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Orbital effect of a magnetic field on the low-temperature state in the organic metal α-(BEDT-TTF)$_2$KHg(SCN)$_4$

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The effect of pressure on the B-T phase diagram of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ is studied. The measured phase lines can be well described by a recent model of a charge-density wave system with varying nesting conditions. A remarkable increase of the transition temperature with magnetic field is found in a certain pressure and field range. We associate this result with a dramatic enhancement of the orbital effect of magnetic field due to a deterioration of the nesting conditions by pressure. Furthermore, we present data which can be interpreted as a first sign of field-induced charge-density waves.

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The organic charge transfer salt α-(BEDT-TTF)$_2$KHg(SCN)$_4$ has a layered crystal structure, consisting of conducting bis(ethylenedithio)tetrathiafulvalene and insulating anion sheets,[1] that leads to a strong anisotropy of the electronic system. Numerous anomalies displayed by this material in magnetic field can reasonably be explained by a density-wave instability of the quasi-one-dimensional (Q1D) part of the electronic system and its interaction with the quasi-two-dimensional (Q2D) conducting band (for a review see Ref. 2).

Extensive studies of the magnetic-field temperature (B-T) phase diagram[3–7] have provided a substantial argument for the charge-density-wave (CDW) nature of the low-temperature state in this compound. Both thermodynamic[3,4,6] and interlayer transport[3,5,7] measurements show a decreasing transition temperature T_p with increasing field. Such a behavior is expected for a well nested CDW system.[8,9] This is due to the competition between the Pauli paramagnetism and the CDW instability in a magnetic field. For a perfectly nested CDW this Pauli effect causes a gradual decrease of the transition temperature with field. In the low-field limit $(B \ll B_c \sim [k_B T_p(B=0)]/\mu_B)$, $\Delta T_p/T_p$ is proportional to B^2.[10] At higher fields, $B \approx B_c$, when the Zeeman energy reaches the value of the zero temperature energy gap, the theory proposes a first order phase transition at low temperatures: the perfectly nested CDW state transforms into a CDW/SDW (where SDW denotes spin density wave) hybrid state with a shifted, field-dependent nesting vector.[8,9] In the present compound the transition temperature is remarkably lower than in most known CDW systems. This gives the unique opportunity of extending the studies of field effects far beyond the low-field limit even in static magnetic fields. Indeed, previous experiments[3,5–7,11] have already demonstrated the existence of a new phase at fields above 24 T and temperatures below 4 K, which can be associated with the CDW/SDW state.

Another, so-called orbital effect of magnetic field must be taken into account in an imperfectly nested density-wave system. Under a magnetic field applied parallel to the open sheets of the Fermi surface, the electrons are forced to move in k space perpendicular to the field along the sheets, causing an oscillatory motion in real space which becomes more restricted to one dimension with increasing the field. This should cause a stabilization of the density-wave state and lead to an increase of T_p with field.[12] Such an increase due to the orbital effect has been observed in SDW systems.[13] Moreover, the orbital quantization was shown to lead to a fascinating macroscopic quantum phenomenon known as field-induced spin-density waves (FISDW) (for a review see Ref. 14). Despite similar predictions,[9] there exists up to now no clear evidence of the orbital effect on T_p in a CDW system.[15] In α-(BEDT-TTF)$_2$KHg(SCN)$_4$, the phase diagram can be fairly well described by a dominant Pauli effect of magnetic field on a CDW state, although a weak dependence of T_p on the magnetic-field orientation[4,6] may be interpreted as an indication of a small orbital effect. The theoretically predicted[9] competition between the two effects was recently suggested by Qualls *et al.*[7] to be a reason for a significant modification of the phase diagram at magnetic fields strongly tilted towards the conducting layers. However, other studies of the present material at high tilt angles[16] reveal a complicated behavior which does not fit into the simple picture proposed in Ref. 7.

In this work we report on a direct manifestation of the orbital effect on the CDW phase diagram obtained by tuning the nesting conditions in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ by quasihydrostatic pressure. The interplane resistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ was measured at temperatures down to 0.4 K in magnetic fields up to 28 T, directed perpendicular to the layers, at different pressures up to $P=4.6$ kbar. Quasihydrostatic pressure was applied using either a conventional clamp cell or a He-pressure apparatus. Several samples from different batches were measured, revealing basically the same behavior.

