic data acquisition scheme. For this purpose, subharmonic lock-in detection¹⁶ is ideal. Here, the electronic signal output from the polarimetric detector is demodulated at half the frequency of the laser repetition rate in a radio frequency (RF) lock-in amplifier to extract only the noise component from the predominating signals at the fundamental repetition rate. Subharmonic demodulation-based FemNoC offers an array of advantages, namely access to broad detection bandwidths and a high dynamic range due to the suppression of the repetition-rate frequency component. While the FemNoC scheme was shown to successfully retrieve the pulse-to-pulse fluctuations, the parameters relevant for an efficient subharmonic detection have not been systematically studied. Indeed, a detailed technical description and a mathematical formulation of the data acquisition procedure have not been put forward.

Here, we provide a full mathematical formulation of the subharmonic lock-in detection for the FemNoC scheme, providing analytical solutions that allow us to predict the optimum combination of parameters for the FemNoC signals. We compare experimental datasets and simulation results to confirm the validity of our model. Parameters that critically influence the signal-to-noise ratio are identified, allowing for significant sensitivity enhancement of the FemNoC signals.

II. EXPERIMENTAL METHOD AND SETUP

A. Overview of the optical FemNoC setup

Our experimental setup rests on the following principle: Two linearly polarized, spectrally distinct femtosecond optical probes are transmitted through the magnetic sample (see Fig. 1). Here, by the magneto-optic Faraday effect, the transient out-of-plane magnetization noise imprints as polarization fluctuations on two femtosecond pulse trains transmitted through the sample. The probes are separated using a dichroic mirror (DM), and the induced polarization noise is then detected with two separate polarimetric detectors. These detectors yield an output signal periodic with the repetition rate f_{rep} of the laser pulse train. We deploy a radio frequency (RF) lock-in amplifier (UHFLI, Zurich Instruments¹⁷)

to extract the fluctuation of the detector output by subharmonic demodulation. This step yields the pulse-to-pulse polarization fluctuations $\Delta\delta\alpha_1(t)$ and $\Delta\delta\alpha_2(t + \Delta t)$ of the two pulse trains, and at the same time, it removes any static background α' as well as spurious $\frac{1}{f}$ -like noise components arising in the detection electronics. In the next step, $\Delta\delta\alpha_1(t)$ and $\Delta\delta\alpha_2(t + \Delta t)$ are multiplied in real time within the RF lock-in amplifier, and the resulting product is averaged subsequently. This procedure results in the correlated polarization noise $\langle \Delta\delta\alpha_1(t) \cdot \Delta\delta\alpha_2(t + \Delta t) \rangle$. At the same time, the uncorrelated background noise (shot noise of the optical probe, Johnson noise in the electronics, etc.) arising in the separate polarimetric detection arms is eliminated. By changing the variable time delay Δt of the pulse trains, the correlation function $\langle \delta M_z(t) \cdot \delta M_z(t + \Delta t) \rangle$ of the transient magnetization fluctuations is retrieved on femtosecond time scales (Fig. 1). The magnetization noise correlation function contains valuable spectroscopic information about the investigated spin system, such as the eigenfrequencies as well as the damping of the thermally populated magnons in the case of a magnetically ordered sample.¹⁵

For our experiment, we exploit a mode-locked Er: fiber laser emitting pulses with a temporal width of 150 fs, a central wavelength of 1.55 μm , and a total energy of 5 nJ at a repetition rate of 40 MHz. This output is first frequency doubled in a periodically poled lithium niobate (PPLN) crystal before being spectrally and spatially separated by a dichroic mirror. The resulting optical probe pulses are linearly polarized and have central wavelengths of 767 and 775 nm with 3–4 nm bandwidths, respectively. Both pulses exhibit energies of ~ 62.5 pJ for a duration of 300 fs (see Fig. 1). One of the pulse trains is sent over an optical delay line, where it is temporally shifted by a variable time-delay Δt with respect to the other femtosecond pulse train. We use a strain-free transmissive microscope objective lens (Nikon, CFI Achrom LWD P 20X) with a numerical aperture of 0.4 and a working distance of 3.9 mm (L1) to tightly focus the pulses onto a spot size below 2 μm on the magnetic sample. Note that in our case, the two probes are aligned to illuminate the same volume of the sample to probe the correlations in time only. In principle, if the probing volumes are spatially separated, information about the spatial correlations could be gathered as well.

In comparison with the ultrafast SNS scheme previously demonstrated in Ref. 9 wherein a single balanced detector receives two femtosecond optical pulses, we here exploit two different spectral bands and separate balanced detection arms for the optical probes. With this scheme, we can reveal the ultrafast correlation dynamics down to femtosecond time delays because the following mechanism is avoided: If a femtosecond laser pulse impinges on a photodetector, a finite dead time in the order of tens of picoseconds emerges due to the persisting photo-carriers. This nonlinearity deteriorates the detected signals when the temporal separation of the two probe pulses is small, thus critically limiting the probing window.

B. Optical probing of the ultrafast magnetization fluctuations

Upon transmission at times t and $t + \Delta t$, transient magnetization fluctuations $\delta M_z(t)$ parallel to the propagation direction of the

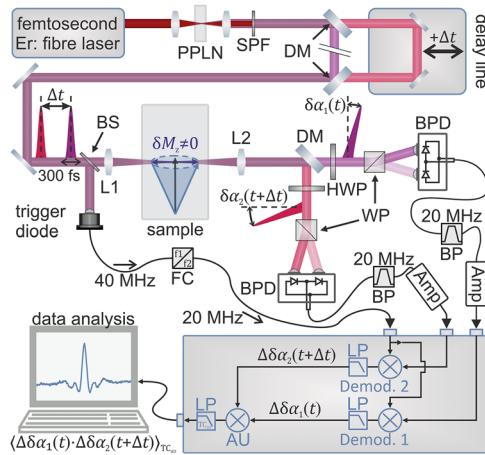


FIG. 1. Schematic illustration of the experimental setup and electronic data processing. An Er: fiber laser generates a 40 MHz pulse train consisting of 150 fs laser pulses with a central wavelength of $1.55 \mu\text{m}$ (dark red beam). The beam is focused onto a periodically poled lithium niobate (PPLN) crystal, where second-harmonic laser pulses with a central wavelength around 775 nm (dark purple beam) are generated. After filtering out the fundamental beam using a short-pass filter (SPF), the second harmonic pulse train is spectrally and spatially separated into two branches with 767 nm (light purple beam) and 775 nm (light red beam) central wavelengths, respectively, using a dichroic mirror (DM, Edmund Optics No. 86-069). The 775 nm beam then passes an optical delay line, where a variable time delay Δt is introduced. Afterward, the two beams are spatially recombined using a second dichroic mirror (DM, Edmund Optics No. 86-104). After second-harmonic generation and spectral separation, the probes exhibit a pulse duration of 300 fs (red and purple pulses). They are focused onto a magnetically ordered sample and, upon transmission, experience a polarization rotation proportional to the out-of-plane spin fluctuations δM_z due to the Faraday effect. The corresponding rotation angles $\delta\alpha_{1,2}$ are measured in separate polarimeters (BPD). The detector outputs are bandpass filtered and amplified before being sent into a radio frequency lock-in amplifier (gray box). In the lock-in amplifier, the pulse-to-pulse fluctuations $\Delta\delta\alpha_{1,2}$ from each branch are extracted using subharmonic demodulation. The two demodulation outputs are then multiplied and averaged in the arithmetic unit (AU) of the lock-in amplifier, yielding the cross correlation function $\langle \Delta\delta\alpha_1(t) \cdot \Delta\delta\alpha_2(t+\Delta t) \rangle_{\text{TC}}$ as a function of the delay time Δt . BS: beam splitter; FC: frequency converter; L1: microscope objective lens; L2: collimation lens; HWP: half-wave plate; WP: Wollaston prism; BPD: balanced photodetector; BP: electronic bandpass filter; Amp: amplifier; Demod.: demodulator; and LP: digital low-pass filter.

