

Microwave magnetoabsorption by free charge carriers in a quasi-two-dimensional organic metal

S. V. Demishev, A. V. Semeno, N. E. Sluchanko, N. A. Samarin,
I. B. Voskoboïnikov, and V. V. Glushkov

Institute of General Physics, Russian Academy of Sciences, 117942 Moscow, Russia

A. E. Kovalev

*Institute of Solid-State Physics, Russian Academy of Sciences, 142432 Chernogolovka,
Moscow Region, Russia*

N. D. Kushch

*Institute of Chemical Physics, Russian Academy of Sciences, 142432 Chernogolovka,
Moscow Region, Russia*

(Submitted 4 July 1995)

Pis'ma Zh. Éksp. Teor. Fiz. **62**, No. 3, 215–219 (10 August 1995)

The structure of magnetoabsorption in the quasi-two-dimensional organic metal $(\text{ET})_2\text{TIHg}(\text{SCN})_4$ at millimeter wavelengths and low temperature was investigated and the contribution from free 2D carriers was separated. It was shown that the cyclotron resonance of the 2D carriers exhibits anomalous behavior: In addition to a resonance with $m = 2.8m_0 = \text{const}$, wide lines ($\omega_c\tau \sim 1$) with $m \sim (1 - 1.4)m_0$ and $m \sim 0.5m_0$, for which the effective mass is frequency-dependent $m = m(\omega)$, which could be due to a strong Fermi-liquid interaction, are observed. © 1995 American Institute of Physics.

1. A large number of theoretical and experimental studies of the problem of cyclotron resonance (CR) in low-dimension organic metals has appeared in the last few years.^{1–6} The first reports of the observation of cyclotron resonance were published in Ref. 2 for the case $\alpha\text{-(ET)}_2\text{KHg}(\text{SCN})_4$ (here ET=bis(ethylenedithio)-tetrathiafulvalene). For electromagnetic radiation frequencies $\nu = \omega/2\pi = 316 - 698$ GHz, two narrow lines, whose resonance frequency depend linearly on the magnetic field H , were observed. The position of these features corresponds to the effective masses $m_1 \sim m_0$ and $m_2 \sim 0.4m_0$, where m_0 is the free-electron mass.² Using the standard formula for the complex conductivity $\sigma(\omega, H)$ of an electron gas in a magnetic field,⁷ the following estimates can be obtained from the linewidths found in Ref. 2: $(\omega_c\tau)_1 \sim 15$ and $(\omega_c\tau)_2 \sim 7$ (here ω_c is the cyclotron frequency, and τ is the relaxation time).

Similar results were later obtained for other two-dimensional organic metals.^{3–5} For example, a resonance with $m \sim m_0$ and $\omega_c\tau \sim 5$ in the case $\alpha\text{-(ET)}_2\text{NH}_4\text{Hg}(\text{SCN})_4$ for $\nu = 45 - 65$ GHz was observed.³ However, the cyclotron-resonance interpretation of the features of the resonance magnetoabsorption^{2–5} in many cases does not agree with experiment. First, in $\alpha\text{-(ET)}_2\text{KHg}(\text{SCN})_4$ the position of the resonances with $m \sim m_0$ and $m \sim 0.4m_0$ does not depend on the orientation of the magnetic field; in addition, these features can be excited even in the case where the magnetic field is parallel to the conducting plane and there is no 2D quantization of the orbital motion.⁶ Second, the

cyclotron mass determined from the quantum oscillations of the magnetoresistance in these systems lies in the range $m_c \sim (3.5-1.4)m_0$, while cyclotron resonance gives $m \sim m_0$.²⁻⁵ Moreover, the only absorption feature corresponding to cyclotron resonance on 2D carriers in α -(ET)₂KHg(SCN)₄ corresponds, according to Ref. 6, to the effective mass $m \sim 2.8m_0$ and $\omega_c \tau \sim 2$, whereas the Shubnikov-de Haas effect gives $m_c \sim 1.5m_0$ (Ref. 8). Third, for $\nu \sim 100$ GHz the parameter $\omega_c \tau$ estimated from the Dingle temperature⁹ is $\omega_c \tau \sim 1$; i.e., the lines interpreted as cyclotron resonance are narrower than the expected width of the cyclotron resonance on free carriers.

The question concerning the observation of cyclotron resonance and the structure of resonance magnetoabsorption attributable to it in organic metals thus remains open. In the present study we investigate this problem experimentally.

2. The measurements were performed on single crystals of α -(ET)₂TlHg(SCN)₄, which is an isostructural analog of α -(ET)₂KHg(SCN)₄ (as far as we know, microwave magnetoabsorption in this compound has never been investigated). The response of a $2 \times 2 \times 0.1$ mm single crystal was recorded with a BWT spectrometer; the spectrometer is described in Ref. 6. The experiments were performed at $T = 1.8$ K in fields $H \leq 70$ kOe in the frequency range $\nu = 50-120$ GHz. The magnetic field and the propagation vector of the electromagnetic wave were directed along the normal direction to the conducting plane of the 2D carriers.

