Spin-split de Haas–van Alphen effect in two-dimensional electron systems

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Spin-split de Haas–van Alphen oscillations in two-dimensional (2D) electron systems are studied experimentally and theoretically. It is shown that large second harmonic observed at low temperatures in the quasi-2D organic metal $\kappa-(ET)_2I_3$ ($ET \equiv BEDT-TTF$) is due to the effect of spin-splitting close to its maximal value. The calculated shape and amplitudes of magnetization oscillations are in remarkable quantitative agreement with the experimental ones. [S0163-1829(98)51944-3]

Two-dimensional (2D), or quasi-2D conductors under high-magnetic fields have become a subject of considerable current interest due to recent reports of peculiar magnetoquantum oscillations in 2D electron systems, such as in $GaN/Ga_xAl_{1-x}N$ heterostructures[1] or in the nearly 2D organic metal $\kappa-(ET)_2I_3$.[2] In particular, the measured de Haas–van Alphen (dHvA) and Shubnikov–de Haas (SdH) oscillations in the latter material exhibit a dramatic appearance at low temperatures of a significant oscillatory structure corresponding to doubling of the fundamental frequency. In what follows we illustrate the observed oscillations in $\kappa-(ET)_2I_3$ by means of dHvA experiments and show by a simple analytical scheme based on the independent electrons model that this behavior is associated with Zeeman spin splitting of the Landau levels under the condition of two dimensionality, an effect that is more pronounced at high fields and low temperatures.

SdH and dHvA experiments were carried out in fields up to 27 T and temperatures between 0.38 and 1.3 K. Crystals were mounted on a rotatable sampleholder and could be tilted from the field orientation perpendicular to the conducting (b,c) planes ($\Theta = 0°$). DHvA experiments were carried out by means of torque technique that could not be applied at $\Theta = 0°$, where only SdH measurements are presented. Figure 1(a) shows dHvA oscillations of $\kappa-(ET)_2I_3$ at $\Theta = 9°$. While at 1.3 K the oscillations corresponding to $F = 3883$ T are dominant, at lower temperatures an additional set of oscillations arises leading to an enhanced second harmonic in the Fourier spectrum.[2] This effect occurs in both SdH and dHvA experiments, and is observed in the entire angular range $0° \leqslant \Theta \leqslant 40°$. The additional set of oscillations, which later will be attributed to Zeeman splitting, *lies always exactly midway* between those that are already present above 1 K.

To account for magnetoquantum oscillations in a 2D metal a model has been considered recently (analytically in Ref. 3 and numerically in Ref. 4) where the electrons on a closed Fermi surface (FS) cylinder may be exchanged with electrons in an open planar sheet of the same FS. The degree of this exchange depends on the energy barrier and the gap in momentum space between the cylinder and the sheet.[3] A rather similar model, but for two coupled closed pockets of carriers, was considered in Ref. 5. A general formula for the dHvA oscillations in 2D metals similar to the classic Lifshitz-Kosevich (LK) formula for three-dimensional (3D) metals,[6] was derived by Shoenberg.[7,8] The effect of Zeeman spin splitting of the Landau levels was considered, however, only in the limiting case of perfectly sharp levels at $T = 0$ K. An analytical method, in which only two levels around the chemical potential were taken into account, was developed for canonical ensemble in Ref. 9 and was generalized in Ref. 3 for a direct summation of an arbitrary number of energy levels around the chemical potential. It yields simple formulas and exact results in the low-temperature–high-magnetic field regimes in terms of the parameter $Q \equiv \hbar\omega_c/k_BT \simeq 1.34B(T)/T(K)\eta_c$, where $\eta_c \equiv m_c/m_e$, m_c is the cyclotron mass, and m_e is the free electron mass. At $Q \geqslant 2\pi^2$, where the LK-Shoenberg (LKS) series[7] fails to de-

