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Spin-density-wave transition and resistivity minimum of the Bechgaard salt $(TMTSF)_2NO_3$ at high magnetic field, where TMTSF is tetramethyltetraselenafulvalene

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Transverse magnetoresistance of the Bechgaard salt (tetramethyltetraselenafulvalene)$_2NO_3$ has been measured up to 36 T, with the magnetic field parallel to the lowest conductivity direction c^*. Data have been recorded at ambient pressure, in the temperature range from 2 to 77 K. Whereas the anion ordering temperature remains field independent, the spin-density-wave transition temperature (T_{SDW}) increases by few tenths of a percent at 32 T and the temperature at which the resistivity minimum occurs (T_{min}) increases by nearly a factor of 3 at 36 T. The field dependence of T_{SDW} is studied in the framework of the mean-field calculations of Bjelis and Maki [Phys. Rev. B **45**, 12 887 (1992)] while the T_{min} variation is discussed on the basis of field-induced electron confinement.

In a previous paper,[1] both oscillatory and semiclassical behaviors of the high-field magnetoresistance (MR) were reported in the ambient pressure spin-density-wave (SDW) ground state of the Bechgaard salt (BS) $(TMTSF)_2NO_3$. Among BS, $(TMTSF)_2NO_3$ exhibits several peculiar features. Indeed, contrary to most other salts with noncentrosymmetric anions, the anion ordering (AO) wave vector is parallel to the best conduction axis a (Ref. 2) which should lead, below $T_{AO}=41$ K, to a compensated two-dimensional (2D) semimetal with electron and hole closed tubes. In addition, the SDW order parameter at ambient pressure and zero temperature is only about 8 K (Ref. 3) and the temperature dependence of the resistivity below the SDW transition temperature ($T_{SDW}=9.4$ K) cannot be satisfactorily accounted for by a thermally activated law. These features both suggest imperfect nesting of electron and hole tubes. Thus smaller closed orbits should be further induced below T_{SDW}. As a matter of fact, in the temperature range below 10 K,[1,4] two series of quantum oscillations with frequency at respectively 63 and 246 T were observed above ~5 and 16 T, respectively. Whereas the 246 T series was ascribed to the so-called fast oscillations usually observed in most of BS, the 63 T series has no counterpart in other BS. It was shown to be linked to the SDW phase[1,4] and might be related to the above-mentioned small carrier pockets,[1,5,6] although the Lifshitz-Kosevitch equation fails to account for both the temperature and field dependence of the oscillations amplitude.[7] On the other hand, the analysis of the semiclassical part of the MR has revealed a significant influence of the magnetic field on T_{SDW} which increases by 14% at 30 T.[1] This behavior which is the signature of the field-induced improvement of the imperfect nesting of the 2D orbits at the SDW transition was satisfactorily accounted for by the mean-field calculations of Montambaux[8] at low magnetic field. However, Bjelis and Maki have proposed more recently[9] a general equation for the field dependence of T_{SDW} which holds whatever the considered magnetic-field range. In the low-field range, this model predicts a quadratic variation of T_{SDW}, in agreement with Ref. 8. On the opposite side, T_{SDW} is predicted to saturate at high field towards the value which corresponds to the SDW transition temperature related to the case of perfect nesting. This feature is in agreement with the idea that magnetic field improves the 1D character of the Fermi surface. However, the above model did not yet receive any quantitative verification since only low magnetic-field experiments[10] or data analysis[1] have been reported up to now. The purpose of this work is thus to present experimental evidence of the field dependence of T_{SDW}, derived within the framework of the Bjelis and Maki model.

