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Spin-Correlated Luminescence of Mechanically Generated Radical Species

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ABSTRACT

A tetraarylsuccinonitrile (TASN) derivative ($\mathbf{1}_2$) substituted with bulky triarylmethane groups dissociated into luminescent cyanodiarylmethyl radical **1** upon thermal or mechanical stimulation. The average dissociation yield after mechanochemical treatment was 2.6%, among the highest reported for small-molecule TASN compounds. Ground $\mathbf{1}_2$ showed a magnetic field effect on luminescence, the first such observation from a radical emitter other than triarylmethyl radicals. This study demonstrates the universality of spin-correlated luminescence among luminescent radicals.

Luminescent organic radicals exhibit unusual photophysical properties and functions [1–4], such as efficient carrier-to-photon conversion [5, 6] and spin-independent energy harvesting [7, 8], depending on their spin multiplicity. The luminescence of assemblies of radicals with multiple accessible spin states can be magnetically modulated; that is, magnetoluminescence (MagLum) [9–22]. The strong correlation between their luminescence and spin states makes MagLum-active molecular systems promising for spintronic, optoelectronic, and quantum applications [11, 23]. Reported radical assemblies exhibiting MagLum can be classified into three groups: monoradicals assembled in a closed-shell host matrix (Figure 1a) [9, 10], organic molecules bearing multiple radical units (e.g., diradicals and triradicals; Figure 1b) [13, 14, 19–21], and radical-based coordination polymers [16]. However, prior explorations have been limited to systems with emitters comprising stable polychlorinated triarylmethyl (TArM) radical units. Clarifying the generality of MagLum among luminescent radicals other than TArM is crucial to expanding the range of molecular systems exhibiting spin-correlated luminescence and to fully understanding the effects of molecular structure and aggregation mode on the MagLum of radicals.

Generation of metastable radicals through controllable stimulation is a useful method to obtain luminescent radical species from readily accessible, chemically stable closed-shell precursors [4, 24]. In contrast to the TArM radicals, such in situ-generated radicals offer diverse molecular skeletons and a wide range of emission colors. Cyanodiarylmethyl (CDAM) radicals are generated from tetraarylsuccinonitrile (TASN) derivatives through homolytic cleavage of the central C–C bond by thermal or mechanical stimuli [25, 26]. The luminescence of CDAM radicals occurs over a broad wavelength range depending on their substituents [27]. As two CDAM radicals are formed from one TASN molecule, we envisage that this class of radicals could provide a new platform for achieving MagLum via in situ mechanical generation and subsequent assembly. However, magnetic and

Dedicated to Professor Tatsuya Nabeshima on the occasion of his 70th birthday.

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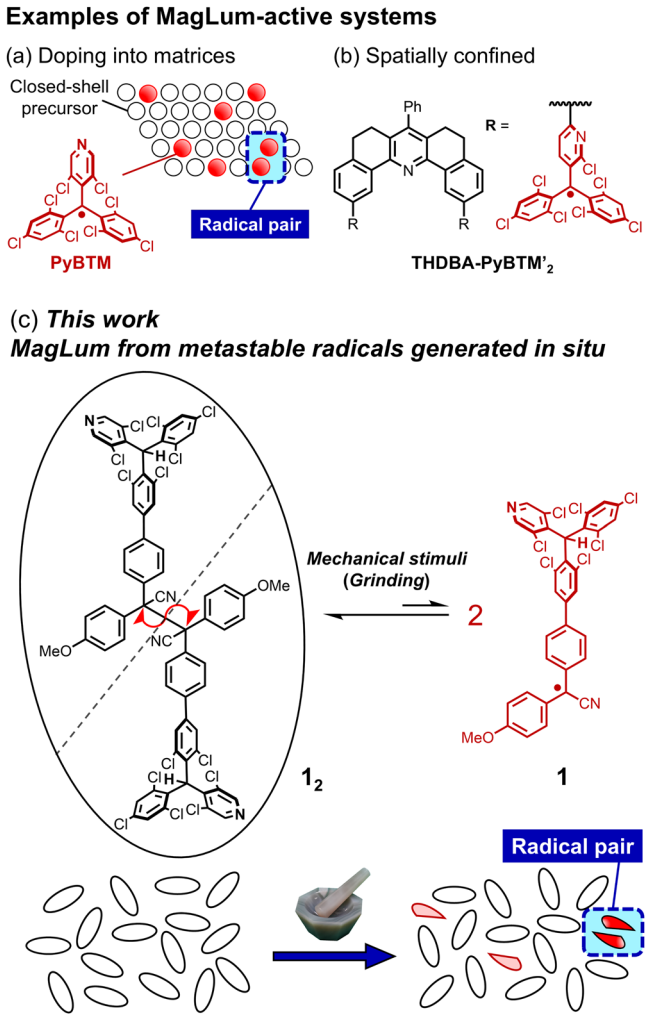