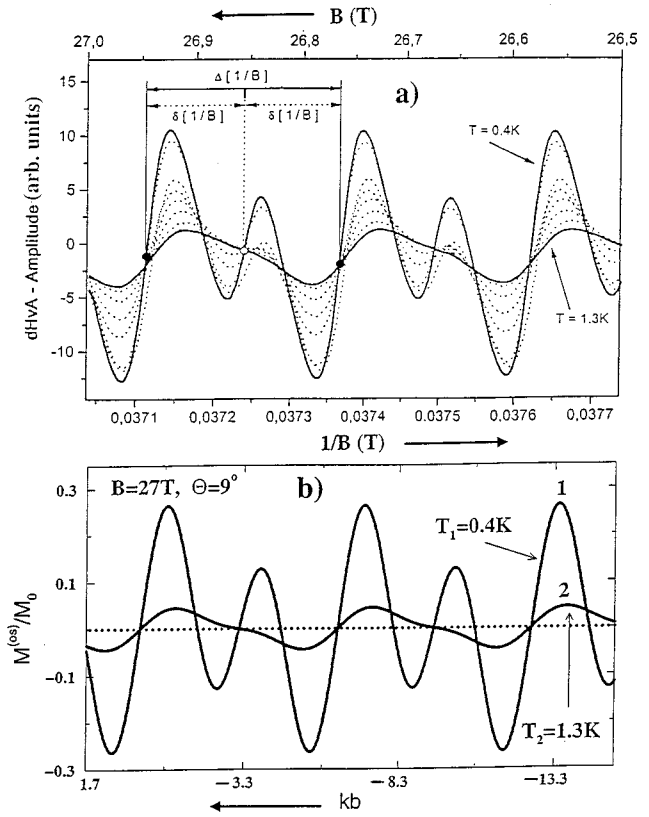

FIG. 1. Spin-split magnetization oscillations in $\kappa-(ET)_2I_3$ at $\Theta=9°$ for temperatures $T_1=0.4$ K and $T_2=1.3$ K. (a) Experiment. (b) As calculated from theory [Eq. (3)] (with $\eta_{c0}=3.9$, $G_0=4.39$, $s_0=0.39$). Parameter Q ranges from 7.0 (at T_2) to 22.9 (at T_1).

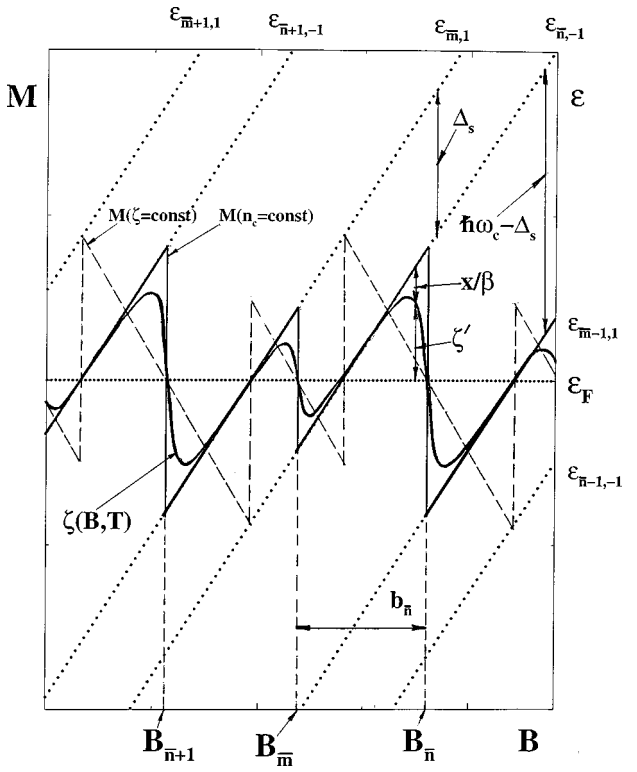

FIG. 2. Schematic magnetization and chemical potential oscillations in a 2D metal with spin-split Landau levels, $\varepsilon_{n,-1}(B)$, $\varepsilon_{m,1}(B)$. Also shown are the ideal $T=0$ K magntization oscillations for a fixed chemical potential (dashed curve), and for constant electron concentration (full curve).

scribe the oscillations via a small number of harmonics, this method has a clear advantage. We shall show that both the harmonics and the levels representations are necessary to account for different aspects of the experimental data in various organic metals.

