Magnetodielectric effects and spin-charge coupling in the spin-liquid candidate κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

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Microwave measurements of the in-plane dielectric function of the spin-liquid candidate κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ revealed anomalies below 300 K that indicate that charge and spin degrees of freedom are correlated down to 1.8 K. If the first anomaly around 100 K can be explained partly by a Debye relaxation model, it signals also the approach of an inhomogeneous high-temperature quantum critical phase (QC$_H$) extending down to 6 K, where a second anomaly is observed at the crossover to the intermediate quantum critical phase (QC$_M$) within which a third anomaly is detected near 3–4 K. The low-temperature anomalies are not only dependent on microwave frequency and power, but they are also strongly modified in a highly anisotropic way by a magnetic field. These dielectric results confirm that a scenario of coupled spin and charge degrees of freedom is indeed valid in this material at low temperatures, as suggested by several theoretical approaches.

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I. INTRODUCTION

The two-dimensional organic charge transfer salts κ-(ET)$_2X$ (ET = BEDT-TTF) constitute a particular class of materials in which a rich variety of quantum phases can be studied due to the interplay between electronic correlations, low dimensionality, and frustrated magnetic interactions. In particular, the discovery of a spin-liquid behavior in κ-(ET)$_2$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 the material does not exhibit any long-range magnetic order down to 32 mK,[4] various anomalies in the physical properties have been observed approaching the spin-liquid state from high temperatures. In the high-temperature range, anomalies were found in the ^{1}H-NMR relaxation rate around 200 K,[5] the thermopower around 150 K,[6] and the dielectric response below 60 K.[7] At much lower temperatures, anomalies due to a strange phase transition near 6 K have been identified on thermodynamic,[1,4] transport,[2] and lattice[8] 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,[1,9–15] or a paired-electron crystal.[16]

Most of the theoretical studies of the spin-liquid properties have been investigated with the anisotropic triangular lattice half-filled band Hubbard model.[17–20] Considering that the degree of frustration $t'/t \sim 0.8$ (t' and t being the interdimer hopping amplitudes)[11,21,22] is smaller than previously thought,[6] one may wonder how, within this model, a quantum spin-liquid (QSL) state can be stabilized over magnetic order. In these geometrically frustrated Mott insulators, however, it has been shown that spontaneous currents and charge redistribution proportional to the scalar spin chirality may exist in the ground state;[23,24] this can lead to the appearance of dipole moments, spontaneous polarization, and multiferroic behavior. The spin-chirality ordering or fluctuations are, indeed, among the consequences of several spin-liquid models.[9,13,25] Other theoretical approaches starting from the quarter-filled band reveal the following simultaneous charge and spin frustration effects: the paired electron crystal model for which charge order and spin gap coexist in two dimensions[16,26] and the dipolar-spin liquid model where quantum electric dipoles couple to spins through the interdimer charge fluctuation.[27] Indeed, thermodynamic measurements near 6 K (Ref. 8) suggest that charge degrees of freedom must be involved in this transition; moreover, the anomalous dielectric behavior below 60 K and the possible antiferroelectric ordering of the dipoles around 9 K (Ref. 7) both require unequal site charges on the dimer unit cell. Thus, although there is still controversy about the presence of a small gap in the spin excitations at low temperatures,[1,2,28] one cannot exclude the possibility that the *gapless spin liquid* may not be the result of geometrical frustration but rather that of strong correlations between spins and charges (dipoles).

To further investigate the possible coupling between electric dipoles and spins in κ-(ET)$_2$Cu$_2$(CN)$_3$, we report an experimental investigation of the in-plane dielectric function at microwave frequencies as a function of temperature and magnetic field. Anomalous dielectric behavior is observed in both high- and low-temperature ranges. At high temperatures, the anomaly shows the typical behavior of the monodispersive Debye relaxation model, and it is thus consistent with a rapidly increasing relaxation time of the electric dipoles with decreasing temperature. At low temperatures below 20 K, several dielectric anomalies which are dependent on microwave frequency and power are systematically observed. Near 6 K, a dielectric anomaly appears consistent with a short-range antiferroelectric (AFE) ordering of the dipoles, which is followed by another dielectric anomaly around 3–4 related instead to inhomogeneous magnetic moments that are highly affected by a magnetic field. These magnetodielectric effects clearly confirm the existence of strong correlations between spins and charges in a highly inhomogeneous spin-liquid state.

II. EXPERIMENT

The κ-(ET)$_2$Cu$_2$(CN)$_3$ single crystals were grown by the standard electrochemical method.[6,29] They have the shape of small platelets whose normal direction is the a^* monoclinic axis. Crystals were selected from three growth batches to verify a possible sample's dependence of the dielectric data. We used a standard microwave cavity perturbation technique[30] to measure the complex dielectric function $\varepsilon^* = \varepsilon' + i\varepsilon''$ along the ***bc*** plane. A copper cavity resonating in the TE$_{102}$ mode at 16.5 GHz and in the TE$_{101}$ at 13 GHz was used. A quartz rod spanning the full length of the cavity allows the insertion of the sample and its precise orientation along the microwave electric field. Following the insertion of the sample, changes in the relative complex resonance frequency $\Delta f/f + i\Delta(1/2Q)$ (Q is the cavity quality factor) as a function of temperature are treated in the depolarization regime.