FIGURE 1 | Schematic illustration of methodologies to achieve MagLum from luminescent radicals: (a) doping into matrices, (b) spatial confinement (previous studies), and (c) in situ generation by mechanical stimuli. The chemical structures of TASN precursor $\mathbf{1}_2$ and luminescent CDAM radical **1** investigated here are shown in (c).

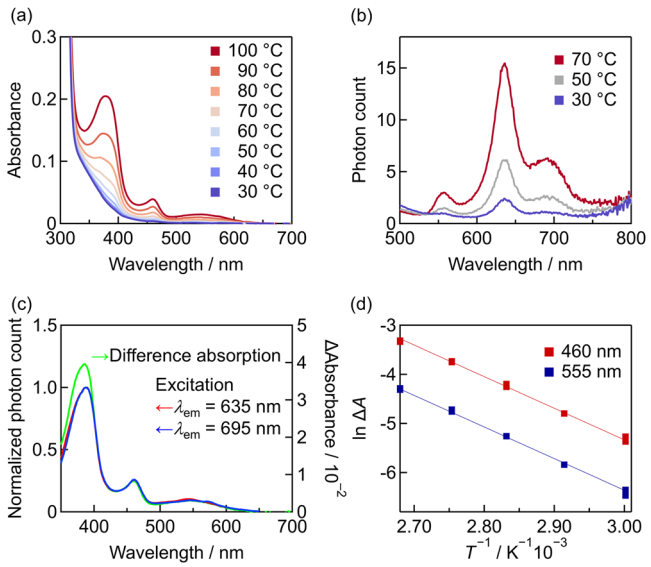


FIGURE 2 | (a) Absorption and (b) emission (λ_{ex} = 380 nm) spectra of $\mathbf{1}_2$ (0.20 mM) in toluene at various temperatures. (c) Normalized excitation (red, λ_{em} = 635 nm; blue, λ_{em} = 695 nm) and difference absorption (green, with respect to the spectrum at 30°C) spectra of $\mathbf{1}_2$ (0.20 mM) in toluene at 70°C. (d) van't Hoff plots for the dissociation of $\mathbf{1}_2$ based on the difference absorption (ΔAbsorbance, Δ*A*) at 460 nm (red) and 555 nm (blue).

electronic interactions among such mechanically generated radicals and the resulting spin-correlated luminescence have not previously been elucidated.

Here we investigated the thermal and mechanochemical generation of luminescent CDAM radical **1** from its TASN precursor $\mathbf{1}_2$ and examined its intermolecular interactions by means of electron spin resonance (ESR) spectroscopy and MagLum investigations (Figure 1c). Our first observation of MagLum from a mechanically generated luminescent radical not related to the TArM demonstrates that the generation by external stimuli of luminescent radicals offers a simple and effective strategy for realizing spin-correlated photofunctionality.

The target compound $\mathbf{1}_2$ was synthesized by the Suzuki-Miyaura cross coupling [28] of a dibromo-substituted TASN derivative ($\mathbf{2}_2$) and a boronic ester derivative of (3,5-dichloro-4-pyridyl)bis(2,4,6-trichlorophenyl)methane, ***αH*-PyBTM** (***αH*-PyBTM-B(Epin)** [29]). The introduction of the bulky ***αH*-PyBTM** unit enabled the exploration of structural effects on bond dissociation in the TASN unit. The presence of this unit might also facilitate derivatization in future studies. Radical **1** was isolated as a diastereomeric mixture and characterized by ^{1}H and ^{13}C NMR, ESI/APCI dual ion source (DUIS) MS, and elemental analysis.

