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Signatures of an Enhanced Valence-Bond-Solid Transition in κ -(BEST)₂Cu₂(CN)₃ probed by Nuclear Magnetic and Electron Paramagnetic Resonance

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We investigated the low-temperature magnetic properties of the organic Mott insulator κ -(BEST)₂Cu₂(CN)₃ using ¹³C nuclear magnetic resonance (NMR) and electron paramagnetic resonance (EPR), where BEST denotes bis(ethylenediseleno)tetrathiafulvalene. The NMR Knight shift and EPR integrated intensity together revealed a pronounced drop in spin susceptibility below $T^* \approx 10$ K, accompanied by a broad hump in the nuclear spin-lattice relaxation rate and the growth of a Lorentzian EPR component. These features closely mirror the “6 K anomaly” previously reported for the isostructural κ -(ET)₂Cu₂(CN)₃, where ET is bis(ethylenedithio)tetrathiafulvalene. Although the two salts share nearly identical electronic correlations and qualitatively exhibit the same magnetic behavior, the combined NMR and EPR results indicate that substituting the heavier BEST for ET enhances the transition temperature to the spin-singlet state. We propose that the increase in molecular mass may induce lattice softening, thereby stabilizing the lattice distortion required for valence-bond-solid order. Our results suggest that lattice dynamics play an important role in the valence-bond-solid transition in κ -type organics.

I. INTRODUCTION

A quantum spin liquid (QSL) is a ground state in which antiferromagnetic (AF) exchange interactions on a frustrated lattice, such as a triangular lattice, suppress long-range magnetic order even at 0 K [1–3]. In a perfectly isotropic triangular lattice, even a slight distortion may relieve frustration and stabilize competing phases, including the AF and valence-bond-solid (VBS) phases [4–8]. Because experiments cannot reach 0 K, direct evidence for a QSL is effectively unattainable. Consequently, identifying the electronic phases that emerge near the isotropic limit provides crucial indirect insights into the physics of QSL. Quasi-two-dimensional organic Mott insulators are ideal for this purpose: spin-1/2 molecular dimers often form two-dimensional triangular networks, and many QSL candidates have been reported [9–16]. In addition, subtle changes in the anisotropy of the triangular lattice introduced by chemical modification [17, 18] are known to stabilize the AF [19–22] and VBS states [23–25].

Among these organic systems, κ -(BEDT-TTF)₂Cu₂(CN)₃ (hereafter κ -ET-CN) [26, 27] was the first material proposed to host a QSL, where BEDT-TTF is bis(ethylenedithio)-tetrathiafulvalene, also referred to as ET (Fig. 1(a)). Although it possesses a nearly isotropic triangular lattice with an interdimer exchange interaction of $J \approx 250$ K, no long-range magnetic order has been detected down to 32 mK using ¹H-nuclear magnetic resonance (NMR) measurements

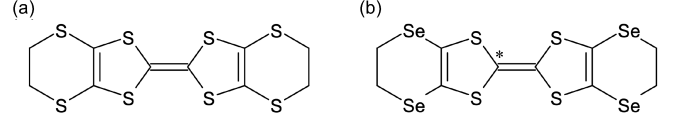


FIG. 1. Molecular structures of (a) BEDT-TTF (ET for short) and (b) BEDSe-TTF (BEST). An asterisk marks the ¹³C-enriched site.

[9]. κ -ET-CN also exhibits the so-called “6 K anomaly.” Thermal expansion [28], nuclear quadrupole resonance (NQR) [29], and inelastic neutron scattering [30] measurements indicate a structural phase transition at $T^* = 6$ K, although diffraction studies have not yielded definitive evidence for such a transition. In addition, ¹³C-NMR [31] and electron paramagnetic resonance (EPR) [32] measurements reveal a pronounced drop in spin susceptibility below T^* , suggesting the presence of a spin gap. Taken together, these observations support the scenario that κ -ET-CN undergoes a VBS transition at 6 K [32].