beams (z -direction) are encoded as a polarization rotation noise of the light,

$$\delta\alpha(t) \propto d \cdot \mu_0 \cdot V(\lambda) \cdot \delta M_z(t), \quad (1)$$

via the magneto-optic Faraday effect.¹⁸ Here, d is the thickness of the sample, μ_0 is the vacuum permeability, and $V(\lambda)$ is the Verdet constant as a function of wavelength λ . Note that the proximity of the center wavelengths of the two readout pulses used here ensures a negligible influence of the dispersion of $V(\lambda)$.

After transmission, a 20 mm focal length lens (L2) collimates the beams, which are then guided into two polarimetric detector arms using a dichroic mirror (DM). Each consists of a half-wave plate (HWP), a Wollaston prism (WP), and a pair of balanced photodetectors (BPD). The WP splits the beam into its s - and p -polarized components, which are subsequently focused onto and

detected by the BPD. The BPD yields a polarization angle $\alpha(t)$ dependent difference current,¹⁹

$$\Delta I(\alpha(t)) = I_s - I_p = sP_0(1 - 2\cos^2(\alpha(t))), \quad (2)$$

where s is the quantum efficiency of the detectors, P_0 is the average laser power, and I_s and I_p are the photocurrents produced by the s - and p -polarized components of the input beam, respectively. Note that the time-dependent polarization angle $\alpha(t) = \langle \alpha \rangle + \delta\alpha(t)$ consists of a static polarization component $\langle \alpha \rangle$ arising, for example, from the initial laser polarization or the static magnetization components along the propagation direction of light, as well as a time-dependent fluctuation part $\delta\alpha(t)$, which contains the transient magnetization fluctuations of the sample introduced in Eq. (1). We use the HWP to rotate the static polarization component of the beams to $\langle \alpha \rangle = \frac{\pi}{4} + \alpha'$, where α' is a small static offset caused by, for example, imperfect balancing. In the case of $\alpha' \ll \delta\alpha(t) \ll 1$, Eq. (2) reduces to

$$\Delta I(\alpha(t)) \approx 2sP_0\delta\alpha(t) \propto \delta M_z(t), \quad (3)$$

indicating that the balanced output is directly proportional to the magnetization fluctuation. In practice, the offset angle $\alpha' \gg \delta\alpha(t)$ and the fluctuations under investigation are consequently dominated by a large static background. Furthermore, additional sources of unwanted background noise arising in the detection circuit impair the measurement, namely the photon shot noise of the probe pulses, as well as Johnson noise and $\frac{1}{f}$ noise. To extract the transient spin noise from such a large spurious background, we use two independent polarimetric detector arms and calculate their cross correlation in a later step. By this method, uncorrelated background noise is eliminated and only the correlated noise due to the magnetization fluctuations remains.

C. Mathematical formulation of the subharmonic lock-in detection and subsequent correlation analysis

In the following, we first mathematically model the process of lock-in detection for general demodulation frequencies. We then focus on the case of subharmonic demodulation at half the repetition frequency $\frac{f_{\text{rep}}}{2}$ and compare it to the conventional harmonic demodulation at f_{rep} . In particular, we show that the subharmonic demodulation grants access to the fluctuation part $\delta V_s(\tau)$ of a time-varying signal $V_s(\tau) = \langle V_s \rangle + \delta V_s(\tau)$, where τ is the general laboratory time, while removing the mean value $\langle V_s \rangle$ and $\frac{1}{f}$ noise components. In contrast, harmonic demodulation reveals $\langle V_s \rangle$ through averaging of $V_s(\tau)$ and thereby diminishing the fluctuation part. Finally, the subsequent correlation analysis is discussed, which yields the correlated noise and removes any uncorrelated background.

1. Lock-in detection of pulse trains

Given their short pulse duration, we model the femtosecond pulsed laser train by a sequence of Dirac delta functions, establishing a time-domain Dirac comb periodic with the sampling time $T_{\text{rep}} = \frac{1}{f_{\text{rep}}}$,

$$\text{III}(\tau) = \sum_{n=-\infty}^{\infty} \delta(\tau - nT_{\text{rep}}). \quad (4)$$

$\text{III}(\tau)$ denotes the sampling function and describes the arrival time of the optical probes impinging on the sample with periodicity T_{rep} at laboratory time τ and $n \in \mathbb{Z}$ as the pulse number. Sampling of a time-varying signal $V_s(\tau)$ in the laboratory time frame τ with a femtosecond pulse train can therefore be approximated by simple multiplication with the sampling function (see Fig. 2, panel 1),

$$V_{\text{mod}}(\tau) = V_s(\tau) \cdot \text{III}(\tau). \quad (5)$$

Upon measurement, this modulated signal $V_{\text{mod}}(\tau)$ is subject to $\frac{1}{f}$ and white noise components arising in the detection electronics (Fig. 2, panel 2). We denote these quantities as $V_{1/f}(\tau)$ and $V_w(\tau)$, respectively. Consequently,

$$V_{\text{out}}(\tau) = V_{\text{mod}}(\tau) + V_{1/f}(\tau) + V_w(\tau) \quad (6)$$

is the total output of the detectors and serves as the input for the lock-in amplifier. In the first step, the lock-in performs a dual-phase demodulation²⁰ on this input: $V_{\text{out}}(\tau)$ is multiplied with a sinusoidal reference function $V_{d,X}(\tau) = A \cdot \cos(\omega_d \tau + \theta)$ and a 90° phase-shifted copy $V_{d,Y}(\tau) = A \cdot \sin(\omega_d \tau + \theta)$. The in-phase and orthogonal quadrature components are

$$X(\tau) = V_{\text{out}}(\tau) \cdot V_{d,X}(\tau) = V_{\text{out}}(\tau) \cdot [A \cdot \cos(\omega_d \tau + \theta)]$$

and

$$Y(\tau) = V_{\text{out}}(\tau) \cdot V_{d,Y}(\tau) = V_{\text{out}}(\tau) \cdot [A \cdot \sin(\omega_d \tau + \theta)], \quad (7)$$

respectively. Here, A is the amplitude, ω_d is the angular frequency, and θ is the relative phase of the reference signal. Mathematically, this mixing can also be described as multiplication with a single complex Euler's function,

$$Z(\tau) = X(\tau) + iY(\tau) = V_{\text{out}}(\tau) \cdot A e^{i(\omega_d \tau + \theta)}. \quad (8)$$

