Combination frequencies of magnetic oscillations in β''-(BEDT-TTF)$_4$(NH$_4$)[Fe(C$_2$O$_4$)$_3$]·DMF

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Interlayer magnetoresistance of the quasi-two-dimensional organic superconductor β''-(BEDT-TTF)$_4$(NH$_4$)[Fe(C$_2$O$_4$)$_3$]·DMF has been investigated in pulsed magnetic fields of up to 55 T in the temperature range from 1.6 K to 4.2 K. According to band-structure calculations, the Fermi surface of this compound originates from hybridized intersecting hole tubes with a cross sectional area equal to the first Brillouin-zone (FBZ) area, leading to two compensated electron and hole orbits. Actually, the Fourier spectrum of the oscillatory magnetoresistance exhibits various frequencies which can be regarded as linear combinations of the two frequencies with the highest amplitude [$F_a=(48\pm2)T$ and $F_b=(241\pm5)T$] corresponding to 1.2% and 6.0% of the FBZ area, respectively. The oscillatory spectrum can be accounted for by three compensated electron and hole orbits and combination frequencies typical of networks of orbits coupled by magnetic breakthrough.

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I. INTRODUCTION

One of the unsolved questions regarding magnetic oscillations in metals deals with combination frequencies that are observed in two-dimensional (2D) multiband systems even though their occurrence is "forbidden" within the semiclassical theory of magnetic breakthrough (MB) by Falicov and Stachowiak.[1] In that respect, the organic charge-transfer salts of the κ-phase,[2] constitute experimental realizations of the linear chain of coupled orbits introduced by Pippard[3] in the early 1960s. Although these compounds, in particular, κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ [where BEDT-TTF stands for bis(ethylenedithio)tetrathiafulvalene], have been extensively studied, the physical origin of some of the observed combination frequencies remains unclear. Actually, besides MB-induced closed orbits, quantum interference (QI),[4,5] oscillation of the chemical potential,[4,6–9] and MB-induced field-dependent broadening of the Landau levels[10,11] have been invoked in order to interpret the data. Numerical computation of the de Haas–van Alphen (dHvA) oscillation spectrum, based on a realistic tight-binding model and including MB, also evidenced combination frequencies at high magnetic field, both in the framework of the canonical and grand canonical ensembles.[12] Similarly, magnetic oscillations in the 2D network of compensated closed orbits provided by the Fermi surface (FS) of the organic metals (BEDT-TTF)$_8$Hg$_4$Cl$_{12}$(C$_6$H$_5$$X$)$_2$ (X=Cl, Br) have revealed, in addition to few closed orbits and one two-arm interferometer, numerous combination frequencies that cannot be accounted for by the semiclassical model.[13]

The molecular salts of the

$$\beta''\text{-(BEDT-TTF)}_4(A)[M(\text{C}_2\text{O}_4)_3]\cdot\text{Solv}$$

(A=H$_3$O; M=Cr, Fe, Ga; Solv = benzonitrile, nitrobenzene, pyridine) family exhibit various ground states and temperature-dependent physical behaviors despite the fact that all of these compounds have a similar structure.[14–19] The crystal structure, which has been described in detail elsewhere[15,19–21], consists of layers of the BEDT-TTF donors with the well-known β''-type packing and anionic layers in between. These layers are built from $[M(\text{C}_2\text{O}_4)_3]^{3-}$ anions and A^+ cations which leave cavities filled by the solvent molecules (Solv). Although the conductivity of these salts originates from the BEDT-TTF donor layers it has been found that changing the nature of the solvent molecules in the anionic layers strongly alters the conductivity. For instance, whereas

$$\beta''\text{-(BEDT-TTF)}_4(\text{H}_3\text{O})[\text{Fe}(\text{C}_2\text{O}_4)_3]\cdot\text{benzonitrile}$$

is superconducting with T_c=8.5 K,[14]

$$\beta''\text{-(BEDT-TTF)}_4(\text{H}_3\text{O})[\text{Fe}(\text{C}_2\text{O}_4)_3]\cdot\text{pyridine}$$

exhibits a metal-insulator transition at 116 K.[16] The characterization of the FS of the different members of this family of 2D conductors is thus extremely interesting because of the possibility to correlate subtle variations in the crystal structure with the different conductivity behaviors.

