Magnetic resonances in the quasi-2D organic conductor $(BEDT\text{-}TTF)_2KHg(SCN)_4$

S. V. Demishev, N. E. Sluchanko, A. V. Semeno, and N. A. Samarin
Institute of General Physics, Russian Academy of Sciences, 117942 Moscow, Russia

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The structure of magnetic resonances in the quasi-2D organic metal $(BEDT\text{-}TTF)_2KHg(SCN)_4$ has been found to be a complex superposition of spin resonances. Included among these resonances are an antiferromagnetic resonance and an electron spin resonance involving conduction electrons. There is also a cyclotron resonance of 2D carriers with effective mass of $2.38m_0$, $2.8m_0$, and $3.29m_0$. These values are larger by a factor of 1.4–1.6 than the effective masses found from the Shubnikov–de Haas effect. The difference may be due to strong Fermi-liquid effects.

1. Organic metals of the family $(BEDT\text{-}TTF)_2X$, where X is a monovalent anion, and BEDT-TTF is bis(ethylenedithio)-tetrathiafulvalene, have attracted research interest for several reasons. For example, by varying the component M in the anion group $MHg(SCN)_4$, where M = K, Pb, NH_4, one can change the low-temperature properties of the sample from those of a superconducting state (M = NH_4) to those of an antiferromagnetically ordered state (M = K) within a given structural class.[1] From the standpoint of the electronic properties, these organic metals constitute a set of conducting planes in which 2D electrons and holes move. The conductivity in the direction perpendicular to these planes is negligible.

There has recently been a substantial effort to study the Fermi surfaces of organic metals[1-3] by working from oscillation effects and magnetooptic effects. A pioneering study[3] of a $(BEDT\text{-}TTF)_2KHg(SCN)_4$ sample revealed an absorption of electromagnetic radiation in the wavelength range 430–950 μm which was resonant in the magnetic field. This absorption was interpreted as a cyclotron resonance of 2D holes. It turned out that the cyclotron resonance corresponds to values of the effective mass m_{CR} which are smaller by a factor 2.4–5 than the values m_{ShdH}, found from the Shubnikov–de Haas effect. This huge discrepancy led to the conclusion that strong Fermi-liquid effects[3] and an associated renormalization of the effective mass occur in organic metals.

Since the effective mass m_{ShdH} corresponds to the case $\omega=0$, while m_{CR} corresponds to $\omega\neq0$ (ω is the frequency of the electromagnetic field), we decided to study the resonant magnetoabsorption at lower frequencies, in particular, in the millimeter wavelength range, in order to test and refine the results of Ref. 3, including the results regarding a possible remormalization of the effective mass. In this letter we briefly report our results.

2. Previous experiments[3-6] to observe resonant magnetoabsorption in organic metals were carried out in a geometry in which the magnetic field **H**, the propagation direction of the electromagnetic wave (**k**), and the normal to the conducting plane (**n**) were parallel. Since crystals of organic metals are usually small (~1 mm × 1 mm ×50–100 μm), the measurements were carried out on mosaic samples. In an experiment of this sort, of course, electron spin resonances may be excited along with a cyclotron resonance. Furthermore, uncontrollable phase shifts which arise in a mosaic sample can seriously complicate the distribution of the wave intensity along the sample which is detected by the detector and can thereby introduce ambiguities in the interpretation of the observed effects.

In an effort to avoid these difficulties, we assembled a special spectrometer. Electromagnetic radiation in the wavelength region $\lambda = 3.5-8$ mm was generated by backward-wave sources with a stability of 10^{-4} and a power level of about 10 mW. This radiation propagated along a waveguide in a cryostat with a superconducting magnet. The condition $\mathbf{H} \| \mathbf{k}$ held. The sample of organic metal was positioned directly on a bolometer. The heat-conducting lubricant Apiezon was used to improve the thermal contact. The sensitivity of the computer-controlled data acquisition system made it possible to detect the response of an individual single crystal of the organic metal. The mosaic nature of the samples thus presented no problems in our case.

To separate ESR and cyclotron-resonance signals, we compared the results found for the case $\mathbf{n} \| \mathbf{H}$ and the case in which the vectors **n** and **H** make an angle α. As an additional test for identifying cyclotron-resonance features, we carried out experiments at $T = 4.2$ and 1.8 K. The literature has no results of this sort on the magnetooptic properties of organic metals. The test samples were $(\mathrm{BEDT\text{-}TTF})_2\mathrm{KHg(SCN)}_4$ single crystals, whose physical properties have been studied in most detail.[1-3]

3. In this measurement arrangement, the radiation is absorbed almost completely by the organic-metal crystal, and the bolometer detects the change in the crystal temperature, which is proportional to the microwave power P that the crystal absorbs. Figures 1 and 2 show some typical results on $P(H)$ for various values of λ and α. There are numerous resonance features [peaks on the $P(H)$ curve] in the test sample, against the background of a smooth component, which is determined by the frequency characteristics of the resonator holding the bolometer and the test sample.

We identified the individual $\omega(H)$ modes making up the complex $P(H,\lambda)$ spectrum. For this purpose we analyzed the shape and amplitude of the resonance in addition to its position. The results are shown in Fig. 3 as a plot of $\hbar\omega = f(H)$; the notation used for the modes corresponds to Figs. 1 and 2.

The $\omega(H)$ modes form two distinct groups. The first contains the modes K, L, M, N, and A, which do not extrapolate to the origin $H=0$, $\omega=0$ (Fig. 3a). Furthermore, the derivative $d\omega/dH$ for the K and L modes is negative; i.e., we have $\omega_{K,L}(H \to 0) \neq 0$. For the modes M, N, and A, there is apparently a threshold magnetic field $H \neq 0$ above which these structural features can be observed. Interestingly, mode A, which is well defined at $T = 4.2$ K, is essentially indistinguishable at $T = 1.8$ K, while modes K, L, M, and N are clearly visible at both temperatures.

