

MAGNETIC FIELD-TEMPERATURE PHASE DIAGRAM OF THE ORGANIC CONDUCTOR α -(BEDT-TTF)₂KHg(SCN)₄

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We present systematic magnetic torque studies of the "magnetic field - temperature" phase diagram of the layered organic conductor α -(BEDT-TTF)₂KHg(SCN)₄ at fields nearly perpendicular and nearly parallel to the highly conducting plane. The shape of the phase diagram is compared to that predicted for a charge-density-wave system in a broad field range.

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Organic metals α -(BEDT-TTF)₂MHg(SCN)₄, where M = K, Tl, or Rb [1], have attracted much attention in the last decade due to their exotic low-temperature electronic state. They are characterized by a layered crystal structure and a unique co-existence of quasi-one-dimensional (Q1D) and quasi-two-dimensional (Q2D) conducting bands [1]. The transition into the low-temperature state is associated with a $2k_F$ nesting instability of the Q1D part of the Fermi surface. Indeed, experiments on the angle-dependent magnetoresistance oscillations [2-5] have revealed a significant change in the electronic system due to a periodic potential with the wave vector close to the doubled Fermi wave vector of the Q1D band. On the other hand, studies of the magnetization anisotropy [6, 7] and μ SR [8] give evidence for a low amplitude modulation of the magnetic moment suggestive of a spin-density wave (SDW). Many of the striking anomalies displayed by these compounds in magnetic field can be fairly well explained by the density-wave instability, taking into account the coexistence of the Q1D and Q2D Fermi surfaces (see e.g. Refs. [3-5, 9, 10]). However, there remain several questions which can hardly be understood within the SDW model. One of the important questions concerns the effect of magnetic field on the low-temperature state.

It is known that magnetic field applied perpendicular to the direction of the spin polarization may stimulate the SDW formation in systems with imperfectly nested Fermi surfaces due to effective reduction of the electron motion to one dimension [11, 12]. This orbital effect leads to a slight increase of the SDW transition temperature as was shown for a Q1D conductor (TMTSF)₂PF₆ [13]. The situation with the α -(BEDT-TTF)₂MHg(SCN)₄ salts is rather controversial in this respect. In agreement with the SDW model, Sasaki et al. [14] reported the transition temperature, T_p , in α -(BEDT-TTF)₂KHg(SCN)₄ to increase in magnetic field perpendicular to the spin polarization plane (which is the highly-conducting *ac*-plane in this compound [15]). On the contrary, other numerous experiments

suggest a reduction of T_p in magnetic field. Some authors [16, 17] claim that the low-temperature state is completely suppressed in this salt and the normal metallic state is restored above the so-called kink transition at $B_{kink} \simeq 24$ T. On the other hand, several works suggest a new phase, different from the normal one, to emerge above B_{kink} [18, 19]. Based on the shape of the “magnetic field – temperature” ($B - T$) phase diagrams [18, 19], Biskup et al. [19] proposed the phase transition to be driven by a charge-density wave (CDW) rather than SDW instability.

It should be noted, that the studies of the high-field region of the $B - T$ diagram of the α -(BEDT-TTF) $_2$ MHg(SCN) $_4$ compounds have been mostly done by use of magnetoresistance technique. Obviously, such experiments are difficult to interpret unambiguously in terms of phase transitions. Therefore a detailed investigation of thermodynamic properties is necessary in order to establish the phase boundaries. So far only few magnetization data at fields above 15 T were presented in two works [14, 18]. However, the conclusions made in these works concerning the field effect on the transition temperature contradict each other. To elucidate the problem, we have carried out a systematic study of the $B - T$ phase diagram of α -(BEDT-TTF) $_2$ KHg(SCN) $_4$ by means of magnetic torque experiments.

Several high quality samples chosen for the experiment were grown by the standard electrochemical method [20] and had a typical mass of 100 to 350 μ g. A cantilever beam magnetometer [7] was used to measure the torque in fields nearly perpendicular and nearly parallel to the highly conducting ac -plane. The measurements were performed at temperatures between 0.4 and 18 K in magnetic fields up to 28 T produced at the High Magnetic Field Laboratory in Grenoble, France.