The crystals were chosen with the approximate shape of a prolate ellipsoid of typical dimensions $1.5 \times 0.8 \times 0.03$ mm^3. Since the low-temperatures properties are found to be dependent on strains, special care was taken to fix the crystal onto the quartz rod. For the majority of samples studied here, the crystal was directly glued onto the quartz rod with the least amount of Apiezon grease (at the top or on the thinnest axis of the ellipsoid). However, to avoid any supplementary strain, a few crystals were inserted in a mylar envelope and immobilized by thin cotton threads to prevent any strain or movement during thermal cycling. After substraction of the envelope contribution to the complex frequency shift, the absolute values of the dielectric function obtained with the ellipsoid approximation for the two fixing methods agree within 30%.

The *in-plane* dielectric function was measured in the 1.7–250 K temperature range with a variable temperature insert (VTI). As the ethylene end groups of the ET molecule remained partially disordered down to a temperature around 200 K, the crystals were cooled very slowly below to allow their full ordering in a staggered conformation around 150 K. Keeping always the microwave electric field oriented along the ***bc*** plane, a static magnetic field up to 16 T could be oriented either parallel or perpendicular to this plane. However, since the change of orientation could only be performed at room temperature by a 90° rotation of the sample on the quartz rod relative to the magnetic field axis, this implies a different thermal cycle of the crystal.

III. RESULTS

The *in-plane* dielectric function was measured on several crystals of different growth batches. If we except an uncertainty of 30% on absolute values due to errors on the determination of the depolarization factor and the crystal's volume, all studied samples show the same temperature profile below 250 K.

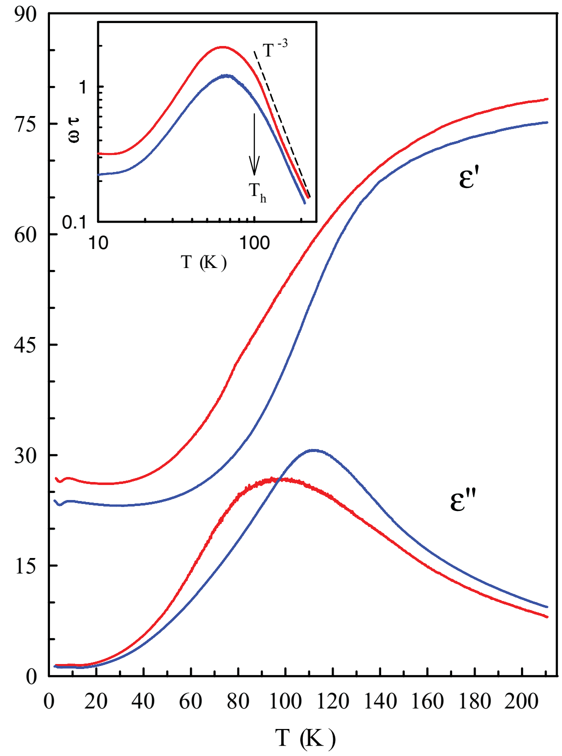

FIG. 1. (Color online) Dielectric function of a single crystal κ-(ET)$_2$Cu$_2$(CN)$_3$ as a function of temperature at 13 GHz (red) and 16.5 GHz (blue). Inset: $\omega\tau(T)$ factor according to Eq. (1); the dashed line indicates the T^{-3} power law.

We present in Fig. 1 a typical temperature dependence of both real ε' and imaginary ε'' parts of the dielectric function obtained at two microwave frequencies. When the temperature decreases below 200 K, the real part ε' at 16.5 GHz shows a steplike decrease centered around 115 K and near saturation at the lowest temperatures; concomitantly, the imaginary part passes through a broad maximum near 115 K. When the frequency is decreased to 13 GHz, both features are shifted to lower temperatures. It is tempting to associate these frequency and temperature dependences to the behavior expected from a monodispersive Debye relaxation model of the electric polarization for which the temperature dependence of the relaxation time is deduced from the ratio

$$\omega\tau(T) = \frac{\varepsilon''(T)}{[\varepsilon'(T) - \varepsilon_\infty]}, \qquad (1)$$

where ε_∞ is the high-frequency dielectric constant and ω is the angular frequency. The $\omega\tau(T)$ curves deduced at both frequencies are shown in the inset of Fig. 1. We observe that both sets of data obey a power law approaching T^{-3} well above 100 K with a value of $\tau \sim 10^{-12}$–10^{-11} s. This $\tau(T)$ profile is much faster than the one deduced for the electronic spin relaxation time, $\sim T^{-1}$, over the same temperature range above $T_h \sim 100$ K[3]. This may indicate that the spin-charge correlations are weak above 100 K. According to the phase diagram of Pratt *et al.*,[3] the system enters the high-temperature quantum critical phase (QC$_H$) below T_h, where inhomogeneous contributions to the dielectric function