Subsequently, low-pass filtering of the input is performed on the mixed signal $Z(\tau)$ in the time interval $[0, TC]$, which we approximate by simple integration as follows:²¹

$$\begin{aligned} \langle Z(\tau) \rangle_{TC} &= \frac{1}{TC} \int_0^{TC} V_{\text{out}}(\tau) \cdot A e^{i(\omega_d \tau + \theta)} d\tau \\ &= \frac{A}{TC} \int_0^{TC} (V_{\text{mod}}(\tau) + V_{1/f}(\tau) + V_w(\tau)) e^{i(\omega_d \tau + \theta)} d\tau \\ &= \frac{A}{TC} \int_0^{TC} \sum_{n=-\infty}^{\infty} \delta(\tau - nT_{\text{rep}}) V_s(\tau) \cdot e^{i(\omega_d \tau + \theta)} d\tau \\ &\quad + \frac{A}{TC} \int_0^{TC} V_{1/f}(\tau) \cdot e^{i(\omega_d \tau + \theta)} d\tau \\ &\quad + \frac{A}{TC} \int_0^{TC} V_w(\tau) \cdot e^{i(\omega_d \tau + \theta)} d\tau. \end{aligned} \quad (9)$$

Next, we exploit the sampling property of the Dirac delta distribution²² $\int f(x) \delta(x - x_0) dx' = f(x_0)$ and choose the time constant $TC = n_{\text{max}} T_{\text{rep}}$ to be a multiple of the interpulse distance T_{rep} to evaluate Eq. (9),

$$\begin{aligned} \langle Z(\tau) \rangle_{TC} &\approx \frac{A}{n_{\text{max}}} \sum_{n=0}^{n_{\text{max}}} \underbrace{V_s(nT_{\text{rep}})}_{V_{s_n}} \cdot e^{i(\omega_d nT_{\text{rep}} + \theta)} \\ &\quad + \frac{A}{TC} \int_0^{TC} \underbrace{V_{1/f}(\tau) \cdot e^{i(\omega_d \tau + \theta)} d\tau}_{\approx V_{1/f}^*(\omega_d)} \\ &\quad + \frac{A}{TC} \int_0^{TC} \underbrace{V_w(\tau) \cdot e^{i(\omega_d \tau + \theta)} d\tau}_{\approx V_w^*(\omega_d)} \\ &= \frac{A}{n_{\text{max}}} \sum_{n=0}^{n_{\text{max}}} V_{s_n} \cdot e^{i(\omega_d nT_{\text{rep}} + \theta)} \\ &\quad + \frac{A}{TC} \cdot V_{1/f}^*(\omega_d) + \frac{A}{TC} \cdot V_w. \end{aligned} \quad (10)$$

We define $V_{s_n} = \langle V_s \rangle + \delta V_{s_n}$ as the time-varying signal $V_s(t)$, probed by the pulse number n . For large time constants ($TC \rightarrow \infty$), the second and third terms of Eq. (10) can be interpreted as the Fourier transform of the background noise time traces evaluated at frequency $\omega = \omega_d$. Therefore, they correspond to the frequency-domain representations $V_{1/f}^*(\omega_d)$ and $V_w^*(\omega_d)$ of $\frac{1}{f}$ and white noise, respectively. The white noise part is frequency-independent and therefore constant for all frequencies $V_w^*(\omega) \equiv V_w = \text{const.}$

2. Harmonic demodulation

In conventional, harmonic demodulation, the goal is to obtain the mean of the time-varying signal $\langle V_s \rangle$ at the reference frequency ω_d . In this case, the reference frequency is chosen to be equal to the angular repetition rate $\omega_{\text{rep}} = 2\pi f_{\text{rep}} = \frac{2\pi}{T_{\text{rep}}} = \omega_d$. To this end, we can neglect the $\frac{1}{f}$ noise component for large repetition rates ω_{rep} , because $V_{1/f}^*(\omega_d \rightarrow \infty) = 0$. Furthermore, long time constants TC are beneficial to determine $\langle V_s \rangle$ with high accuracy, because the number of pulses $n_{\text{max}} = \frac{TC}{T_{\text{rep}}}$ increases over which the averaging is performed. Consequently, the fluctuation part δV_{s_n} vanishes for $TC \rightarrow \infty$ and $\langle V_{s_n} \rangle_{n_{\text{max}}} \rightarrow \langle V_s \rangle$. Inserting these expressions into Eq. (10) gives

$$\begin{aligned} \langle Z(\tau) \rangle_{TC} &\approx \frac{A}{n_{\text{max}}} \sum_{n=0}^{n_{\text{max}}} V_{s_n} \cdot e^{i(2\pi n + \theta)} + \frac{V_w A}{TC} \\ &= \langle V_{s_n} \rangle_{n_{\text{max}}} \cdot A e^{i\theta} + \frac{V_w A}{TC} \\ &\approx \langle V_s \rangle \cdot A e^{i\theta} + \frac{V_w A}{TC}. \end{aligned} \quad (11)$$

Thus, harmonic demodulation suppresses the fluctuation part for long time constants TC , providing the mean of the time-varying signal $\langle V_s \rangle$ multiplied by a phase factor $A e^{i\theta}$ (Fig. 2, panel 3a). Still, a non-zero white noise background $\frac{V_w A}{TC}$ persists in the case of finite TC .

3. Subharmonic demodulation

We now turn to the case of subharmonic demodulation. To access the fluctuation part $\delta V_s(\tau)$ of the pulse train, the lock-in input is mixed with reference sinusoidal functions with a sub-