It can be shown¹⁰ that in the arrangement chosen for the measurements the microwave power P absorbed in the measuring cell can be represented as a sum of two contributions

$$P = P_1 + P_2, \quad (1)$$

$$P_1 = P_0(1 - R(Z_c/Z_0)), \quad (2)$$

$$P_2 = P_0 f(Z_c/Z_0) \text{Re}(Z_c/Z_\sigma), \quad (3)$$

where P_0 is the incident power, P_1 is the power absorbed in the cell with no sample, and R is the reflection coefficient. The function P_2 describes the additional absorption associated with the organic-metal sample, and $f(Z_c/Z_0)$ describes the change produced in the absorption process when the sample is inserted into the cell. In Eqs. (1)–(3) Z_c , Z_0 , and Z_σ are the effective impedances of the waveguide, the empty cell, and the sample, respectively.

The typical experimental data on the magnetic-field-dependence of P with $\nu = \text{const}$ are shown in Fig. 1. When the sample is inserted into the measuring cell, an additional wide feature appears in the magnetoabsorption (compare curves 1 and 2). The field-dependence $P_1(H)$ arises as a result of the change introduced in the impedance Z_0 by the magnetoresistance of the bolometer which is used to record the absorbed power. Since the functions $R(Z_c/Z_0)$ and $f(Z_c/Z_0)$ are continuous, the quantity P_1 depends monotonically and virtually linearly on H (Fig. 1, curve 1), and the nonmonotonic character of the total power $P(H)$ (Fig. 1, curve 2) is determined by the particular features of the absorption $P_2(H)$, which are associated with the organic metal (Fig. 1, curve 3). Estimates show that the structure of $P_2(H)$ cannot be explained simply by the magnetoresistance of

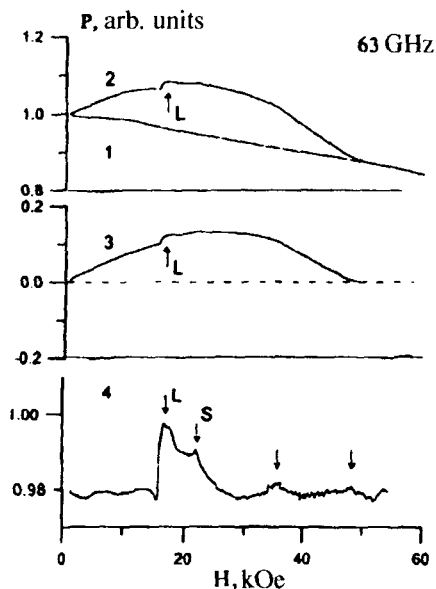


FIG. 1. Structure of magnetoabsorption in α -(ET)₂TIHg(SCN)₄ at $\nu=63$ GHz. Curve 1—Empty cell; 2—cell + sample; 3—absorption on free 2D carriers; 4—different magnetic resonances (marked by arrows); S—ESR, L—antiferromagnetic resonance.

the sample at $\nu=0$, since in fields $H \leq 70$ kOe α -(ET)₂TIHg(SCN)₄ is characterized by a monotonic magnetoconductance which has no features in the magnetic field.¹¹

Narrower lines with a smaller amplitude (arrows in Fig. 1) are observed against the background formed by the wide features in $P_2(H)$. To separate these features we approximated $P_2(H)$ by a smoothed-out curve $\tilde{P}_2(H)$ and then calculated the ratio $P_2(H)/\tilde{P}_2(H)$ (Fig. 1, curve 4). Obviously, the experimental apparatus made it possible to record the resonance features at a level of 0.3-0.4% of $P_2(H)$.

Proceeding from an analysis of the linewidth (Sec. 1) and the data of Ref. 6, the structure of the magnetoconductance in α -(ET)₂TIHg(SCN)₄ can be interpreted as follows. The wide features in $P(H)$ with an amplitude of $\sim 10\%$ are due to absorption by 2D carriers and the narrower lines with amplitude $\leq 2\%$ are due to magnetic resonances (Fig. 1).

The structure of magnetic resonances in α -(ET)₂TIHg(SCN)₄ will be described in detail in a separate paper. The data obtained by us show that this structure is in many ways reminiscent of the structure of the resonances in α -(ET)₂KHg(SCN)₄ and is apparently formed as a result of the superposition of ESR and AFMR (the possibility of antiferromagnetic ordering at low temperatures is indicated by the results obtained in Ref. 11). In what follows we shall consider the wide features in $P(H)$, since they are related to the cyclotron resonance problem.