Understanding the origin of this anomaly is essential to elucidate how AF, QSL, and VBS phases compete with frustrated Mott insulators. The electronic properties of κ -(ET)₂X (X represents a monovalent anion) salts are typically described using a universal phase diagram parameterized by the ratio of on-site Coulomb repulsion and bandwidth (U/W) [33], which successfully accounts for the Mott transition [34, 35] and proximity of AF and superconductivity [36]. However, this phase diagram does not consider lattice contributions, even though the spin and lattice are coupled in the VBS transition [24]. Conventionally, κ -(ET)₂X salts are explored through anion X substitution [18]; however, this approach alters both the U/W ratio and crystal structure, making it difficult

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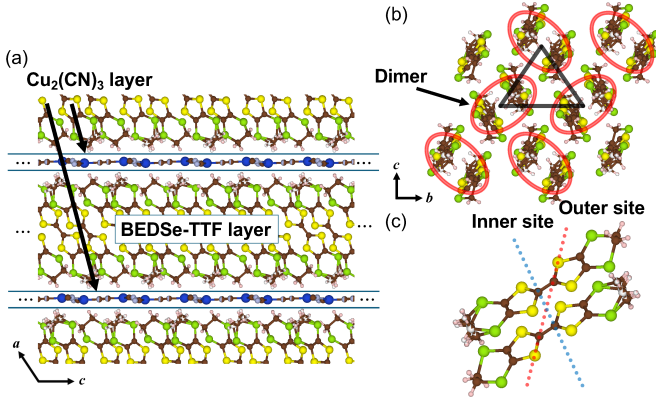


FIG. 2. (a) Crystal structure of κ -BEST-CN viewed along the b axis. (b) κ -type molecular arrangement in the donor layer. The circles and triangle represent dimers formed by face-to-face molecules and their triangular arrangement, respectively. (c) Two crystallographically inequivalent ^{13}C -sites (inner and outer) in a dimer.

to isolate the lattice contribution across different materials.

To isolate the contribution of the lattice degree of freedom, we focus on the isostructural analog κ -(BEDSe-TTF) $_2\text{Cu}_2(\text{CN})_3$ (hereafter κ -BEST-CN) [37], where BEDSe-TTF is bis(ethylenediseleno)tetrathiafulvalene (BEST) (Fig. 1(b)). This material has the same structure as the κ -ET-CN: anion and donor layers alternate along the a axis (Fig. 2(a)), and within the donor layer, the dimers of the BEST molecules adopt a κ -type arrangement that forms a triangular lattice (Fig. 2(b)) [37]. Resistivity measurement indicates that κ -BEST-CN resides in the same electronic phase as κ -ET-CN, and is positioned on the slightly pressurized side near the Mott boundary [37]. However, impurity moments are more pronounced at lower temperatures, contaminating bulk susceptibility measurements and obscuring whether κ -BEST-CN exhibits an anomaly analogous to the “6 K anomaly”. In this study, we performed ^{13}C -NMR and EPR measurements to determine whether κ -BEST-CN exhibits an analogous low-temperature anomaly, where the lattice is perturbed by the substitution of the BEST without significantly altering the crystal structure and electronic correlations. Direct comparison with κ -ET-CN allows us to sharpen the characterization of well-known 6 K anomaly.

II. EXPERIMENTAL

Single crystals of κ -BEST-CN were prepared following a procedure described in the literature [37]. For the ^{13}C -NMR experiments, single-site ^{13}C -enriched BEDSe-TTF molecules (with the labelled carbon indicated by an asterisk in Fig. 1(b)) were synthesized from substituted thioketones and naturally abundant ketones [38] to avoid the Pake-doublet problem [39].

NMR experiments were performed on seven aligned thin single crystals under a magnetic field of 6.08 T that was parallel to the a^* axis ($\mu_0 H \parallel a^*$). The NMR spectra were obtained through the fast Fourier transformation of the spin-echo signals. The typical $\pi/2$ pulse width was 3 μs . The NMR shifts were referenced to tetramethylsilane, and the shifts and linewidths were determined by fitting the spectra to two Lorentzian functions above 5 K and one Lorentzian function below that temperature. The nuclear spin-lattice relaxation time T_1 was measured using the saturation-recovery method. Above 5 K, T_1 was determined by fitting the magnetization recovery of each of the two separate peaks to a single exponential function. Below 5 K, where the two peaks overlap, T_1 was obtained by fitting the integrated magnetization $M(t)$ to the following composite function:

$$\frac{M_0 - M(t)}{M_0} = \frac{1}{2} \exp \left[- \left(\frac{t}{T_1} \right)^\beta \right] + \frac{1}{2} \exp \left[- \left(\frac{t}{\alpha T_1} \right)^\beta \right], \quad (1)$$

where M_0 is the equilibrium magnetization and β is the stretched exponent. We found that β is nearly unity within experimental uncertainty above 5 K and decreases upon cooling below this temperature. α is the ratio of the squared hyperfine coupling constants transverse to the quantization axis at the two sites. Because the hyperfine-coupling tensor for $2p_z$ spins is anisotropic, this transverse coupling is distinct from the longitudinal component that determines the Knight shift. From the high-temperature data where the two lines are resolved (above 6 K), we experimentally determined $\alpha \approx 2$ and fixed 2.0 in the low-temperature analysis.

