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Near quantum limit SdH and dHvA wave forms in low-dimensional metals

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Abstract

We describe experiments in steady magnetic fields up to 33 T at low-temperatures on low-dimensional organic metals where the Landau level separation becomes greater than either the thermal or the impurity broadening. High precision, simultaneous Shubnikov–de Haas and de Haas–van Alphen measurements allow direct correlation of the oscillations of the magnetization with the dissipation as the Landau levels cross the Fermi level. From these results, we can determine with reasonable certainty the functional form of the density of states, and their relative values in the open- and closed-orbit bands.

Keywords: Organic conductors; Fermi surfaces; de Haas–van Alphen oscillation; Shubnikov–de Haas oscillation

The electronic behavior of the materials α-(BEDT-TTF)$_2$MHg(SCN)$_4$, where M is K or NH_4, continues to be an active area of interest due to the complexity which arises from the open- and closed-orbit band nature of the Fermi surface, as shown in Fig. 1.

Of specific interest is the nature of the Shubnikov–de Haas (SdH) and de Haas–van Alphen (dHvA) oscillations in high magnetic fields. Here the near quantum limit is realized in these materials at 30 T and 0.5 K, since $\hbar\omega_c$ is of order 20 K for an effective mass of 2 m_0. This is greater than either the impurity energy ($\hbar/\tau \approx 1$ K) or the interlayer bandwidth energy (≈ 5 K or less). Hence, the wave forms show that the Landau level separation is greater than any other characteristic energy. A result of this is that in high fields the standard Lifshitz and Kosevich (LK) treatment of the SdH oscillation amplitudes [1] of both materials yield spurious effective mass (m^*) parameters which appear to be field dependent. A second issue is the difference in the two materials in terms of ground-state properties [2] and anomalous wave form behavior [3,4] in high magnetic fields. The extreme quantum limit is reached at a magnetic field of order of the fundamental frequency of the SdH orbit under investigation, which is between 500 and 700 T in these materials for the fundamental closed orbital.

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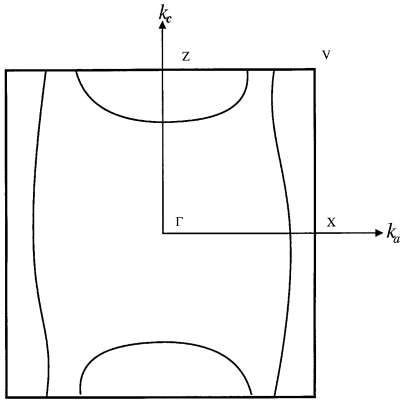

Fig. 1. Fermi surface for α-(BEDT-TTF)$_2$MHg(SCN)$_4$. It is characterized by 1D open sheets and 2D closed orbits.

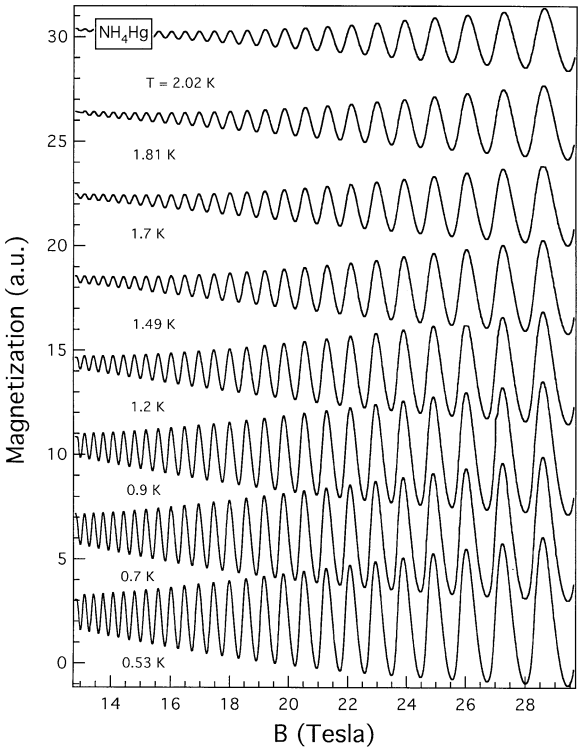

Fig. 2. de Haas–van Alphen effect in α-(BEDT-TTF)$_2$ NH$_4$Hg(SCN)$_4$ up to 33 T. Here $m^* = 2.45m_0$; $T_D = 0.55$ K, and $F_0 = 580$ T.

