# Ultrasonic investigation of the transition at 6 K in the spin-liquid candidate $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

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(Received 15 October 2013; revised manuscript received 17 January 2014; published 29 January 2014)

An ultrasonic measurement has been performed on the spin-liquid organic candidate $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ at low temperatures. A softening anomaly was observed on the longitudinal velocity along a direction perpendicular to the organic planes with a consistent one on the attenuation. The velocity anomaly appears as a frequency dependent softening peak below 20 K with a maximum value found near 5–6 K. A magnetic field affects predominantly the peak only below a temperature $T_p = 5.9$ K that is associated with the 6 K transition found on other physical properties. We suggest to relate the frequency dependent velocity softening at low temperatures to a spinon-phonon coupling, softening that is reduced below 5.9 K due to a pairing instability transition of the spinons.

DOI: 10.1103/PhysRevB.89.045138 PACS number(s): 75.10.Kt, 74.70.Kn, 63.20.kk, 62.65.+k

## I. INTRODUCTION

The discovery of a spin-liquid behavior in $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ has motivated new experimental and theoretical studies dedicated to the identification of the nature of the spin-liquid state [1–3]. Although there is no sign of long range magnetic order down to 32 mK [4] despite an exchange interaction $J \approx 250$ K, anomalies due to a strange phase transition near 6 K have been identified on thermodynamic [1,4], transport [2], lattice [5], and microwave [6] properties. These have been tentatively explained by a crossover from a thermally to a quantum disordered state [1], an instability of the quantum spin liquid [1,7–13], or a paired-electron crystal [14].

Most theoretical studies of the spin-liquid properties have been investigated with the anisotropic triangular lattice half-filled band Hubbard model [15–18]. The spin-chirality ordering or fluctuations are among the consequences of several spin-liquid models [7,11,19]. Other theoretical approaches based on simultaneous charge and spin frustration effects [14,20,21] are proposed on the basis of anomalous dielectric behavior below 60 K, the possible antiferroelectric ordering of the dipoles around 9 K [22], magnetodielectric effects [6], and thermodynamic measurements [5] near 6 K. Although there is still controversy about the presence of a small gap in the spin excitations at low temperatures [1,2,23], one cannot exclude the possibility that the gapless spin liquid may not be the result of geometrical frustration but that of strong correlations between spins and charges. However, since the absence of charge order was recently reported [24], spatial and temporal variations of the charge distribution need to be explored in more detail.

The existence of low lying excitations in organic spin-liquid materials $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ and EtMe$_3$Sb[Pd(dmit)$_2$]$_2$, as revealed in thermal conductivity measurements [1], appears to support the picture of mobile particles called spinons, which form a Fermi surface and which are coupled to $U(1)$ gauge fields [25,26]. It was then suggested that the 6 K transition in $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ could result from an instability of the Fermi surface, which leaves a finite density of states intrinsically or due to impurities [10]. To test this picture of spinons in the organic salts, a few experiments have been suggested, such as metalliclike spin transport [27] and spinon-phonon coupling [28], for which specific temperature dependencies of the ultrasonic attenuation were predicted.

In this paper we address, for the spin-liquid candidate $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, the coupling of acoustic longitudinal phonons with low-temperature excitations in the vicinity of the 6 K transition by using an ultrasonic wave propagation technique. The velocity of the phonons propagating along a direction perpendicular to the **bc** plane presents a frequency dependent softening peak near 6 K with a consequent variation in the attenuation. A magnetic field of 16 T is found to affect these elastic anomalies only below a characteristic temperature $T_p \simeq 5.9$ K. These ultrasonic data will be discussed along with the model of fermionic spinons coupled to phonons, the strange 5.9 K transition then signaling a pairing instability of the spinons.

## II. EXPERIMENT

The ultrasonic pulse echo method requires samples with parallel shiny surfaces on which piezoelectric transducers can be glued. Moreover, these surfaces must be sufficiently far apart to separate in time the multiple reflected echoes. The $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ single crystals grow electrochemically as very thin platelets whose parallel faces are only separated by 25 $\mu$m on average. Several of these crystals have been tested: Although an elastic anomaly over the appropriate low-temperature range was detected, the data were not reliable on different experimental parameters (pulse width, time delay and width of the detection gate, and frequency). Fortunately, a single platelet crystal with a thickness around 100 $\mu$m was obtained and the ultrasonic method could be used for the longitudinal mode. No transverse mode could be studied with such a thin crystal because of mode conversion at the different interfaces and thus superposition of several acoustic echoes.