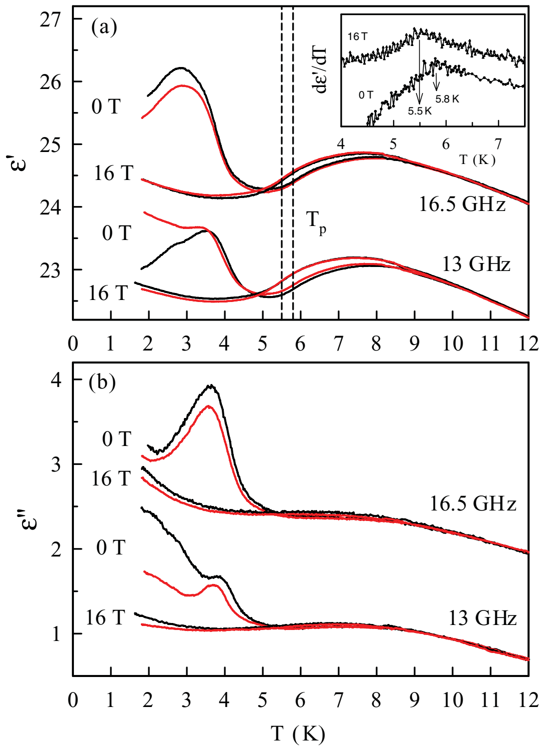

FIG. 2. (Color online) Dielectric function at 13 and 16.5 GHz below 12 K in 0 and 16 T magnetic-field values and at two microwave powers, −31 dbm (black) and −26 dbm (red). T_p indicates the boundary between phases QC_M and QC_H. (a) Real part ε' and (b) Imaginary part ε''. Inset: temperature derivative ($d\varepsilon'/dT$) of the dielectric constant at 0 and 16 T.

progressively build up and the Debye relaxation model is no longer valid.

The absolute value of the *in-plane* dielectric constant, $\varepsilon' \sim 80$, at high temperature is one order of magnitude larger than the *cross-plane* one, while the conductivity $\sigma = \omega\varepsilon_0\varepsilon''$ is higher by two to three orders of magnitude.[7] Such an anisotropy of the conductivity is fully consistent with the anisotropy generally found in the κ-(ET)$_2X$ salts.

In the low-temperature range, several dielectric anomalies are observed. Their temperature profile is very peculiar since it is not only dependent on frequency but also on average microwave power and on magnetic field. We present a typical example of such profiles below 12 K in Fig. 2. The average microwave power has been fixed to −31 dbm (0.8 μW), a value typically used in this particular microwave technique. As the temperature is decreased in zero magnetic field, the dielectric constant ε' goes through a maximum just below 8 K before decreasing down to 5 K, where it starts to increase rapidly and shows another maximum [Fig. 2(a)]; the dissipation ε'' presents a similar behavior over the same temperature range [Fig. 2(b)]. The temperature profile of both parts of the dielectric function is highly dependent on frequency below 5 K, especially on ε'', which shows quite different dissipation peaks. To understand the origin of these low-temperature anomalies, we applied a 16 T magnetic field parallel to the **bc** plane. As shown in Fig. 2, the magnetic field affects the dielectric function in the approximate temperature range 1.8–10 K: the field not only shifts the maximum near 8 K to lower temperatures, but it depresses completely the anomalies below 5 K. Another interesting observation over the same range is the dependence upon the microwave power. When the power is increased by 5 db (Fig. 2), although the amplitude of the microwave electric field E_0 has merely been multiplied by 1.8, we observe a small downshift of the maximum near 8 K together with a depression of the low-temperature anomalies, these effects being much smaller for a 16 T field. Let us mention that usually no power dependency is observed on conventional materials with such small power values.[33]

Because of the weakness of the anomalies, one may argue that these are due to impurities or a very small portion of the sample consistent with inhomogeneities. Since these anomalies appear directly linked to the *curious* 6 K feature that has been previously identified on thermodynamic, transport, and elastic properties,[1,2,4,8] we suggest here to assign them to a weak coupling between charge degrees of freedom and spins. In the inset of Fig. 2, we plotted the temperature derivative ($d\varepsilon'/dT$) curves at 16.5 GHz. In zero magnetic field, the sharp peak obtained at $T_p \simeq 5.8 \pm 0.1$ K mimics perfectly the *in-plane* expansivity data α_c,[8] which showed overall agreement with magnetic susceptibility and specific-heat data. Manna *et al.*[8] have associated their 6 K feature in the thermal expansivities to a second-order phase transition that cannot be explained only by spins. The authors suggested that charge degrees of freedom could be involved in the transition. Our data appear to agree completely with this suggestion. Moreover, a magnetic field of 16 T shifts down this transition by a few tens of degrees to 5.5 K, indicating a direct link with the spins. According to the phase diagram of κ-(ET)$_2$Cu$_2$(CN)$_3$,[3] T_p defines the boundary between the QC_H phase above 5.8 K and the QC_M (intermediate quantum critical) below. From Fig. 2, it is thus clear that new dielectric anomalies which are highly affected by both magnetic field and/or microwave power appear within the QC_M phase, confirming then a definite coupling between charge and spin.

