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Two-dimensional Fermi liquid with fixed chemical potential

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The observation of magnetic quantum oscillations in three-dimensional (3D) metals is well proven,1 within the framework of the Lifshitz–Kosevich (LK) theory,2 to be a powerful tool for obtaining important band-structure parameters by means of quantitative analyses of experimental data. This is in contrast to two-dimensional (2D) conductors in which a qualitatively different behavior is expected. For an ideal 2D metal, i.e., a metal with only one 2D Fermi surface (FS), at sufficiently high-magnetic fields, B, a sawtooth-like de Haas-van Alphen (dHvA) signal (with sharp drops of the magnetization M for rising fields) is predicted. While an analytical formula for the 2D electron gases applicable when \( \hbar \omega_c \gg k_B T \) (where \( \omega_c = eB/m_c \) is the cyclotron frequency, \( m_c \) is the effective cyclotron mass, \( k_B \) is the Boltzmann constant, and \( T \) is the temperature) was derived in Ref. 3, important damping effects, such as those arising from quasiparticle scattering, were not included. Such effects can considerably broaden the Landau levels and thereby mask the sawtooth-like dHvA signal.

In reality, however, these damping effects are not absent. Thus, for instance, the dHvA signal in dithio-tetra-thia-fulvalene is predicted. While an analytical formula for the 2D electron gases applicable when \( \hbar \omega_c \gg k_B T \) (where \( \omega_c = eB/m_c \) is the cyclotron frequency, \( m_c \) is the effective cyclotron mass, \( k_B \) is the Boltzmann constant, and \( T \) is the temperature) was derived in Ref. 3, important damping effects, such as those arising from quasiparticle scattering, were not included. Such effects can considerably broaden the Landau levels and thereby mask the sawtooth-like dHvA signal.

In reality, however, these damping effects are not absent. Thus, for instance, the dHvA signal in dithio-tetra-thia-fulvalene is observed, which is to be expected when the chemical potential is pinned at a constant value. While this proves the existence of the theoretically predicted quasi-one-dimensional band, it further implies that this band has an exceptionally large density of states.

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number of organic metals.\textsuperscript{9,12–15} The applicability of the LK formula in the case of the SdH effect is, nevertheless, problematic owing to complications involving the conductivity tensor, and other novel effects that appear to be unique to magnetotransport.\textsuperscript{12,16} Hence, a reliable comparison of experimental results with theoretical predictions should be done with true thermodynamic quantities such as the magnetization. Here, we provide experimental evidence for a unique situation where the magnetic quantum oscillations of a 2D metal are qualitatively different from the theoretical expectation\textsuperscript{3} and from the observed behavior in semiconductor-based 2D electron gases\textsuperscript{5,6} and bulk organic metals.\textsuperscript{4,8,9} Rather unusually, the measured dHvA signal has exactly the form to be expected for a system in which the chemical potential is pinned at a constant value, i.e., the 2D grand canonical ensemble.\textsuperscript{1,4}

The single crystals of the organic superconductor $\beta^\ast$-(BEDT-TTF)$_2$SF$_6$CH$_2$CF$_2$SO$_3$ ($T_c=4.4$ K) used here were grown by electrocrystallization.\textsuperscript{15} In contrast to earlier crystals\textsuperscript{11,18} the quality of the samples here (which we shall refer to as 1 and 2) were much better, as evidenced by the lower Dingle temperatures (see below). The dHvA effect was measured by means of a capacitance cantilever torque magnetometer mounted on a rotatable sample holder in a $^3$He cryostat at the Grenoble High Magnetic Field Laboratory in fields of up to 28 T.

Figure 1(a) shows the raw torque signal, $\tau=M\times\vec{B}$, of sample 1 at different temperatures. The pronounced peak below about 1.5 T originates from the diamagnetic response of the sample in its superconducting state. For rising fields, a type-II superconductor exhibits diamagnetism, i.e., a negative magnetization. For falling fields, a positive magnetization occurs below the irreversibility point, with the hysteresis being due to vortex pinning effects [see Fig. 2(c)]. Here, we discuss only the torque dHvA data taken on the rising field.