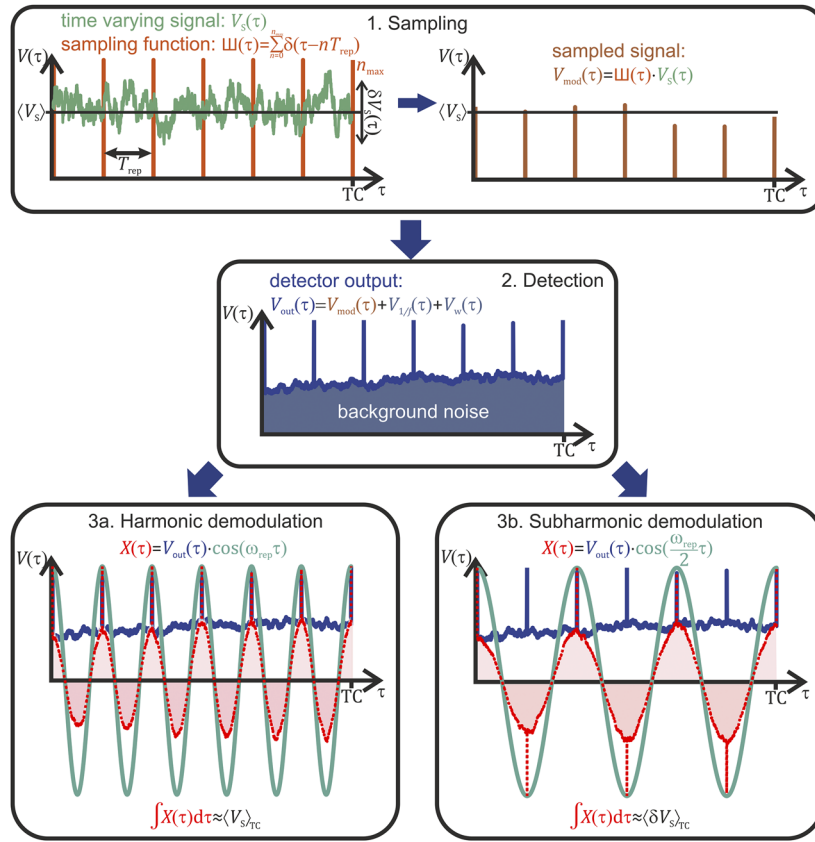


FIG. 2. Difference between the harmonic and subharmonic demodulation of a sampled time-varying signal $V_s(\tau) = \langle V_s \rangle + \delta V_s(\tau)$. $V_s(\tau)$ consists of a mean value $\langle V_s \rangle$ and a fluctuation part δV_s . In a first step (1. sampling), $V_s(\tau)$ is sampled with a sampling function $\Psi(\tau)$, which consists of $n_{\max}$ periodic Dirac pulses equally spaced by the repetition time T_{rep} within the observation interval $[0, TC]$. This yields the sampled or modulated time trace $V_{\text{mod}}(\tau)$ and emulates ultrafast probing of a fluctuating material using a pulse train of femtosecond laser pulses. Upon detection of this modified pulse train (2. detection), $V_{\text{mod}}(\tau)$ is afflicted by $\frac{1}{f}$ noise $V_{1/f}(\tau)$ and white background noise $V_w(\tau)$, respectively. The detector output $V_{\text{out}}(\tau) = V_{\text{mod}}(\tau) + V_{1/f}(\tau) + V_w(\tau)$ then serves as the input for the demodulation process in a lock-in amplifier. Here, $V_{\text{out}}(\tau)$ is multiplied with a reference cosine function with the pulse train frequency $\omega_{\text{rep}} = \frac{2\pi}{T_{\text{rep}}}$ to yield the mean value $\langle V_s \rangle_{TC}$ of the investigated time-varying signal $V_s(\tau)$ in the observation window $[0, TC]$ upon integration of the product (3a. harmonic demodulation). Alternatively, multiplication with a cosine function of half the pulse train frequency $\frac{\omega_{\text{rep}}}{2}$ and subsequent integration grant access to the fluctuation part $\langle \delta V_s \rangle_{TC}$ (3b. subharmonic demodulation).

harmonic repetition frequency $\omega_d = \frac{\omega_{\text{rep}}}{m}$, where $m \in \mathbb{N} \setminus \{0\}$. We will see that in this case, small time constants TC are beneficial to measure δV_s . Consequently, the approximation of the background noise by a Fourier transform in Eq. (10), where large time constants were assumed, is no longer valid. In addition, we assume that $\frac{1}{f}$ background fluctuations that predominantly appear below hundreds of kHz are much slower than the fluctuations of interest here, which are encoded in the optical probe pulse trains at tens of megahertz repetition rates. Therefore, within the corresponding time scale of tens of nanoseconds, they can be approximated as constant ($V_{1/f}(\tau) \approx V_{1/f} = \text{const}$). Low-pass filtering this constant $\frac{1}{f}$ component therefore yields

$$\begin{aligned} \frac{A}{TC} \int_0^{TC} V_{1/f}(\tau) \cdot e^{i(\omega_d \tau + \theta)} d\tau \\ \approx \frac{A V_{1/f} e^{i\theta}}{TC} \int_0^{TC} e^{i\left(\frac{2\pi}{m T_{\text{rep}}}\right) \tau} d\tau \\ = -i \frac{A V_{1/f} m e^{i\theta}}{2\pi n_{\max}} \left(e^{2\pi i \frac{n_{\max}}{m}} - 1 \right). \end{aligned} \quad (12)$$

Equation (12) shows that in order to avoid a static background arising from the $\frac{1}{f}$ noise component, the time constant must fulfill the boundary condition $TC = m k T_{\text{rep}}$, where $k = \frac{n_{\max}}{m} \in \mathbb{N}$ is the number of subharmonic cycles within one time constant. If this applies, $e^{2\pi i \frac{n_{\max}}{m}} - 1 = e^{2\pi i k} = 0$ and, consequently, the

$\frac{1}{f}$ term can be neglected. In contrast, this does not apply to the white noise component $\frac{A}{TC} \int_0^{TC} V_w(\tau) \cdot e^{i(\omega_d \tau + \theta)} d\tau := V'_w(\tau)$, because of its frequency independence. Therefore, a finite white noise contribution also prevails in the case of the subharmonic demodulation.

In the following, we restrict the discussion to the first subharmonic reference frequency at half the repetition rate $\omega_d = \frac{\omega_{rep}}{2}$ ($m = 2$). As we show in more detail in S3 of the [supplementary material](#), this choice avoids preliminary averaging of the fluctuation part and maximizes the output fluctuation amplitude. It is thus more advantageous compared to the case of larger m values, in agreement with Ref. 16.

Inserting $\omega_d = \frac{\omega_{rep}}{2}$ into Eq. (9) leads to

$$\begin{aligned} \langle Z(\tau) \rangle_{TC} &\approx \frac{Ae^{i\theta}}{n_{max}} \sum_{n=0}^{n_{max}} V_{s_n} \cdot e^{i\pi n} + V'_w(\tau) \\ &= \frac{Ae^{i\theta}}{n_{max}} \sum_{n=0}^{n_{max}} (V_{s_{2n}} - V_{s_{2n+1}}) + V'_w(\tau) \\ &= \frac{2Ae^{i\theta}}{n_{max}} \sum_{n=0}^{n_{max}} (\langle V_s \rangle + \delta V_{s_{2n}} - \langle V_s \rangle - \delta V_{s_{2n+1}}) + V'_w(\tau) \\ &= 2Ae^{i\theta} \langle \delta \Delta V_{s_{2n}} \rangle_{\frac{n_{max}}{2}} + V'_w(\tau), \end{aligned} \quad (13)$$

where $\langle \delta \Delta V_{s_{2n}} \rangle_{\frac{n_{max}}{2}} = \sum_{n=0}^{\frac{n_{max}}{2}} (\delta V_{s_{2n}} - \delta V_{s_{2n+1}})$ is the mean pulse-to-pulse difference (pulse-to-pulse fluctuation) of $\frac{n_{max}}{2}$ pulse pairs sampled within the detection time window $[0, TC]$. From Eq. (13), we can see that by choosing the time constant to be $TC = 2T_{rep}$, the demodulation output directly reflects pulse-to-pulse fluctuations. Remember that δV_s contains information of the investigated magnetization fluctuation, whereas the white noise $V'_w(\tau)$ part is constituted by spurious background noise. However, usually $V'_w(\tau) \gg \delta V_s$, which is why an additional correlation analysis is performed to extract δV_s from the dominant background.