Recently, Prokhorova *et al.*[20] have reported three new metallic salts of the above family. As reported below, some hints of a superconducting ground state are found for one of them, β''-(BEDT-TTF)$_4$(NH$_4$)[Fe(C$_2$O$_4$)$_3$]$\cdot$DMF (where DMF stands for the dimethylformamide solvent). The FS of this organic metal originates from one quasi-2D elliptic hole tube with a cross section area equal to the first Brillouin zone (FBZ) area. In the following, the corresponding orbit is referred to as the $\odot$ orbit. In the extended zone scheme, $\odot$ tubes intersect in the $\Gamma' M'$ direction[22] (see Fig. 1). Removals of degeneracy yield the resulting FS which is composed of one electron (E) and one hole (H) tube, located around X' and M', respectively. The calculated cross section of these compensated orbits is 8.8% of the FBZ area. Nevertheless, as pointed out in Ref. 20, the Fermi level lies in a very small energy gap in the region close to Y' so that an additional small orbit labeled o in Fig. 1(b) may be present in this region. In that respect, it can be mentioned that the calculated FS of the isostructural compound β''-(BEDT-TTF)$_4$(NH$_4$)$_{0.75}$K$_{0.25}$[Cr(C$_2$O$_4$)$_3$]$\cdot$DMF exhibits quite similar feature.[20] Provided at least one of the MB gaps Δ_{oE} and Δ_{EH} between the orbits of this multiband system is not too large in view of the strength of the magnetic field will give rise to a network of coupled orbits. As reported in this paper, this latter picture actually holds for this compound.

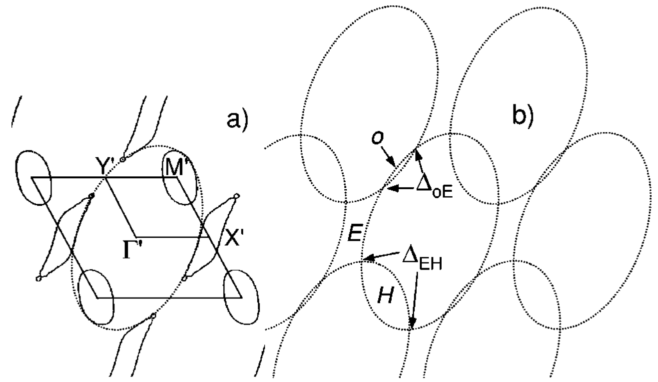

FIG. 1. Fermi surface of β''-(BEDT-TTF)$_4$(NH$_4$)[Fe(C$_2$O$_4$)$_3$] $\cdot$DMF according to band-structure calculations (Refs. 20 and 22) [Fig. 1(a)] and schematic representation of intersecting elliptic tubes in the extended zone scheme [Fig. 1(b)]. The cross section of the ellipses in dotted line is equal to the FBZ area. Latin labels are discussed in the text.

II. EXPERIMENT

The studied crystal is a hexagonal platelet with approximate dimensions $0.6\times0.4\times0.25$ mm^3, the largest faces being parallel to the conducting *ab* plane. Four terminal magnetoresistance experiments were performed in pulsed magnetic field of up to 55 T (pulse decay time 0.18 sec) in the temperature range from 1.6 K to 4.2 K. A rotating sample holder allowed us to change the direction of the magnetic field with respect to the conducting plane. Electrical contacts were made to the crystal using annealed Pt wires of 20 μm diameter glued with graphite paste. Alternating current (2 μA, 20 kHz) was injected parallel to the c^* direction (interlayer configuration). A lock-in amplifier with a time constant of 100 μs was used to detect the signal across the potential leads.