The sign of the torque signal depends on the relative orientation of $B$ with respect to the cantilever, and on the sign of the capacitance change as the cantilever is bent. The observation of the diamagnetic peak in the low-field torque enables the unequivocal determination of the sign of $M$, which is important for the wave-shape analysis of the dHvA oscillations discussed in detail below.

The dHvA signal is clearly apparent as the oscillating part superimposed on a steady background of the torque, increasing with field. The steady part of the torque is proportional to $B^2$, arising from an anisotropic field-independent susceptibility $\chi$ of the sample. In general, $\chi$ is a tensor and the magnetization is proportional to the product of this tensor with $\vec{B}$. Thus, the absolute value of the torque has the form $\tau=\Delta\chi B^2 \sin \theta \cos \varphi$, where $\Delta\chi$ is the susceptibility difference between two principal axes of the $\chi$ tensor and $\varphi$ is the angle between $B$ and one of principal tensor axes. In the present material, the principal axes do not coincide with the crystal axes. One of these axes occurs at $\theta=30^\circ$, where $\theta$ is the angle between $B$ and the normal to the conducting layers, as evidenced by the vanishing of the magnetic torque at this angle. The origin of this unusual anisotropic susceptibility is unclear, but might be a remnant of the local magnetism of the molecular bands.

The torque acting on a sample with an anisotropic FS is proportional to the derivative of the extremal area of the FS with respect to $\Theta$, i.e., proportional to $dF/d\Theta$, where $F=F_0/\cos\Theta$ is the dHvA frequency of a 2D cylindrical FS. Hence, for $\Theta=0$, the amplitude of the dHvA signal in the magnetic torque is rather small. This has been utilized in the measurement of sample 2 shown in Fig. 2(a) to exclude any nonlinearities of the torque signal due, e.g., to the torque interaction effect, and to ensure the reliability of the data.\textsuperscript{19} The steady part of the torque at $\Theta=-0.7^\circ$ follows perfectly the $B^2$ behavior up to the highest fields, implying that the torquemeter operates unequivocally in the linear regime and that the relatively small dHvA signal is undisturbed by experimental artifacts. Another possible distortion of the dHvA signal might in principal be caused by magnetic interaction.\textsuperscript{1} Here, this can only be of marginal importance owing to the small magnetization and the very small dHvA frequency.

The angular dependence of the dHvA frequency with $F_0$
However, at low temperatures and high fields, clear deviations from the conventional 3D LK theory are observed in both samples. Figure 1(b) shows the field dependence of the fundamental dHvA amplitude extracted from the Fourier transformations made over short intervals in \(1/B\) of the data of sample 1 at \(T=0.43\) K. The dashed line depicts the behavior expected for a 3D metal with \(T_D=1.3\) K, as obtained from a best fit to the 3D LK formula at low-magnetic fields. At higher fields, the data lie clearly below the fitted line; note the logarithmic scale of the ordinate. A fit of the 3D LK formula at higher fields would yield a much lower \(T_D\), but leaving the low-field data well below the fit line. A completely analogous behavior is observed for sample 2. A further observation is the apparent field-dependent effective cyclotron mass, \(m_c\). At low fields, below \(\sim 12\) T, \(m_c = 1.9 m_e\) is found to be in good agreement with earlier low-field data. At higher fields, however, the fit to the LK formula results in an appreciably reduced effective mass, i.e., \(m_c = 1.6 m_e\) for an average field of \(B\sim 20\) T [Fig. 1(c)]. A similar apparent mass reduction with increasing magnetic field was observed previously with dHvA data of another organic superconductor, where it could be explained by the influence of chemical-potential oscillations on the wave form in a 2D spin-split Fermi liquid. A remarkable new aspect of the data reported here, is the clear observation of a “inversed sawtooth” of the dHvA signal with this numerical model showing to be distinguished from theory [solid line]. The parameters used are those obtained from previous measurements and from the low-field high-temperature data of the present experiment, i.e., \(m_c = 1.9 m_e\), \(g m_e/m_e = 3.9\) (which is independent from the theory employed), and \(T_D = 0.4\) K. Indeed, the latter parameter can be analytically extracted from the present data by means of fits to Eq. (1) and fits with the low-field estimate using the 3D LK formula. It is the exceptionally high quality of sample 2 that gives rise to the inverted-sawtooth shape of the dHvA signal, which then becomes less pronounced—but still detectable—for samples with higher scattering rates, i.e., for sample 1 with \(T_D = 1.3\) K. Note that, with the exception of a simple scaling factor, no free parameter is necessary to obtain the almost perfect agreement of data and 3D theory (1).

