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Stretched exponential spin relaxation in organic superconductors

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Proton NMR measurements on the organic superconductor κ-$(ET)_2Cu[N(CN)_2]Br$ [where ET represents bis(ethylenedithio)tetrathiafulvalene] ($T_C = 11.6$ K) exhibit stretched exponential spin-lattice relaxation below $T \approx 25$ K, suggestive of an inhomogeneous magnetic phase that develops in the normal state and coexists with superconductivity. The onset of this phase coincides approximately with a large normal-state Nernst signal reported previously. By contrast, the closely related superconductor κ-$(ET)_2Cu(NCS)_2$ ($T_C = 10.5$ K) shows single exponential spin-lattice relaxation and a conventional Nernst effect. The temperature range of $T_C < T <$ 30 K encompasses several phenomena in the κ-$(ET)_2X$ conductors, including changes in conduction-electron-spin resonance, electronic phase separation, and the onset of antiferromagnetic order. Analogous behavior in $La_{2-x}Sr_xCuO_4$ suggests that a spin glass or density wave may develop in κ-$(ET)_2Cu[N(CN)_2]Br$.

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The observation of a large normal-state Nernst signal in κ-$(ET)_2Cu[N(CN)_2]Br$ (Ref. 1) reinforces the correspondence between the κ-$(ET)_2X$ (Ref. 2) organic superconductors and the copper oxides.[3,4] In the cuprates, large Nernst signals were first attributed to normal-state vortices,[5,6] but this interpretation has been challenged by models involving enhanced Gaussian fluctuations[7] and Fermi-surface reconstruction.[8,9] If magnetic fluctuations of some kind are responsible for the Nernst signal, they may be detectable via nuclear-spin relaxation. With this motivation in mind, we have examined proton NMR in κ-$(ET)_2Cu[N(CN)_2]Br$ ($T_C = 11.6$ K) and κ-$(ET)_2Cu(NCS)_2$ ($T_C = 10.5$ K). Since ethylene group protons in the ET molecule are weakly coupled to conduction electrons, they are sensitive probes of vortex motion and, in principle, other sources of nonhyperfine spin relaxation.[10–13] The separation of hyperfine relaxation from other mechanisms is more difficult to achieve in the copper oxides.[14] We find that, below $T \sim 25$ K, the spin-lattice relaxation in κ-$(ET)_2Cu[N(CN)_2]Br$ develops a stretched exponential time dependence whose exponent β exhibits no apparent change at T_C. The temperature and field dependence of β shows a striking correspondence to the Nernst coefficient reported earlier.[1] By contrast, spin relaxation in κ-$(ET)_2Cu(NCS)_2$, which exhibits no anomalous Nernst effect, shows single exponential time dependence. The temperature range of $T_C < T < 30$ K encompasses several phenomena observed in the κ-$(ET)_2X$ family, including changes in conduction-electron-spin resonance,[15] electronic phase separation,[16,17] and the onset of antiferromagnetic order κ-$(ET)_2Cu[N(CN)_2]Cl$ at $T_{\text{Néel}} = 27$ K.[18] We discuss similarities to $La_{2-x}Sr_xCuO_4$ where experiments have also demonstrated stretched exponential relaxation[19] and a Nernst signal far above T_C.[5,6]

Single crystals of κ-$(ET)_2Cu[N(CN)_2]Br$ and κ-$(ET)_2Cu(NCS)_2$ with natural isotopic abundances were grown using methods previously described.[20,21] The samples were mounted on sapphire with a small amount of proton-free grease[22] and were cooled in a gas flow cryostat. It is well established that the rate of cooling through 80 K has a significant effect on the transport and superconducting properties of κ-$(ET)_2Cu[N(CN)_2]Br$.[23–27] To ensure a controlled experiment, we followed the same cooling procedure for all runs. Except where indicated, the sample was cooled at $dT/dt = 0.3$ K/min from 100 K down to 40 K. As we show later, a change of a factor of 60 in the cooling rate made no significant difference to our main result. NMR measurements were taken with a homebuilt probe and spectrometer in fields of 1, 1.5, 2, and 3 T. Unless otherwise indicated, the static field B_0 was perpendicular to the conducting planes, and the rf (H_1) field was parallel to the conducting planes. Nutation curves for spin rotation angles out to 450° were weakly decaying sinusoids, indicative of uniform spin excitation throughout the sample. A saturating comb solid echo sequence $[90_x]^{50} - t - 90_x - \tau_e - 90_y - \tau_e -$ solid echo was used to measure spin-lattice relaxation. The 90_x comb (10^{-4}-s pulse spacing) ensures uniform saturation. The solid echo ($\tau_e = 10^{-5}$s) refocuses nearest-neighbor dipolar coupling which is large (∼40 kHz) for ethylene protons.[28]