III. RESULTS

The temperature dependence of the interlayer resistance of the studied crystal, normalized by its room-temperature value, is plotted in Fig. 2. A metallic behavior is observed down to $\sim$38 K while a resistance increase is observed at lower temperature, followed by a resistance decrease below $\sim$2 K that is suppressed by a very low magnetic field (see the inset of Fig. 2). Such a behavior is very similar to the temperature dependence of the resistivity of β''-(BEDT-TTF)$_4$(H$_3$O)[Ga(C$_2$O$_4$)$_3$](C$_5$H$_5$N).[21] Indeed, this monoclinic salt, which is isostructural to the compound studied, displays a resistance minimum, although at higher temperature ($\sim$150 K), which is followed by a superconducting transition below 2 K.

The interlayer magnetoresistance, measured with the magnetic field applied perpendicular to the conducting plane, and the corresponding Fourier spectra are displayed in Fig. 3. At low temperature, the two oscillations with the highest amplitude are at frequencies $F_a = (48\pm2)T$ and $F_b = (241 \pm 5)T$ that correspond to 1.2% and 6.0% of the FBZ area, respectively. In addition to these two frequencies, two oscillations with frequencies of $(192\pm2)T$ and $(283\pm5)T$ which can be ascribed to F_{b-a} and F_{b+a}, respectively, are observed in the Fourier spectra. A frequency $F_{2b} = (480 \pm 20)T$, which is equal to $2F_b$ within the error bars, is observed more clearly in the high field range of the magnetoresistance [see Fig. 3(a)]. The splitting of the b oscillation observed above $\sim$3 K in Fig. 3(b), which is not observed for a magnetic field tilted away from c^* [see Fig. 4(b)], arises likely from some artifact with no physical meaning.

In the framework of the 2D Lifshits-Kosevich (LK) model[23] and neglecting for the moment any eventual contribution of MB and spin damping, the field and temperature

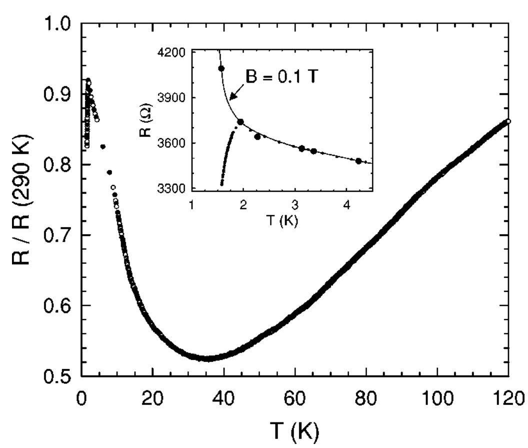

FIG. 2. Temperature dependence of the normalized interlayer resistance. The inset shows the lowest temperature part of the interlayer resistance both in zero magnetic field and at 0.1 T.