We use a pulsed ultrasonic interferometer [29] to measure the variation of longitudinal acoustic velocity along the direction perpendicular to the parallel faces ($\boldsymbol{a}^*$ axis) relative to its value at a fixed temperature $T_0 = 30$ K, $\Delta V/V = [V(T) - V(T_0)]/V(T_0)$, and the corresponding variation of the attenuation. The ultrasonic technique is used in the transmission mode and, because of the small thickness of the crystal along the $\boldsymbol{a}^*$ axis ($\simeq$100 $\mu$m), the sample is glued with a silicone seal onto a $CaF_2$ delay line in order to separate the first transmitted acoustic echo (through the crystal-$CaF_2$ assembly) from the electric pulse. The longitudinal acoustic pulses are generated with $LiNbO_3$ piezoelectric transducers resonating at 30 MHz and odd overtones bonded to the crystal and the $CaF_2$ delay line with a silicone seal. Because of the monoclinic structure, the acoustic mode is not pure but it is dominated by the compressibility modulus $C_{11}$; after subtraction of the contribution of the $CaF_2$ crystal [29], the velocity variations are thus related to the variations of the compressibility along a direction perpendicular to the two-dimensional $(ET)_2$ planes. The velocity variations were not corrected to take into account the thermal expansion since these effects are usually negligible. We could also measure the variation of the attenuation $\Delta\alpha$ by monitoring the amplitude of the first transmitted echo and subtracting the contribution of the $CaF_2$ delay line. A magnetic field up to 16 T could be applied along the $\boldsymbol{a}^*$ axis.

## III. RESULTS

As the temperature is decreased from room value, a large amount of the acoustic signal is progressively lost due the deterioration of the interfaces between the organic crystal, the piezoelectric transducer, and the $CaF_2$ line, resulting from the large thermal expansion coefficient of these organic crystals compared to the other materials. For our ultrasonic measurements on the thin $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ crystal, we could only get reliable data at two frequencies of 150 and 210 MHz. To achieve this, the time duration of the electric pulse fed to the emitting transducer was kept at 100 ns to minimize phase mixing between the first transmitted acoustic echo through the crystal-$CaF_2$ line and multiple reflected echoes inside the crystal to the detriment of available power.

When decreasing the temperature from room value, the velocity increases smoothly, in agreement with the stiffening of the interplane cohesion forces, without indication of any instability down to 25 K. At lower temperatures, instead of the expected saturation of the velocity, we observe near 5–6 K a frequency dependent anomaly that is shown in Fig. 1. Below 30 K, both sets of data present an identical temperature increase down to 18 K, where the two curves separate from each other. At each frequency, $\Delta V/V$ first goes through a wide

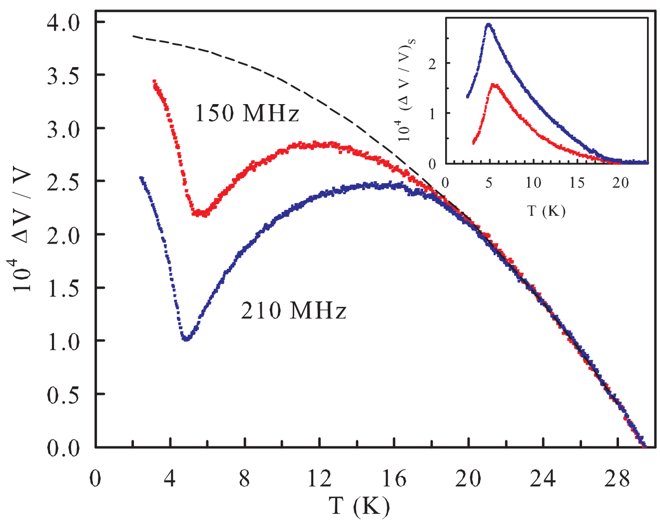


FIG. 1. (Color online) Relative variation of the longitudinal velocity along the $\boldsymbol{a}^*$ axis for $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ as a function of temperature at 150 MHz (red) and 210 MHz (blue). The dashed line suggests the normal elastic behavior. Inset: Softening peaks obtained after subtraction of the velocity data from the normal elastic curve.