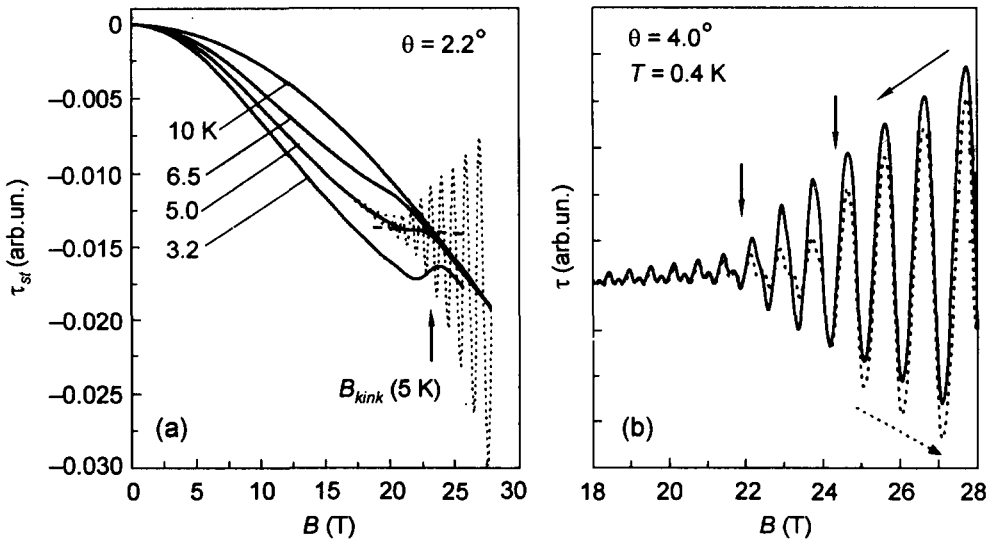


Fig.1. Torque as a function of magnetic field applied nearly perpendicular to the ac -plane: (a) – steady part of the torque at different temperatures; the dotted curve represents the total signal from the sample, with the dHvA oscillations at $T = 5.0$ K; (b) – up (dotted line) and down (solid line) field sweeps of the torque at low temperature

We first focus on field directions almost perpendicular to the layers. Typical field dependences of the steady part of the torque $\tau_{st}(B)$ are shown in Fig.1a, for the angle

θ between the magnetic field and the normal to the ac -plane equal to 2.2° . At high temperatures ($T \geq 8$ K) we find an almost temperature insensitive quadratic dependence of the torque on magnetic field. On lowering the temperature below 8 K the quadratic term increases at small fields, but above 4 T the dependence becomes weaker than quadratic and at high fields the curves bend to merge with the high temperature curve. The field at which the torque returns back to its normal behaviour coincides with the kink field B_{kink} as determined in other experiments [14, 16–19]. In addition to the steady part of the torque, de Haas-van Alphen (dHvA) oscillations were observed. At 10 K these oscillations were resolved only at the highest fields, but at 5.0 K their amplitude was already comparable to $\tau_{st}(B)$ as shown by a dashed line in Fig.1a. To extract $\tau_{st}(B)$ we used a Fourier filter. In contrast to the measurements at higher temperatures, the curve at 3.2 K does not return to the high temperature part at B_{kink} but stays below. For temperatures below 3 K the dHvA amplitude becomes so strong that the steady torque cannot be extracted reliably any more. In Fig.1b we show a trace of a field sweep from 18 T to 28 T and back made at 0.4 K. There is a clear transition from a low field state (characterized by a splitting of the oscillation amplitude) to a high field state (characterized by a higher oscillation amplitude and the absence of splitting). This transition shows a strong hysteresis of the dHvA amplitude in the field interval marked by fat arrows in Fig.1b. Furthermore, there is a significant shift between up and down sweep curves in the high field part indicating a complex magnetic state.