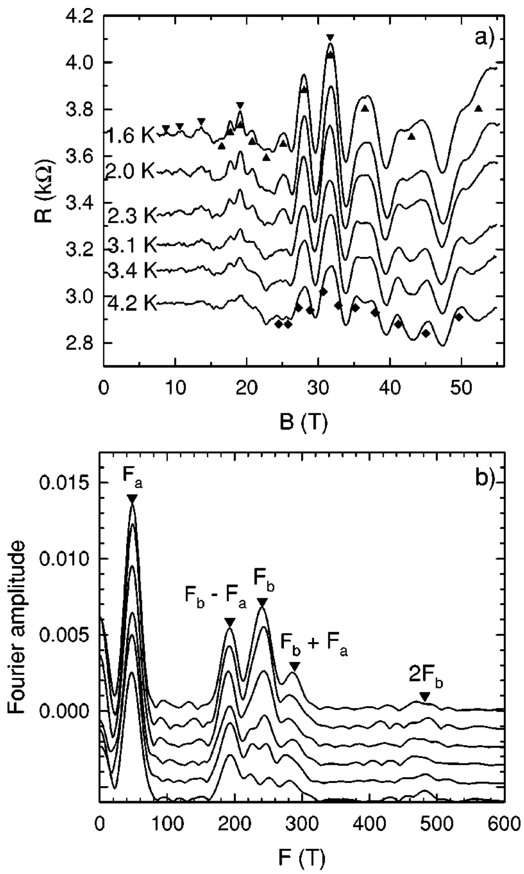

FIG. 3. Interlayer magnetoresistance at various temperatures (a) and corresponding Fourier spectra of the oscillatory magnetoresistance calculated in the range 10–55 T (b). The curves have been shifted from each other for clarity. Down triangles, up triangles and diamonds in (a) are marks calculated with $F_a = 48T$, $F_b = 241T$, and $F_{2b} = 482T$, respectively. Down triangles in (b) are marks calculated with $F_a = 48T$ and $F_b = 241T$.

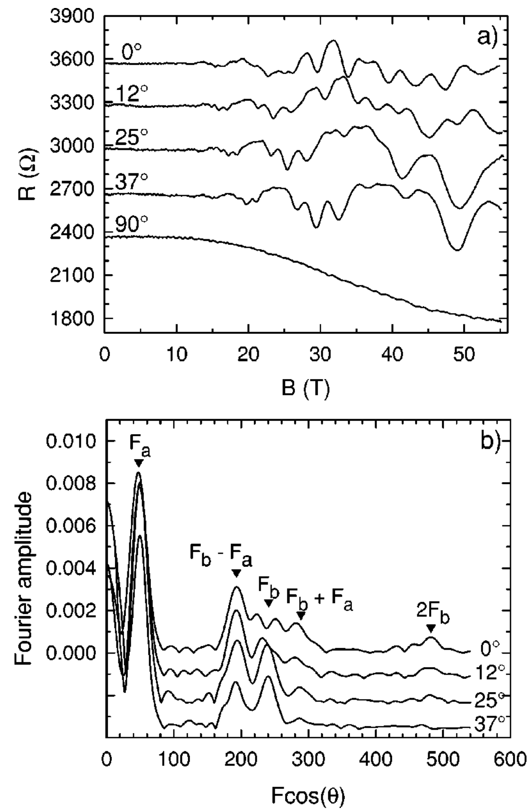

FIG. 4. Interlayer magnetoresistance (a) and corresponding Fourier spectra of the oscillatory magnetoresistance calculated in the range 10–55 T (b) for various directions of the magnetic field (θ is the angle between the field direction and the normal to the conducting *ab* plane) at 4.2 K. The curves have been shifted with respect from each other for clarity. Down triangles in (b) are marks calculated as in (b).

dependence of the oscillations amplitude (A) is given by $A \propto R_T R_D$. The thermal (R_T) and Dingle (R_D) damping factors are given by $R_T = u/\sinh(u)$ where $u = 2\pi^2 k_B m_0 T m_c / \hbar e B$ and $R_D = \exp(-u_D)$ where $u_D = u T_D / T$. The LK model formally accounts for the field and temperature dependence of the various oscillations mentioned above (see, e.g., Fig. 5 for the temperature dependence of the amplitude). The effective masses (m_c) and Dingle temperatures (T_D) deduced from the data analysis are given in Table I. Remarkably, even though the amplitude of the $2b$ oscillation exhibits a significant magnetic field dependence in the explored temperature range [$A \propto \exp(-B_0/B)$ with $B_0 = (140 \pm 30)T$] it remains temperature independent. This latter feature accounts for an apparent zero effective mass.