Figure 1 shows the magnetization recovery versus delay time for κ-$(ET)_2Cu[N(CN)_2]Br$ at 9 K and $B_0 = 1$ T. Previous investigators have fit recoveries to a sum of two exponentials, the longer of which was considered intrinsic to the sample.[10,12] However, we find that a stretched exponential (solid curve),

$$M(t) = M_0(1 - \exp[-(t/T_1)^{\beta}]) \quad (1)$$

provides a better fit to our data over the entire recovery period. Figure 2 shows a typical recovery in κ-$(ET)_2Cu(NCS)_2$. A single exponential generally provided a superior fit. Fits to a stretched exponential yielded $\beta \approx 1$ at all temperatures.

Stretched exponential relaxation is observed in a wide variety of disordered systems.[29,30] In NMR, it may arise from a distribution of spin-lattice relaxation times.[19,31] Small β implies a broad distribution, whereas, $\beta = 1$ corresponds to the single exponential relaxation expected for a homogeneous system.

Figure 3 shows $1/T_1$ and β for κ-$(ET)_2Cu[N(CN)_2]Br$ for 4.2 K $< T <$ 300 K, in a field of 1 T. For temperatures above 25 K, the stretching exponent β approaches unity, indicating uniform spin-lattice relaxation. In this region, the

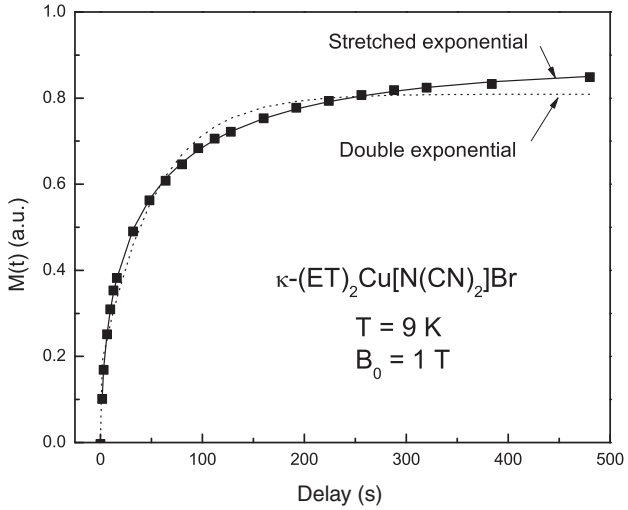

FIG. 1. κ-$(ET)_2Cu[N(CN)_2]Br$ magnetization recovery with fits to double exponential (dashed curve) and stretched exponential (solid curve) with $T_1 = 44$ s, $\beta = 0.52$.

double exponential fit is nearly indistinguishable from the stretched exponential, and our values for T_1 agree with previous measurements.[10–12,23,32] Beginning at $T = 25$ K, β begins to fall, reaching a minimum of $\beta \sim 0.5$. Below 7.5 K, magnetic fluctuations from the vortex liquid lead to more homogeneous spin relaxation and an increasing β. For still lower temperatures, β decreases as the vortex lattice forms, and relaxation once again becomes inhomogeneous.

$1/T_1$ exhibits several different regimes of relaxation, shown in Fig. 3. The large peak near 250 K appears as the exponentially activated correlation time for the ethylene group motion crosses the Larmor frequency.[10,13,33] The smaller peak near 160 K coincides with large changes in central ^{13}C and ^{1}H linewidths.[16,34,35] The weak maximum near 50 K coincides with a sharp change in $1/T_1$ for ^{13}C nuclei that is associated with the opening of a spin gap.[34–37] Nearly all transport coefficients[3] as well as the conduction-electron-spin