EPR measurements were performed using a Bruker E500 X-band spectrometer equipped with Oxford ESR900 and ESR910 cryostats. Several single crystals were aligned such that the magnetic field was almost parallel to the a^* axis.

III. RESULTS AND DISCUSSION

A. ^{13}C -NMR

Figure 3 shows the NMR spectra of κ -BEST-CN (left) and κ -ET-CN (right) [31] at various temperatures. Because the inversion center of both salts is at the center of the dimer [26, 37], two distinct peaks arising from the crystallographically inequivalent ^{13}C site were expected [31, 40], as shown in Fig. 2(c). Indeed, the spectra of both salts consisted of two peaks corresponding to the inner and outer sites. In both salts, the linewidth shows little change above 30 K but increases significantly below this temperature. Nevertheless, no clear spectral splittings indicative of AF ordering were observed down to the lowest measured temperature. Below 10 K, the two peaks approach each other and merge into a single broad peak.

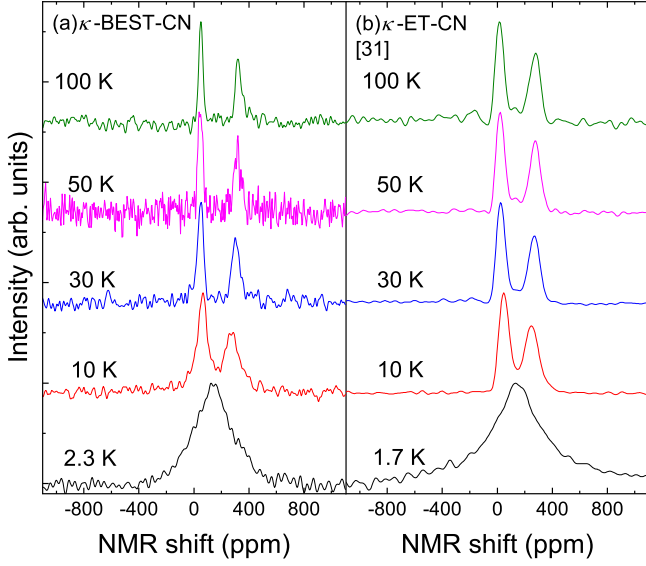


FIG. 3. ^{13}C -NMR spectra of κ -BEST-CN (left) and κ -ET-CN (right, Ref. [31]) at several temperatures with $\mu_0 H \parallel a^*$.

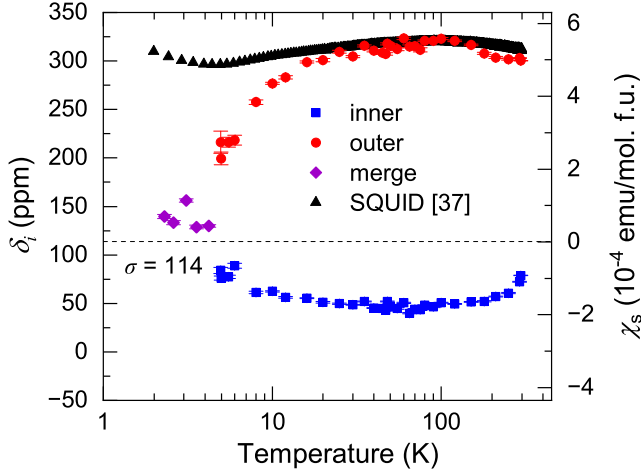


FIG. 4. Temperature dependence of NMR shifts for inner (squares) and outer (circles) sites and merge components (diamonds) in κ -BEST-CN (left axis). The spin susceptibility measured using a SQUID magnetometer (triangles, Ref. [37]) is plotted on the right axis.

