



1.0



1.1



1.25

4.5
5.0
5.6
6.3
7.1
8.0
9.0
10.0



2.8



3.2



3.6



4.0



2.5



2.2



2.0



1.8



1.4



1.6

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FERMIOLOGY AND De HAAS-van ALPHEN EFFECT OF β -(ET)₂IBr₂⁺

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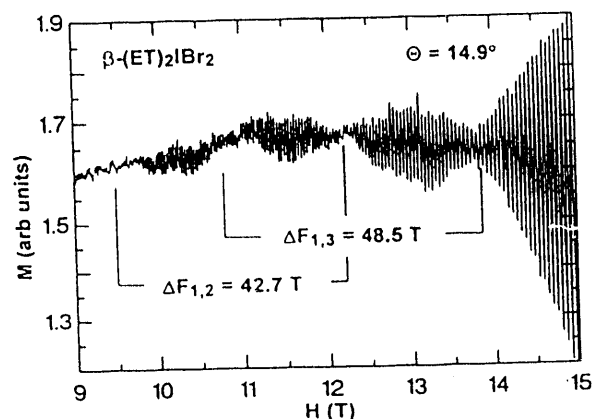
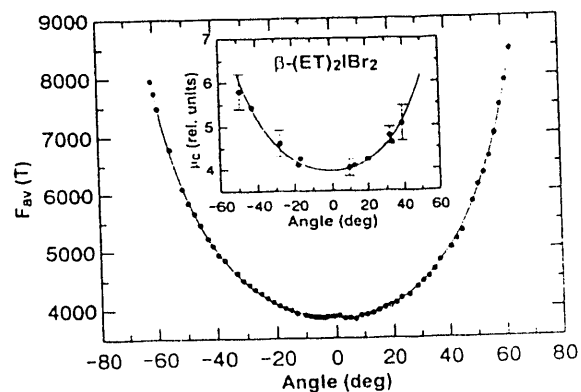
FERMIOLOGY AND DE HAAS - VAN ALPHEN EFFECT OF β -(ET)₂IBr₂J. Wosnitzer,^a G. W. Crabtree,^b K. D. Carlson,^b H. H. Wang,^b and J. M. Williams^b^aPhysikalisches Institut, Universität Karlsruhe, Engesserstr. 7, 7500 Karlsruhe, Germany^bMaterials Science and Chemistry Divisions, Argonne National Laboratory, Argonne, Illinois 60439*

The Fermi surface of the organic superconductor β -(ET)₂IBr₂ investigated by measurements of the de Haas - van Alphen (dHvA) effect has been found to have the typical two-dimensional cylindrical form. A small amount of corrugation could be quantitatively determined by the distinctive angular dependence of beating nodes. The existence of up to four almost identical frequencies in the dHvA signal may be explained by magnetic interaction effects within the samples. Due to the $1/\cos(\Theta)$ -behavior of the effective mass spin-splitting zeros could be detected.

Organic superconductors of the type (ET)₂X, where ET stands for bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF) and X for a monovalent anion, are characterized by their extremely two-dimensional (2D) character. This has been verified for a variety of ET compounds by direct observation of a cylindrical Fermi surface (FS) via de Haas - van Alphen (dHvA) and Shubnikov - de Haas (SdH) experiments [1,2]. In β -(ET)₂IBr₂, however, from previous SdH measurements considerable confusion concerning the exact topology of the FS exist [3,4] so that a clarification was highly desirable.

Altogether six different crystals of β -(ET)₂IBr₂ grown by the standard electrocrystallization technique were investigated. The magnetization M showed for almost all investigated angles and in all samples a beating behavior of the dHvA signal. Fig. 1 shows M between 9 and 15 T for $\Theta = 14.9^\circ$, where Θ denotes the angle between the field and the normal to the highly conducting (a,b)-plane. At this angle four nodes of the dHvA signal are visible, two with nearly zero and two with finite amplitude. This distinctive behavior can be explained with the existence of two pairs of frequencies, F_1 to F_4 , with an average frequency $F_{av} = (3976 \pm 20)$ T and with differences $\Delta F_{1,2} (= F_2 - F_1) = \Delta F_{3,4} = (42.7 \pm 0.3)$ T and $\Delta F_{1,3} = \Delta F_{2,4} = (48.5 \pm 0.3)$ T. Systematic investigation of the angular dependence of the dHvA signal showed for F_{av} the behavior expected for a cylindrical FS, namely $F_{av} = F_0/\cos(\Theta)$ with $F_0 = (3842 \pm 10)$ T shown in Fig. 2. The corresponding area of the FS $A_{av} = (2\pi e/\hbar) \cdot F_{av} = 3.67 \cdot 10^{15}$ cm⁻², i. e., $\approx 53\%$ of the first Brillouin zone, is consistent with band structure calculations [5] and in excellent agreement with the SdH value of Ref. [4]. The ori-

gin of the discrepancy to the result of Ref. [3], where approximately half the value stated above and an unsystematic angular change of the SdH frequency was reported, is not clear.