The electronic thermodynamic potential of a 3D metal in which the conduction electrons have a 2D energy dispersion (i.e., with a cylindrical FS) under magnetic field B, tilted with respect to the a^* axis (i.e., to the direction perpendicular to the easy conducting planes) at angle Θ, is[10]

$$\frac{\Omega}{V}=\frac{A}{\beta}B\sum_{\sigma}\sum_{n=0}{}^{\frac{1}{2}}\ln\{1+e^{(\zeta-\varepsilon_{n,\sigma})\beta}\}\,, \tag{1}$$

where $A\equiv 2\cos\Theta/(\phi_0 a^*)$, $\phi_0=hc/e$ is the flux quantum, $\beta\equiv 1/k_BT$, $\zeta(B,T)$ is the chemical potential, V is the volume, and a^* is the lattice constant in the anisotropic direction. In this expression the spin-split Landau levels are $\varepsilon_{n,\sigma}=(n+1/2)\hbar\omega_c+(g/2)(\sigma/2)\hbar\omega_e$, $\sigma=\pm 1$, $\hbar\omega_c=\mu_c B$, $\mu_c=e\hbar/m_c c$, $m_c=m_{c0}/\cos\Theta$, m_{c0} is the cyclotron mass at zero tilt angle, $\hbar\omega_e=\mu_e B$ is the Zeeman splitting energy for free electrons, $\mu_e=e\hbar/m_e c$, and g is the electron g factor (for free electrons $g=2$).

We define an effective spin-splitting energy as the energy difference between two adjacent levels $(\bar{n},\bar{m})$ with opposite projections of spin (Fig. 2): $\Delta_s\equiv|\varepsilon_{\bar{n},-1}-\varepsilon_{\bar{m},1}|=|I(G)-G|\hbar\omega_c$, where $I=[G]=\bar{n}-\bar{m}$ is the integer part of $G\equiv(g/2)\eta_c$. We may introduce spin-splitting parameter $s\equiv\Delta_s/\hbar\omega_c=|I-G|$: $0\leqslant s\leqslant 0.5$. For example, for $G=3.6$ or 4.4, $I=4$ and the spin-splitting parameter is $s=0.4$. For an integer G, the effective spin splitting is zero and spin-polarized levels are degenerate; for half integer G, $s=0.5$ and the spin-polarized levels are equidistant, separated by the energy $\hbar\omega_c/2$.

It can be proved quite generally that for a 2D metal, with a constant number of electrons in the conduction band, the oscillating part of the chemical potential is related to the magnetization oscillations by the formula[3]

$$\frac{M^{(\mathrm{os})}(B)}{M_0}=\frac{2}{\hbar\omega_c}\,\zeta'(B)+O(\hbar\omega_c/\varepsilon_F)\,. \tag{2}$$

Here $M_0=\varepsilon_F/\phi_0 a^*$, and the chemical potential, $\zeta'(B)$, being measured from the Fermi energy, that is, $\zeta(B)=\varepsilon_F+\zeta'(B)$ (see Fig. 2). At $T=0$ K this oscillatory pattern has the sawtooth form shown in Fig. 2, which is the spin-split version of the purely orbital function first derived by Peierls.[11] Note that the intersection points between this pattern and the average M base line are equidistant. This symmetry property characterizes also the corresponding pattern obtained in the grand canonical ensemble (see Fig. 2). The two patterns differ by the sign of the slope in the smooth sectors. Temperature smearing makes the difference less transparent. In fact, the quantum oscillations observed in $\kappa-(ET)_2I_3$ do not show a sawtooth shape at $T=0.4$ K.