maximum, then decreases to reach a narrow minimum below 6 K, and increases further down to the lowest temperature of 2 K. On the one hand, the maximum is the result of a velocity softening whose amplitude and temperature range increase with frequency. On the other hand, the narrow minimum below 6 K results from a low-temperature stiffening whose amplitude is practically constant in frequency. To analyze more precisely this softening peak, we must try to evaluate what should be the *normal* elastic behavior for this crystal without the low-temperature anomaly. Since there is no frequency dependence of $\Delta V/V$ above 20 K, we have extrapolated (second degree polynomial) these data to low temperatures, as indicated by the dashed line plotted in Fig. 1. Keeping in mind a certain amount of uncertainty at the lowest temperatures, the velocity data were subtracted from this extrapolation curve to yield the softening peaks $(\Delta V/V)_S$ presented in the inset (Fig. 1). In an attempt to identify the interactions responsible for these peaks below 20 K, we have investigated the effects of a magnetic field.

These softening peaks are shown in Fig. 2 below 10 K for two magnetic field values, 0 and 16 T. At 150 MHz in zero magnetic field, the softening increases smoothly below 10 K and, in spite of the noise, we can clearly identify a break in the increasing rate near 5.9 K (indicated by a vertical dashed line), a temperature that corresponds to $T_p(0)$ identified in a recent microwave experiment on similar samples [6]. Below 5.9 K, $(\Delta V/V)_S$ is practically flat down to 5.5 K, where the maximum occurs; then it decreases rapidly first and more slowly below 4 K, with a temperature dependence that suggests saturation below 2 K. At a higher frequency of 210 MHz, the softening below 10 K is more pronounced, so the break in the increasing rate is replaced by an inflection point at $T_p(0)$, then a maximum is found near 4.9 K, and finally the softening decreases to saturation below 2 K with an amplitude almost identical to the 150 MHz data. If the link between the break in the increasing rate and $T_p(0)$ is genuine, we should observe a

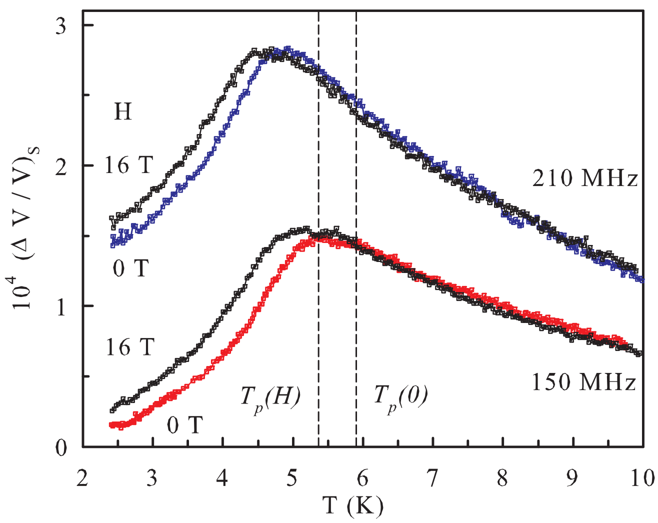


FIG. 2. (Color online) Softening peaks $(\Delta V/V)_S$ for $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ as a function of temperature below 10 K: $H = 0$ T, 150 MHz (red) and 210 MHz (blue); $H = 16$ T (black). Vertical dashed lines indicate the transition temperatures $T_p(0)$ and $T_p(H)$.