IV. DISCUSSION

The temperature and field dependence of the interlayer resistance displayed in Fig. 2 is in agreement with a superconducting ground state, as it is the case for other monoclinic salts of this family.[21,24] A large resistance rise is observed in the temperature range from $\sim$38 K down to the onset of the superconducting transition (see Fig. 2) and a negative magnetoresistance is evidenced in Fig. 4(a) for magnetic field applied parallel to the conducting *ab* plane. Analogous behavior of the temperature[25] and magnetic field[26] dependencies of the resistivity have already been observed in the $(BEDO\text{-}TTF)_2ReO_4 \cdot H_2O$ superconductor which is known to undergo several phase transitions as external parameters such as temperature, magnetic field and pressure are varied. It can be remarked that the FS of this latter compound bears similarities with that of β''-$(BEDT\text{-}TTF)_4(NH_4)[Fe(C_2O_4)_3] \cdot DMF$, namely, compensated electron and hole orbits originating from intersecting $\odot$ orbits.[27]

According to band-structure calculations,[20] electron and hole orbits are compensated. Only one frequency should then be observed in the Fourier spectrum unless an additional orbit [labeled *o* in Fig. 1(b)] is present in the region close to the point Y' of the FBZ, as mentioned in the introduction. As a matter of fact, the oscillatory spectrum involves several oscillations whose frequencies are linear combinations of F_a and F_b. Provided the FS schematized in Fig. 1(b) is still valid at low temperature, it is likely that F_a, which corresponds to 1.2% of the FBZ area, only, is linked to the small *o* orbit. Assuming, as suggested in Ref. 20, that the FS can be regarded as arising from $\odot$ orbits intersecting in both directions $\Gamma' M'$ and $\Gamma' Y'$, the *o* orbit should have a hole char-

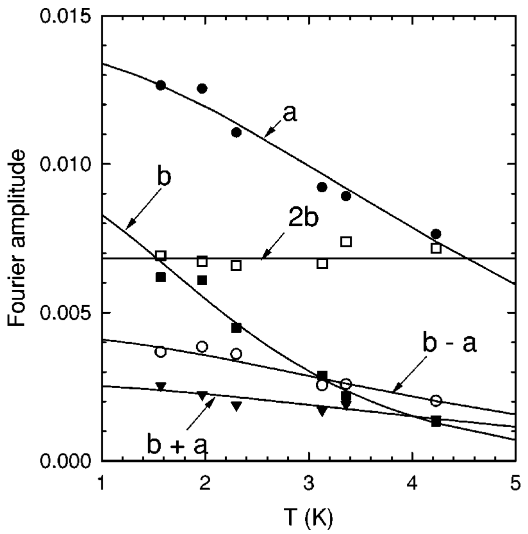

FIG. 5. Temperature dependence of the amplitude of the various oscillations evidenced in Fig. 3. The mean value of the magnetic field is $\overline{B}=17$ T except for the $2b$ oscillation for which $\overline{B}=33$ T.

acter. Nevertheless, in the case where the FS region located around Y' is of electron character, a set of two quasi-one-dimensional sheets parallel to the $\Gamma' M'$ direction should be generated rather than a closed orbit. In this case, a MB-induced closed orbit could nevertheless be generated in high enough magnetic field. In addition, it cannot be excluded that an electron closed tube arises around Y', given that the above picture might be oversimplified.

It should be mentioned that the frequency linked to the $\odot$ orbit ($F_{\odot}=4007$ T) whose cross sectional area is equal to the FBZ area cannot be detected in the data, even at the highest magnetic fields explored. The reduction of the oscillation amplitude due to MB is accounted for by a damping factor R_{MB} involving the probabilities of magnetic breakthrough (p_i) and Bragg reflection (q_i) at the MB junctions. The probability amplitudes p_i and q_i are usually given by $p_i^2=\exp(-B_i/B)$ and $q_i^2=1-p_i^2$ where the MB field B_i is proportional to the square of the MB gap Δ_i [the index i stands for oE or EH, see Fig. 1(b)]. The drastic damping of the amplitude of the $\odot$ oscillation may be due to either the large number of MB junctions involved ($R_{MB}(\odot)=p_{oE}^4 p_{EH}^4$), a large value of at least one of the MB gap [e.g., Δ_{EH} in view of the FS of Fig. 1(a)], or to a large effective mass ($m_c\sim 2.4$, in the framework of the Falicov-Stachowiak model), in view of the large Dingle temperature values reported in Table I.