Although the determination of transition points from the magnetoresistance is not straightforward, reasonable estimates in an applied magnetic field can be made via Kohler's rule, which is a similarity law for the magnetoresistance.[17] It

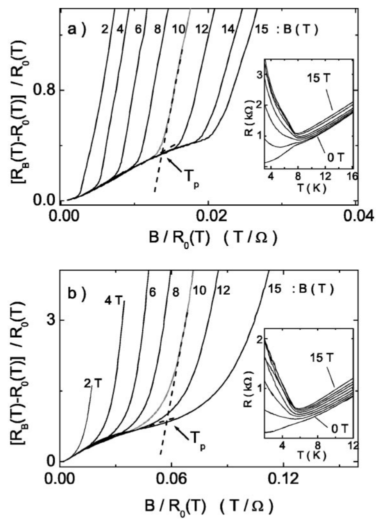

FIG. 1. Kohler plots at (a) ambient pressure and (b) 1.6 kbar. The corresponding resistance versus temperature curves at fixed magnetic fields are shown in the insets.

is based on the assumption that the scattering processes do not depend on magnetic field. If one further expects the zero-field resistance R_0 to be inversely proportional to the scattering time τ, the magnetoresistance can be expressed as a general function of B/R_0:

$$[R_B(T)-R_0(T)]/R_0(T)=F[B/R_0(T)], \tag{1}$$

that constitutes Kohler's rule. Kohler's rule has already been found to work well in several organic metals (see, e.g., Ref. 18). Figure 1 shows typical scaling plots, so-called Kohler plots, obtained from temperature sweeps at fixed magnetic fields which are depicted in the inset. It is clearly seen that at higher temperatures the curves follow one general function, in accordance with Kohler's rule. This suggests that this rule is valid for the normal metallic (NM) state of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at the given orientation and range of the applied field. At lower temperatures all the curves start to diverge dramatically. This is consistent with an earlier report on a strong violation of Kohler's rule in the low temperature (LT) state of this material.[19] We therefore ascribe the deviation from Kohler's rule to the phase transition from the NM to the LT state. As a characteristic temperature of the transition T_p, we take the temperature corresponding to the crossing point of linear extrapolations from the NM and LT parts of the Kohler plots as shown in Fig. 1 for $B=10$ T. The dependence of T_p on magnetic field is shown in Fig. 2 (empty circles) for three different pressures. We note that the same behavior is obtained for the temperatures corresponding to the maximum curvature of the Kohler plots or a typical kink in their derivatives. Thus, even though we cannot assert an exact definition of the absolute value of the critical temperature from the above procedure, we believe that the curves in Fig. 2 reflect the correct dependence of the real critical temperature on magnetic field and hydrostatic pressure.

At ambient pressure, the observed monotonic shift of the transition temperature to lower values with increasing field is

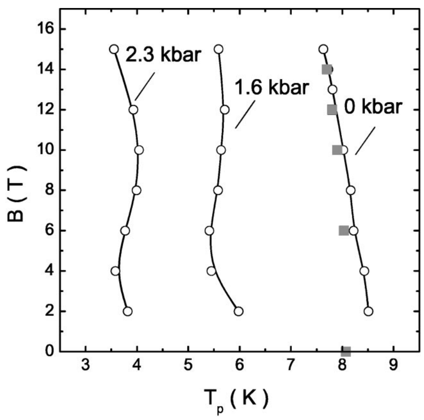

FIG. 2. Phase boundaries between the LT and NM states obtained from the Kohler plots at three different pressures (sample No. 1, circles) and from earlier specific heat measurements at ambient pressure (Ref. 4) (squares).

consistent with data obtained by specific heat[4] (squares in Fig. 2) and magnetic torque experiments.[6] Under pressure the phase boundary moves to lower temperature; no transition has been detected at 4.6 kbar in agreement with previous studies.[20] Furthermore, pressure causes a remarkable change in the shape of the phase boundary: in a certain range the field clearly *stabilizes* the LT state. This result can be readily understood in terms of a competition between the orbital and Pauli effects of magnetic field on the CDW state. At ambient pressure, when the nesting is good, the Pauli paramagnetic effect dominates, leading to a constant decrease of T_p with increasing field. An applied pressure deteriorates the nesting conditions, thereby suppressing the zero-field T_p. At the same time, the orbital motion in a magnetic field perpendicular to the ac plane acts to effectively reduce the dimensionality of the electronic system, resulting in a relative increase of T_p. Thus, with an increasing pressure, hence warping of the Fermi surface, the orbital effect becomes more pronounced and may even become dominant as seen in Fig. 2. However, if the characteristic frequency of the orbital motion, $\omega_c=eB\nu_F^{(1D)}d/\hbar c$ (ν_F, Fermi velocity; d, length of the unit cell along the open sheets of the Fermi surface; c, velocity of light), is sufficiently high, $\hbar\omega_c>k_BT_p(0)$, the contribution from the orbital effect to $T_p(B)$ saturates[11] and the CDW state should be eventually suppressed due to the Pauli effect. This qualitative consideration is found to be in very good agreement with the evolution of the B-T diagram of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ under pressure.