Experimental details were published previously.[1,4] To avoid cracks, a slow cooling rate was achieved (~0.1

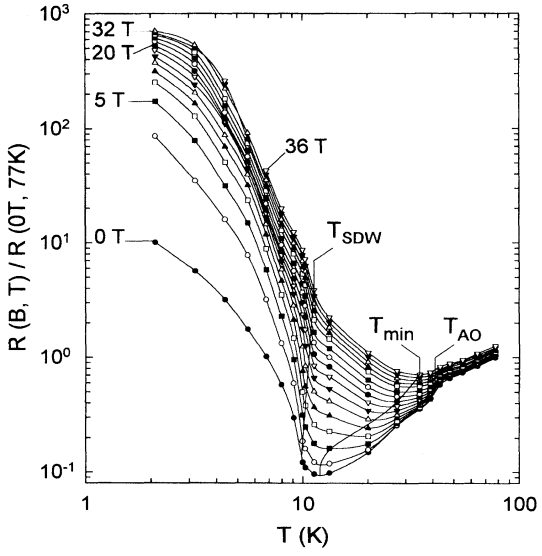

FIG. 1. Temperature dependence of the normalized resistance of $(TMTSF)_2NO_3$ recorded at several magnetic-field values. From the bottom to the top, the value of the magnetic field is 0, 2.5, 5, 7.5, 10, 12.5, 15, 17.5, 20, 22.5, 25, 28, 30, 32, 34, and 36 T (some of these values are indicated on the figure). Solid lines are guides for the eye.

K min^{-1}) from 300 K down to 2 K so that the standard temperature dependence of the resistance was obtained at zero field,[3,11] as can be seen in Fig. 1 where a metallic behavior is observed down to 12 K, followed by a steep resistance increase due to the condensation of the SDW phase. The temperatures at which SDW and AO transitions take place were conventionally estimated[3] from the extrema of $d[\ln(R)]/d(1/T)$. This parameter exhibits a minimum at $T_{AO}=41$ K and a maximum at $T_{SDW}=9.4$ K,[1] both in good agreement with data of the literature.[2,3] Magnetoresistance measurements were carried out in pulsed magnetic fields up to either 32 or 36 T during the decay period (1.2 s) of the field, in the transverse configuration ($B\perp a$) with B parallel to c^* and 50 kHz ac current parallel to the best conduction direction (a axis).

While T_{AO} remain unaffected by magnetic field up to 36 T, Fig. 1 shows that T_{SDW} increases: from 9.4 K in zero field up to 10.7 K at 32 T as evidenced in Fig. 2. As predicted,[8,9] a quadratic behavior is observed at low field, followed by a saturating behavior. According to Maki,[12] and for magnetic field B applied parallel to c^*, the general equation given in Ref. 9 can be approximated as

$$-\ln\left(\frac{T_{SDW}}{T_{SDW_0}}\right)=\frac{1}{2}\left(\frac{\varepsilon_0}{\omega_b}\right)^2\left\{\mathrm{Re}\left[\Psi\left(\frac{1}{2}+\frac{i\omega_b}{2\pi T_{SDW}}\right)\right]-\Psi\left(\frac{1}{2}\right)\right\}$$
$$-\frac{1}{32}\left(\frac{\varepsilon_0}{\omega_b}\right)^4\left\{4\ \mathrm{Re}\left[\Psi\left(\frac{1}{2}+\frac{i\omega_b}{2\pi T_{SDW}}\right)\right]-\mathrm{Re}\left[\Psi\left(\frac{1}{2}+\frac{i\omega_b}{\pi T_{SDW}}\right)\right]-3\Psi\left(\frac{1}{2}\right)\right\}. \quad (1)$$

The relevant parameters entering this equation are the SDW transition temperature corresponding to the perfect nesting case (T_{SDW_0}), the cyclotron pulsation (ω_b), and the energy parameter which characterizes the deviation from perfect nesting (ε_0), defined as[13]

$$\varepsilon_0=t_b^2/t_a\sqrt{2}, \quad (2)$$

where t_a and t_b are transfer integrals in a and b directions, respectively. ω_b is written as

$$\omega_b=\nu beB, \quad (3)$$

where ν is the Fermi velocity in the a direction.