It remains to correlate the other frequencies observed in Fig. 3 with the orbits E and H and to eventual MB orbits. Assuming the compound is compensated[20] and F_a is linked to the o orbit (of hole character), it can be inferred from the data that $(b-a)$ and b oscillations [which have higher amplitude than $(b+a)$] correspond to the H and E orbit, respectively. Besides, the $b+a$ oscillations can likely be regarded as combination frequency due to e.g., MB-induced Landau levels broadening rather than Shubnikov–de Haas (SdH) frequencies linked to, eventually MB-induced, closed orbits (see Refs.[10–12]).

TABLE I. Experimental effective masses (m_c) and Dingle temperatures (T_D) relevant to the observed oscillation series.

oscillation	m_c	T_D (K)
a	0.58 ± 0.06	3.6 ± 0.8
$b-a$	0.7 ± 0.1	6.5 ± 1.5
b	1.07 ± 0.10	5.5 ± 2.0
$b+a$	0.5 ± 0.1	17 ± 4
$2b$	$0^{+0.15}_{-0}$	

Indeed, since within the above hypotheses, the relation $F_o+F_H=F_E$ should hold, the H and E orbits can then be linked to either (i) F_b and F_{b+a} or (ii) F_{b-a} and F_b, respectively. Given the value of F_{b+a}, the effective mass of the oscillation $b+a$ is very small when compared to that linked to the oscillation b (see Table I). In addition, the Dingle temperature $T_D(b+a)$ is much higher than $T_D(b)$ which makes unlikely the hypothesis (i). Based on the hypothesis (ii), $b+a$ might correspond to a $H+2o$ type MB orbit. However, this latter statement cannot be considered as a result of the FS topology. The $b+a$ oscillation should then be regarded as a combination frequency rather than a conventional SdH orbit, as stated above. Since its temperature dependence is consistent with a zero effective mass (see Fig. 5), the $2b$ oscillation cannot be regarded as due to, e.g., spin splitting effect on the b oscillation and has also a non-SdH origin. Although a zero effective mass is often the signature of a QI path,[28,13] the FS in Fig. 1 cannot yield a QI path with the F_{2b} frequency.

It should be noted that, in the case where the o orbit is of electron character, and following analogous arguments, we come to the conclusion that the F_{b-a} and F_b frequencies are linked to the E and H orbits, respectively, while the $b+a$ and $2b$ oscillations are still not due to SdH orbits unless the calculated FS strongly differs from the actual FS at low temperature.

Frequency combinations are often attributed to the oscillation of the chemical potential of 2D Fermi liquid in magnetic field. Nevertheless, the oscillation of the chemical potential is significantly damped by defect scattering[7] and by the smearing of the Fermi-Dirac function at temperatures above ~ 1 K.[7,8] According to the data in Table I, the scattering time is of the order of few 10^{-13} s only, which makes a predominant contribution of the chemical potential oscillation to the data unlikely. Consequently, the combination frequencies could be mainly due to MB-induced Landau levels broadening[11,3] even though no MB orbits are directly in evidence, probably because they are masked by the contribution of the "basic" orbits. Indeed, assuming as above that the o orbit is of hole character and keeping in mind that electron and hole orbits have "opposite signs," the MB orbit $E+o$ corresponds to the same frequency F_{b-a} as the H orbit. Nevertheless, the contribution of the MB orbit should be masked by that of the H orbit due to a higher effective mass [$m_c(E+o)=m_c(b)+m_c(a)$, in the framework of the semiclassical model of Falicov and Stachowiak[1]] and a reduced value of the damping factor [$R_{MB}(E+o)=p_{oE}^2 q_{oE}^2 q_{HE}^2 \leqslant 0.25$].