Figure 4 shows the temperature dependence of the NMR shifts for the inner and outer sites of κ -BEST-CN together with a merged shift at low temperatures. For comparison, the bulk spin susceptibility χ_s measured by the SQUID magnetometer [37] is plotted on the right-hand axis. NMR shift δ_i (i = inner or outer sites) can be expressed as follows:

$$\delta_i = K_i + \sigma = A_i \chi_s + \sigma, \quad (2)$$

where K_i , A_i , and σ are the Knight shift, hyperfine-coupling constant, and chemical shift, respectively. Because the local fields from neighboring molecules are different for the two sites, K_i and A_i are site-dependent. By

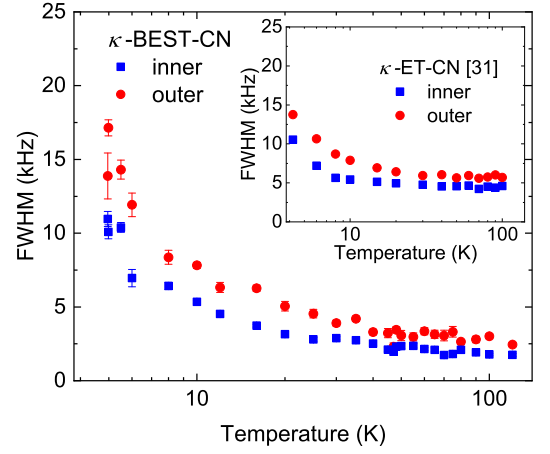


FIG. 5. Temperature dependence of the NMR linewidth (FWHM) in κ -BEST-CN. The inset shows the results for κ -ET-CN [31].

contrast, σ is expected to be identical for the inner and outer sites because they are chemically equivalent within the molecule.

Above 20 K, NMR shifts δ_i vary slightly and are proportional to the bulk spin susceptibility (Fig. 4). The inner and outer sites have negative and positive hyperfine coupling constants, respectively. Below 20 K, δ_{outer} decreases while δ_{inner} increases, indicating a decrease in the Knight shift. Despite the significant changes in δ_i at low temperatures, the bulk susceptibility shows a weak temperature dependence, which is most plausibly attributed to a Curie-like contribution from impurity spins that dominates the bulk measurement [37].

The NMR linewidth, defined by the full width at half maximum (FWHM), broadens for κ -BEST-CN below ~ 30 K and more significantly below 8 K (Fig. 5). The discrepancy between the bulk susceptibility and NMR shifts at low temperatures suggests that impurity spins contribute to the broadening of the FWHM. Low-temperature broadening, together with the convergence of the two NMR peaks, forces us to treat the spectrum as a single line below 5 K, making it difficult to accurately estimate the linewidth in this temperature range. For comparison, the inset of Fig. 5 presents the temperature dependence of the NMR linewidth for κ -ET-CN [31], also exhibiting the line broadening with decreasing temperature.

Here, we estimate σ and A_i to determine the contribution of the local spin susceptibility. Solving Eq. (2) for the inner and outer sites simultaneously yields [41, 42]

$$\delta_{\text{outer}} = \frac{A_{\text{outer}}}{A_{\text{inner}}} \delta_{\text{inner}} + \left(1 - \frac{A_{\text{outer}}}{A_{\text{inner}}}\right) \sigma. \quad (3)$$

Figure 6 is a plot of δ_{inner} versus δ_{outer} in the range of 7 – 60 K. According to Eq. (3), the slope and intercept of this plot yield σ and the ratio $A_{\text{outer}}/A_{\text{inner}}$. A least-squares fit, indicated by the dashed line in Fig. 6 yields

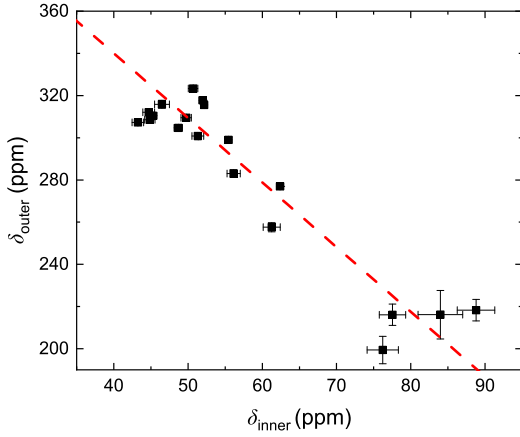


FIG. 6. $\delta_{\text{inner}} - \delta_{\text{outer}}$ plot of κ -BEST-CN. Red dashed line is the least-squares fit.