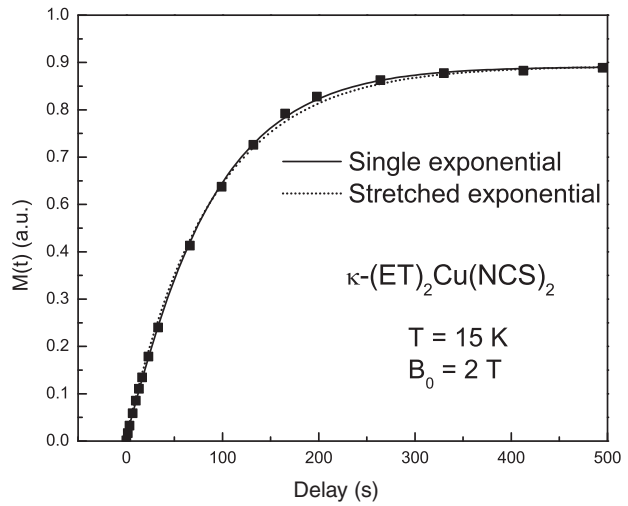

FIG. 2. κ-$(ET)_2Cu(NCS)_2$ magnetization recovery with fits to single exponential (dashed curve) and stretched exponential (solid curve) with $T_1 = 87$ s, $\beta = 1.07$.

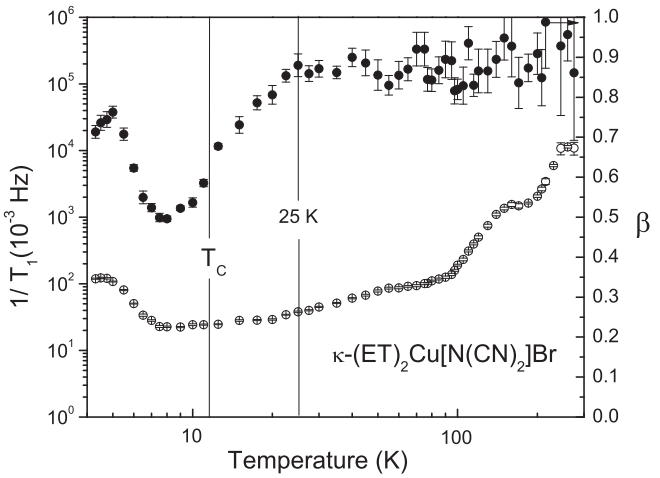

FIG. 3. Stretching exponent β (filled circles) and $1/T_1$ (open circles) in κ-$(ET)_2Cu[N(CN)_2]Br$ for $B_0 = 1$ T.

resonance signal change dramatically near 50 K.[15,38,39] The diverse physical mechanisms responsible for these changes in $1/T_1$ have no apparent effect on β. The relaxation remains homogeneous down to $T = 25$ K, below which β drops and the slope of $1/T_1$ versus T shows a slight change.

Figure 4 shows corresponding data for κ-$(ET)_2Cu(NCS)_2$. In this case, single exponential ($\beta \sim 1$) relaxation holds throughout the entire temperature range. The large peak near 250 K and the weaker peak near 50 K remain. The peak near 160 K is less well defined, and there is no vortex peak. The latter is consistent with a much lower irreversibility temperature in this material.[40]

Neither material showed any change in spin-lattice relaxation at T_C, consistent with a very weak hyperfine coupling to the conduction-electron system. In both cases, $1/T_1$ was independent of position in the NMR line. Despite different anions and crystal structures, κ-$(ET)_2Cu[N(CN)_2]Br$ (orthorhombic) and κ-$(ET)_2Cu(NCS)_2$ (monoclinic) have very similar superconducting properties,[41,42] making it unlikely that the unusual spin relaxation seen only in κ-$(ET)_2Cu[N(CN)_2]Br$ is a superconducting fluctuation

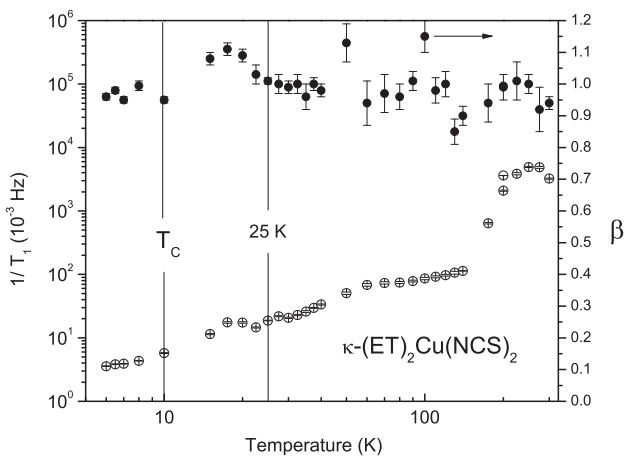

FIG. 4. Stretching exponent β (filled circles) and $1/T_1$ (open circles) in κ-$(ET)_2Cu(NCS)_2$ for $B_0 = 1$ T.