In this work we address the ratio of the open- and closed-orbit density of states in the two materials, and also explicitly show the relationship between the SdH and dHvA wave forms. High-quality

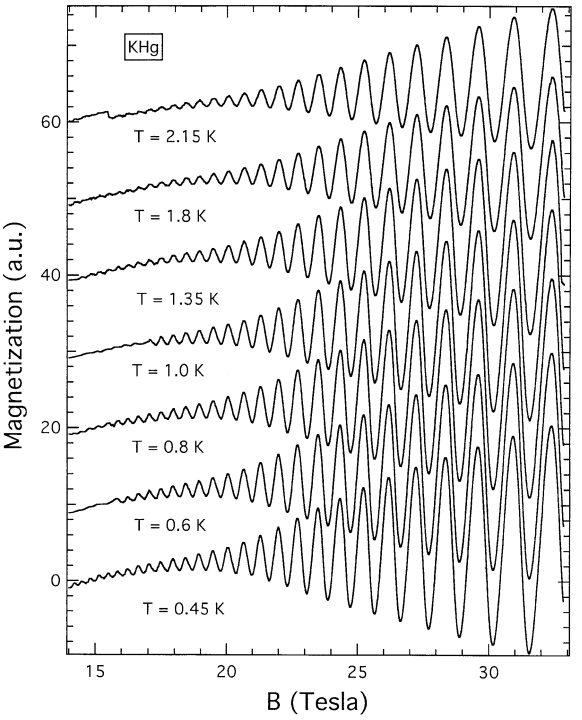

Fig. 3. de Haas–van Alphen effect in α-(BEDT-TTF)$_2$ KHg(SCN)$_4$ up to 33 T. Here $m^* = 1.85m_0$; $T_D = 2.0$ K, and $F_0 = 680$ T.

dHvA measurements on both $M = \mathrm{K}$ and $\mathrm{NH_4}$ materials, using a cantilever magnetometer method, are shown in Figs. 2 and 3 as a function of temperature in high magnetic fields. From these measurements, we may estimate the ratio of the density of states, following the method of Harrison et al. [5]. The magnetization is a thermodynamic function of state which can be written analytically as

$$M[B, T, \mu] = M[B, T, \mu] = -\partial\Omega/\partial B|_{\mu,T}.$$

The thermodynamic potential can be written as $\Omega = -kT\int g[\varepsilon, B]\ln[1 + \mathrm{e}^{(\mu-\varepsilon)/kT}]\,\mathrm{d}\varepsilon$.

Therefore, a numerical solution can be calculated for $g[\varepsilon, B]$, the density of states, based on a correct choice of the form and magnetization data. It has been shown in Ref. [5] that in this system, g_{1D} can be considered as a constant and independent of magnetic field and ε as a first approximation. Therefore, $g[\varepsilon, B]$, can be written as $g[\varepsilon,B] = g_{1D} + g_{2D}[\varepsilon, B]$.

The form of $g_{2D}[e, B]$ has been experimentally shown to have a Lorentzian broadening distribution form [6]. Hence, we may write the DOS as

$$g_{2D}[\varepsilon, B] = (2eB/\hbar\pi)\sum\Gamma((E - E_n)^2 + \Gamma^2)^{-1},$$

where Γ is the broadening and is given by $\Gamma = \hbar/2\tau$ and E_n is the energy of the Landau level of index n given by $E_n = (n + 1/2)\hbar\omega_c$.