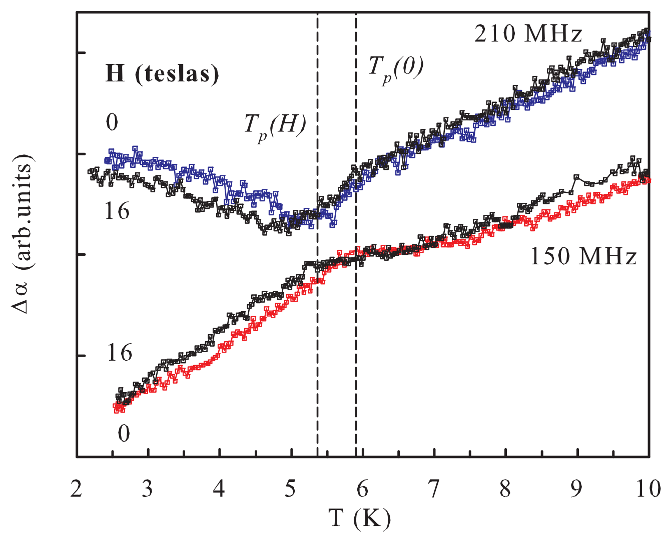


FIG. 3. (Color online) Variation of the attenuation $\Delta\alpha$ below 10 K at two frequencies: $H = 0$ T, 150 MHz (red) and 210 MHz (blue); $H = 16$ T (black). Vertical dashed lines indicate the transition temperatures $T_p(0)$ and $T_p(H)$.

downshift of $T_p$ and substantial effects over this temperature range when a magnetic field is applied [6]. This is indeed the case at both frequencies, as shown in Fig. 2: In a field of 16 T, the softening weakly decreased on the high-temperature side of the peaks but was markedly enhanced on the low-temperature side, so the peaks appear downshifted by 0.5 K [$T_p(H)$]. It is thus clear that the temperature $T_p(0) \simeq 5.9$ K establishes a boundary below which the character or/and the density of the excitations responsible for this elastic anomaly are modified.

On thin crystals, the measurement of the variation of the attenuation is delicate since it is simply obtained by monitoring the amplitude of the first transmitted acoustic echo, which can be more affected than the velocity by overlapping multiple reflected echoes in the crystal. With a 100 ns pulse, however, we should expect to minimize the overlap and obtain a reliable temperature dependence of the variation of the attenuation $\Delta\alpha$. These data at the two frequencies are presented in Fig. 3 for 0 and 16 T magnetic fields. Although the data are quite noisy because the attenuation is measured only over a 100 $\mu$m distance, they are satisfactorily reproducible. At the two frequencies, $\Delta\alpha$ decreases monotonically with temperature from 10 K down to $T_p(0)$, where a change of behavior is observed. Surprisingly, this change of behavior is strongly frequency dependent: At 150 MHz in zero magnetic field, the attenuation decreases more rapidly below $T_p(0)$ down to the lowest temperature while it rather increases at 210 MHz below 5 K after a small drop at $T_p(0)$. Similarly to the velocity data, a magnetic field of 16 T hardly affects $\Delta\alpha$ above $T_p(0)$ but markedly modifies the curves below by shifting the anomaly to $T_p(H)$. This temperature dependence of $\Delta\alpha$ is likely intrinsic to the material, since we do not see why extrinsic frequency effects would be confined only below $T_p$; this issue could be clarified only when attenuation will be measured on a thicker crystal.

## IV. DISCUSSION

Important softening anomalies on the compressibility modulus perpendicular to the organic layers were previously reported for all members of the $\kappa$-(BEDT-TTF)$_2X$ family. For metallic and superconducting compounds at ambient pressure, $X =$ Cu[N(CN)$_2$]Br and Cu(SCN)$_2$, wide elastic anomalies centered respectively near 38 and 50 K were attributed to a coupling of longitudinal acoustic phonons with antiferromagnetic (AF) fluctuations [29]. An identical anomaly was also observed above 35 K for the antiferromagnet member, $X =$ Cu[N(CN)$_2$]Cl, when submitted to $\sim$300 bars of pressure that shifts the compound into the metallic portion of the pressure-temperature phase diagram [30]. These measurements indicated clearly that this peculiar softening anomaly is closely related to the Mott critical point and to the pseudogap crossover line of the phase diagram. It was also shown that the anomaly survives above 30 K in the AF Mott insulating portion of the diagram at low pressures, an observation indicating that electron-electron interactions are still active in this temperature range above the Néel temperature [30]. No frequency and magnetic field effects could be detected on these softening anomalies.

For the spin-liquid candidate [$X =$ Cu$_2$(CN)$_3$], the softening anomaly appears to be different from the other compounds in several respects. The anomaly peaks at a much lower temperature (6K) over a narrower range and its amplitude is almost two orders of magnitude smaller ($\Delta V/V \sim 10^{-4}$ instead of $10^{-2}$). Moreover, the anomaly occurs in clear coincidence with the outcome of a second order phase transition at $T_p \simeq$ 6K, below which substantial magnetic field effects are observed. Finally, if we add the frequency effects, these observations suggest a different origin for the elastic softening anomaly, most likely related to a spin-liquid state, apparently not related to the Mott transition, even though all these systems present similar electronic properties. The true nature of the observed spin-liquid state in $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ remains elusive in spite of several experimental results indicating a coupling between charge, spin, and lattice degrees of freedom, and in spite of various theoretical proposals and numerical calculations [11,25,26,31–34]. We will now examine a few scenarios that most likely could explain the softening of the lattice compressibility along a direction perpendicular to the $(BEDT\text{-}TTF)_2$ organic planes at low temperatures around 6K.