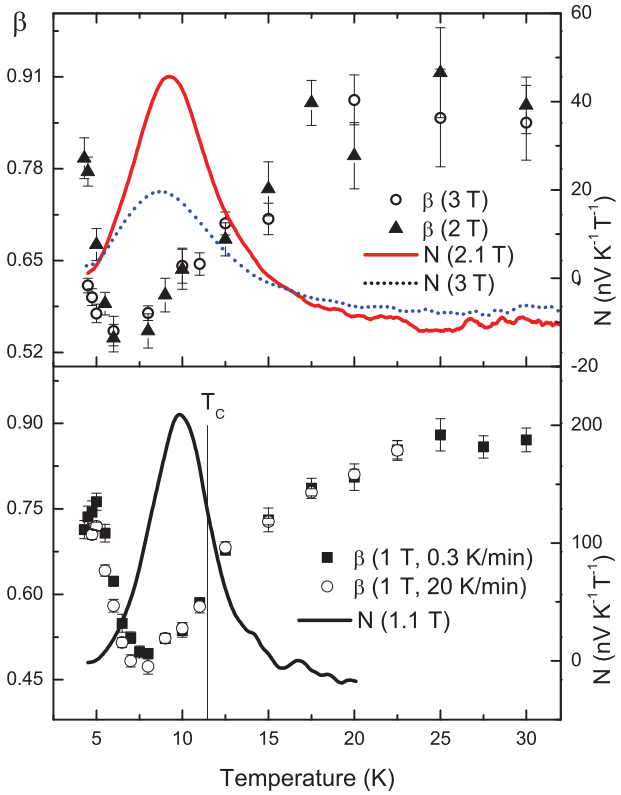

FIG. 5. (Color online) Top panel: β in κ-$(ET)_2Cu[N(CN)_2]Br$ for $B_0 = 2$ T (filled triangles), 3 T (open circles), and Nernst coefficient N for 2.1 T (solid line) and 3 T (dotted line). Lower panel: β for $B_0 =$ 1 T with fast cooling (filled squares) and slow cooling (open circles). Solid line shows N for $B_0 = 1.1$ T.[1]

effect.[43] In addition, fluctuations would not be expected to produce stretched exponential relaxation.

Electronic phase separation is a potential source of inhomogeneous relaxation. Separation into macroscopic metallic and antiferromagnetic regions was clearly demonstrated in deuterated κ-$(ET)_2Cu[N(CN)_2]Br$ with both scanning infrared spectroscopy[16] and NMR.[17] The large internal field of the antiferromagnetic clusters resulted in ^{13}C NMR line splitting that developed below 30 K (Ref. 17) and a broadening of the ^1H NMR line below 15 K.[44] Our data showed no splitting or broadening of the ^1H line near 25 K. In separately measured κ-$(ET)_2Cu[N(CN)_2]Br$ samples enriched with ^{13}C, we observed single exponential relaxation at all temperatures and no line splitting, although the ^{13}C linewidth showed a weak local minimum near 25 K. If the stretched exponential behavior was due to phase separation, one might also expect it to depend upon the cooling rate, but that is not the case. The lower panel of Fig. 5 shows essentially no difference in β for cooling rates of $dT/dt = 0.3$ K/min and $dT/dt = 20$ K/min. We, therefore, do not attribute the change at 25 K to macroscopic phase separation.

Figure 5 shows the field dependence of β for $B_0 =$ 1–3 T applied normal to the conducting planes. We have also plotted the Nernst coefficient N for $B_0 = 1.1$, 2.1, and 3 T provided to us by the Oxford group.[1] The correspondence between N and β is notable, each quantity showing an onset near 25 K, an extremum in the superconducting state, and no discernible