4. Correlation analysis

Finally, we perform a cross correlation analysis of the two sampling pulse trains, which are detected and demodulated in identical, but separate optical and electronic channels. Channels 1 and 2 each yield the output $\langle Z_{1,2}(\tau) \rangle_{TC} \approx 2Ae^{i\theta_{1,2}} \langle \delta \Delta V_{1,2,s_{2n}} \rangle_{\frac{n_{max}}{2}} + V'_{1,2,w}(\tau)$ [see Eq. (13)] after the demodulation process. Here, θ_1 and θ_2 are the phases of pulse trains 1 and 2 relative to the reference sinusoidal functions, respectively. In the next step, the real parts $\text{Re}(\langle Z_{1,2}(\tau) \rangle_{TC}) = \langle X_{1,2}(\tau) \rangle_{TC} = 2A \cos(\theta_{1,2}) \cdot \langle \delta \Delta V_{1,2,s_{2n}} \rangle_{\frac{n_{max}}{2}} + V'_{1,2,w}(\tau)$ are multiplied within the lock-in amplifier using its real-time calculation function. In the following, this function is called *arithmetic unit* (AU), following the notation of the RF lock-in used in this study. The resulting product is then integrated over a sufficiently long time window TC_{AU} to obtain the correlation function between the two channels,

$$\begin{aligned} &\langle \langle X_1(\tau) \rangle_{TC} \cdot \langle X_2(\tau) \rangle_{TC} \rangle_{TC_{AU}} \\ &= \frac{4A^2 \cos(\theta_1) \cos(\theta_2)}{TC_{AU}} \int_0^{TC_{AU}} \\ &\quad \times \left[\langle \delta \Delta V_{1,s_{2n}} \rangle_{\frac{n_{max}}{2}} \langle \delta \Delta V_{2,s_{2n}} \rangle_{\frac{n_{max}}{2}} + V'_{1,w}(\tau) V'_{2,w}(\tau) \right. \\ &\quad \left. + \langle \delta \Delta V_{1,s_{2n}} \rangle_{\frac{n_{max}}{2}} V'_{2,w}(\tau) + V'_{1,w}(\tau) \langle \delta \Delta V_{2,s_{2n}} \rangle_{\frac{n_{max}}{2}} \right] d\tau. \end{aligned} \quad (14)$$

The background noise of the distinct channels $V'_{1,2,w}(\tau)$ and the cross terms between the background noise and pulse-to-pulse fluctuations $\delta \Delta V_{1/2,s_{2n}}$ are uncorrelated, and so, their correlation term vanishes for large TC_{AU} ,

$$\begin{aligned} &\int_0^{TC_{AU}} V'_{1,w}(\tau) V'_{2,w}(\tau) d\tau \\ &= \int_0^{TC_{AU}} \langle \delta \Delta V_{1,s_{2n}} \rangle_{\frac{n_{max}}{2}} V'_{2,w}(\tau) d\tau \\ &= \int_0^{TC_{AU}} V'_{1,w}(\tau) \langle \delta \Delta V_{2,s_{2n}} \rangle_{\frac{n_{max}}{2}} d\tau = 0. \end{aligned} \quad (15)$$

As the final output of the subharmonic demodulation and correlation analysis, we thus obtain

$$\begin{aligned} &\langle \langle X_1(\tau) \rangle_{TC} \cdot \langle X_2(\tau) \rangle_{TC} \rangle_{TC_{AU}} \\ &= \frac{4A^2 \cos(\theta_1) \cos(\theta_2)}{TC_{AU}} \int_0^{TC_{AU}} \langle \delta \Delta V_{1,s_{2n}} \rangle_{\frac{n_{max}}{2}} \\ &\quad \times \langle \delta \Delta V_{2,s_{2n}} \rangle_{\frac{n_{max}}{2}} d\tau \\ &= 4A^2 \cos(\theta_1) \cos(\theta_2) \langle \langle \delta \Delta V_{1,s_{2n}} \rangle_{\frac{n_{max}}{2}} \langle \delta \Delta V_{2,s_{2n}} \rangle_{\frac{n_{max}}{2}} \rangle_{TC_{AU}} \\ &= 4A^2 \cos(\theta_1) \cos(\theta_2) \langle CCR_{\delta \Delta V} \rangle_{TC_{AU}}. \end{aligned} \quad (16)$$

Here, $\langle CCR_{\delta \Delta V} \rangle_{TC_{AU}}$ is the mean momentary cross correlation between the pulse-to-pulse fluctuation sampled with pulse trains 1 and 2. By delaying the pulse trains with a variable ultrashort time delay Δt , $CCR_{\delta \Delta V}(\Delta t)$ directly reflects the magnon noise autocorrelation in the ultrafast time scale.

III. RESULTS AND DISCUSSION

Section II C shows that there are several essential parameters that critically influence FemNoC signals. Here, we systematically investigate the dependences of $CCR_{\delta \Delta V}(\Delta t)$ on each of the parameters and propose a protocol to optimize the signal-to-noise ratio from both experiment and simulation. In the experiment, we measure the real part $\langle \langle X_1(\tau) \rangle_{TC} \cdot \langle X_2(\tau) \rangle_{TC} \rangle_{TC_{AU}} = 4A^2 \cos(\theta_1) \cos(\theta_2) \cdot \langle CCR_{\delta \Delta V} \rangle_{TC_{AU}}$ [see Eq. (16)] of the correlated magnon noise in a 10 μm thick antiferromagnetic $\text{Sm}_{0.7}\text{Er}_{0.3}\text{FeO}_3$ sample^{15,23} held at 294.2 K, i.e., close to the temperature at which the sample undergoes a spin-reorientation transition and the magnon noise is enhanced.¹⁵ Furthermore, we apply an external magnetic field of 28 ± 5 mT along the out-of-plane direction to suppress the stochastic picosecond random telegraph noise reported in Ref. 15, thus providing more robust and reproducible datasets between multiple measurements. We sample the noise correlation function by varying the time delays of the two

pulse trains from $\Delta t = -85$ ps to $\Delta t = +85$ ps in 2 ps steps. Each data point retrieved for a fixed time delay is the result of averaging $\sim \frac{TC_{AU}}{T_{rep}}$ correlation samples. Between measurements of data points, a dwell time of $2TC_{AU}$ is implemented. Unless specified otherwise, the following measurement parameters were used: $\theta_1 = 0^\circ$, $\theta_2 = 20^\circ$, $TC = 50$ ns, and $TC_{AU} = 1.0$ s. Furthermore, each of the two inputs $\langle X_1(\tau) \rangle_{TC} [V]$ and $\langle X_2(\tau) \rangle_{TC} [V]$ is subject to a scaling factor of $\frac{10}{\sqrt{V}}$, which yields the correlation function $100 \cdot \langle \langle X_1(\tau) \rangle_{TC} \cdot \langle X_2(\tau) \rangle_{TC} \rangle_{TC_{AU}} [V]$ (also in the unit of V) as our final output from the lock-in amplifier.

The experimental observations are compared to simulations resting on the mathematical formalism introduced in Sec. II C. The details of the simulation are summarized in S1 of the [supplementary material](#). In the simulation, two periodic Dirac combs with a correlated amplitude modulation are prepared, thereby emulating the polarization rotation noise imprinted on the probing pulse trains by the correlated magnon noise. Next, uncorrelated $\frac{1}{f}$ and white noise contributions are added in the time domain, corresponding to the spurious background arising in the detection circuit. The white noise time trace is modeled via selecting random numbers from a Poissonian distribution and subsequently applying a scaling factor. The $\frac{1}{f}$ noise is modeled by first generating a white noise time trace in the same way as above and subsequent application of a $\frac{1}{f}$ filter and a scaling factor. For further information, see S1 and S2 of the [supplementary material](#). In a next step, the pulse trains are subject to subharmonic demodulation followed by a correlation analysis as described in Sec. II C. This procedure results in the correlation amplitude for a given Δt . Repeating the simulation at various time delays Δt emulates the autocorrelation of the magnetization obtained in the experiments. To account for the stochastic nature of this simulation, the data provided in this article are the average of five independent iterations, for each of which the background noise contributions are generated independently.

A typical experimental waveform of the magnon correlation is depicted in Fig. 3. The time trace shows a temporally symmetric function peaking at $\Delta t = 0$, followed by a gradual decrease and oscillation with a period of tens of picoseconds. As we have shown in detail in Ref. 15, this oscillation is ascribed to the quasi-ferromagnetic mode magnon dynamics,²⁴ enhanced around the spin reorientation transition. It proves that our FemNoC system can correctly retrieve the thermal fluctuations of magnetization. In the following, we use such waveforms to verify the parameter dependences predicted by Eq. (16).