To investigate the thermoresponsive properties of $\mathbf{1}_2$ in solution, its absorption and photoluminescence spectra in toluene were measured at various temperatures (Figure 2a,b). At 30°C, it showed strong absorption below 350 nm with a weak tail at 350–500 nm. With increasing temperature, new absorption bands appeared around 380, 460, and 545 nm, which were attributed to dissociated radical species; that is, **1** (Figure 2a). Recovery of the initial spectrum by cooling suggests that heating established a dimer–monomer equilibrium without any detectable decomposition reactions. The emission spectrum of the solution also showed clear temperature dependence. At 30°C, weak emission bands were observed at 557, 635, and 695 nm upon photoexcitation at 380 nm. The intensity of these emissions substantially increased with increasing temperature (Figure 2b), but the original spectrum was recovered by cooling. We note that the complete reversibility of the dimer–monomer equilibrium eliminates the possibility of the conversion of **1** into a permanent TArM radical via hydrogen atom transfer from the ***αH*-PyBTM** unit to the CDAM radical unit.

Excitation spectra at 70°C indicate the origin of the photoluminescence (Figures 2c, S4, and S7). The excitation spectra at λ_{em} = 635 and 695 nm are nearly identical and in good agreement with the difference absorption spectrum at 70°C with respect to that at 30°C, within the 400–700 nm range. These results indicate that the new absorption bands at 460 and 555 nm and the emission bands at 635 and 695 nm originated from the same chemical species, namely, **1**. On the other hand, the excitation

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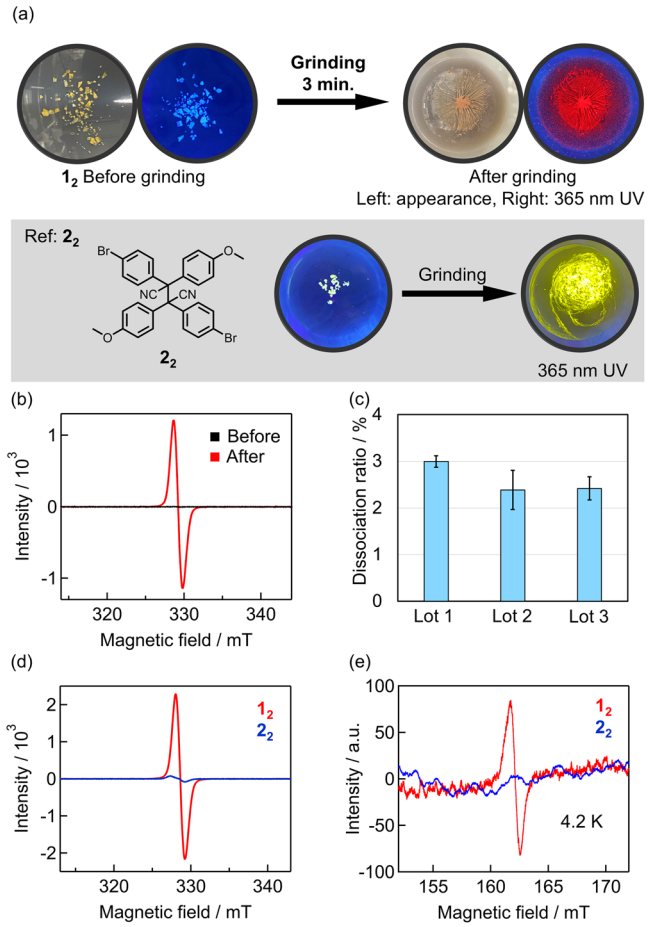


FIGURE 3 | (a) Photographs of $\mathbf{1}_2$ and $\mathbf{2}_2$ before and after grinding. (b) ESR spectra of $\mathbf{1}_2$ at room temperature before and after grinding. (c) Inter-lot reproducibility of the dissociation ratio of ground $\mathbf{1}_2$. (d,e) ESR spectra of ground $\mathbf{1}_2$ and $\mathbf{2}_2$ in (d) the $\Delta m_s = \pm 1$ region at room temperature and (e) the $\Delta m_s = \pm 2$ region at 4.2 K.