$A_{\text{outer}}/A_{\text{inner}} = -3.1(5)$ and $\sigma = 114(6)$ ppm. This σ value is almost the same as κ -ET-CN, 117 ppm, which was estimated from measurements in the same magnetic field orientation [31]. These nearly identical values are reasonable because the BEDSe-TTF molecule retains the same tetrathiafulvalene (TTF) core as BEDT-TTF, and the chemical shift reflects the local chemical environment. A_{outer} was estimated from $A_{\text{outer}} = (\delta_{\text{outer}} - \sigma)/\chi_s$ above 20 K, yielding $A_{\text{outer}} = 2.07(1)$ kOe/ μ_B . Using $A_{\text{outer}}/A_{\text{inner}} = -3.1(5)$, we also obtain $A_{\text{inner}} = -0.67(11)$ kOe/ μ_B . These values are comparable to those of κ -ET-CN, $A_{\text{outer}} = 1.59$ kOe/ μ_B and $A_{\text{inner}} = -0.97$ kOe/ μ_B [31], which is consistent with the isostructural nature of the compounds and their shared TTF core.

We can trace the low-temperature evolution of the spin susceptibility using the A_i and σ determined above. Figure 7(a) plots χ_s of κ -BEST-CN normalized to its value at 20 K. The triangles, circles, and squares represent χ_s measured using SQUID, NMR, and EPR (discussed later) magnetometers. The merged NMR shift is used below 5 K. The $\chi_s = (\delta_{\text{outer}} - \sigma)/A_{\text{outer}}$ from NMR decreases steeply below ~ 10 K. Because the Knight shift reflects the local susceptibility at the nuclear site, it is essentially free of the impurity contributions present in the SQUID data, revealing the intrinsic decrease in χ_s .

Figure 7(b) shows χ_s of κ -ET-CN derived from its NMR shift [31], SQUID [9] and EPR [32]. In both κ -ET-CN and κ -BEST-CN, the steep suppression of the local spin susceptibility, weak temperature dependence of the bulk susceptibility, and pronounced broadening of the NMR line at low temperatures (Fig. 5) were qualitatively the same. Although the Knight shift alone does not have sufficient temperature resolution to determine where they begin to decrease, we compare the combined NMR and EPR results in Sec. III B.

We now examine the temperature dependence of $(T_1 T)^{-1}$ to probe low-energy spin fluctuations [43]. Figure 8 shows $(T_1 T)^{-1}$ for both salts. In κ -ET-CN,

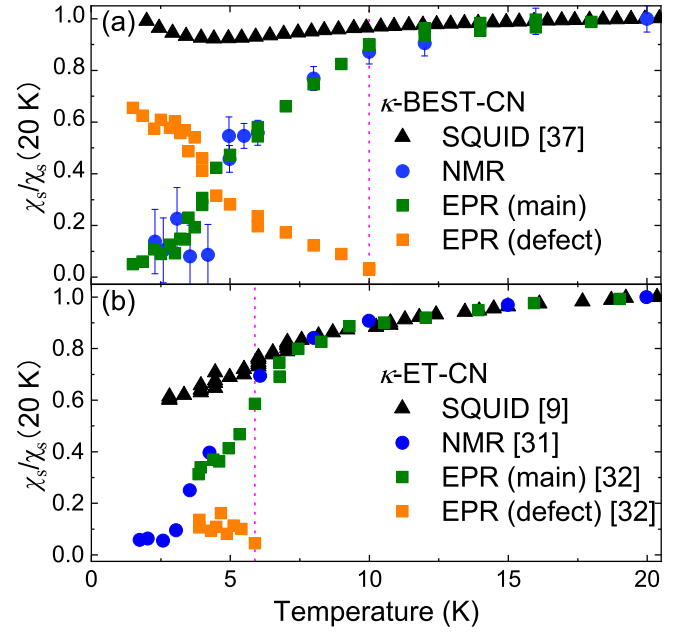


FIG. 7. Temperature dependence of normalized spin susceptibility, $\chi_s/\chi_s(20 \text{ K})$, for (a) κ -BEST-CN [37] and (b) κ -ET-CN [9, 31, 32] measured by SQUID, NMR, and EPR (main and defect components). Vertical dashed lines denote the onset temperature of the EPR defect component, from which we estimate $T^* \approx 10$ K for κ -BEST-CN and $T^* \approx 6$ K for κ -ET-CN.