These low-temperature QC_M and QC_H phases possess an inhomogeneous character. We compare in Fig. 3 the temperature profiles of the low-temperature anomalies at 16.5 GHz in zero magnetic field for samples submitted to different conditions of internal and/or external strains at constant microwave power: sample A, in a mylar envelope and fast cooling rate; sample B, in a mylar envelope and slow cooling rate; sample C, in a mylar envelope and slow cooling rate; sample D, same as C but held with grease and second slowest cooling rate; sample E, same as D but third slowest cooling rate. We observe that external strains (C and D) and repeated thermal cycles to room temperature (D and E) depress the low-temperature anomalies with a tendency to increase slightly T_p. Different internal strains due to the crystal growth process modify also the anomalies (B and C). From these observations, we conclude that the following anomalies are truly intrinsic to this compound: local extremum of the dielectric constant near 9 K, maximum ($d\varepsilon'/dT$) rate at T_p, and further increase of the dielectric function within the QC_M phase; however, their relative sensitivity to strains and thermal cycling are consistent with an inhomogeneous character of the QC_H and QC_M phases. To enhance this sensitivity to strain, we completely dipped a crystal in silicone fluid (Dow Corning) to mimic an increase of pressure when the temperature is

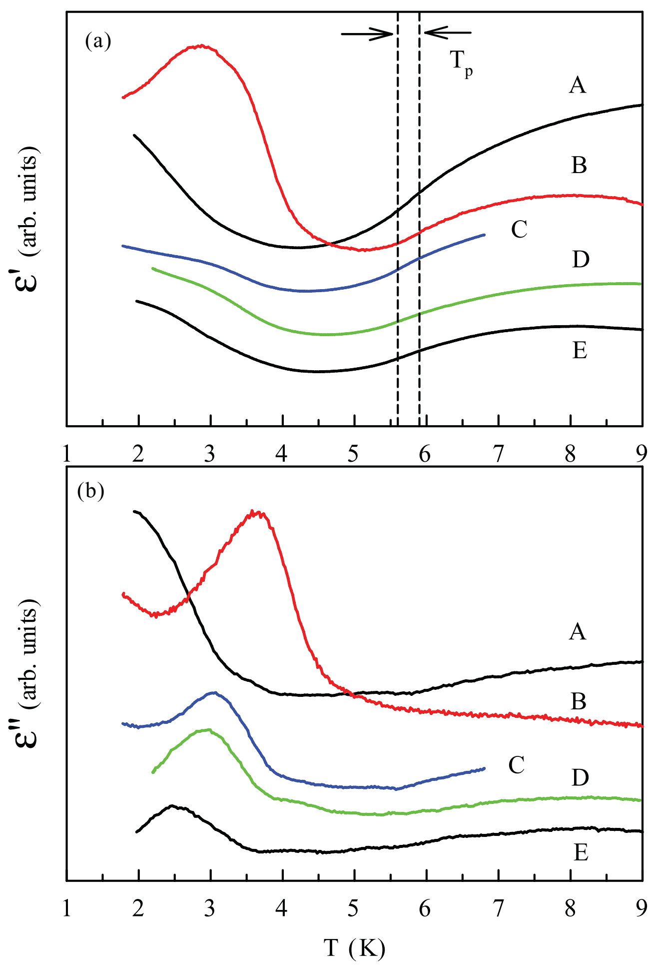

FIG. 3. (Color online) Sample, strain, and thermal cycle dependences of the low-temperature dielectric anomalies at 16.5 GHz and constant microwave power (−31 dbm). Sample A (mylar, fast cool), sample B (mylar, slow cool), sample C (mylar, slow cool), sample D (same as C, grease, second slow cool), sample E (same as C and D, grease, third slow cool). The vertical dashed lines indicate the variation of T_p.