Fig. 1. dHvA signal of β -(ET)₂IBr₂ at $\Theta = 14.9^\circ$.Fig. 2. Angular dependence of the average dHvA frequency and the cyclotron effective mass (inset). The solid lines are $1/\cos(\Theta)$ fits.

The angular change of the frequency differences $\Delta F_{1,2}$ and $\Delta F_{1,3}$, on the other hand, shows quite unusual behavior [2,6]. The nonmonotonic angular dependence of $\Delta F_{1,3}$ is consistently describable by the asymmetric energy dispersion curve [7]

$$\varepsilon_k = \frac{\hbar}{2m_b}(k_x^2 + k_y^2) - 2t \cdot \cos(c'k_z + u_x k_x + u_y k_y). \quad (1)$$

The interlayer transfer integral, t , for hopping along a vector $\mathbf{h} = (u_x, u_y, c)$ is obtained to be $t/\varepsilon_F \approx 1/300$ with the Fermi energy ε_F [2,6].

For the frequency difference $\Delta F_{1,2}$ we found a symmetric angular dependence described by $\Delta F_{1,2} = 24.6\text{T} + 17.75\text{T}/\cos(\Theta)$. The effective masses and the scattering times of the orbits of F_1 and F_2 , respectively F_3 and F_4 are identical seen by the vanishing dHvA amplitude at the corresponding nodes (Fig. 1). This suggests magnetic interaction as a possible mechanism for the observed behavior. An antiferromagnetic exchange with a staggered magnetization of the form $M = \pm M_0 \pm M_1 \cdot H \cdot \cos(\Theta)$ can qualitatively explain the angular dependence of $\Delta F_{1,2}$.

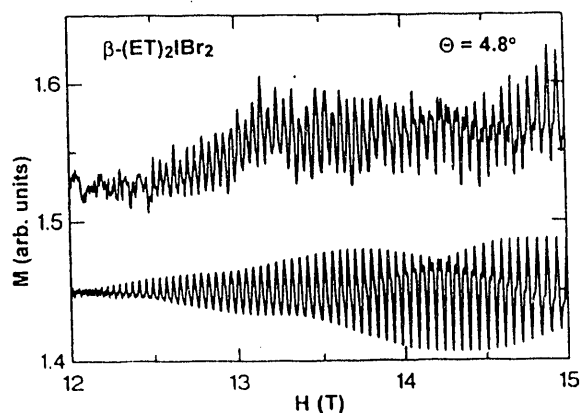


Fig. 3. dHvA signal at $\Theta = 4.8^\circ$. Upper trace as measured, lower trace calculated (see text).

The angular change of the cyclotron effective mass, m_c , (see inset of Fig. 2) can be described by $m_c = m_{c0}/\cos(\Theta)$ with $m_{c0} = (4.0 \pm 0.1)m_e$ understandable by the 2D FS. This $1/\cos(\Theta)$ -dependence is reflected by a peculiar change of the dHvA amplitude. Due to the spin-splitting factor [8], $\cos(\pi n g m_b / 2m_e)$, at those angles where the g-factor times the bare electron mass without electron-phonon interaction $g m_b = 2(n-0.5)m_e$ (with $n = 1, 2, \dots$)

the amplitude of the fundamental frequency ($r = 1$) becomes zero, whereas the amplitude of the second harmonic ($r = 2$) becomes maximal. This condition seems to be fulfilled close to $\Theta = \pm 5^\circ$ where small but still visible fundamental dHvA oscillations are observed shown for $\Theta = 4.8^\circ$ in Fig. 3. Between 14 and 15 T in the measured signal (upper trace in Fig. 3) a clear double peak structure with relatively to each other shifting peaks is seen. Obviously, for one set of frequencies (e. g. for $F_3 = 3850.3$ T and $F_4 = 3892.7$ T) only the second harmonics are visible ($g m_b = 7m_e$) and the amplitudes of the other frequencies ($F_1 = 3798.8$ T and $F_2 = 3841.2$ T) are close to zero but the fundamentals are still appreciable ($g m_b$ close but smaller $7m_e$). A calculation of the dHvA signal using the Lifshitz-Kosevich formula [8] with suitable parameters ($T_D = 1$ K, $T = 0.5$ K, $g = 2$) is plotted as the lower trace in Fig. 3. A very good agreement between calculation and measurement is obtained. However, the amplitude of the second harmonics of F_3 and F_4 are somewhat larger than expected. This might be related to the pronounced 2D character of the FS, thereby increasing the harmonic content of the dHvA signal observed also at other angles in larger harmonics than by the simple 3D theory expected.

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