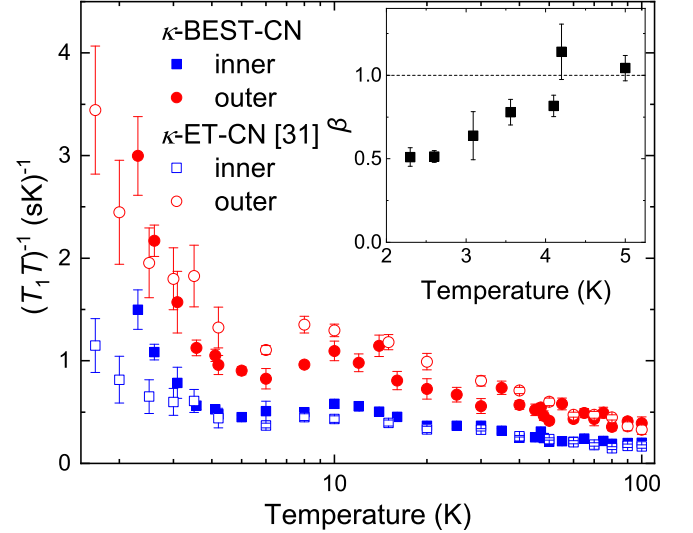


FIG. 8. Temperature dependence of $(T_1 T)^{-1}$ for κ -BEST-CN and κ -ET-CN [31]. The inset shows the temperature dependence of stretched exponent β of κ -BEST-CN.

$(T_1 T)^{-1}$ increases upon cooling and exhibits a maximum at 7–8 K [31]. Below this temperature, the nuclear-magnetization recovery can no longer be described by a single exponential [31, 44], indicating the onset of spatial inhomogeneity. κ -BEST-CN shows a similar trend: $(T_1 T)^{-1}$ increases with decreasing temperature and ex-

hibits a broad hump around 10–12 K, which is in qualitative agreement with the downturn of χ_s derived from NMR. In addition, the stretched exponent β of κ -BEST-CN, which is nearly unity at higher temperatures, drops toward ~ 0.5 below 5 K, as shown in the inset of Fig. 8. The features of the hump in $(T_1T)^{-1}$ and the inhomogeneous relaxation are suggestive of a reduction in magnetic fluctuations due to spin-singlet formation, accompanied by the emergence of unpaired spins that fail to participate in the singlet state. Because of the decrease in β , we refrain from a detailed analysis of $(T_1T)^{-1}$ below 5 K. Even so, previous work on κ -ET-CN showed that T_1^{-1} is nearly constant down to 1 K similar to κ -BEST-CN and then decreases as $T^{3/2}$ below 1 K [44]. Determining whether κ -BEST-CN exhibits the same asymptotic behavior will require measurements at lower temperatures and is left for future work.

B. X-band EPR

EPR, which probes electron spins directly, offers an alternative method for examining the 6 K anomaly [32]. To test whether κ -BEST-CN exhibits a transition analogous to that of κ -ET-CN, we measured EPR spectra below ~ 100 K (Fig. 9(a)). Upon cooling below 40 K, the linewidth decreased. Between 100 K and 10 K, the spectra were well reproduced by a slightly asymmetric Dysonian line shape. In κ -BEST-CN, the room-temperature resistivity is not insulating but moderate ($\sim 3 \times 10^{-3} \Omega\text{cm}$) and is only about one order of magnitude higher than that in the pressure-induced metallic state [37], indicating a weakly semiconducting regime rather than a strongly insulating one. Indeed, optical conductivity measurements on κ -ET-CN report no hard Mott gap, unlike κ -(ET)₂Cu[N(CN)₂]Cl, suggesting itinerant electrons [45, 46]. Therefore, a slight Dysonian-like asymmetry of κ -BEST-CN is consistent with finite resistivity in the low temperature region.

Attempts to fit the spectra with a single Dysonian function failed below 10 K, as reflected by an increase in the reduced chi square, suggesting the emergence of an additional component. As reported for κ -ET-CN [32], the 6 K anomaly is accompanied by the growth of a Lorentzian contribution attributed to unpaired spins that do not form spin singlets. A satisfactory fit to the spectra of κ -BEST-CN in the low-temperature region was also obtained by combining Dysonian and Lorentzian components. Figure 9(b) shows an example of a two-component fit at 5 K, which reproduces the obtained spectrum well.