It is unlikely that charge degrees of freedom could be responsible for the velocity softening peaks observed near 6K. Though there is a well defined anomaly in the temperature dependence of the in-plane microwave constant [6] at $T_p$, significant charge effects on the compressibility modulus are not expected in this compound if we consider their absence in organic systems where real charge ordering (CO) transitions occur [35]. A coupling between the spin degrees of freedom and the lattice remains then the most probable mechanism to render an account of these elastic anomalies.

Since there is no long range magnetic order in this system down to 32 mK, the softening peaks could arise from a coupling (magnetoelastic) between acoustic phonons and magnetic fluctuations. In one-dimensional systems, the theoretical model predicts a softening peak centered near a temperature of the order of the intrachain exchange interaction $J$, in perfect agreement with experiment [36]. The model is not applicable here since the softening peak is observed near 6K when the exchange interaction is approximatively 250 K. Moreover, the frequency dependence of the softening peak, its position, and its amplitude cannot be explained by the model. It is known that the electronic spin relaxation rate, which is constant from 100 K, decreases markedly below $T_p \simeq 6$ K [3]. However, we do not see how an increase of the spin relaxation time at 6K could yield a peak in the velocity softening and, in addition, with the wrong frequency dependence.

One defining property of quantum spin liquids is the existence of deconfined spinons, which are exotic uncharged excitations usually with spin 1/2, obeying bosonic or fermionic statistics and may or may not have a gap. When spinons form a Fermi surface, they are generally accompanied by an emergent gauge field [25,26]. Observations of metal-like specific heat and thermal conductivity [37] have given strong support to this *fermionic spinon* picture. A spin transport experiment was recently proposed to probe these spinons [27]. Moreover, the observation of the 6K transition on various properties suggests that the Fermi surface may be unstable due to a pairing instability [10]. Besides, low-temperature thermal conductivity measurements in magnetic fields were recently explained by a model of a strongly correlated quantum spin liquid (SCQSL) located near the fermion condensation phase transition [38]. In relation to ultrasonic measurements, the spinon-phonon interaction was theoretically investigated with the intention of identifying the pairing transition of the spinons [28]. It was shown that the ultrasonic transverse attenuation should be particularly sensitive to the gauge field fluctuations. Although the effects of the spinon-phonon interaction on the ultrasonic velocity have not been addressed theoretically, they could be a plausible explanation for the softening peak observed on the longitudinal velocity.

This spinon-phonon interaction implies the existence of low energy excitations, magnetic in nature, that can couple to acoustic phonons, as observed here. So, the increase of the softening below 20 K could coincide with the progressive appearance of a spinon Fermi surface; considering the low energy scale of the acoustic phonons, the observed frequency dependence is surprising but not excluded, considering that these fermionic quasiparticles are highly mobile [37]. Then, the modification of the softening at $T_p \simeq 5.9$ K could signal a pairing instability of the Fermi surface and a possible nodal gap structure that is analogous to $d$-wave superconductivity [10]; the reduction of the softening below $T_p$ appears consistent with a reduction in the density of states of the spinons that is not expected to be dependent on frequency, as observed. The nonzero value of $(\Delta V/V)_S$ observed as $T \to 0$ (Fig. 2) is consistent with a finite intrinsic density of states in the nodal gap scenario or with impurities [11]. The application of a magnetic field not only reduces $T_p$, but increases the quasiparticle density of states so as to enhance back the softening only below 5.9 K, an observation consistent with the reported magnetic effects on the thermal conductivity at low temperatures [37,38]. In-plane magnetic field measurements were not possible during these experiments, but they are expected to be identical according the previously observed isotropic field dependence of $T_p(H)$ [6]. According to Zhou *et al.* [28], the attenuation should decrease below the critical temperature $T_p$ when the quasiparticles are gapped. The decrease of $\Delta\alpha$ at 150 MHz (Fig. 3) below $T_p$ and its shift with field could be consistent with this picture; however, at 210 MHz, the decrease of attenuation below $T_p$, which is replaced by an increase below 5 K, deviates from the prediction.