3. We shall now analyze the evolution of the shape of the curves $P(H)$ as a function

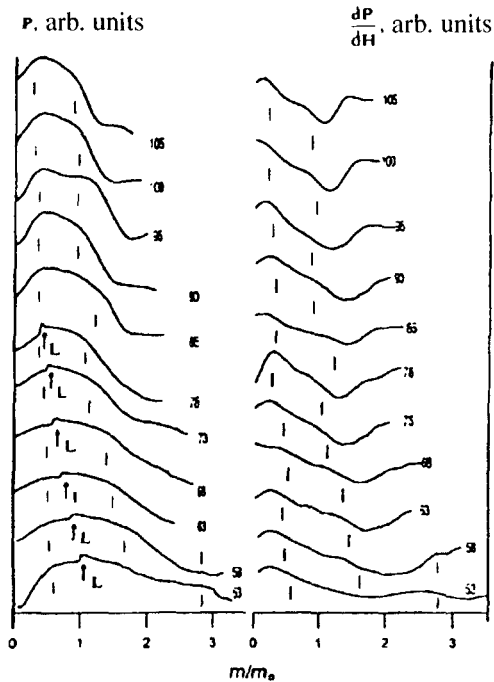


FIG. 2. $P(H)$ and dP/dH at different frequencies. The vertical bars mark the positions of cyclotron-resonance-like features; the arrows mark the magnetic resonances. The numbers on the curves correspond to the frequency in GHz.

of frequency (Fig. 2; to compare the data, the magnetic-field scale is converted into an effective-mass scale according to the formula $m = eH/2\pi\nu c$). We see that a feature in $P(H)$ can generally be represented as a superposition of two wide maxima which become sharper in the region $\nu \sim 100$ GHz (Fig. 2).

Since $P_1(H)$ is quasilinear, to determine the position of the features we smoothed the curves $P(H)$ in order to eliminate the distortions introduced by narrow magnetic resonances. We then calculated the derivative dP/dH . The absorption maxima were found from the inflection point on the curves dP/dH (Fig. 2; the vertical bars mark the positions of the features).

Two features are observed in the entire experimental frequency range: one near $m \sim 0.5m_0$ and the other near $m \sim (1 - 1.5)m_0$. The position of these features depends on the frequency. In addition, a resonance with $m \sim 2.8m_0$ was observed near the lower limit of the frequency range. This resonance is identical to the one registered in Ref. 6. Just as in the case of $\alpha\text{-(ET)}_2\text{KHg(SCN)}_4$, this resonance is seen as a wide maximum in $P(H)$ for $\nu \leq 53$ GHz and it becomes rapidly narrower at $\nu = 58$ GHz (Fig. 2). At high frequencies, this feature is shifted outside the admissible range of magnetic fields; this made it impossible for us to follow its further evolution. The position of the different features in $P(H)$, which are converted into an effective mass, for different frequencies is summarized in Fig. 3.

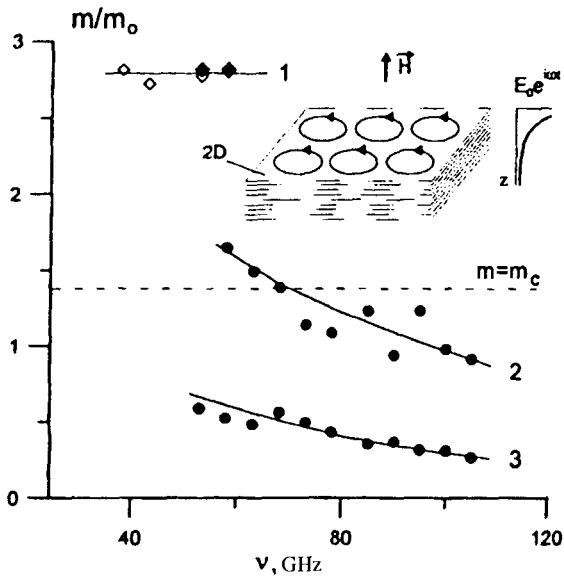


FIG. 3. Effective masses in $\alpha\text{-(ET)}_2\text{TIHg(SCN)}_4$. Curve 1—Resonance with $m = \text{const}$; 2, 3—frequency-dependent branches corresponding to the main absorption feature. For curve 1 the dark dots are for $\alpha\text{-(ET)}_2\text{TIHg(SCN)}_4$ and the light dots are for $\alpha\text{-(ET)}_2\text{KHg(SCN)}_4$ from Ref. 6. Inset: Geometry of the experiment on cyclotron resonance in an organic metal.

4. The physical picture of the cyclotron resonance in an organic metal must be substantially different from the case of a normal metal. In a normal metal at cyclotron resonance the carriers move in a substantially nonuniform alternating electric field E , while in an organic metal, because of the two-dimensional character of the carrier motion, $E = \text{const}$ for each conducting plane, although the field decreases with the distance from the surface (the geometry of the experiment on cyclotron resonance in an organic metal is shown in the inset in Fig. 3).