A. Demodulation phase dependence

First, we measure the correlation amplitude at a relative time delay of $\Delta t = 0$ (see Fig. 3) for various phases $\theta_{1,2}$ of the reference sinusoidal function used in the subharmonic demodulation of the lock-in channels 1 and 2. They are varied for 2π in steps of $\frac{\pi}{12}$. Since only the real part of the correlation function [see Eq. (8)] is measured in the experiment, the signal-to-noise is expected to strongly depend on $\theta_{1,2}$. The result is plotted in the left panel of Fig. 4. It clearly shows the $\cos(\theta_1) \cos(\theta_2)$ behavior expected from Eq. (16). A tiny shift of the experimental data toward positive θ_2 is assigned to slightly different phases of the inputs of channels 1 and 2 arising,

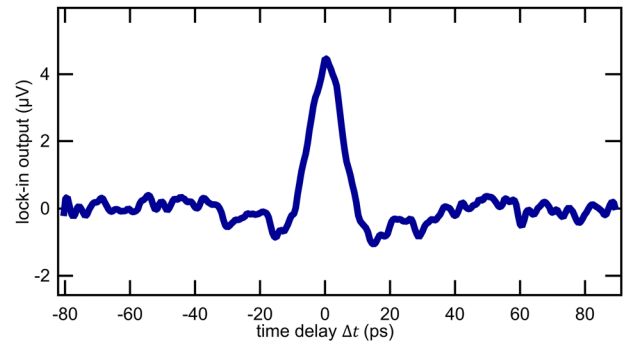


FIG. 3. Typical correlated noise waveform recorded for antiferromagnetic $\text{Sm}_{0.7}\text{Er}_{0.3}\text{FeO}_3$ at 306.65 K. The correlated noise amplitude depends on the time delay Δt of the two optical femtosecond probes.

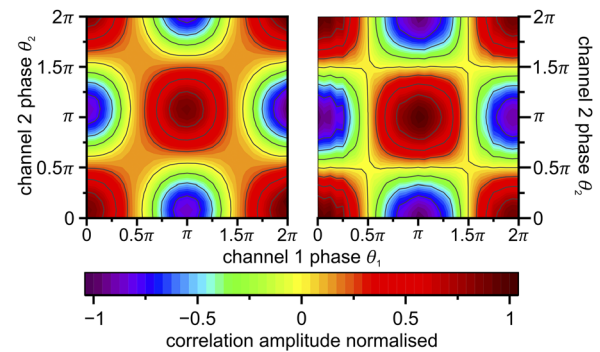


FIG. 4. Normalized correlated noise amplitude as a function of phases θ_1 and θ_2 of the inputs of channels 1 and 2. The experimental data (left) are recorded using the measurement parameters specified in the text, while the simulation data (right) are the result of the simulation (see the [supplementary material](#)). The plots are normalized to the interval $[-1, 1]$.

for example, from minor differences in the electronic circuit. The simulation based on the introduced mathematical formalism clearly replicates the characteristics of the experimental data, as depicted in the right panel of Fig. 4. As is evident from this observation, the phases of the reference to demodulate each channel need to be chosen to match the extrema positions in Fig. 4 to maximize the output.

B. Amplitude dependence

Second, the influence of the fluctuation amplitude on the correlated noise output is investigated. As can be seen in Eq. (16), the FemNoC signal is a product of the polarization noises demodulated from two signal channels. Therefore, the output correlation signal amplitude should scale quadratically to the strength of the correlated noises. To verify this feature, we perform a numerical simulation in which the standard deviation σ_{corr} of the simulated noisy pulse train is varied. The results are plotted in Fig. 5(b) (see the [supplementary material](#) for details). The correlation amplitude uniformly increases for a larger amplitude of σ_{corr} . By plotting the correlation amplitude for $\Delta t = 0$ as a function of σ_{corr} [Fig. 5(d)], it is evident that

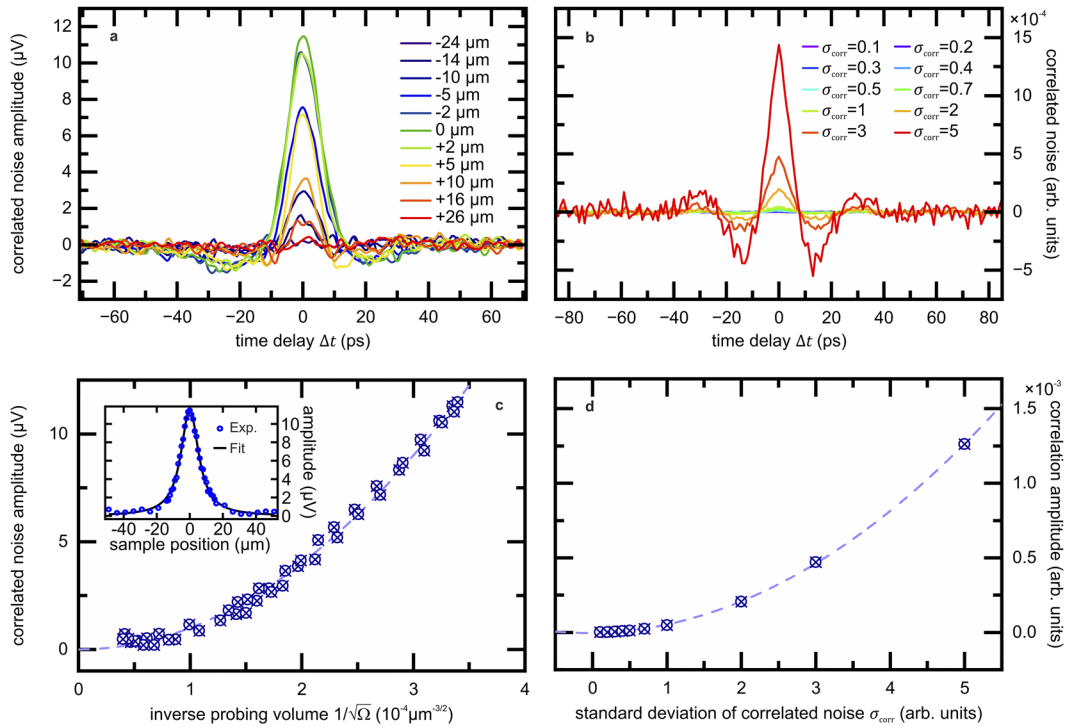


FIG. 5. Dependence of the correlated noise on the pulse-to-pulse fluctuation amplitude. (a) The experimental data showing the correlated noise waveforms for different longitudinal sample positions z relative to the laser focus. (b) The simulated noise correlation waveforms (see the supplementary material). (c) Correlated noise amplitude for time delay $\Delta t = 0$ as a function of the square-root of the inverse probing volume $\frac{1}{\sqrt{\Omega}}$. The correlated noise clearly follows a parabolic trend as indicated by the $\frac{A}{\Omega}$ fit (purple dashed line) in accord with Ref. 15. Here, A is a fitting constant. The z -dependent illuminated volume $\Omega(z)$ is calculated by fitting the amplitude- z -position dependence (inset) with the fitting function $\frac{1}{\Omega(z)} = \frac{1}{\pi w_0^2 \left[d + \frac{d^3/12 + dz^2}{z_R^2} \right]}$ (Ref. 15). Here, d is the sample thickness and z_R is the Rayleigh length. (d) Calculated correlation amplitude for $\Delta t = 0$ as a function of the standard deviation of the correlated noise σ_{corr} . The dashed line is the result of quadratic fit of the form $A \cdot \sigma_{\text{corr}}^2$, where A is a fitting constant.

the graph is fitted excellently by a quadratic function as expected from Eq. (16).