Figure 9(c) displays the temperature dependence of the integrated intensity of each component. The Dysonian component dropped sharply below ~ 10 K, while the Lorentzian component emerged simultaneously, suggesting that the intrinsic decrease in spin susceptibility in κ -BEST-CN can be distinguished from the increase due to defect-induced unpaired spins. These behaviors closely resemble those observed in κ -ET-CN [32].

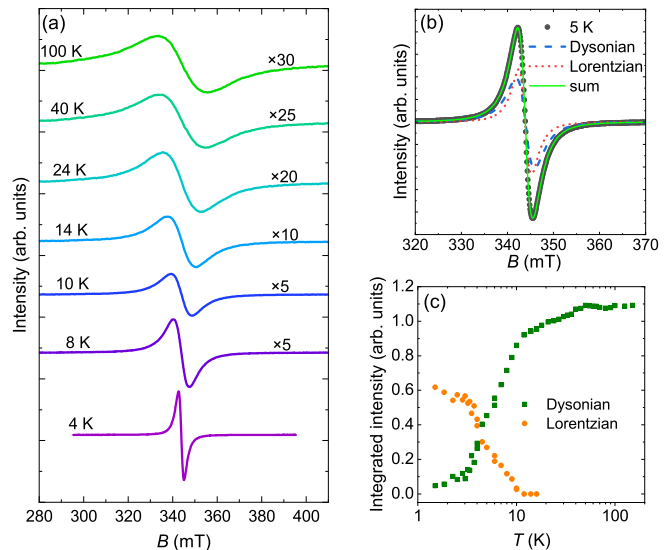


FIG. 9. (a) Temperature evolution of the EPR spectra for κ -BEST-CN. For clarity, the spectra at $T \geq 8$ K are multiplied by the factors noted at the right. (b) Spectrum at 5 K (circles) together with fit components from the Dysonian (dashed) and Lorentzian (dotted), and their summation (solid). (c) Temperature dependence of the integrated EPR intensity of κ -BEST-CN for the Dysonian (squares) and Lorentzian (circles) components.

In κ -ET-CN, $T^* = 6$ K is well established by NQR and thermal expansion measurements [28, 29]. In EPR, the Lorentzian component emerges at this temperature, where the local susceptibility also begins to deviate from the bulk susceptibility, as shown by the vertical dashed line in Fig. 7(b) [32]. This behavior may reflect residual spins that fail to participate in singlet formation and become effectively decoupled from the singlet background. Applying the same criterion to κ -BEST-CN, we estimate $T^* \approx 10$ K (Fig. 7(a)), suggesting a higher T^* than in κ -ET-CN. To test whether the emergence of the defect-spin signal is sample dependent (as expected for extrinsic impurities) or reproducible, we performed the same EPR analysis on different κ -BEST-CN single crystals. We obtained essentially identical temperature dependences of the Dysonian and Lorentzian components, including the onset at $T^* \approx 10$ K (see Fig. S1 in Supplemental Material [47]).

Assuming the Curie law for defect-induced $S = 1/2$ spin, the defect-spin fraction of κ -ET-CN is estimated as $\sim 0.07\%$ from the susceptibility [9, 32] at 4 K ($= 0.66T^*$), the lowest temperature available in Ref. [32], and the defect-spin fraction of κ -BEST-CN is estimated as $\sim 0.10\%$ from the susceptibility at 6 K (corresponding to the temperature of $0.66T^*$). These values are comparable, suggesting that the unpaired spins are induced by the same mechanism.

Comparing the susceptibility of κ -ET-CN and κ -BEST-CN shown in Figs. 7(a) and 7(b), we find that the behavior below their respective T^* is qualitatively

the same: the local susceptibility (EPR/NMR) deviates from the bulk susceptibility (SQUID) and then decreases significantly toward zero on cooling. The Knight shift probes the local spin susceptibility at the nuclear site. Dilute paramagnetic defects predominantly cause an inhomogeneous local-field distribution and thus mainly contribute to line broadening rather than to the NMR shift. Therefore, the Knight shift provides a comparatively defect-insensitive measure of the intrinsic susceptibility. The consistent temperature evolution between the EPR intensity and the Knight shift provides an independent cross-check of our EPR-based analysis.

