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Fermi surface and magnetic properties of low-dimensional organic conductors

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We report systematic high-magnetic-field and low-temperature measurements on several members of the α-(BEDT-TTF)$_2$MHg(SCN)$_4$ family of low-dimensional organic conductors. For M = K, transport, high-pressure and magnetic studies show the presence of a magnetic phase which is removed either with high magnetic fields or with high pressure. For M = Rb, transport measurements indicate the existence of a magnetic phase similar to the case for M = K, but with a higher transition temperature and critical field. For both M = K and Rb the split Landau level phenomenon is observed. We also report the pressure dependence of the Fermi surface for M = NH$_4$ and K.

α-(BEDT-TTF)$_2$MHg(SCN)$_4$ is a family of low-dimensional organic conductors (where M = NH$_4$, K, or Rb) [1] which has proved to be of great interest due to the remarkable difference in low-temperature physical properties with only small variations in the anion structure through the substitution of M. The material α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ is a superconductor below 1.5 K [2] and in high magnetic fields exhibits a single Shubnikov–De Haas (SDH) frequency of 568 tesla [3]. In striking contrast, the material α-(BEDT-TTF)$_2$KHg(SCN)$_4$ is not a superconductor, but has been shown to exhibit a magnetic phase transition below 8 K [4] to a new magnetic state, which is most likely of an antiferromagnetic nature. This material has been studied by a number of researchers [5–12]. This state is removed at very high magnetic fields (the critical field H_k is about 24 tesla) and also by high pressure. The SDH and the De Haas–Van Alphen (DHVA) effects show that the Landau levels are split, and the oscillatory effects are affected by the behavior of the magnetic phase, especially when the phase line is crossed in field. Finally, for α-(BEDT-TTF)$_2$RbHg(SCN)$_4$, we will report below recent results which show that this material also exhibits the Landau level splitting behavior, and has a low-temperature magnetic phase with a critical temperature of 11 K, and a critical field in excess of 30 tesla.

In fig. 1 representative magnetoresistance (SDH) and torque magnetization measurements (DHVA) are shown, done for α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at low temperatures up to 30 tesla. The splitting of the Landau levels is clearly observable, and the 'kink' field H_k is evident near 24 tesla. Other unusual features in fig. 1 include the dramatic change in the torque magnetization signal above H_k, the pronounced hysteresis which appears, and the highly field and hysteresis dependent wave forms of the oscillations.

A magnetic phase diagram based on the magnetic field dependence of the magnetoresistance

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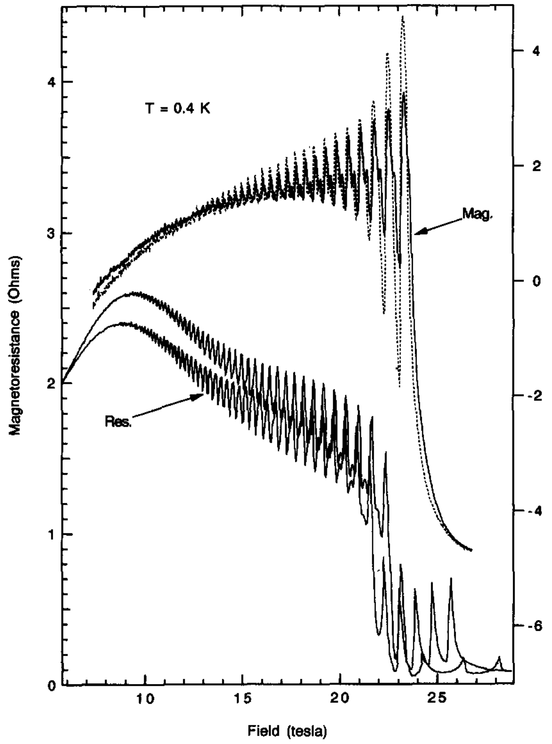