In order to make Eq. (1) more easily tractable, an analytical approximation of $\mathrm{Re}\{\Psi(\frac{1}{2}+ix)-\Psi(\frac{1}{2})\}$, where Ψ is the digamma function, was calculated. Keeping in mind that

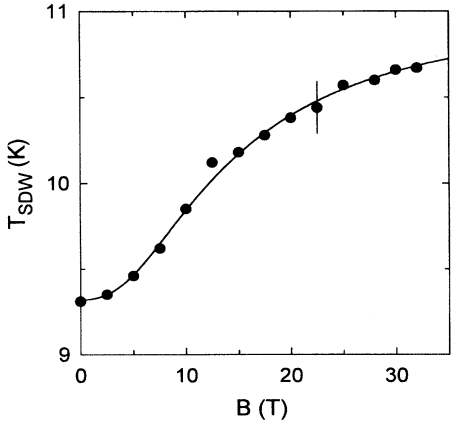

FIG. 2. Magnetic-field dependence of the spin-density-wave transition temperature (T_{SDW}). The full line is the best fit to Eq. (1) (see text).

$$\Psi(\tfrac{1}{2}+ix)-\Psi(\tfrac{1}{2})=2\Psi(2ix)-\Psi(ix)+\gamma \quad (4)$$

and

$$\mathrm{Re}[\Psi(ix)]=-\gamma+x^2\sum_{n=1}^{+\infty}\frac{1}{n(n^2+x^2)}, \quad (5)$$

where $\gamma=-\Psi(\frac{1}{2})-2\ln(2)$ and $\Psi(\frac{1}{2})=1.963\,51$, the following analytical expression can be obtained:

$$\mathrm{Re}\left[\Psi\left(\frac{1}{2}+ix\right)-\Psi\left(\frac{1}{2}\right)\right]$$
$$=8x^2\left\{\frac{1}{1+4x^2}+\frac{1}{3(9+4x^2)}+\frac{1}{5(25+4x^2)}\right.$$
$$\left.+\frac{11}{147(49+4x^2)}+\frac{1}{3(49+4x^2)^2}\right\}$$
$$+\frac{1}{2}\ln\left[1+\frac{4x^2}{49}\right]. \quad (6)$$

It is worthwhile to notice that Eq. (6) is accurate within 4×10^{-5} in the whole magnetic-field range covered by the experiments. The solid line in Fig. 2 is the best fit of Eq. (1) to experimental data: a very good agreement is obtained. The fit yields $\nu=(2.4\pm0.3)\ 10^5$ ms^{-1}, $T_{SDW_0}=11.0\pm0.2$ K, and $\varepsilon_0=13\pm4$ K. It can be noticed first that a reasonable value is obtained for the Fermi velocity. On the other hand, T_{SDW_0} is in excellent agreement with the value measured for $(TMTSF)_2PF_6$ for which the nesting can be regarded as perfect. Indeed, for $(TMTSF)_2PF_6$ crystals with best resistivity ratio, the SDW transition temperature is 11.1 K.[3,14] Let us consider now the energy parameter ε_0 which accounts for the deviation from perfect nesting. This parameter is directly linked to the transfer integrals t_a and t_b [see Eq. (2)]. Unfortunately, no data about transfer integral values in the SDW state of $(TMTSF)_2NO_3$ are presently available in the litera-

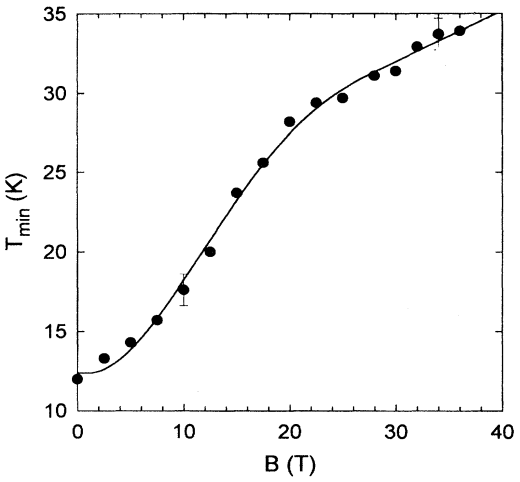

FIG. 3. Magnetic-field dependence of the resistivity minimum temperature (T_{min}). The solid line is a guide to the eye.