From the magnetization results, the Dingle temperature T_D and effective mass m^* for the materials were calculated using LK formalism. We emphasize that the LK was applied in the low-field limit (below 20 T for NH_4 and just above the "kink field" at 22.5 T for K in the metallic phase) where the uncertainties in the m^* parameter are the least. These in turn were used for the DOS numerical calculations. Using the foundation laid out above, numerical fits for the magnetization were made, and the ratio g_{1D}/g_{2D} was calculated for the two materials. (Note that the vaule for g_{2D} is the field averaged DOS.) The results were as follows: $g_{1D}/g_{2D} \approx 0.5$ for the K material and $g_{1D}/g_{2D} \approx 2.0$ for the NH_4 material. Our results indicate that in the K material the dominant factor is the DOS of the 2D orbits, whereas it is the DOS of the 1D orbits in the NH_4 material. It may be that this difference in DOS ratios determines the superconducting (NH_4) or density wave (K) ground states, and may also affect the very different wave forms seen in SdH studies of the two materials in the high-field and low-temperature limit. Additionally, M = K shows possible signs of the quantum-Hall effect whereas $M = NH_4$ does not. One possible explanation may lie in the role of the DOS ratio.

We now turn to simultaneous measurements of the SdH and dHvA oscillations in the material α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$, as shown in Fig. 4. Measurements were performed on the same crystal as the results in Fig. 2 above. Here 4-terminal (12 μm gold wire) AC resistance along the b-axis was measured simultaneously with the capacitance of the cantilever magnetometer upon which the

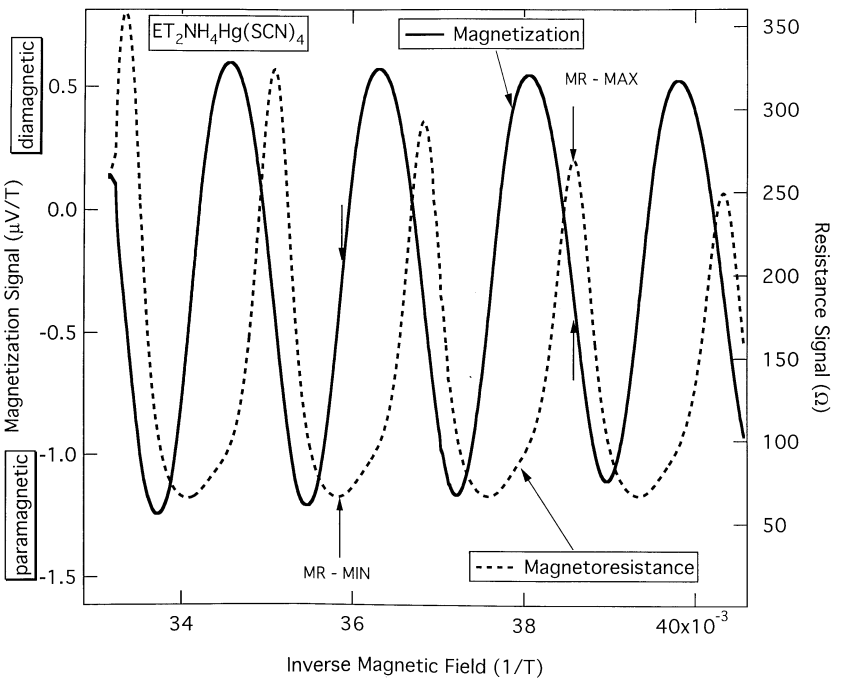

Fig. 4. Simultaneous SdH and dHvA measurements on α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ versus inverse magnetic field at 0.5 K. The crossing points of the magnetization signal correspond to the maxima and minima of the SdH signals. Although the magnetization signals are nearly sinusoidal, the SdH wave forms are asymmetric in the minima.

sample was mounted with the *b*-axis about 5°. away from the field direction. The purpose of this is to allow for an unambiguous determination of the sign of the signal (dia- versus paramagnetic). No cross talk between the two measurements was observed, and the magnetization signal was identical to that previously observed with no electrical connections. From this data we may observe that (a) the resistivity peaks when the magnetization crosses from para- to diamagnetic, as expected when the Fermi level sits between Landau levels, and has a minimum when the Fermi level is within a Landau level for the dia- to paramagnetic transition; (b) in all our dHvA measurements we see no evidence for saw tooth wave forms, nor any evidence for quantum-Hall effect signatures as have been reported in pulsed field magnetization studies. We have, however, seen in pulsed field studies [7] and in the present study (Fig. 4) a small anomalous feature near the mimima in the SdH wave forms above about 20 T. Similar, but more dramatic anomalies are seen in the K material [3,8], and it is possible that the differences in the DOS in the two cases as discussed above is responsible for the differing wave forms.

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