Considering the magnitude of the key parameter Q in these experiments (i.e., when $23\geqslant Q\geqslant 7$, corresponding to the magnetic fields and temperatures used, see Fig. 1), it

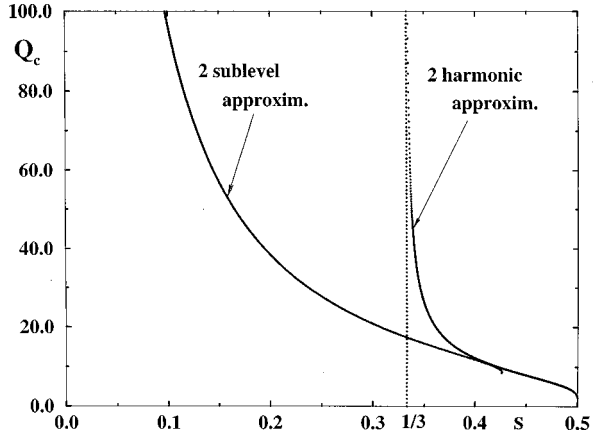

FIG. 3. The critical value Q_c for the appearance of the spin-splitting oscillation as a function of s, as obtained from the two-harmonics approximation (3) and from the level approximation (6). For $Q>Q_c(s)$ the slope of $M(B)$ at $B=B_{\bar{m}}$ is negative, and changes sign for $Q=Q_c$. Note the breakdown of the two harmonics approximation at $s=\frac{1}{3}$.

seems reasonable to describe the data in terms of the first two harmonics of the Shoenberg formula,[7] namely,

$$M^{(\mathrm{os})}(b)/M_0 = -a_1 \sin\, kb + a_2 \sin 2kb, \tag{3}$$

where $a_1 = 4\pi\, \cos(\pi s)/[Q \sinh(2\pi^2/Q)]$ and $a_2 = 4\pi\, \cos(2\pi s)/[Q \sinh(4\pi^2/Q)]$.

The parameters b and k, appearing in Eq. (3), are defined as follows: We first define a set of magnetic fields $B_{\bar{n}}$ and $B_{\bar{m}}$ at which the chemical potential coincides with its zero field value (i.e., with ε_F) and is located midway between two adjacent spin-polarized levels (see Fig. 2). Now $b = B - B_{\bar{n}}$ is defined for any B within the corresponding quasiperiod and $k \equiv \pi/b_{\bar{n}}$, where $b_{\bar{n}} \equiv B_{\bar{n}} - B_{\bar{m}}$ is half the quasiperiod.

Figure 1(b) shows the calculated magnetization oscillations [Eq. (3)]. Here the mass ratio at $\Theta = 0°$ was taken to be $\eta_{c0} = m_{c0}/m_e = 3.9$ and s_0 was used as an adjustable parameter; we find very good agreement with the experimental data for $G_0 = 4.39$, i.e., for $s_0 = 0.39$. The extrema of these oscillations can be readily derived from Eq. (3); their positions $b_{m,\pm} \equiv B_{\max,\pm} - B_{\bar{n}}$ are given by $\cos kb_{m,\pm} = (1/8a_2)(a_1 \pm \sqrt{a_1^2 + 32a_2^2})$ where the minus sign corresponds to the main component and the plus to the spin-splitting one. The latter disappears when the absolute value of the right-hand side of the above equation becomes larger than 1, that is, when Q becomes smaller than the critical value $Q_c = 2\pi^2/\ln[y + \sqrt{(y^2-1)}]$, with $y \equiv \cos 2\pi s/\cos \pi s$. Thus, only for values of s between 0.5 and 1/3, Q_c is well defined (see Fig. 3). The divergence of Q_c at $s = 1/3$ reflects the breakdown of the harmonic approximation for the spin-splitting component. To describe this component in the range $0 \leqslant s \leqslant 1/3$ it is necessary to invoke the opposite approximation based on the level representation.