In contrast, experimentally controlling the fluctuation amplitude is not a trivial task. For this purpose, we exploit the fact that the FemNoC signal is strongly dependent on the optical probing volume $\Omega(z) = \pi w_0^2 \left[d + \frac{d^3/12 + dz^2}{z_R^2} \right]$ on the sample.¹⁵ Here, w_0 is the laser spot size in the focus, d is the sample thickness, and z_R is the Rayleigh length. By enlarging the probe spot size, spatially incoherent magnon fluctuations are smeared out, and thus, the polarization noise decreases. Specifically, polarization noise of a single probing pulse train is expected to decrease with the square-root of the inverse probing volume $\frac{1}{\sqrt{\Omega}}$.⁶ Consequently, the correlation waveform is expected to scale as $\frac{1}{\Omega}$.¹⁵ We assess this aspect by studying the correlation waveform for varying longitudinal sample positions z relative to the laser focus. The observed waveforms are shown in Fig. 5(a). The corresponding correlation amplitude at $\Delta t = 0$ ps is plotted in Fig. 5(c) as a function of $\frac{1}{\sqrt{\Omega}}$, as well as z -position. The measured correlated noise amplitude drastically changes for longitudinal sample positions relative to the laser focus, and ~ 20 μm

away from the confocal position, the magnon noise is no longer visible. Its amplitude clearly follows the expected $\frac{1}{\Omega}$ dependence. Note that the aforementioned scaling is expected from uncorrelated spin systems, such as paramagnets.⁶ In correlated spin systems, such as ferro- or antiferromagnets, this scaling hints at the existence of mutually incoherent oscillators with spatial extents smaller than the probe spot size.¹⁵ However, a detailed physical picture for this characteristic length scale, and its impact on the volume scaling when the probe spot size comes into the same order of magnitude, remains an open question. Nevertheless, this observation suggests that minimizing the probing volume is crucial for enhancing the signal strength in FemNoC experiments. Tight focusing optics and careful adjustment of the confocal plane with respect to the sample position are warranted.

C. Time constant dependence: TC

We now focus on another critical parameter that influences the FemNoC signals, which is the time constant of the subharmonic demodulation TC . The measured correlation waveforms for various

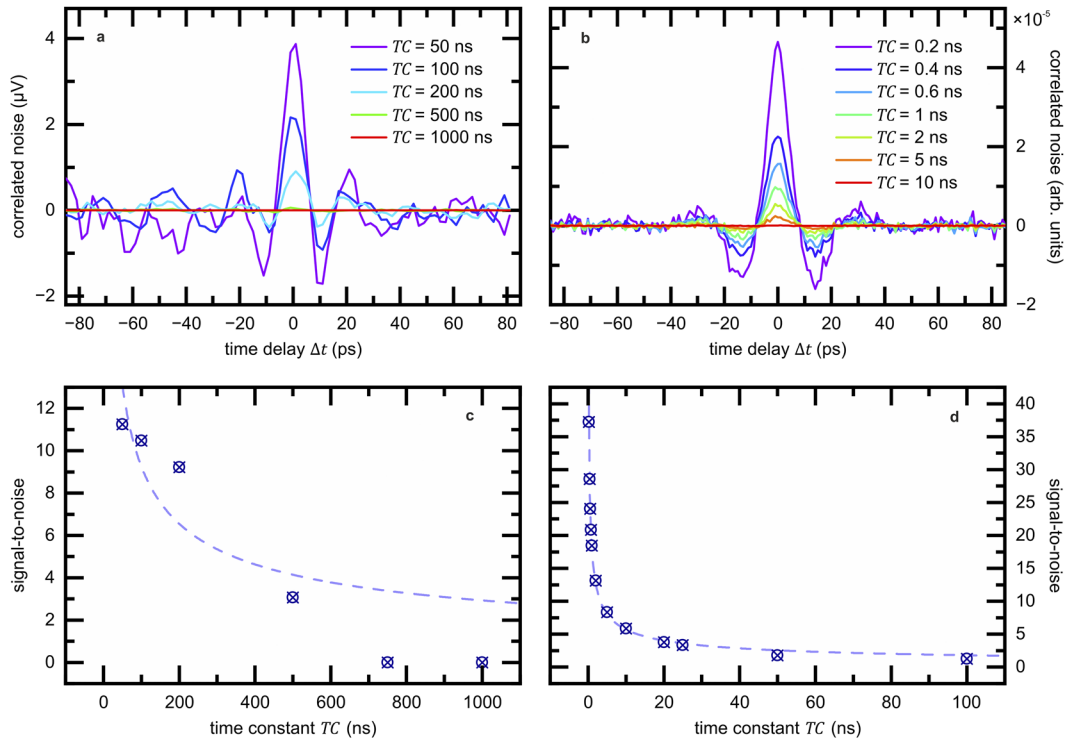


FIG. 6. Correlated noise amplitude and signal-to-noise ratio as a function of the subharmonic demodulation time constant TC . (a) The experimental data are recorded using the measurement parameters specified in the text, while the calculated waveforms (b) are the result of the simulation (see the [supplementary material](#)). (c) and (d) Calculated signal-to-noise ratio of experiment and simulation, respectively. The dashed line is the result of an inverse square-root fit of the form $\frac{A}{\sqrt{TC}}$, where A is a fitting constant.

TC are plotted in Fig. 6(a). For increasing TC , the absolute amplitude of the waveforms rapidly decreases. This behavior is in good agreement with Eq. (13) and is interpreted as the preaveraging of the pulse-to-pulse fluctuation amplitude. At the same, it is interesting to note that the signal-to-noise ratio also decreases at larger TC . This observation is understood by considering the statistics of the pulse-to-pulse fluctuations: Due to their stochastic nature, averaging reduces their standard deviation as $\frac{1}{\sqrt{TC}}$. Because the fluctuation amplitude enters the correlation function once for each channel, the total amplitude dependence of the correlation output is $CCR(0) \propto \frac{1}{TC}$. In contrast, the root-mean-square (RMS) of the correlation function scales with $\sim \frac{1}{\sqrt{TC}}$, whereby the signal-to-noise ratio is $SNR = \frac{CCR(0)}{RMS} \propto \frac{1}{\sqrt{TC}}$ as observed in the experimental data [Fig. 6(c)]. The simulation reproduces well the trend of the experiment, clearly following a $\frac{1}{\sqrt{TC}}$ shape [Figs. 6(b), 6(d)]. This result suggests that indeed preaveraging of the fluctuation amplitude for large time constants is the reason for a decrease in signal-to-noise. Consequently, the time constants of the subharmonic demodulation process ideally need to be chosen to be $TC = 2T_{rep}$ to include the direct and unaveraged pulse-to-pulse fluctuation, which then serves as the input of the correlation analysis. Note that, for large time constants, the simulation slightly differs from the inverse square-root fit. This finding

is attributed to the calculation procedure of the SNR (see S1 of the [supplementary material](#)).