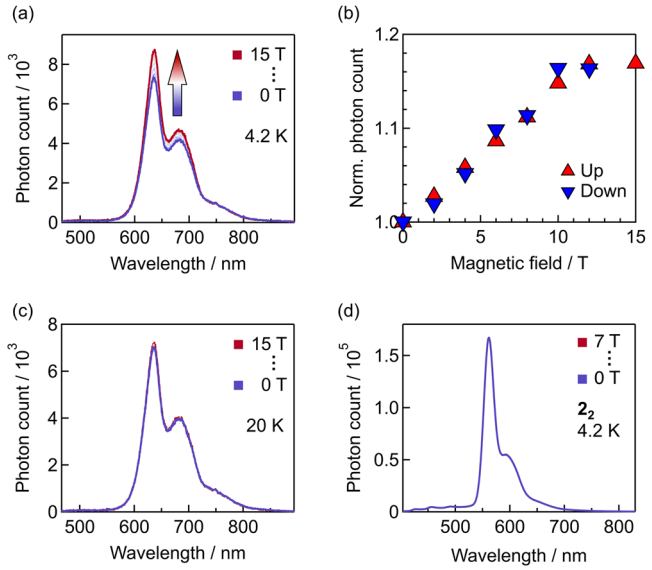


FIGURE 4 | (a, c) Emission spectra of ground $\mathbf{1}_2$ at (a) 4.2 and (c) 20 K. (λ_{ex} = 375 nm) (b) Magnetic field dependence of the ratio of emission photon counts at 636.1 nm at 0 T. (d) Emission spectra of ground $\mathbf{2}_2$ at 4.2 K under various magnetic fields (λ_{ex} = 375 nm).

spectrum at λ_{em} = 557 nm does not match those at λ_{em} = 635 and 695 nm, indicating the presence of a different thermoresponsive luminophore. This compound was assigned as **2**, which probably came from trace contamination of the residual precursor $\mathbf{2}_2$ in $\mathbf{1}_2$ (Figures S4–S7).

The obtained temperature-dependent absorption data allowed us to estimate the bond dissociation enthalpy of the central C–C bond in $\mathbf{1}_2$ as the enthalpy change (ΔH_{UV}) for the dissociation equilibrium [25]. The van't Hoff plot of the absorption difference at 460 and 555 nm gives a dissociation enthalpy of 107 ± 2 kJ mol^{-1} (Figure 2d), comparable to that of tetraphenylsuccinonitrile (**TPSN**, 109.6 kJ mol^{-1}) [25].

We then investigated the mechanochemical generation of radical species by grinding. The precursor $\mathbf{1}_2$ existed as a yellow solid that exhibited no discernible ESR signal before grinding. After grinding, however, the color of the solid turned light brown (Figure 3a), and a distinct ESR signal was detected (g = 2.003; Figure 3b). The ground solid also exhibited red photoluminescence upon irradiation with 375 nm ultraviolet light. These results clearly indicate the mechanochemical generation of radical **1**. Similar mechanofluorochromic behavior was observed from the ground solid of $\mathbf{2}_2$, which displayed yellow emission (Figure 3a).

ESR measurements using an external standard estimated the average yield of mechanochemical dissociation to be 2.6% (Figure 3c). This value is several orders of magnitude greater than those for **TPSN** (2.93×10^{-3}%) and $\mathbf{2}_2$ (3.72×10^{-2}%, Figure 3d) and is approximately 10 times that for **TASN-urea** (0.48%) [30], which was the highest yield among small-molecule TASN systems. Otsuka and coworkers have reported the increased dissociation yield of mechanochemical radical generation by connecting polymer chains to the TASN units [24, 26, 31, 32]. The dissociation yield of $\mathbf{1}_2$ was even comparable to that of macromolecular systems such as a polystyrene-decorated TASN derivative (5.08%) [26]. Although the exact reasons for the exceptionally high dissociation yield of $\mathbf{1}_2$ among TASN derivatives remain unclear, it is possible that the bulky ***αH*-PyBTM** substituents facilitate the efficient transfer of mechanical stress to the central C–C bond of the TASN core during grinding. Further explorations of the enhanced mechano-reactivity of TASN derivatives are underway.

To investigate whether effective magnetic interactions exist between the radicals generated in the solid state, ESR measurements at 4.2 K were performed in the $\Delta m_s = \pm 2$ region for ground samples of $\mathbf{1}_2$, $\mathbf{2}_2$, and **TPSN**. A half-field signal was clearly observed from ground $\mathbf{1}_2$, but not from $\mathbf{2}_2$ or **TPSN** (Figures 3e, S8, and S9), suggesting two important facts. First, at a low dissociation ratio (i.e., for $\mathbf{2}_2$ and **TPSN**), the mechanochemically generated radicals are isolated to such an extent that intermolecular magnetic interactions can be neglected. Second, the molecules of **1** in $\mathbf{1}_2$ generated by grinding have effective intermolecular magnetic interactions with each other, probably owing to the high radical concentration.