Fig. 1. Magnetoresistance and torque magnetization of α-$(BEDT-TTF)_2KHg(SCN)_4$ at low temperatures. Magnetization: dashed line, down-sweep. Resistance: lower curve, down-sweep.

for different temperatures is presented in fig. 2. The main magnetic phase is expected to be an antiferromagnetic spin density wave phase, and there is strong evidence that the low-temperature high-field phase may be of a ferromagnetic character. We note that ferromagnetism has recently been observed in the related compound κ-$(BEDT-TTF)_2Cu[N(CN)_2]Cl$ [13]. Recent work by Kouno et al. [14] has established the pressure dependence of the magnetic phase, with a critical pressure of about 6 kbar where the phase is completely removed. Another aspect of the phase diagram involves an additional phase line within the main phase (which marks the onset of low-field hysteresis near 10 tesla) which has recently been reported [12].

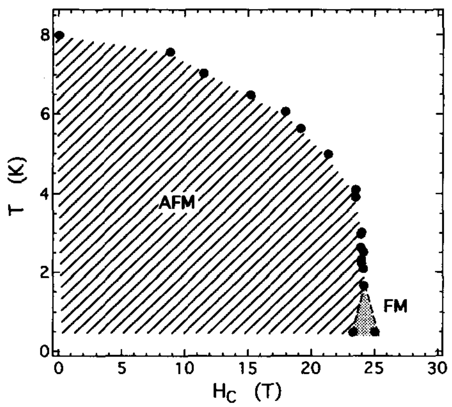

Fig. 2. Magnetic phase diagram of α-$(BEDT-TTF)_2KHg(SCN)_4$.

The behavior of the SDH and DHVA oscillations within and above the magnetic phase boundary deserves special attention. We have investigated the applicability of the Lifshitz–Kosevich (L–K) formalism to the problem. Here there are key parameters, the g-factor, the Dingle temperature (T_D) and the Fermi surface topology (i.e. the number of frequencies), all of which may have some magnetic field dependence, and all of which may change in some important way when the magnetic phase line is crossed. The magnetic-field-dependent g factor was first considered by Sasaki et al. [15] and the possibility of a doubling of the closed-orbit Fermi surface due to nesting has been discussed by Doporto et al. [9]. We have recently considered the magnetic field dependence of T_D and g. Here it is possible that there are two terms for T_D, one for each spin orientation. Below H_k, T_D and g may change smoothly with magnetic field. Above H_k both T_D and g could change very rapidly with the field. L–K simulations using models for the field dependence of these parameters can come quite close to predicting the experimental results. However, there are many details, such as the mechanism of magnetic interaction, which may also play an important role. More work will be needed to provide a completely satisfactory simulation of the oscillatory behavior.

We feel that there is a reasonably good explanation for the nonoscillatory background resistance in terms of the open and closed orbits determined from band structure calculations for this material [16]. The open- and closed-orbit conductivity terms are added in parallel. In the event that the open orbits nest (and produce a spin-density wave state) then only the closed-orbit conductivity will remain dominant. With increasing magnetic field the closed-orbit magnetoresistance will increase. However, at high enough magnetic fields, if the SDW state is reduced and eventually removed at H_k, then the open-orbit conductivity will reappear and shunt the magnetoresistance. This model would explain the maximum in the magnetoresistance and the precipitous drop at H_k.

The temperature dependence of the resistance in α-(BEDT–TTF)$_2$KHg(SCN)$_4$ is shown in fig. 3 for different magnetic fields. The shoulder near 8 T for zero fields is the onset of the low-temperature magnetic phase. The large variation in magnetoresistance with increasing field is evident in the sequence of measurements shown.