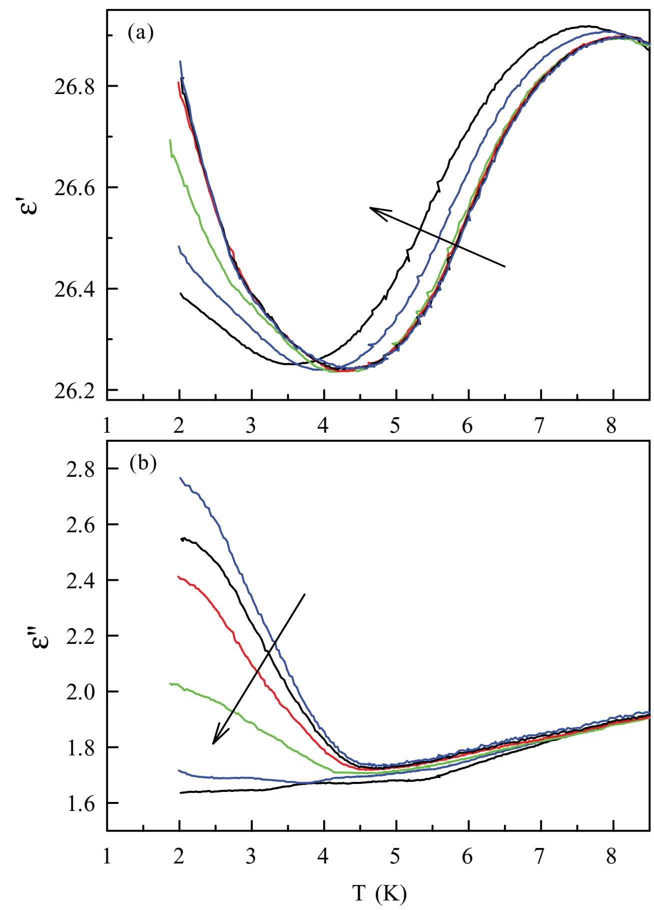

FIG. 4. (Color online) Microwave power dependence of the low-temperature anomalies at 16.5 GHz below 10 K. Minimum power 0.032 μW: the arrow indicates a power increase by steps of 5 db. (a) real part ε' and (b) ε''.

decreased (not shown here), and we noticed an important depression of the QC_M phase anomalies and an increase of T_p from 5.9 K to ∼6.7 K, in perfect agreement with the preceding observations.

An example of the effects produced by a variation of the microwave power at 16.5 GHz is presented in Fig. 4. Both parts of the dielectric function are affected by a 22 db increase of power indicated by the arrow (the microwave electric field E_0 is multiplied by ∼13): the maximum of ε' around 8 K and the concomitant transition temperature T_p are downshifted, and the low-temperature anomalies within the QC_M phase are progressively depressed. For the lowest values of the power, however, all the curves appear superposed one onto the other as if the intrinsic temperature profile has been reached. Such power effects appear to mimic the ones produced by a magnetic field of 16 T (Fig. 2). On all samples investigated, the power dependency of the dielectric function happens only below ∼10 K when approaching the boundary between the QC_H and QC_M phases at T_p.

Most existing experimental data on κ-$(ET)_2Cu_2(CN)_3$ did not reveal significative magnetic field effects: a small enhancement of the thermal conductivity in a 10 T field,[2] an absence of sensitivity to an 8 T field for the specific heat,[1] a dielectric response independent of magnetic field up to 15 T,[7] and features in the expansion coefficient α_c that are unaffected by a field of 8 T applied along the **c** axis.[8] However, muon spin rotation experiments did reveal a rich magnetic phase diagram at low temperatures for the perpendicular configuration.[3] The microwave dielectric anomalies reported here, which are highly dependent on the amplitude of the magnetic field and on its orientation with respect to the **bc** plane, indeed confirm the presence of magnetic phases. Their observation in a temperature range where other experiments fail to show substantial sensitivity to a magnetic field has to be attributed to an appropriate time scale of the charge and spin fluctuations.

We present in Fig. 5 the temperature dependence of the dielectric function below 8 K for different values of the magnetic field up to 16 T oriented parallel to the **bc** plane. For this crystal in zero field, the anomaly on the dissipative part ε'' shows a peak around 3.6 K when the real part ε' increases rapidly. The inset of Fig. 5(b) establishes a clear correspondence between the dissipation peak ε'' and the

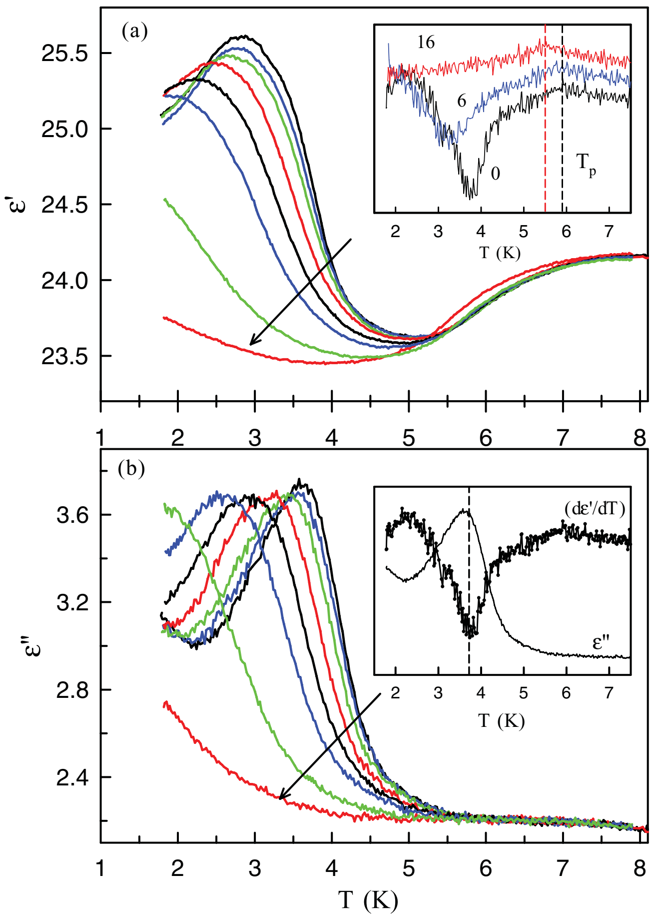

FIG. 5. (Color online) Dielectric anomalies at 16.5 GHz (0.8 μW power) as a function of temperature at different values of the magnetic field (0, 1.0, 2.0, 4.0, 6.0, 8.0, 12.0, and 16.0 T) applied parallel to the **bc** plane: (a) real part ε' [inset: $(d\varepsilon'/dT)$ at selected field values]; (b) imaginary part ε'' [inset: comparison of $(d\varepsilon'/dT)$ and ε'' in zero field]. The arrows indicate the increasing field.