In Fig. 3 we present the phase lines obtained from the investigation of several samples under different pressures in magnetic field up to 27 T. The data points correspond to one single experiment for each pressure. The circles are taken from the Kohler plots as described above. The triangles represent the so-called kink transition (that is supposed to be a transition from the low-field CDW to the high-field CDW/SDW hybrid state[5–8]), which was recorded in the field sweeps of the magnetoresistance at $P=0$ and 1.8 kbar. The solid line at ambient pressure illustrates the general behavior observed in previous works.[3–6] As a whole, the phase diagrams are strikingly similar to those predicted by Zanchi

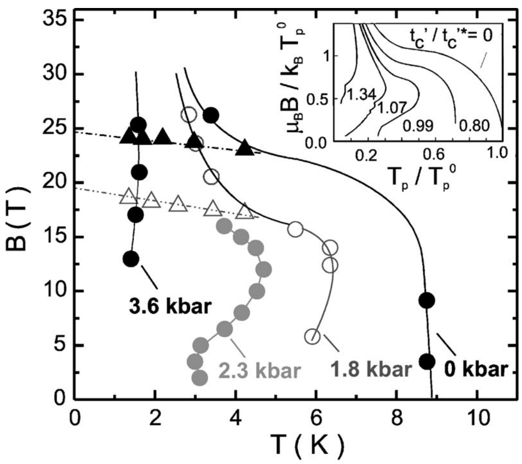

FIG. 3. *B-T* phase diagrams measured at 0, 1.8, 3.6 kbar (sample No. 2) and 2.3 kbar (sample No. 3). Circles: boundary between the NM and the LT states; triangles: kink transition. Inset: theoretically proposed phase diagrams of a CDW system at different nesting conditions (Ref. 9).

et al.[9] for a CDW system with varying nesting conditions. The latter are shown in the inset in Fig. 3. Here, the imperfect nesting is introduced by the second order transfer integral t_c' entering the dispersion relation:

$$\epsilon = v_F(|k_x| - k_F) - 2t_c\cos(k_c c) + 2t_c'\cos(2k_c c), \qquad (2)$$

and $t_c'^*$ is a critical value of t_c' at which the CDW is completely suppressed at zero field [$t_c'^*$ can be estimated as $\cong k_B T_p^0(0)$ where $T_p^0(0)$ is the zero-field transition temperature at $t_c'=0$ (Ref. 21)]. The transition between the low-field CDW and high-field hybrid CDW/SDW states was analyzed so far only for a perfectly nested system ($t_c'=0$).[9] Therefore the phase lines in the inset in Fig. 3 do not include this transition. It would be highly interesting to extend these studies for the case of finite t_c' and to compare the theory with the pressure dependence of the experimentally observed kink transition.

An explicit comparison between the experiment and theory cannot be done at this stage: On the one hand, the boundaries can change to some extent depending on the values of the coupling constants. On the other hand, the theoretical model[9] ignores such factors as fluctuations and the presence of the additional, Q2D conducting band which can also lead to a modification of the phase diagram. Nonetheless, certain conclusions based on the qualitative similarity between the experiment and theoretical predictions shown in Fig. 3 can be made. At $P=3.6$ kbar the NM state persists down to at least 1.4 K at fields below 10 T, whereas clear deviations from Kohler's rule are detected at $B>12$ T. This behavior obviously corresponds to $t_c'/t_c'^*>1$. The competition between the orbital and Pauli effects is expected to be the most pronounced at $t_c'/t_c'^*=1.0\pm0.1$.[9] It is this region in which both $T_p(0)$ and the shape of the phase line are extremely sensitive to t_c'. Our data in Fig. 3 suggest that the pressure of 2.3 kbar corresponds to $t_c'/t_c'^*$ almost exactly equal to 1.

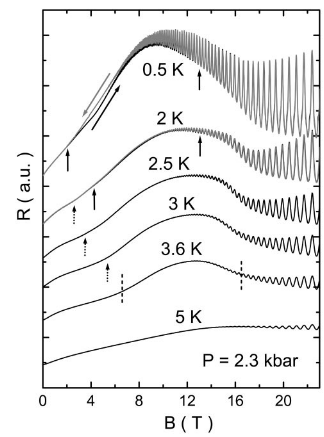

FIG. 4. Magnetoresistance of sample No. 3 under $P=2.3$ kbar, at several temperatures. Vertical dashed lines correspond to the phase boundaries in Fig. 3. The features marked by arrows are discussed in the text.