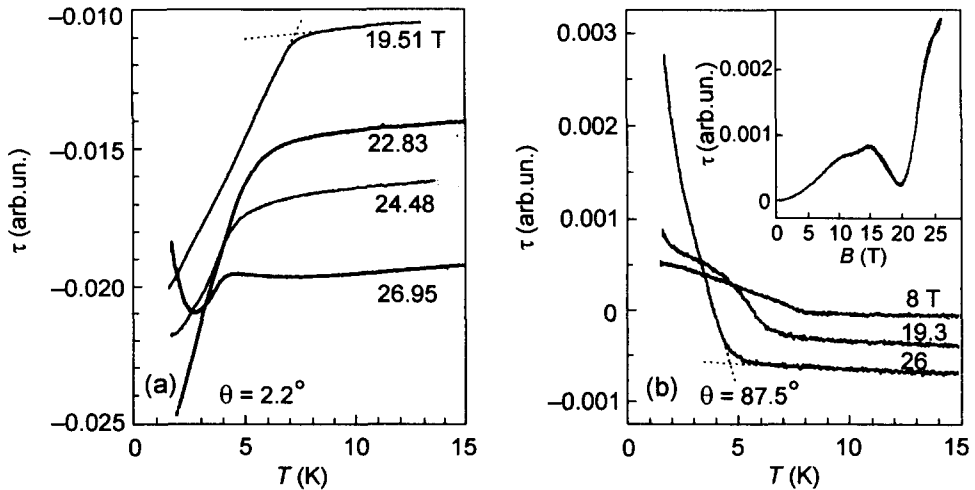


Fig.2. Temperature sweeps of the torque at $\theta = 2.2^\circ$ (a) and 87.5° (b) at different fields. The inset shows the field dependence of the torque at $\theta = 87.5^\circ$, $T = 1.3$ K

To clarify the latter point, we performed temperature sweeps at constant fields. For these experiments it is of crucial importance to suppress the influence of the oscillatory part [18]. We therefore performed these sweeps at field values, at which the dHvA contribution to the temperature dependence is nearly zero. The results are shown in Fig.2a. Despite a small remanent dHvA contribution there is still a clear transition into a new state even at the highest field.

In order to determine anisotropy effects in the phase diagram, we performed torque experiments at fields almost parallel to the layer plane. The phase transition is clearly

seen in temperature sweeps. Typical examples taken at different fields at $\theta = 87.5^\circ$ are given in Fig.2b. The field dependence of the torque below 4 K shows a complex behavior with a strong hysteresis between up and down field sweeps [21]. This behavior is drastically different from the feature observed at the kink transition at low angles. An example of a field sweep at 1.3 K is shown in the inset in Fig.2b.

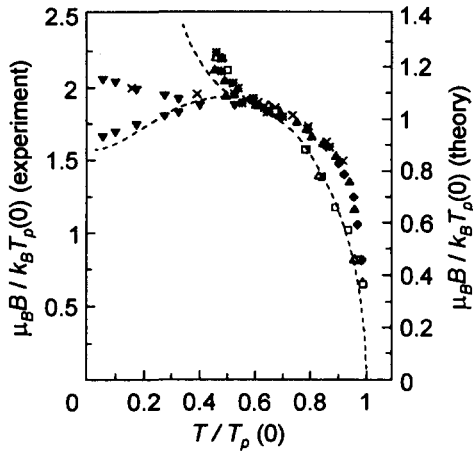


Fig.3. Phase diagram of α -(BEDT-TTF)₂KHg(SCN)₄. Different symbols correspond to the transition points obtained from: the $\tau_{st}(T)$ sweeps at $\theta = 2.2^\circ$ (stars, sample #1), 6.5° (solid diamonds, sample #2), 11.8° (solid up-triangles, sample #3), 87.5° (open squares, sample #1), and 89.5° (open up triangles, sample #1); $\tau_{st}(B)$ sweeps at $\theta = 2.2^\circ$ (crosses, sample #1); and characteristic changes in the dHvA signal at $\theta = 4.0^\circ$ (solid down-triangles, sample #4). The dashed lines represent the phase diagram predicted for a CDW system with a perfectly nested Fermi surface [23]

The results of our studies can be summarized by plotting a $B - T$ phase diagram as shown in Fig.3. Here the data obtained on 4 samples having slightly different T_p (ranging from 8.0 to 8.4 K) are presented. That is why the temperature and field are given in reduced units $T/T_p(0)$ and $\mu_B B/k_B T_p(0)$, respectively [here $T_p(0)$ is the extrapolated critical temperature at zero field]. The definition of the transition points is illustrated in Figs.1 and 2.