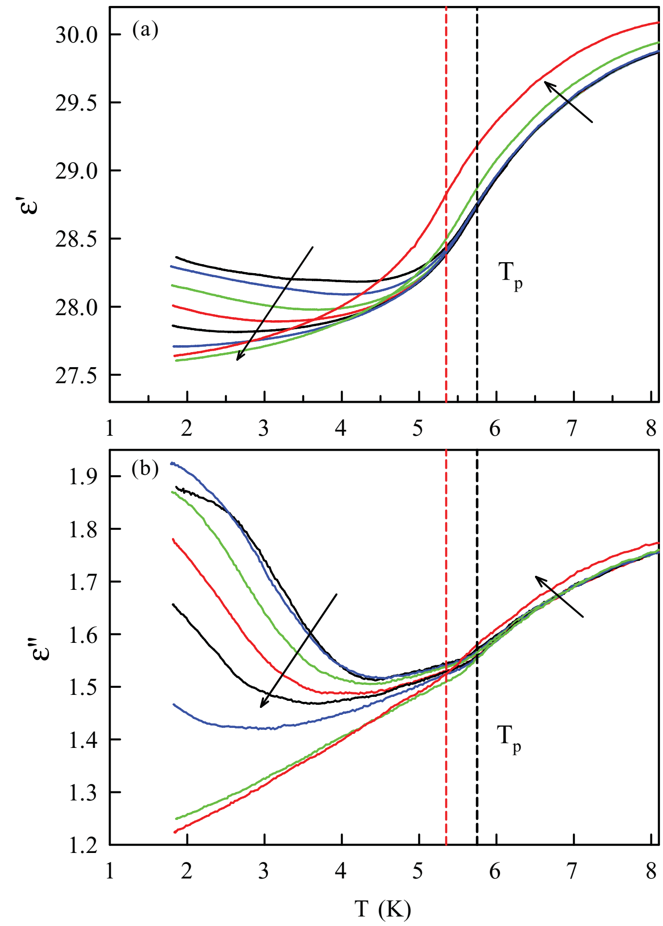

FIG. 6. (Color online) Dielectric anomalies at 16.5 GHz (0.8 μW power) as a function of temperature at different values of the magnetic field (0, 0.5, 1.0, 1.5, 2.0, 3.0, 8.0, and 16.0 T) applied perpendicular to the **bc** plane: (a) real part ε'; (b) imaginary part ε''. The arrows indicate the increasing field. The dashed lines indicate the transition temperature T_p in 0 and 16 T magnetic field.

temperature derivative of the dielectric constant $(d\varepsilon'/dT)$, a correspondence that gives us a criterion to follow the anomalies as the field is increased. The magnetic field progressively shifts down the anomalies until they are almost completely depressed at 16 T. In the inset of Fig. 5(a), the derivative $(d\varepsilon'/dT)$ reveals not only a downshift of the 3.6 K anomaly but also a progressive decrease of T_p. A very similar behavior in magnetic field is observed for all samples, although a well-defined anomaly is not always obtained. For example, there is no such anomaly for sample A shown in Fig. 3: for this particular sample, we had to follow instead the displacement of the ε' minimum around 4.5 K for an increasing field. The criterion used to quantify the magnetic field effects is thus dependent on the particular temperature profile of the anomalies. Recently, a field-induced length change anomaly has been reported near 8.7 K for a magnetic field $H = 1$ T oriented along the in-plane **b** axis.[31] Although the field was not oriented intentionally along a particular direction of the **bc** plane, we did not find measurable traces of such an anomaly in our microwave experiments in the temperature range above 7 K (Fig. 5), where the spin-charge coupling is vanishingly small.

When the magnetic field is oriented perpendicularly to the **bc** plane, the anomalies are completely depressed for much smaller values, as shown in Fig. 6 for another sample (different from the one in Fig. 5). A field of only 3 T is sufficient to produce the same depression of the low-temperature anomalies as the one observed for a parallel 16 T field. However, the downshift of T_p is found to be the same. We can summarize the effects of the magnetic field by establishing a tentative H-T phase diagram shown in Fig. 7, which includes the data obtained on three samples. As previously discussed, the zero-field location of the lines is dependent on strain and microwave power, but their curvature as the field is increased is reproduced on all samples. The $T_p(H)$ line is not dependent on frequency and appears isotropic as long as the average microwave power is approximatively the same: the differences observed in $T_p(0)$ (Fig. 7) are due to different strain and microwave power conditions. This $T_p(H)$ line establishing a well-defined boundary between the QC_H and QC_M phases is depressed quasiquadratically with field. The low-temperature anomalies within the QC_M phase yield lines that are dependent on the criterion used to define the anomaly. Since their temperature profiles depend on frequency, power, and strain,