In the geometry considered by us the impedance of the sample has the form $Z_\sigma \sim (\sigma(\omega, H)\delta)^{-1}$, and therefore $P_2 \sim \text{Re}\sigma(\omega, H)$. An estimate of the thickness δ of the skin layer gives the value 20–100 μm , which is much greater than in a normal metal (the data of Ref. 12 were used to obtain the estimates).

As a result, cyclotron resonance in an organic metal is found to be closer to a semiconductor, in which the absorption coefficient is also proportional to $\text{Re}\sigma(\omega, H)$. The structure of the function $P(H)$ also agrees well with this assumption (Fig. 1). It is well known that for $\omega_c = (eH/mc) \gg \omega$ $\text{Re}\sigma(\omega, H) \rightarrow 0$ (Ref. 7) and $P_2 \rightarrow 0$; i.e., excess absorption arises in an interval of magnetic fields which is bounded from above, as is observed experimentally (Fig. 1).

In summary, the features in $P(H)$ are apparently determined by the cyclotron resonance of free charge carriers (Fig. 3). It is interesting that besides the resonance with $m \sim 2.8m_0 = \text{const}$ (Fig. 3, curve 1), two additional branches with $m(\omega) \neq \text{const}$ are observed (Fig. 3, curves 2 and 3). These branches determine the main absorption feature

(Figs. 1 and 2). We underscore the fact that cyclotron resonance with a constant effective mass $m_c = 1.4m_0 = \text{const}$, which follows from quantum oscillation effects in $\alpha\text{-(ET)}_2\text{TiHg(SCN)}_4$,¹¹ is not observed in the experimental frequency range, and in the region $m \sim 1.4m_0$ the experiment gives $m = m(\omega)$ (Fig. 3).

The possibility for the occurrence of a cyclotron resonance with $m = m(\omega)$ in organic metals has, to the best of our knowledge, not been previously studied. This anomalous behavior is probably due to strong Fermi-liquid effects, which cause the effective mass to depend on the frequency.¹³ As a result, cyclotron resonance in an organic metal will be much more complex from the theoretical and experimental standpoints than in the cases of a standard metal or semiconductor. This hypothesis can be checked by studying in detail the structure of $P(H)$ in an oblique magnetic field. This is a problem for future investigations.

We wish to thank B. A. Volkov, L. A. Fal'kovskii, S. V. Sharov, A. A. Volkov, and J. Singleton, W. Heiss, and S. Blandell for helpful discussions. We also thank M. V. Kartsovnik for reading the manuscript and making valuable remarks. This work was performed as part of the International Projects INTAS 93-2400 and INTAS 94-1788. Financial support for a number of aspects of this work was provided by the Royal Society (Great Britain) and the Russian National Program "Fullerenes and Atomic Clusters."

¹L. P. Gor'kov and A. G. Lebed', *Phys. Rev. Lett.* **71**, 3874 (1993).

²J. Singleton, F. L. Pratt, M. Doporto *et al.*, *Phys. Rev. Lett.* **68**, 2500 (1992).

³S. Hill, A. Wittlin, J. van Bentum *et al.*, *Synth. Met.* **70**, 821 (1995).

⁴S. Hill, J. Singleton, F. L. Pratt *et al.*, *Synth. Met.* **55-57**, 2566 (1993).

⁵J. Singleton, F. L. Pratt, M. Doporto *et al.*, *Physica B* **184**, 470 (1993).

⁶S. V. Demishev, N. E. Sluchanko, A. V. Semeno, and N. A. Samarin, *JETP Lett.* **61**, 313 (1995).

⁷J. Hajdu and G. Landwehr in *Strong and Ultrastrong Magnetic Fields and Their Applications*, edited by F. Herlach, Springer-Verlag, N. Y., 1985 [Russian translation, Mir, Moscow, 1988, p. 69].

⁸F. L. Pratt, J. Singleton, M. Doporto *et al.*, *Phys. Rev. B* **45**, 13904 (1992).

⁹J. Caulfield, S. J. Blundell, M. S. L. de Croo de Jough *et al.*, *Phys. Rev. B* **51**, 8325 (1995).

¹⁰I. V. Lebedev, *Microwave Technology and Devices* [in Russian], Vysshaya shkola, Moscow, 1970, Vol. 1, p. 181.

¹¹M. V. Kartsovnik, A. E. Kovalev, and N. D. Kushch, *J. Phys. I France* **3**, 1187 (1993).

¹²T. Sasaki and N. Toyota, *Solid State Commun.* **75**, 93 (1990).

¹³T. Li, *Phys. Rev. B* **48**, 4991 (1993).

Translated by M. E. Alferieff