The low-angle data in Fig.3 are qualitatively consistent with the $B - T$ diagrams obtained from earlier magnetoresistance [18, 19] and torque [18] measurements in tilted fields: Firstly, the transition temperature continuously decreases with increasing the field; secondly, the low-temperature state is different from the normal non-magnetic state even above the kink transition. Quantitatively, our data are in perfect agreement with those obtained from specific heat measurements at $B \leq 14$ T [22]. These results are obviously in conflict with the SDW model. On the other hand, they can be compared to what is expected for a CDW [23]. At low field the CDW₀ phase with an optimal zero-field wave vector is stable below T_p . As the field increases, the Zeeman splitting of the subbands with antiparallel spins leads to the deterioration of the nesting conditions and, consequently, suppression of T_p [24]. However, when the Zeeman splitting energy reaches the value of the zero-temperature energy gap, a formation of a spatially modulated CDW_z state with a longitudinally shifted wave vector is expected. This state is analogous to the Fulde-Ferrel-Larkin-Ovchinnikov state predicted for superconductors [25] and persists to considerably higher fields than the conventional CDW₀. The phase diagram proposed by Zanchi et al. [23] for a CDW system with perfect nesting is shown by dashed lines in Fig.3. Apart from different field scales, the phase diagrams are remarkably similar to each other.

Assuming the CDW model, the deviation of the actual phase boundary for fields nearly perpendicular to the plane to higher temperatures at $T_p/T_p(0) > 0.6$ can be ascribed to a significant orbital effect of the magnetic field. This effect is important for an imperfectly

nested Fermi surface and leads to a relative increase of T_p [12, 23]. In our case, when the warping of the open Fermi surface sheets is much stronger within the ac -plane than in the interlayer direction, the orbital effect should be anisotropic: its contribution decreases as the angle θ approaches 90° . Indeed, the critical temperature of the transition into the low-temperature low-field state is found to be systematically lower at $\theta \simeq 90^\circ$, lying perfectly on the theoretical line (Fig.3). This implies that the orbital effect is absent for the in-plane field direction.

In the high-field region, the phase lines determined at different field orientations seem to converge, suggesting an isotropic effect of magnetic field on the transition temperature into the low-temperature high-field state. For a definite conclusion, more detailed studies at different angles are needed.

The considerable difference between the field scale in the phase diagram obtained from the experiment and that predicted by the CDW model is not very surprising. Indeed, the model calculations [23] are made within a mean-field approximation neglecting fluctuation effects. The latter may significantly lower $T_p(0)$ with respect to the mean-field value. Furthermore, the imperfect nesting which likely occurs in the present system has a stronger suppressing effect on $T_p(0)$ than on the critical field [23]. Both these factors lead to an underestimation of the actual critical fields.

Finally, we note that the field dependence of the torque at high angles has no simple explanation within the proposed model. The non-monotonic torque with a hysteresis between up and down field sweeps observed at $\theta \gtrsim 60^\circ$ [21] is reminiscent of multiple phase transitions. As the angle approaches 90° , the features become less pronounced though still persist to the angles as high as 88 - 89° (see inset in Fig.2b). In principle, an additional phase transition into a CDW_y state with a transversally shifted wave vector may be expected at high angles at which the orbital effect is sufficiently suppressed [23]. Still, it cannot account for the whole structure of the torque at high angles and its complicated angular dependence. Obviously, the applied model [23] is too oversimplified to explain all the field effects. For a more adequate description it seems very important to include the Q2D band into consideration. In particular, it was recently shown that oscillations of the chemical potential due to the quantization of the 2D orbits have a significant impact on the CDW gap [26]. On the other hand, the magnetization anisotropy itself, revealing an "easy-plane" spin polarization at low temperatures [15], indicates a non-trivial magnetic structure linked to the probable CDW.

In conclusion, we have presented a $B - T$ phase diagram of α -(BEDT-TTF)₂KHg(SCN)₄ built on the basis of magnetization measurements. The shape of the diagram and the effect of the field orientation are suggestive of a CDW formation accompanied by imperfect nesting of the Q1D part of the Fermi surface. If this is true, the high-field phase would represent the first example of a CDW with a spatially modulated wave vector.

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