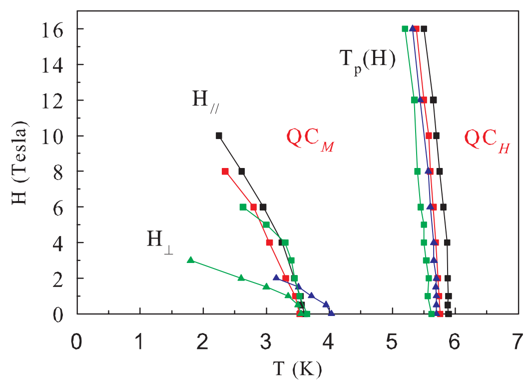

FIG. 7. (Color online) *Tentative* H-T phase diagram of κ-$(ET)_2Cu_2(CN)_3$ at low temperatures. Magnetic field parallel (□), perpendicular (△): Sample 1 at 16.5 GHz (black) and 13 GHz (red); sample 2 at 16.5 GHz (blue); sample 3 at 16.5 GHz (green).

we expect these lines to be scattered over a wide temperature range between 2.5 and 4.5 K depending on the criterion used to characterize a particular anomaly. Although these lines are depressed for either the parallel or the perpendicular field configuration, there is an important anisotropy relative to their quasiquadratic dependence. The perpendicular line seems to extrapolate to zero temperature at a field below 4 T, while a field greater than 16 T is expected for the parallel line. How this diagram is affected by pressure remains to be investigated, but our study of the strain effects suggests an increase of $T_p(0)$ with a drastic depression of the QC_M phases anomalies.

IV. DISCUSSION

In agreement with recent theoretical approaches,[16,26,27] our microwave measurements confirm the presence of electric dipoles in κ-$(ET)_2Cu_2(CN)_3$ from room temperature down to 1.8 K. Although the in-plane dielectric constant ε' remains high over the full temperature range, it shows a sensitivity to the spin degrees of freedom through a sequence of anomalies. When the temperature is decreased from room temperature, the dielectric function reveals a relaxation time that increases much faster (T^{-3}) than the electronic spin relaxation rate,[3] indicating a weak coupling between spin and charge degrees of freedom. This increase is, however, stopped near $T_h \sim 100$ K when, according to the phase diagram of Pratt *et al.*,[3] the system enters the QC_H phase for which the electronic spin relaxation rate is almost constant. Entering this phase, the dielectric constant undergoes an important decrease and dielectric inhomogeneities progressively build up. When the temperature has decreased well below T_h, there is a clear tendency to an enhanced electric polarization which accelerates as the QC_H-QC_M phase boundary at $T_p \sim 6$ K is approached. The observation of a local maximum of ε' around 8 K and its further decrease showing a maximum rate in $(d\varepsilon'/dT)$ at T_p is consistent with a short-range AFE ordering of the electric dipoles, as suggested by the out-of-plane dielectric measurements.[7] The dielectric transition around 6 K is accompanied by a change of the magnetic susceptibility $(d\chi/dT)$,[4,8] which conforms well with thermodynamic quantities, and by a decrease of the spin fluctuation rate[3]; this indicates that spin degrees of freedom are involved to some extent in the transition. As the low-temperature phases (below T_h) are inhomogeneous, the short-range order and the transition temperature T_p are dependent on thermal cycling and internal/external strains.

When entering the QC_M phase below T_p, the dielectric constant ε' is enhanced at microwave frequencies with a concomitant increase of the losses (ε''). Because of the dependence on the amplitude and frequency of the microwave electric field in addition to strains, which affects their temperature profile, it is difficult to determine precisely the exact number of dielectric anomalies appearing within the QC_M phase. The results presented here suggest at least two anomalies that are related to a strengthening of the short-range AFE ordering due to the important coupling to the spin degrees of freedom. Even if these are not considered as phase transitions like the one at T_p because their temperature location depends on frequency, they appear in a temperature range where the **c** axis expansivity data revealed a smaller and reproducible anomaly around 3 K (Ref. 8) within the QC_M phase.

The short-range AFE ordering at T_p and the dielectric enhancement within the QC_M phase are both dependent upon the spins degrees of freedom, and a possible explanation is the outcome of an anisotropic short-range antiferromagnetic (AFM) order consistent with the emergence of inhomogeneous moments reported in NMR experiments.[32] In zero field, the dielectric anomalies express an enhancement of electric polarization due to a buildup of a short-range AFM order; several anomalies indicate necessarily successive transformations of this AFM order. An important magnetic field, either parallel or perpendicular to the **bc** plane, likely depresses the AFM order, inhibits the spin-dipole coupling channel, and then enhances the AFE fluctuations with a consequent shift of T_p to smaller temperatures. Why are these magnetic field effects highly anisotropic? If there exists an easy-plane anisotropy coinciding with the plane of the triangle, a canting of the spins away from the **bc** plane could be a good candidate: if the effect of canting is interpreted as a reduction of frustration, we can understand why a perpendicular field could increase frustration more rapidly than a parallel one when the spins become finally collinear, and delay the AFE transition to lower temperatures.