Figure S2 in Supplemental Material [47] also compares the NMR- and EPR-derived $\chi_s/\chi_s(20\text{ K})$ of the two salts directly. Although the Knight shift alone does not resolve the difference in T^* , the combined NMR and EPR data suggest that the suppression of spin susceptibilities in κ -BEST-CN occurs at a higher temperature than in κ -ET-CN. Thus, $T^* \approx 10\text{ K}$ should be regarded as an estimate from the combined spectroscopic results; an accurate determination requires additional NQR or thermal expansion measurements.

C. BEDSe-TTF substitution effects

BEDSe-TTF retains the same TTF skeleton with high electron density [48] as BEDT-TTF; therefore the highest occupied molecular orbitals of the two molecules, responsible for the electronic properties, are expected to be nearly identical. Consistently, ^{13}C -NMR chemical shifts and hyperfine coupling constants of κ -BEST-CN and κ -ET-CN are almost indistinguishable. Electrical resistivity measurements showed that κ -BEST-CN lies slightly on the positive-pressure side of κ -ET-CN, by a chemical pressure on the order of 50 MPa [37, 49]. A shift of the same order of magnitude, although opposite in sign, was reported for κ -(BEST) $_2$ Cu[N(CN) $_2$ Br] [50]. Taken together, these observations indicate that substituting BEDSe-TTF for BEDT-TTF perturbs U/W only marginally.

A positive pressure effect has been reported to decrease the VBS transition temperature near the Mott boundary [51]. If the higher T^* suggested by the combined NMR and EPR data reflect an enhancement of the VBS transition, this enhancement is unlikely to be explained by a simple pressure effect. A possible origin of the higher T^* is the difference in molecular mass: BEDSe-TTF weighs 572.3, which is approximately 50% more than 384.7 for BEDT-TTF. Because the VBS transition involves lattice distortion, its transition temperature is determined by the balance between the electronic energy gain and the elastic cost of the lattice. The heat capacity data for κ -[(BEST) $_x$ (ET) $_{1-x}$] $_2$ Cu[N(CN) $_2$ Br] showed that the Debye temperature decreases with increasing x , implying lattice softening by heavier molecules [52]. If a comparable lattice softening occurs in κ -BEST-CN, the elastic cost of the distortion accompanying the VBS transition

is reduced. Although it is reasonable to infer that a similar lattice softening effect occurs in κ -BEST-CN, future direct measurements of its Debye temperature via experiments such as low-temperature specific heat would provide definitive evidence for this scenario. Therefore, we suggest that the reduced lattice energy caused by Se substitution may contribute to the enhancement in T^* . An analogous mass-shifted transition was reported for the Peierls transition in α' -(ET) $_2$ IBr $_2$ upon isotopic substitution [53].

Finally, direct evidence for the VBS transition in κ -ET-CN is still lacking, because conventional helium-flow X-ray diffraction becomes technically challenging at temperatures well below 6 K. If the higher T^* is confirmed, κ -BEST-CN may offer a practical opportunity to test the proposed VBS state by low-temperature diffraction experiments.

IV. SUMMARY

^{13}C -NMR and X-band EPR measurements on κ -BEST-CN reveal signatures of a spin-singlet transition with T^* estimated to be approximately 10 K from the combined spectroscopic results, higher than that of κ -ET-CN ($T^* = 6\text{ K}$). Both salts exhibit the same qualitative signatures at T^* : a rapid drop in spin susceptibility, linewidth broadening, a hump in $(T_1T)^{-1}$, and the emergence of a Lorentzian EPR component. These behaviors are consistent with a VBS state in which spin-singlet formation coexists with a small fraction of defect spins. Transport data showed that the electronic correlation ratio U/W is nearly identical in both compounds, suggesting that the higher T^* in κ -BEST-CN may be associated with the lattice degree of freedom. We suggest that the 50% increase in molecular mass introduced by the BEDSe-TTF donor induces lattice softening that stabilizes the distortion, without substantially changing U/W . These findings suggest the importance of extending the conventional U/W phase diagram to include the lattice degree of freedom and motivate future thermodynamic and structural measurements to determine T^* and test the proposed VBS state in κ -BEST-CN.

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The data that support the findings of this paper are openly available [54].

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