The nonmonotonic field dependence of T_p at 2.3 kbar is also reflected in isothermal field sweeps of the magnetoresistance which are shown in Fig. 4. In contrast to a smooth behavior at $T\geqslant5$ K, the magnetoresistance at $T=3.6$ K exhibits a clear enhancement due to entering the LT state. According to the phase diagram in Fig. 3, the LT state occurs at 3.6 K in the field range between $\approx$ 6.5 and 16.5 T as indicated in Fig. 4 by dashed lines. The low-field feature rapidly weakens and shifts to lower fields (as marked by the dotted vertical arrows in Fig. 4) as the temperature is reduced below 3 K.[22] The decrease of the magnetoresistance background at high fields manifests a field-induced transition into the high-field modification of the LT state (kink transition) or into the NM state and can be observed in the field sweeps at any temperature below 5 K.

At $T<2.5$ K a hysteresis in the magnetic-field sweeps emerges in a broad interval as marked by solid vertical arrows in Fig. 4. It is accompanied by a change of the slope of the magnetoresistance in a certain field range as clearly seen at the 0.5 K curve at 4–5 T. Both the hysteresis and nonmonotonic behavior of the magnetoresistance become even more pronounced at higher pressure as shown in Fig. 5 for $P=3$ kbar, $T=1.4$ K. These anomalies, at first glance sur-

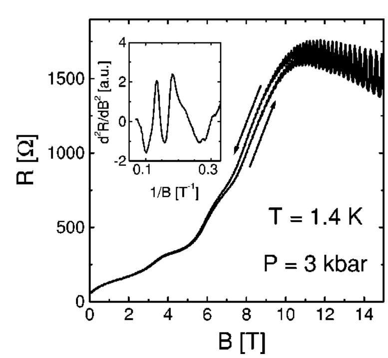

FIG. 5. Magnetoresistance of sample No. 4 at 1.4 K under $P=3$ kbar. Inset: The second derivative $d^2R(B)/dB^2$ taken after filtering out the Shubnikov–de Haas signal, vs inverse field.

prising, may turn out to be a sign of an interesting quantum phenomenon. For certain nesting conditions, namely when t_c' becomes comparable to $t_c'^*$, theory[9] predicts a cascade of field-induced CDW (FICDW) transitions. This phenomenon is analogous to already well known FISDW,[14] where the quantized adjustment of the nesting vector serves to keep the Fermi energy level between the Landau levels, in order to stabilize the SDW in a varying magnetic field. Comparing our results with the theoretical prediction,[9] we suggest that the features displayed in Fig. 4 (for the lowest temperatures) and Fig. 5 may be a manifestation of the FICDW phenomenon. In this context, the magnetoresistance behavior at 3 kbar (Fig. 5) can be described as follows: The increase of the slope at 2.7 T corresponds to the boundary between the NM and FICDW regions. Due to the relatively high temperature,[23] no clear features can be resolved between 3 and 5 T. The enhancement of the magnetoresistance at ≈ 5 T is the first direct indication of switching between different FICDW subphases. As the field further increases, the transition anomaly (at ≈ 7.4 T) becomes sharper and exhibits a pronounced hysteresis. If we associate the transition points with the maxima in the d^2R/dB^2 dependence shown in the inset to Fig. 5, and assume them to be periodic in $1/B$, another transition at $B \cong 12.5$ T might be expected. However, no clear anomaly has been found between 10 and 15 T. On the one hand, this may be an indication that the system is already in the $n=0$ state [i.e., the longitudinal component of the nesting vector is $q_\| = 2k_F$ (Ref. 14)] above 7.4 T. On the other hand, the FICDW transition above 10 T should be influenced by (i) the orbital quantization of the Q2D band reflected in strong Shubnikov–de Haas oscillations; and (ii) the Pauli effect of the magnetic field which is expected to induce the kink transition well below 20 T at the given pressure. Further theoretical and experimental studies should clarify how these two mechanisms interfere with the field-induced quantization of the nesting vector.

Finally, we conclude that although the data shown in Figs. 4 and 5 are not yet sufficient to claim unambiguously the detection of FICDW, the similarities between the experimentally obtained phase diagram and that proposed theoretically for a CDW system[9] make the present material a promising candidate for the realization of this new quantum phenomenon.

Summarizing, the B-T phase diagrams of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at different pressures can be consistently interpreted in terms of the interplay between the Pauli and orbital effects of the magnetic field on a CDW system with varying nesting conditions. The orbital effect at pressure of $\simeq 2$ kbar is clearly manifested by a remarkable increase of the transition temperature in magnetic field. The nonmonotonic hysteretic behavior of the magnetoresistance at pressures corresponding to $t_c'/t_c'^* \gtrsim 1$ provides an argument for the existence of FICDW subphases.

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[22] Noteworthy, although the phase diagram presented in Fig. 3 suggests $T_p(B) \gtrsim 3$ K, the low-field feature can be resolved in the field sweeps at temperatures down to at least 2 K. Taking into account a smooth character of the changes in the magnetoresistance during the NM-LT transition, one could suggest that the existence of this feature below T_p is due to the proximity to the phase boundary.

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