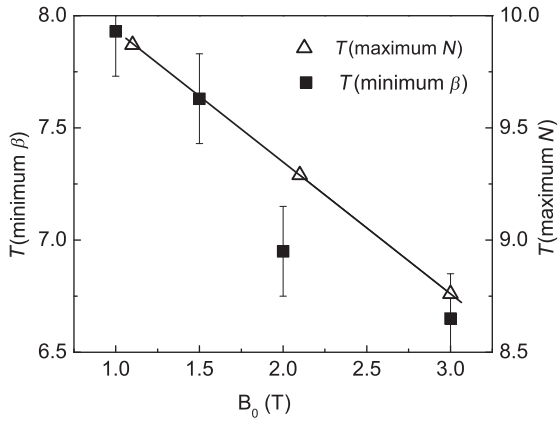

FIG. 6. T (minimum β) and T (maximum N) versus B_0 in κ-$(ET)_2Cu[N(CN)_2]Br$. Straight line is a fit to T (maximum N) versus B_0.

change at T_C. Figure 6 shows the field dependence of two quantities: (1) the temperature at which β reaches a minimum and (2) the temperature at which N reaches a maximum. Both quantities are determined by vortex motion. Below T_C, a large N results from the flow of entropy-carrying vortices down a temperature gradient, in turn, inducing a transverse electric field via the Josephson effect. The rapid increase in β and $1/T_1$ below T(minimum β) is due to the increasing strength of magnetic-field fluctuations from the vortex liquid, which scale as $1/\lambda^4$ where λ is the penetration depth.[45,46]

Vortex behavior in organic superconductors is highly anisotropic.[10,47,48] If the stretched exponential relaxation was due to vortices, we would expect this anisotropy to be reflected in the NMR. Figure 7 shows β and $1/T_1$ in κ-$(ET)_2Cu[N(CN)_2]Br$ for two orthogonal magnetic-field orientations. Below T_C, both β and $1/T_1$ vary strongly with orientation with the peak in β and $1/T_1$ occurring near the irreversibility temperature corresponding to the perpendicular field component.[10–13,49] However, above T_C, there is no significant orientation dependence. This would appear to rule out vortices as a source of spin relaxation above T_C. Normal-state vortices may still be present and observable

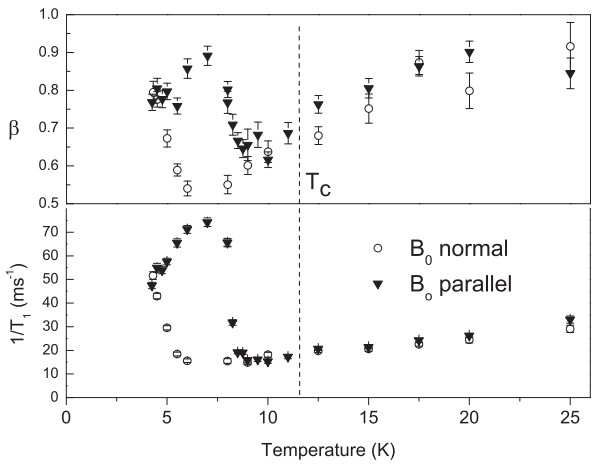

FIG. 7. (Top) β versus temperature for $B_0 = 2$ T normal (circles) and parallel (triangles) to conducting planes. (Bottom) $1/T_1$ versus temperature for the same two field orientations (κ-$(ET)_2Cu[N(CN)_2]Br$).

via Nernst measurements, but they no longer generate field fluctuations on the time scale of nuclear-spin precession.