We have investigated the pressure dependence of the SDH oscillation frequency in α-(BEDT–TTF)$_2$MHg(SCN)$_4$ for M = K and M = NH$_4$. In both cases pressures of approximately 8 to 10 kbar changes the closed-orbit areas of the Fermi surface by as much as 15%. Our results are shown in fig. 4. There is some indication of a maximum in the SDH frequency with pressure for α-(BEDT–TTF)$_2$NH$_4$Hg(SCN)$_4$, but more work is needed to verify this dependence. We note that α-(BEDT–TTF)$_2$NH$_4$Hg(SCN)$_4$ does not exhibit splitting of the Landau levels, either at zero pressure or at high pressures. In marked contrast, the splitting behavior in α-(BEDT–TTF)$_2$NH$_4$Hg(SCN)$_4$ disappears at 4 kbar, but at 8 kbar we have noted an onset of splitting above about 21 tesla [11]. It is possible that there are considerable changes in the Fermi surface, not only in the extremal area, but in the effective dimensionality and the band structure.

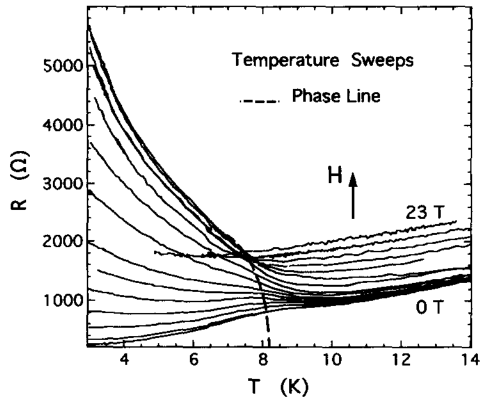

Fig. 3. Temperature dependence of the resistance of α-(BEDT–TTF)$_2$KHg(SCN)$_4$ for different magnetic fields.

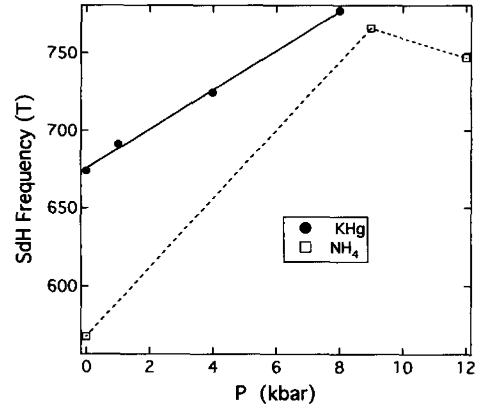

Fig. 4. Pressure dependence of the SDH oscillation frequency for M = K and NH$_4$.

Recently, we have investigated the magnetoresistance and SDH effect in α-(BEDT–TTF)$_2$RbHg(SCN)$_4$. A resistance anomaly similar to that shown in fig. 3, but at a higher temperature of 10 K, has been previously reported [17]. We find a striking similarity in behavior with α-(BEDT–TTF)$_2$KHg(SCN)$_4$. The magnetoresistance is large with a maximum at around 15 tesla at low temperatures, the Landau levels show the splitting behavior (the fundamental frequency is 676 tesla and the effective mass is 1.5m_0) and there is a kink field H_k which is observable at higher temperatures below 30 tesla. Our results are shown in fig. 5.

In fig. 6, a tentative magnetic phase diagram for α-(BEDT–TTF)$_2$RbHg(SCN)$_4$ is presented. Here the differences from α-(BEDT–TTF)$_2$ KHg(SCN)$_4$ are apparent. The transition temperature is near 11 K, not 8 K, and the kink field at low temperatures is probably near 35 tesla, not 24 tesla. Higher magnetic fields will be needed to explore the low-temperature behavior of the kink field in this case.