D. Time constant dependence: TC_{AU}

Finally, we investigate the influence of the time constant TC_{AU} on the FemNoC signal. TC_{AU} determines the time window over which the product of the subharmonic outputs is averaged. The correlated noise waveform measured for different time constants TC_{AU} of the lock-in's arithmetic unit is plotted in Fig. 7(a). For increasing TC_{AU} , the waveforms become less noisy, in accord with Eq. (16), which can be interpreted to result from additional averaging of the pulse-to-pulse fluctuation correlation $\left\langle \langle \Delta V_{1,s_{2n}} \rangle_{\frac{n_{max}}{2}} \langle \Delta V_{2,s_{2n}} \rangle_{\frac{n_{max}}{2}} \right\rangle_{TC_{AU}}$ between the two channels. The signal-to-noise as a function of TC_{AU} is shown in Fig. 7(c). The graph clearly follows a square-root trend, as expected from the averaging behavior. The simulated autocorrelation waveforms in Fig. 7(b) progressively become smoother by increasing the number of averaged pulse pairs, comparable to their experimental counterparts. The simulated signal-to-noise ratio [Fig. 7(d)] also follows a distinct square-root trend. This finding shows that $TC_{AU} \rightarrow \infty$ is beneficial for the correlation-based spin noise spectroscopy measurements. Nevertheless, large TC_{AU} come with the trade-off of

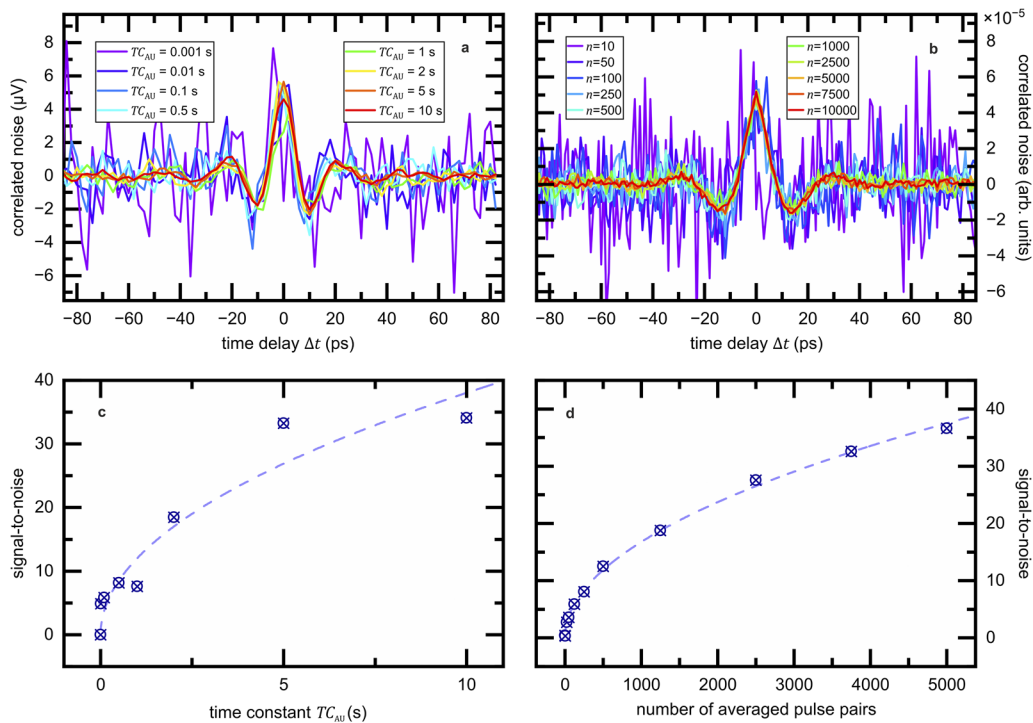


FIG. 7. Correlated noise amplitude and signal-to-noise ratio as a function of the arithmetic unit's time constant TC_{AU} . (a) The experimental data are recorded using the measurement parameters specified in the text, while the calculated waveforms (b) are the result of the simulation (see the [supplementary material](#)). (c) and (d) Calculated signal-to-noise ratio of experiment and simulation, respectively. The dashed line is the result of square-root fit of the form $A\sqrt{TC_{AU}}$, where A is a fitting constant.

an increased measurement time and eventual drifts need to be considered for selecting an optimum solution in any experimental implementation. Note that the amount of the averaged pulse pairs, and with that the signal-to-noise, can also be increased using higher repetition rate lasers. At the limit of ultra-high repetition rates, however, caution needs to be taken because when the interpulse distance of the probe pulse trains becomes comparable to the coherence time of the investigated dynamics, the measured correlation function may be distorted.

IV. CONCLUSION

In summary, we present a thorough technical description of subharmonic demodulation-based femtosecond noise correlation spectroscopy (FemNoC). We introduce a mathematical representation of the FemNoC data acquisition scheme and determine parameters that critically influence the FemNoC measurements. The impact of the parameter values on the FemNoC signals is then verified by simulations based on a mathematical formalism and comparison with experimental datasets. Finally, we identify a set of ideal measurement parameters that promise to maximize the signal-to-noise ratio of FemNoC experiments: First, the relative phases $\theta_{1,2}$ of the reference function need to be carefully adjusted to enhance the output of the subharmonic demodulation. Further-

more, the absolute fluctuation amplitude is to be maximized, e.g., by carefully placing the experimental sample into a strongly focused laser beam or choosing an appropriate pair of probe wavelengths. Finally, to avoid preaveraging of the subharmonic component, the time constant of the demodulation TC should correspond to twice the repetition rate of the laser $TC = 2T_{rep}$, whereas the time constant of the arithmetic unit should be as long as possible and limited only by practical constraints to increase the number of correlation samples.

SUPPLEMENTARY MATERIAL

The following sections of the [supplementary material](#) (S) accompany this article: “S1: Simulation of correlated spin noise measurement” gives detailed information about the simulations performed in this work. “S2: Simulation dependence on the background noise model” analyzes how the signal-to-noise of the simulated data changes depending on the used background noise model. “S3: General derivation of mathematical formalism” discusses subharmonic demodulation beyond the first subharmonic frequency. Here, a mathematical formalism for general subharmonic frequencies is derived and the signal-to-noise of the simulated data as a function of subharmonic frequency is investigated.

ACKNOWLEDGMENTS

This research was supported by the Overseas Research Fellowship of the Japan Society for the Promotion of Science (JSPS), Zukunftskolleg Fellowship from the University of Konstanz, JSPS KAKENHI (Grant Nos. JP21K14550, JP23K17748, and JP24H00317), and the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation)—Project No. 425217212-SFB 1432.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

M. A. Weiss: Data curation (lead); Formal analysis (lead); Investigation (lead); Methodology (lead); Project administration (lead); Software (lead); Writing – original draft (lead). **F. S. Herbst:** Formal analysis (supporting); Writing – review & editing (supporting). **S. Eggert:** Resources (equal). **M. Nakajima:** Resources (equal). **A. Leitenstorfer:** Conceptualization (equal); Resources (equal); Writing – review & editing (supporting). **S. T. B. Goennenwein:** Conceptualization (equal); Resources (equal); Supervision (equal); Writing – review & editing (supporting). **T. Kurihara:** Conceptualization (equal); Investigation (supporting); Project administration (equal); Supervision (equal); Writing – original draft (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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