The presence of effective magnetic interactions between the radicals in ground $\mathbf{1}_2$ encouraged us to investigate its spin–

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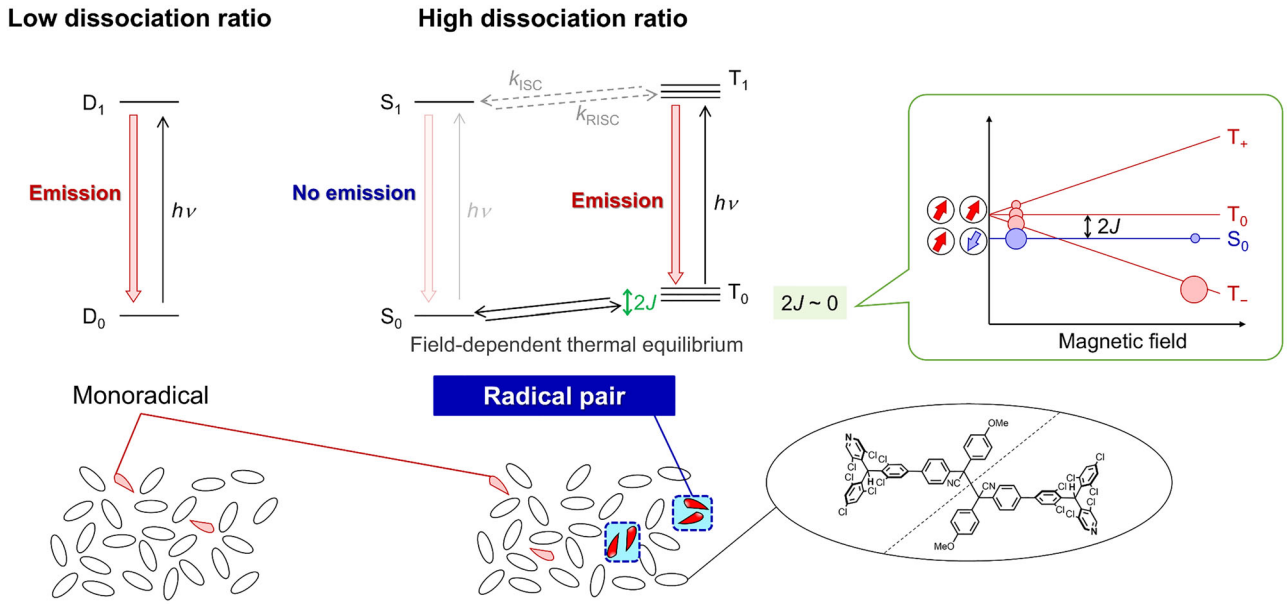


FIGURE 5 | Plausible mechanisms of the spin-correlated luminescence of the stimuli-generated radicals.

luminescence-correlated properties. Figure 4a shows the emission spectra of ground $\mathbf{1}_2$ under a magnetic field at 4.2 K. The intensity of the emission peak at 636 nm increased by 17% in a magnetic field of 15 T. This marks the first observation of MagLum from a stimulus-generated luminescent radical and, notably, from an emissive organic radical not related to TArM systems. The magnetic field effect (MFE) of ground $\mathbf{1}_2$ saturated at ~12 T at 4.2 K (Figure 4b). At 20 K, the effect was negligible (Figure 4c). Equivalent investigations of the ground solids of $\mathbf{2}_2$ and **TPSN** found no MFE, even at 4.2 K (Figures 4d and S10).

To elucidate the mechanism of MagLum in the present system, the emission decay of the ground solid was measured at 4.2 K under various applied magnetic fields (Figures S11–S16). The decay curves obtained at λ_{em} = 635 nm reveal that the lifetime and amplitude of the luminescence increased by 6.4% and 14%, respectively, at 15 T. Hence, the increase in emission intensity was due primarily to changes in amplitude, similar to the behavior of MagLum in assemblies of TArM radicals, thus indicating their similar MagLum mechanisms.