Finally, we have made some preliminary AC

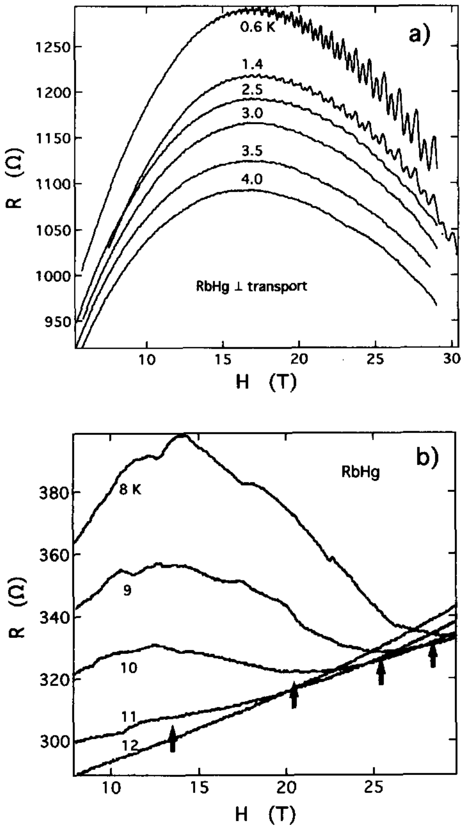

Fig. 5. Magnetoresistance of α-$(BEDT-TTF)_2RbHg(SCN)_4$ at low temperatures (a) and at higher temperatures (b) where the critical field H_k (arrows) is evident.

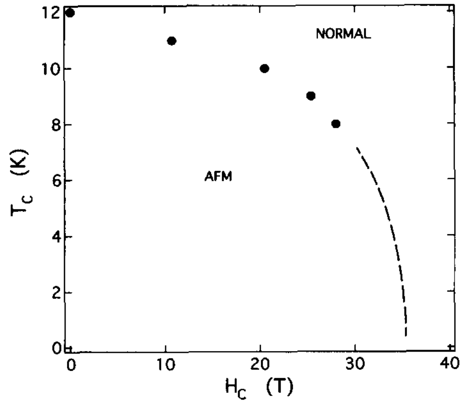

Fig. 6. Magnetic phase diagram of α-$(BEDT-TTF)_2RbHg(SCN)_4$. Note: the dashed line above 30 tesla is an estimate of the high-field phase-line behavior.

specific-heat measurements to search for calorimetric evidence for a phase transition along the $H_k(T)$ phase boundary in α-$(BEDT-TTF)_2KHg(SCN)_4$. Both constant-field and constant-temperature scans across the phase boundary have been made, but we have observed no noticeable changes in the specific heat which would indicate a phase transition. There may be several reasons why, for instance, no specific heat jump is observed. For constant field, both resistance and susceptibility measurements show a very gradual change near 8 K. Secondly, the phonon contribution at such high temperatures may swamp the electronic contribution. For constant temperature (near 0.5 K) we observed a large Schottky term at low fields which vanished by 6 tesla, but at H_k the variations in signal were very small in comparison and no distinctive feature was observed. We estimate that if there is a jump or variation in the specific heat as the kink field is crossed, it is probably less than 5 mJ/g K. More work will be needed to determine the precise nature of the electronic and magnetic contributions to the specific heat in the magnetic phase of the material.

In conclusion, we have presented some recent results on the high-magnetic-field- and pressure-dependent properties of the α-$(BEDT-TTF)_2MHg(SCN)_4$ family of low-dimensional organic conductors. WE find that for both M = K and Rb there is a low temperature magnetic phase with similar split Landau level behavior. The critical temperature and field for M = Rb are considerably higher than for M = K. In general, we find that the L-K formalism is applicable to predict the behavior of the SDH and DHVA oscillations in these materials, but that parameters such as the g factor and the Dingle temperature may be magnetic-field-dependent, and could change dramatically above the critical field H_k. More work is needed to produce simulations in complete accord with the experimental data. We have also investigated the pressure dependence of the Fermi surface for M = K and NH_4, and find that it increases as much as 15% at 8 kbar. In no case did α-$(BEDT-TTF)_2NH_4Hg(SCN)_4$ exhibit split Landau levels.

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