V. SUMMARY AND CONCLUSION

At low temperature, the spectrum of the oscillatory magnetoresistance of the β''-(BEDT-TTF)$_4$(NH$_4$)[Fe(C$_2$O$_4$)$_3$] $\cdot$DMF superconductor exhibits various frequencies that can be regarded as linear combinations of $F_a = (48 \pm 2)T$ and $F_b = (241 \pm 5)T$. Although the calculated FS is composed of one electron and one hole orbit, it has been suggested that additional small orbits may also be present in the FBZ.[20] In agreement with this statement, the oscillatory magnetoresistance at low temperature can be consistently interpreted on the basis of three compensated electron and hole orbits with area 1.2%, 4.8%, and 6.0% the FBZ area. These areas correspond to the frequencies F_a, F_{b-a}, and F_b, respectively. Assuming the compound is compensated, the *o* orbit, which is certainly linked to F_a, is of the same character as the orbit linked to F_{b-a}. Assuming further, in agreement with the intersecting orbits scheme displayed in Fig. 1(b) that the *o* orbit is of hole character, F_{b-a} and F_b correspond to the *H* and *E* orbits, respectively.

Although the LK model formally accounts for all the observed oscillations, the field and temperature dependence of the oscillations' amplitude with frequencies F_{b+a} (very small effective mass and very large Dingle temperature) and F_{2b} (zero effective mass) suggest that they do not correspond to conventional MB orbits. Remarkably, the frequency combination F_{b+a} is observed at relatively large temperature (at least up to 4.2 K) even though the scattering time is as low as few 10^{-13} s. This suggests that it mainly result from MB-induced Landau levels broadening which leads to combination frequencies as predicted by the model of coupled orbits network by Pippard[3,11] and by numerical simulations[12] rather than oscillation of the chemical potential. Indeed, it is known that this latter phenomenon is significantly damped by defect scattering[7] and by the smearing of the Fermi-Dirac function at temperatures above than ~ 1 K.[7,8]