The dissociation yields of ground $\mathbf{1}_2$ and $\mathbf{2}_2$ were 2.6% and 0.037%, respectively, which correspond to luminescent radicals being present at approximately 5.2 wt% and 0.074 wt% in the solids. The lack of MagLum in ground $\mathbf{2}_2$ suggests that the radicals generated by mechanical C–C bond cleavage are spatially separated and behave as isolated radicals in terms of photophysics. On the other hand, the presence of MagLum in ground $\mathbf{1}_2$ suggests partial assembly of **1** in ground $\mathbf{1}_2$. These findings are consistent with the ESR measurements discussed above. Previous studies have supported the relationship between radical concentration and degree of assembly: crystals of the closed-shell matrix ***αH*-PyBTM** doped with 4 wt% luminescent radical **PyBTM** showed MagLum, whereas a corresponding 0.1 wt% doped sample did not [9, 15]. In addition, a sample of ground $\mathbf{1}_2$ aged for 17 days had its dissociation ratio decrease to 1.1% and showed negligible MagLum (Figure S21). Overall, ground $\mathbf{1}_2$ appeared to exhibit MagLum owing to its high dissociation yield.

The mechanism of the observed MagLum can be explained by the radical pair model proposed in our previous studies (Figure 5) [9, 19, 21]. In the ground state, magnetically interacting radical pairs can exist in either singlet (S) or triplet (T_+, T_0, T_-) spin states. As the relative populations of these states are governed by a Boltzmann distribution, Zeeman splitting under a magnetic field increases the population of the triplet (T_-) state and decreases the population of the S state. These population changes in the ground state eventually result in enhanced photoluminescence from the bright triplet states. According to this mechanism, photoluminescence from spin-S radical pairs would decrease under a magnetic field. However, such emission decline was not observed in the measured range. One possible explanation is that the spin-S radical pairs are nearly nonemissive. It is also possible that the reduced emission from the spin-S radical pairs and the enhanced emission from the spin-T radical pairs counterbalance each other, giving the appearance of no (or positive) net change.

The MagLum behavior of ground $\mathbf{1}_2$ saturated around 12 T at 4.2 K, indicating that the spin state of the radical pairs was fully populated to T_- under these conditions. From this saturation behavior, magnetic exchange interactions (J) between neighboring radicals in ground $\mathbf{1}_2$ were estimated to be $|J/k_B| < 5$ K. We note that the diradical **THDBA-PyBTM′**$_2$ (Figure 1b), which exhibits a small exchange interaction ($2J/k_B = 8.3 \pm 11.3$ K), also shows saturation of its MFE at approximately 12 T [19]. However, the increase in the emission intensity of ground $\mathbf{1}_2$ (17%) at 15 T is slightly smaller than that observed for **THDBA-PyBTM'**$_2$ (22%). This discrepancy likely originates from the coexistence of non-aggregated (i.e., isolated) **1** in ground $\mathbf{1}_2$, because the emission from such magnetic field-insensitive species offsets the effect of magnetic-field-dependent behavior. The coexistence of isolated **1** can also explain the effect of magnetic field on emission lifetime, if we assume that isolated and assembled **1** have similar but distinct lifetimes. In such a scenario, the application of a magnetic field would increase the proportion of the longer-lived radical pairs, leading to an apparent extension of lifetime [21].

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In conclusion, a novel mechanoresponsive TASN derivative $\mathbf{1}_2$ was synthesized. Its average mechanochemical dissociation yield was 2.6%, which is among the highest reported small-molecule TASN systems. This exceptionally high yield allowed the radicals (**1**) generated in the ground solid to partially assemble and thereby magnetically interact with each other to show distinct MagLum at 4.2 K. This is the first observed MagLum from a radical emitter not related to TArM radicals. This study demonstrates the universality of spin-correlated luminescence from radical assemblies, independent of their structure. Our findings also demonstrate that mechanochemical radical generation is a simple and highly effective method to realize MagLum-active systems and thus offers a notable contribution to the expansion of the chemical diversity of MagLum species for future applications. Moreover, integrating stable radical skeletons with mechanochemical radical generation may open new avenues for developing radical-based, stimuli-responsive materials with spin-correlated luminescence functionality. Such study based on the present molecular design is going on in our group.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

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

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.

The authors have cited additional references within the Supporting Information [33].

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