Finally, the suggestion of the existence of spinons at low temperatures requires one to modify the explanation for the dielectric anomaly appearing at the same temperature $T_p(0)$ [6], considering that the spinons are uncharged quasiparticles. As indicated by Sedlmeier *et al.* [24], the absence of modification in the charge distribution around 6K indicates that the dielectric relaxation is not simply due to the formation of electric dipoles on the $(BEDT\text{-}TTF)_2$ dimers; it could possibly originate from the interaction of the molecular layers with the polymeric anion sheet, which will be indirectly affected by the instability of the Fermi surface.

## V. CONCLUSION

We have performed a pulsed ultrasonic experiment on a 100 $\mu$m thick $\kappa$-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ organic crystal which is considered as a spin-liquid candidate for which the *spinon Fermi sea* model could apply. We have identified a important frequency dependent softening peak near 6K on the velocity of longitudinal waves propagating perpendicularly to the organic $(BEDT\text{-}TTF)_2$ planes. Although the attenuation is delicate to interpret on thin crystals because of possible extrinsic effects, an anomaly in the temperature dependence was also obtained consistently with the velocity data. Neither a charge-lattice coupling nor a classical magnetoelastic coupling appear appropriate to render an account of these elastic anomalies. On the one hand, charge ordering identified on the dielectric constant near 6K is not expected to produce measurable effects on the compressibility modulus. On the other hand, although the softening peak has a magnetic character, the classical model of the magnetoelastic coupling cannot predict a softening peak located near 6K when the exchange interaction is 40 times larger at 250 K. Relaxation effects were also discarded because of inconsistencies with the frequency dependence. We suggest to interpret our data below 20 K along a spinon-phonon coupling that is modified at $T_p \simeq 5.9$ K due to the loss of gapped quasiparticles. If we want to pursue further the interpretation of the ultrasonic data along a spinon-phonon coupling model, two important issues must be addressed. First, the ultrasonic experiments must be performed on much thicker crystals in order to extend the frequency range, to get more precision on the attenuation, and to successfully measure the transverse attenuation which is predicted to be sensitive to the gauge field fluctuations at the transition. Second, the theoretical treatment of the spinon-phonon coupling must be extended to evaluate the effects produced on diverse elastic moduli and, in particular, the compressibilty modulus perpendicular to the organic planes.

## ACKNOWLEDGMENTS

The authors thank P. A. Lee (MIT) for useful comments, and acknowledge stimulating discussions with C. Bourbonnais (Sherbrooke) and the technical support of Mario Castonguay. This work was supported by grants from the Fonds Québécois de la Recherche sur la Nature et les Technologies (FQRNT) and from the Natural Science and Engineering Research Council of Canada (NSERC).


[1] S. Yamashita, Y. Nakazawa, M. Oguni, Y. Oshima, H. Nojiri, Y. Shimizu, K. Miyagawa, and K. Kanoda, Nat. Phys. **4**, 459 (2008).

[2] M. Yamashita, N. Nakata, Y. Kasahara, T. Sasaki, N. Yoneyama, N. Kobayashi, S. Fujimoto, T. Shibauchi, and Y. Matsuda, Nat. Phys. **5**, 44 (2008).

[3] F. L. Pratt, P. J. Baker, S. J. Blundell, T. Lancaster, S. Ohira-Kawamura, C. Baines, Y. Shimizu, K. Kanoda, I. Watanabe, and G. Saito, Nature (London) **471**, 612 (2011).

[4] Y. Shimizu, K. Miyagawa, K. Kanoda, M. Maesato, and G. Saito, Phys. Rev. Lett. **91**, 107001 (2003).

[5] R. S. Manna, M. de Souza, A. Brühl, J. A. Schlueter, and M. Lang, Phys. Rev. Lett. **104**, 016403 (2010).

[6] M. Poirier, S. Parent, A. Côté, K. Miyagawa, K. Kanoda, and Y. Shimizu, Phys. Rev. B **85**, 134444 (2012).

[7] G. Baskaran, Phys. Rev. Lett. **63**, 2524 (1989).