An important ingredient for the validity of Eq. (1) is the assumption of a fixed chemical potential, or equivalently, the existence of a charge-carrier reservoir. Band-structure calculations do after all predict the existence of a pair of quasi-1D electron surfaces in addition to the 2D hole pocket that gives rise to the dHvA signal. It is therefore most likely that it is this 1D band that acts as charge-carrier reservoir causing the chemical potential to be pinned. The influence of an additional density of states (DOS), originating from 1D bands, has been shown to modify the dHvA wave shape. Simulations of the dHvA signal with this numerical model show that the 1D DOS has to be at least approximately five times larger than the 2D DOS at the Fermi level. However, the numerical model reproduces the experimental data somewhat less accurately than Eq. (1). In particular, it is the steep slopes of the rising magnetization that cannot be reproduced perfectly by the numerical model.

Until now, there has been no direct evidence for the existence of these 1D bands in the present material. Only some rather indirect indications, such as a negligible Hall effect and the enormous background magnetoresistance, have
been reported. The present results show clearly that \( \beta^\prime \)- (BEDT-TTF)\(_2\)SF\(_5\)CH\(_2\)CF\(_2\)SO\(_3\) is a compensated metal with a large 1D DOS. This provides additional support for the validity of the predicted band structure and makes speculations on a possible density-wave transition questionable.

Although the expression for the 2D grand canonical ensemble reproduces the experimental dHvA data at low temperatures rather well, the temperature dependence of the signals, i.e., the apparent field dependence of \( m_c \), cannot be explained. The 2D grand canonical ensemble predicts the same temperature dependence of the dHvA signal as the 3D formula. In the numerical model, a notable apparent field-dependent effective cyclotron mass is only expected close to a spin-splitting zero, which is not the case here. The origin for the reduced \( m_c \) found for both samples, independent of quality, is unclear but might be caused by a crossover from the 2D theory at low temperatures to the 3D behavior at higher temperatures. It would be of considerable interest to investigate the origin of this apparent field-dependent \( m_c \).

There is a debate on the actual Landau-level broadening due to impurity scattering; e.g., whether the Landau levels have Gaussian or Lorentzian line shapes (see Ref. 5 for references). The very clean dHvA signal in our case can be well reproduced by Eq. (1) assuming a Lorentzian broadening. As we checked by means of the numerical model, a field-independent Gaussian Landau-level profile gives a less good description of the experimental data since it results in a much weaker slope for the increasing branch of the dHvA signal.

The pronounced two dimensionality of the present material is equally reflected in strong anharmonicities of the magnetoresistance oscillations at higher fields and lower temperatures. This is partially understood since the SdH effect, i.e., the relative conductivity oscillations, is expected to be proportional to \( B^2 dM/dB \). This leads to strongly peaked nonsinusoidal SdH signals, as have been observed experimentally. However, the large background magnetoresistance in strong fields, with an apparent field-induced insulting behavior, prohibits a reliable comparison of the experimental magnetoresistance oscillations with theory as is possible here for the dHvA effect.

In conclusion, we have found clear experimental evidence for a 2D Fermi liquid in a bulk organic material. In contrast to previously reported scenarios, we observed magnetic quantum oscillations that are characteristic of a 2D grand canonical ensemble, which can only be possible if the chemical potential is fixed. This, therefore, can be considered as proof for a field-independent fixed chemical potential, indicating the existence of a large 1D DOS at the Fermi energy of the present material.

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17 U. Geiser, J.A. Schluter, H.H. Wang, A.M. Kini, J.M. Williams,


19 Torque interaction was indeed observed for another sample in a separate experiment. This resulted in a steepening of the falling side of the dHvA signal.

20 Higher values than five for the 1D DOS result in only marginal changes of the dHvA signal.

21 The low-field SdH data in Ref. 18 are still in line with the conventional 3D LK theory.