So, let us derive now expressions for the chemical potential and magnetization oscillations in the region $Q > 2\pi^2$ where a small number of levels is sufficient to describe the oscillations in the entire range of the spin-splitting parameter $0 < s \leqslant 0.5$. Following the method developed in Ref. 3 we can write an equation for the chemical potential in the case when spin splitting is taken into account. In the limit $\hbar\omega_c(B) \ll \varepsilon_F$, for magnetic fields inside any quasiperiod, $B_{\bar{n}+1} \leqslant B \leqslant B_{\bar{n}}$ (see Fig. 2):

$$-\frac{b}{2b_{\bar{n}}} = \frac{F}{B} - \frac{F}{B_{\bar{n}}} = \frac{\frac{1}{2}}{1+e^{x}} + \frac{\frac{1}{2}}{1+e^{x+sQ}} + \sum_{k=1}^{\bar{n}} \left(\frac{\frac{1}{2}}{1+e^{kQ+x}} - \frac{\frac{1}{2}}{1+e^{kQ-x}} \right) + \sum_{k=1}^{\bar{m}} \left(\frac{\frac{1}{2}}{1+e^{kQ+x+sQ}} - \frac{\frac{1}{2}}{1+e^{kQ-(x+sQ)}} \right), \tag{4}$$

where $x(B,T) \equiv [\varepsilon_{\bar{n},-1}(B) - \zeta(B)]\beta$, $F = n_c/A = \varepsilon_F/\mu_c$ is the fundamental frequency of oscillations. Note that inside the quasiperiod $B \sim B_{\bar{n}} \ll F$, $b_{\bar{n}} = B_{\bar{m}} B_{\bar{n}}/2F \ll B_{\bar{n}}$. Note also that the variable $x(B,T)$ may be represented by $x(B,T) = (Q/2)[1 - s + b/b_{\bar{n}} - (2/\hbar\omega_c)\zeta'(b)]$. Solving Eq. (4) for chemical potential (neglecting the sums) and using Eq. (2) we get for the amplitudes (maximal values) of the main (Mn) and spin-splitting (ss) components:

$$\frac{M_{\mathrm{Mn}}^{(\mathrm{os})}}{M_0} = 1 - s - \frac{2}{Q} - \frac{2}{Q} \ln\left[\frac{Q}{2} (1 + e^{-sQ}) - 2 \right], \tag{5}$$

$$\frac{M_{\mathrm{ss}}^{(\mathrm{os})}}{M_0} = s - \frac{2}{Q} - \frac{2}{Q} \ln\left[\frac{Q}{2} (1 + e^{-sQ}) - 2 \right]. \tag{6}$$

Note that for $s = 0.5$ the amplitudes of the main and the spin-splitting components coincide. This is a general property, which reflects the equivalence of the energy levels corresponding to the spin and the orbital quantum numbers in this case. According to Eq. (6) the spin-splitting extrema exist as long as Q is larger than the critical value Q_c, satisfying $sQ_c \approx 2 + 2\ln(Q_c/2 - 2)$, so that the parameter determining the appearance of these extrema is sQ rather than Q, which controls the damping of the main extrema. For $s = 1/3$ this equation yields $Q_c \approx 18 \approx 2\pi^2$, implying that in the range $0 \leqslant s \leqslant 1/3$ the temperatures and magnetic fields at which the spin-splitting extrema can be observable correspond to values of Q well within the range of validity of the level approximation (see Fig. 3).

An important parameter that can be varied in the experiment is the tilt angle Θ and, consequently, the cyclotron mass, which influences both the spin-splitting parameter s and the damping parameter Q. The dependence of the amplitudes (5) and (6) on Θ is therefore through $s = |I([G_0/\cos\Theta]) - G_0/\cos\Theta|$, $G_0 \equiv (g/2)\eta_{c0}$, and $Q = \hbar\omega_{c0} \cos\Theta/k_B T$ (η_{c0} is the mass ratio m_c/m_e and ω_{c0} is the cyclotron frequency for $\Theta = 0°$). Figure 4 shows our results for main and spin-split amplitudes. It is seen that near $\Theta = 0°$ there is a broad region where the spin-splitting component is comparable to the main component. At maximal spin splitting ($s = 0.5$) the two components are identical. At angles corresponding to $s = 0$ the spin-splitting component is absent and the main component reaches its maximal value. Comparison of these calculations with the experimental data[2] for $\Theta = 0°$ (SdH), $\Theta = 9°$ and $\Theta = 16°$ (dHvA) shows very good agreement (see also Fig. 1). It should be noted that our approach to calculation of the angular dependence is based