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[1] L.M. Falicov and H. Stachowiak, Phys. Rev. **147**, 505 (1966).
[2] For example, J.M. Williams, J.R. Ferraro, R.J. Thorn, K.D. Carlson, U. Geiser, H.H. Wang, A.M. Kini, and M-H. Whangbo, *Organic Superconductors (Including Fullerenes): Synthesis, Structure, Properties and Theory* (Prentice Hall, Englewood Cliffs, NJ, 1992), Chap. 8.
[3] A.B. Pippard, Proc. R. Soc. London **270**, 1 (1962).
[4] N. Harrison, J. Caulfield, J. Singleton, P.H.P. Reinders, F. Herlach, W. Hayes, M. Kurmoo, and P. Day, J. Phys.: Condens. Matter **8**, 5415 (1996).
[5] M.V. Kartsovnik, G.Yu. Logvenov, T. Ishiguro, W. Biberacher, H. Anzai, and N.D. Kushch, Phys. Rev. Lett. **77**, 2530 (1996).
[6] A.S. Alexandrov and A.M. Bratkovsky, Phys. Rev. Lett. **76**, 1308 (1996); Phys. Lett. A **234**, 53 (1997).
[7] A.S. Alexandrov and A.M. Bratkovsky, Phys. Rev. B **63**, 033105 (2001); *ibid.* **65**, 035418 (2002).
[8] K. Kishigi and Y. Hasegawa, Phys. Rev. B **65**, 205405 (2002).
[9] T. Champel, Phys. Rev. B **65**, 153403 (2002).
[10] J.Y. Fortin and T. Ziman, Phys. Rev. Lett. **80**, 3117 (1998).
[11] V.M. Gvozdikov, Yu V. Pershin, E. Steep, A.G.M. Jansen, and P. Wyder, Phys. Rev. B **65**, 165102 (2002).
[12] J.H. Kim, S.Y. Han, and J.S. Brooks, Phys. Rev. B **60**, 3213 (1999); S.Y. Han, J.S. Brooks, and J.H. Kim, Phys. Lett. A **85**, 1500 (2000).
[13] C. Proust, A. Audouard, L. Brossard, S.I. Pesotskii, R.B. Lyubovskii, and R.N. Lyubovskaia, Phys. Rev. B **65**, 155106 (2002); D. Vignolles, A. Audouard, L. Brossard, S.I. Pesotskii, R.B. Lyubovskii, M. Nardone, E. Haanappel, and R.N. Lyubovskaya, Eur. Phys. J. B **31**, 53 (2003).
[14] A.W. Graham, M. Kurmoo, and P. Day, J. Chem. Soc., Chem. Commun. **1995**, 2061.
[15] M. Kurmoo, A.W. Graham, P. Day, S.J. Coles, M.B. Hursthouse, J.L. Caulfield, J. Singleton, F.L. Pratt, W. Hayes, L. Ducasse, and P. Guionneau, J. Am. Chem. Soc. **117**, 12 209 (1995).
[16] S. Turner, P. Day, K.M.A. Malik, and M.B. Hursthouse, Inorg. Chem. **38**, 3543 (1999).
[17] L. Martin, S.S. Turner, P. Day, K.M.A. Malik, S.J. Coles, and M.B. Hursthouse, Chem. Commun. (Cambridge) **1999**, 513.
[18] S. Rashid, S.S. Turner, P. Day, J.A.K. Howard, P. Guionneau, E.J.L. McInnes, F.E. Mabbs, R.J.H. Clark, S. Firth, and T. Biggs, J. Mater. Chem. **11**, 2095 (2001).
[19] L. Martin, S.S. Turner, P. Day, P. Guionneau, J.A.K. Howard, K.M.A. Malik, M.B. Hursthouse, M. Uruichi, and K. Yakushi, Inorg. Chem. **40**, 1363 (2001).
[20] T.G. Prokhorova, S.S. Khasanov, L.V. Zorina, L.I. Buravov, V.A. Tkacheva, A.A. Baskakov, R.B. Morgunov, M. Gener, E. Canadell, R.P. Shibaeva, and E.B. Yagubskii, Adv. Funct. Mater. **13**, 403 (2003).
[21] H. Akutsu, A. Akutsu-Sato, S.S. Turner, D. Le Pevelen, P. Day, V. Laukhin, A.-K. Klehe, J. Singleton, D.A. Tocher, M.R. Probert, and J.A.K. Howard, J. Am. Chem. Soc. **124**, 12 430 (2002).
[22] In Ref. 20 [see Fig. 1(a)], the FS is considered on the basis of a unit cell with vectors $a' = a$, $b' = (a+b)/2$, and $c' = c$. This unit cell contains four BEDT-TTF molecules.
[23] D. Shoenberg, *Magnetic Oscillations in Metals* (Cambridge University Press, Cambridge, 1984); N. Harrison, R. Bogaerts, P.H.P. Reinders, J. Singleton, S.J. Blundell, and F. Herlach, Phys. Rev. B **54**, 9977 (1996).
[24] L. Martin, S.S. Turner, P. Day, P. Guionneau, J.A.K. Howard, D.E. Hibbs, M.E. Light, M.B. Hursthouse, M. Uruichi, and K. Yakushi, Inorg. Chem. **40**, 1363 (2001).
[25] S. Kahlich, D. Schweitzer, P. Auban-Senzier, D. Jérome, and H.J. Keller, Solid State Commun. **83**, 77 (1992).

[26] A. Audouard, V.N. Laukhin, C. Proust, L. Brossard, and N.D. Kushch, J. Phys. I **7**, 599 (1997); C. Proust, A. Audouard, V.N. Laukhin, L. Brossard, M. Honold, M.S. Nam, E. Haanappel, J. Singleton, and N.D. Kushch, Eur. Phys. J. B **21**, 31 (2001).
[27] S.S. Khasanov, B.Zh. Narymbetov, L.V. Zorina, L.P. Rozenberg, R.P. Shibaeva, N.D. Kushch, E.B. Yagubskii, R. Rousseau, and E. Canadell, Eur. Phys. J. B **1**, 419 (1998).
[28] R.W. Stark and C.B. Friedberg, Phys. Rev. Lett. **26**, 556 (1971); N. Harrison, R.G. Goodrich, J.J. Vuillemin, Z. Fisk, and D.G. Rickel, *ibid.* **20**, 4498 (1998).