How is the dependence on microwave power explained within this picture? An increase of power translates into an important increase of the in-plane microwave electric field that likely inhibits the spin-dipole coupling channel; then, the short-range AFE fluctuations appear enhanced and the short-range AFM fluctuations evolve independently. We thus understand why an increase of microwave power mimics an increase of magnetic field by inhibiting the spin-dipole coupling channel as observed in Figs. 4 and 5. The coupling of the spins to the electric dipoles relieves frustration and favors short-range AFE and AFM orders. The dielectric anomalies observed within the QC_M phase in zero field could be related to successive partial reliefs of magnetic frustration. Their unique observation in the microwave frequency range relies on the appropriate time scale of the AFE and AFM fluctuations.

Our dielectric measurements are qualitatively consistent with the theoretical approaches predicting charge and spin frustration effects. Among these approaches, the model implying spontaneous orbital currents or nonuniform charge distribution proportional to a scalar spin chirality[23,24] appears possibly adequate to explain the low-temperature microwave

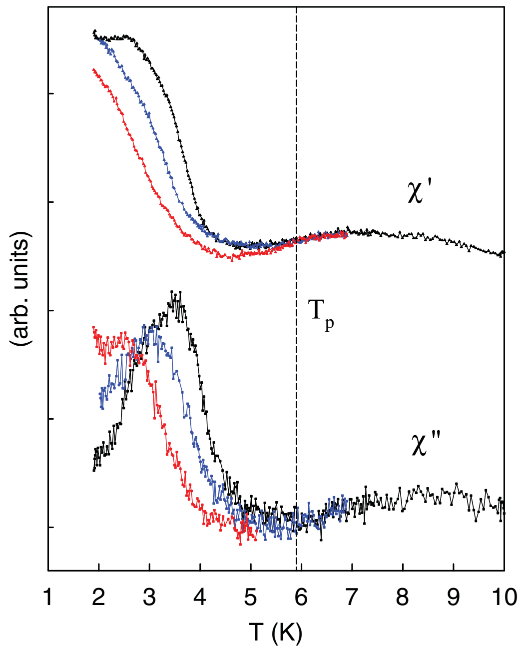

FIG. 8. (Color online) Magnetic susceptibility function perpendicular to the **bc** plane at 16.5 GHz as a function of temperature. The transition temperature T_p is indicated by the dashed line. A magnetic field is oriented in-plane: 0 (black), 4 (blue), and 8 T (red).

anomalies and the anisotropic magnetic field effects. Although this spin chirality implies a magnetic state which breaks time-reversal invariance, it does not show the usual long-range magnetic order and could be an example of a time-reversal–broken spin liquid. According to this approach, low-energy magnetic states contribute comparably to the dielectric and magnetic responses functions, and the anisotropic magnetic field effects could be explained by considering easy-plane anisotropy. Indeed, as for the dielectric function, anomalies are also observed on the perpendicular microwave magnetic susceptibility, $\chi = \chi' + i\chi''$, within the QC_M phase, as shown in Fig. 8. Although these magnetic susceptibility anomalies are difficult to observed because of a low sensitivity of the microwave technique for magnetic measurements on platelet samples, we observe a small maximum of χ' just below 8 K and a further increase around 3.5 K, compatible with either a ferromagnetic component or orbital moments created by spontaneous orbital currents within the triangle's plane; there is a concomitant peak on the imaginary part χ'. These features are progressively shifted and depressed to lower temperatures as a parallel field is increased in a fashion similar to the dielectric anomalies. Because of the weakness of these anomalies, we could not verify a dependency on the amplitude of the microwave magnetic field. Consistent with an in-plane anisotropy of the AFM order, no measurable in-plane magnetic susceptibility could be observed on any of the studied samples. In a recent report, Nakajima *et al.*[34] reveal at zero field below 3 K a microscopic phase separation between paramagnetic islands and a singlet sea. In our microwave experiment, the observation of the inhomogeneous character of the QC_M phase and the anisotropic short-range AFM order are fully consistent with this report, and they are thus considered as intrinsic features of this spin-liquid compound.

V. CONCLUSION

The microwave measurements of the in-plane dielectric function confirm that a spin-charge coupling dominates the low-temperature electronic properties of the spin-liquid system κ-$(ET)_2Cu_2(CN)_3$. From these measurements, the strange 6 K feature expresses a short-range AFE transition that is strongly affected by the spin-charge coupling. Although frustration prevents any long-range magnetic order, anomalies of the dielectric function within the QC_M phase below 6 K are possibly due to short-range in-plane AFM ordering of the spins with a ferromagnetic component or to orbital moments along the perpendicular direction that originate from spontaneous orbital currents. The degree of frustration in these low-temperature inhomogeneous phases can be affected by a magnetic field and/or the amplitude of the microwave electric field. These dielectric measurements confirm the pertinence of novel theoretical approaches in which nontrivial charge degrees of freedom that survive in the dimer Mott insulator must be taken into account to characterize the spin-liquid state of this organic compound.

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