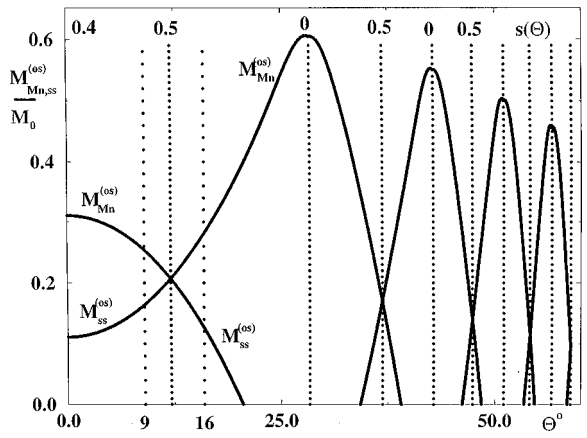

FIG. 4. Angular dependence of the magnetization oscillations amplitudes. Upper curve—the main amplitude, with maxima at $s(\Theta)=0$. Lower curve—the spin-splitting amplitude, having maxima at $s(\Theta)=0.5$, where it coincides with the minima of the main amplitude. At $\Theta=9°$ and $\Theta=16°$ the calculated amplitudes are in good agreement with the experimental results from $\kappa-(ET)_2I_3$ (Ref. 2). The parameters used at $\Theta=0$ are $s_0=0.4$, $G_0=4.4$ ($g=2.2$, $\eta_{c0}=4$), $Q_0=22.2$ ($B_{\bar{n}}=26.5$ T, $T=0.4$ K).

on the level approximation and gives results in all range of the spin-splitting parameter, $0<s<0.5$. As can be seen from Fig. 3, the conventional, ''harmonic ratio'' method[8,12] can be applied only at $1/3<s<0.5$. Furthermore, we obtain ''windows'' where the spin-split component does not even exist, while the two-harmonic approximation (3) fails to describe the spin-splitting effect.

In summary, a simple analytical scheme, based on the independent quasiparticles approximation, is presented to account for our dHvA and SdH measurements on the extremely 2D metal $\kappa-(ET)_2I_3$. In the high-temperature–low-field regimes corresponding to $Q=<2\pi^2$, the two-harmonics approximation of the LKS theory[7,8] properly describes the shape and amplitudes of the oscillations only at relatively large values of the spin-splitting parameter, $0.35<s<0.5$. Fortunately, the experimental data fall within this region and comparison with theory is quantitative. We have also considered theoretically values of the spin-splitting parameter significantly smaller than that found in $\kappa-(ET)_2I_3$. For these values of s the spin-splitting oscillation is observable only in the intermediate and high-fields–low-temperatures regimes, where $Q>2\pi^2$ and the LKS theory fails to describe the oscillations quantitatively via a few number of harmonics. In this region the opposite, few levels approximation gives correct results.

It should be noted that, in contrast to the nearly symmetric oscillations observed in $\kappa-(ET)_2I_3$, the oscillations observed in the same range of Q in a different crystalline phase of the same compound, that is, $\Theta-(ET)_2I_3$, have the nearly ideal sawtooth form characteristic of a 2D electron system.[13] The difference in the shape of oscillations can be ascribed to the different barriers separating the closed and the open Fermi-surface orbits in these phases. Such a barrier is likely to be very small in the κ structure[12,14] in comparison to the Θ structure[12,13] due to the presence of center of symmetry in the former. The absence of spin-splitting structure in $\Theta-(ET)_2I_3$,[13] indicates a small spin-splitting parameter for that phase.

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