ture. Nevertheless, the deduced value of ε_0 can be compared to that deduced from band-structure calculations related to $(TMTSF)_2PF_6$ at a temperature of 4 K.[15] These calculations yield $\varepsilon_0 = 16$ K,[16] which is close to and even larger than the value deduced for $(TMTSF)_2NO_3$, and thus not consistent. This disagreement could be accounted for assuming either ε_0 [as defined in Eq. (2)] is not a relevant enough parameter or available band-structure calculations for BS are not fully reliable. It must be remarked that, in the case of perfect nesting, T_{SDW} should remain magnetic-field independent which, according to Eq. (1), would yield $\varepsilon_0 = 0$. In other words, Eq. (1) suggests that a SDW phase transition with perfect nesting of Fermi surface can only take place in BS for which $t_b = 0$. This is in conflict with the theoretical model[17] which predicts that electronic instability cannot be observed at finite temperature in perfect 1D compounds. In any case, accurate band-structure calculations are needed to solve this discrepancy. Nevertheless, it can be noticed that, although this model neglects any influence of both electron correlations and AO, it nicely accounts for experimental data and yields satisfactory values for Fermi velocity and T_{SDW_0}.

Finally, it can be seen in Fig. 1 that the temperature T_{min} at which the resistivity minimum occurs ($T_{min} = 12$ K in zero field) strongly increases as B increases. The field dependence of T_{min} is displayed in Fig. 3. A similar behavior has already been observed in slowly cooled $(TMTSF)_2ClO_4$ (Ref. 18) for which a SDW phase is stabilized at low field thus giving rise to a resistivity minimum, the temperature at which it occurs increasing up to 22 K at 26 T. This phenomenon could be due to the predicted[9,19] field-induced growth of antiferromagnetic fluctuations. However, this interpretation seems to be unlikely since in the present case the field dependence of the width of the SDW transition[1] is not significant enough to account for such a large increase of T_{min}. An alternate explanation[18] could involve the field-induced decrease of the transverse interactions. Indeed, in tetramethyltetrathiafulvalene (TMTTF) salts, for which ε_0 is typically lower by a factor of 2 than in BS,[16] the condensation of the charge degrees of freedom occurs at a temperature T_ρ which is higher than the condensation temperature of the spin degrees of freedom leading to a resistivity minimum around T_ρ. In the present case, the features of the temperature dependence of the resistance at high field (see Fig. 1) are quite similar to those observed at zero field in, e.g. $(TMTTF)_2Br$, although its T_ρ is well higher [$T_\rho = 100$ K (Ref. 20)] than T_{min} in the present case ($T_{min} = 34$ K at 36 T). This could be the signature of an increased separation of charge and spin degrees of freedom in $(TMTSF)_2NO_3$. Since this separation is a general character of strongly correlated electron gas, it is suggested that the effect of a field applied parallel to c^* is to evidence the electron correlation properties through the field-induced confinement in the ab plane.

As a summary, we have shown that the mean-field calculations of Bjelis and Maki[9,12] nicely account for the field-induced increase of the SDW transition temperature of $(TMTSF)_2NO_3$ up to 32 T. Satisfactory values of both T_{SDW_0} and Fermi velocity have been deduced from the analysis. However, the deduced value of the energy parameter ε_0 is not consistent with the available data of transfer integrals in BS. This suggests that ε_0 might not be the most relevant parameter to account for the deviation from perfect nesting in BS. We have also evidenced that magnetic field strongly increases the temperature of the resistivity minimum. As proposed in Ref. 18, this latter phenomenon is likely related to field-induced electron confinement.

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