[8] J. Liu, J. Schmalian, and N. Trivedi, Phys. Rev. Lett. **94**, 127003 (2005).

[9] B. Kyung and A.-M. S. Tremblay, Phys. Rev. Lett. **97**, 046402 (2006).

[10] T. Grover, N. Trivedi, T. Senthil, and P. A. Lee, Phys. Rev. B **81**, 245121 (2010).

[11] S.-S. Lee, P. A. Lee, and T. Senthil, Phys. Rev. Lett. **98**, 067006 (2007).

[12] V. Galitski and Y. B. Kim, Phys. Rev. Lett. **99**, 266403 (2007).

[13] H. Kawamura and S. Miyashita, J. Phys. Soc. Jpn. **53**, 4138 (1984).

[14] H. Li, R. T. Clay, and S. Mazumdar, J. Phys.: Condens. Matter **22**, 272201 (2010).

[15] T. Mizusaki and M. Imada, Phys. Rev. B **74**, 014421 (2006).

[16] L. F. Tocchio, A. Parola, C. Gros, and F. Becca, Phys. Rev. B **80**, 064419 (2009).

[17] H.-Y. Yang, A. M. Laüchli, F. Mila, and K. P. Schmidt, Phys. Rev. Lett. **105**, 267204 (2010).

[18] L. Balents, Nature (London) **464**, 199 (2010).

[19] T.-K. Ng and P. A. Lee, Phys. Rev. Lett. **99**, 156402 (2007).

[20] S. Dayal, R. T. Clay, H. Li, and S. Mazumdar, Phys. Rev. B **83**, 245106 (2011).

[21] C. Hotta, Phys. Rev. B **82**, 241104 (2010).

[22] M. Abdel-Jawad, J. Terasaki, T. Sasaki, N. Yoneyama, N. Kobayashi, Y. Uesu, and C. Hotta, Phys. Rev. B **82**, 125119 (2010).

[23] S. Yamashita, T. Yamamoto, Y. Nakazawa, M. Tamura, and R. Kato, Nat. Commun. **2**, 275 (2011).

[24] K. Sedlmeier, S. Elsässer, D. Neubauer, R. Beyer, D. Wu, T. Ivek, S. Tomić, J. A. Schlueter, and M. Dressel, Phys. Rev. B **86**, 245103 (2012).

[25] O. I. Motrunich, Phys. Rev. B **72**, 045105 (2005).

[26] S.-S. Lee and P. A. Lee, Phys. Rev. Lett. **95**, 036403 (2005).

[27] C.-Z. Chen, Q.-F. Sun, F. Wang, and X. C. Xie, Phys. Rev. B **88**, 041405(R) (2013).

[28] Y. Zhou and P. A. Lee, Phys. Rev. Lett. **106**, 056402 (2011).

[29] K. Frikach, M. Poirier, M. Castonguay, and K. D. Truong, Phys. Rev. B **61**, R6491 (2000).

[30] D. Fournier, M. Poirier, M. Castonguay, and K. D. Truong, Phys. Rev. Lett. **90**, 127002 (2003).

[31] W. LiMing, G. Misguich, P. Sindzingre, and C. Lhuillier, Phys. Rev. B **62**, 6372 (2000).

[32] H. Morita, S. Watanabe, and M. Imada, J. Phys. Soc. Jpn. **71**, 2109 (2002).

[33] S. Yunoki and S. Sorella, Phys. Rev. B **74**, 014408 (2006).

[34] Y. Qi, C. Xu, and S. Sachdev, Phys. Rev. Lett. **102**, 176401 (2009).

[35] M. Poirier, A. Langlois, C. Bourbonnais, P. Foury-Leylekian, A. Moradpour, and J.-P. Pouget, Phys. Rev. B **86**, 085111 (2012).

[36] B. Dumoulin, P. Fronzes, M. Poirier, A. Revcolevschi, and G. Dhalenne, Synth. Met. **86**, 2243 (1997).

[37] M. Yamashita, N. Nakata, Y. Senshu, M. Nagata, H. M. Yamamoto, R. Kato, T. Sibauchi, and Y. Matsuda, Science **328**, 1246 (2010).

[38] V. R. Shaginyan, A. Z. Msezane, K. G. Popov, G. S. Japaridze, and V. A. Khodel, Europhys. Lett. **103**, 67006 (2013).