Measurements were also performed on ^{13}C-enriched κ-$(ET)_2Cu[N(CN)_2]Br$ samples. Both central ^{13}C sites showed single exponential relaxation at all temperatures. ^{13}C relaxation is dominated by hyperfine coupling to conduction electrons and changes abruptly at T_C.[34,35] Hyperfine coupling to protons is far weaker as evidenced by a much longer T_1 and the lack of any change T_C. Protons are, however, sensitive to the onset of antiferromagnetic order at $T_{\text{Néel}} = 27$ K in κ-$(ET)_2Cu[N(CN)_2]Cl$.[18] This suggests that the stretched exponential relaxation of protons may come from a disordered magnetic phase, possibly a spin glass, developing below 25 K. This may change the Fermi surface enough to affect the Nernst signal but not enough to affect the rapid relaxation of ^{13}C by conduction electrons. The temperature dependence of β shown in Fig. 5 is reminiscent of a spin glass where simulations[50] and experiments[51] show that $\beta(T)$ falls monotonically beginning at a characteristic ordering temperature and approaches its asymptotic minimum near the glass transition. In κ-$(ET)_2Cu[N(CN)_2]Br$, a plausible magnetic ordering temperature would be $T_{\text{Néel}} = 27$ K observed in the closely related compound κ-$(ET)_2Cu[N(CN)_2]Cl$.[18] We searched for glasslike behavior by fixing the sample temperature at 15 K after rapid cooling and looking for an evolution of the NMR signal. The spectrum shape and location were unchanged over a period of 2500 min. However, measurements over a wider range of temperatures and cooling rates would be necessary to identify a spin-glass phase. The possibility of magnetic phase has some support in recent muon spin resonance measurements of the local electronic spin susceptibility in κ-$(ET)_2Cu[N(CN)_2]Br$. These show a sharp peak near 15 K and structure extending up to ~25 K.[52] The situation here has analogies to underdoped $La_{2-x}Sr_xCuO_4$, which exhibits a Nernst effect far above T_C (Refs. 5 and 6) stretched exponential ^{139}La nuclear quadrupole resonance spin relaxation[19] and spin-glass-like magnetic phases both inside and outside the superconducting dome.[53] Interestingly, Knight shift anomalies in $La_{2-x}Sr_xCuO_4$ (Ref. 54) show evidence for a two component electronic fluid,[55] similar to the situation in heavy fermion compounds.[56,57] The appearance near 25 K of a magnetic phase in κ-$(ET)_2Cu[N(CN)_2]Br$ may be a manifestation of a second component.

Density waves or stripes could also lead to inhomogeneous relaxation that is correlated with a Nernst signal. In Nd-doped $La_{2-x}Sr_xCuO_4$, a Nernst signal similar to that in κ-$(ET)_2Cu[N(CN)_2]Br$ appears when charge stripes are present but is featureless when they are absent.[9] Stripes have been proposed for the κ-$(ET)_2X$ materials,[58] but these compounds reside at half-filling and appear to have conventional quasiparticles, casting doubt on a density wave reconstruction of the Fermi surface at 25 K. However, quantum oscillation experiments in κ-$(ET)_2Cu[N(CN)_2]Br$ require high fields (30 T) where magnetic breakdown is apparently dominant.[59] For some probes, breakdown may obscure Fermi-surface modifications from a density wave with a small energy gap, whereas, the Nernst coefficient may be particularly sensitive to these changes.

On an effective pressure-temperature diagram, κ-$(ET)_2Cu[N(CN)_2]Br$ resides closer to the antiferromagnetic phase than κ-$(ET)_2Cu(NCS)_2$ and, therefore, has stronger electronic correlations.[3] This fact is probably the origin of the differences in the behavior reported here and in the Nernst behavior.[1] However, some time ago, Klutz *et al.*[60] reported deviations from exponential recovery below 25 K in collections of unoriented crystallites of κ-$(ET)_2Cu(NCS)_2$. It is possible that strains incurred through packing of grains can lead to greater electronic inhomogeneity than obtained from a single-crystal κ-$(ET)_2Cu(NCS)_2$, accounting for their observation. They attributed the inhomogeneous NMR relaxation to nonuniform spin excitation due to a finite skin depth. Our data do not support a skin-depth picture. The rf field in our experiment was parallel to the conducting planes. For this orientation, the skin depth (~3 mm) is larger than the κ-$(ET)_2Cu[N(CN)_2]Br$ sample ($1.2 \times 0.7 \times 0.6$ mm). As shown in Fig. 5, the measurements at several different frequencies did not exhibit a systematic change in onset temperature as would be expected from a skin-depth model. Moreover, the NMR nutation curves did not show the dependence upon a spin rotation angle characteristic of skin-depth limiting.[61] Since our κ-$(ET)_2Cu(NCS)_2$ sample had lower resistivity and was larger ($1.7 \times 2.7 \times 0.5$ mm), any finite skin-depth effect would be enhanced over κ-$(ET)_2Cu[N(CN)_2]Br$. The fact that the onset temperature occurred near 25 K in both experiments is evidence for a common magnetic energy scale in the κ-$(ET)_2X$ organics.

In conclusion, we find a correspondence between stretched exponential spin relaxation and the large normal-state Nernst signal observed in κ-$(ET)_2Cu[N(CN)_2]Br$. Both effects are absent in the more weakly correlated superconductor κ-$(ET)_2Cu(NCS)_2$. The lack of field orientation dependence argues against vortices as the source of the anomalous relaxation. The data point toward a disordered magnetic phase, a spin-glass or a density wave, that develops below 25 K and coexists with superconductivity.

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