This behavior does not correspond to the case of cyclotron resonance or ESR, in which we would have $\omega(H \to 0) = 0$. We believe that this behavior may be due to the excitation of an antiferromagnetic resonance.[7] Qualitatively, the structure of the modes in

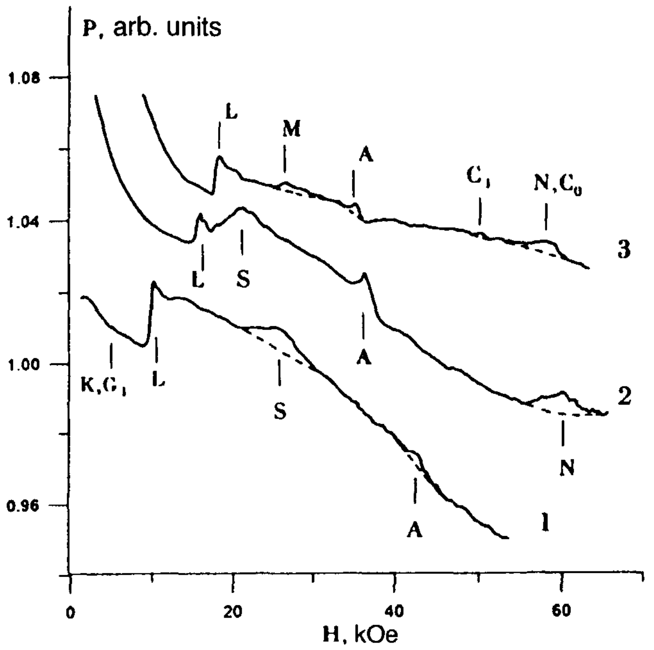

FIG. 1. Magnetic resonances of (BEDT-TTF)$_2$KHg(SCN)$_4$ at $T=4.2$ K in the geometry $\mathbf{H}\|\mathbf{n}\|\mathbf{k}$ ($\alpha=0$). Curve 1—$\lambda=3.84$ mm; 2—4.83 mm; 3—5.16 mm.

Fig. 3a corresponds to possible oscillations of an easy-axis antiferromagnet.[7] The modes L and N can be identified with oscillations of the noncollinear state $^{+}1_{\perp}$ and the collapsed state $^{+}2_{\perp}$. The modes K and M can be identified with oscillations of the antiparallel state $^{+}0_{\|}$ and the noncollinear flipped state $^{+}1_{\|}$, respectively. These suggestions agree qualitatively with the data of Ref. 1, according to which the organic metal (BEDT-TTF)$_2$KHg(SCN)$_4$ is an antiferromagnet with an easy axis in the ac conducting plane for $T<T_N\sim 10$ K. On the other hand, the simultaneous excitation of longitudinal and transverse modes in the geometry $\mathbf{H}\|\mathbf{k}\|\mathbf{n}$ suggests that the easy axis may be inclined with respect to the ac plane. Corresponding experiments with a tilted sample revealed that with $\alpha=44°$ the K, L, M, and N lines undergo essentially no shift. In the case of an antiferromagnetic resonance, we would expect such a behavior if the easy axis made an angle $\alpha\sim 22°$ with respect to the normal.

The strong temperature dependence of mode A apparently reflects a change in the magnetic structure of the sample in the interval $1.8<T<4.2$ K. We intend to carry out a detailed study of the temperature dependence of this mode and also of the angular distributions of the K, L, M, and N modes in future research.

The second group of modes satisfies the condition $\omega(H\rightarrow 0)=0$ (Fig. 3b). Modes G_0 and S coincide, within the experimental errors, with modes seen in Ref. 3. In addition to the fundamental mode G_0, we observe two more modes, G_1 and G_2, in weak magnetic fields. The frequencies of the two are approximately multiples of G_0. We found that

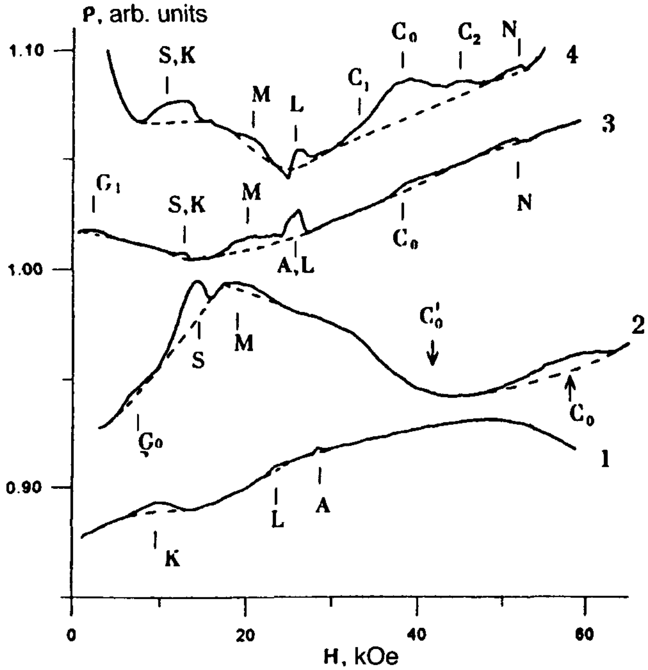

FIG. 2. Effect of the temperature and the inclination of the sample on the magnetic resonances of $(BEDT\text{-}TTF)_2KHg(SCN)_4$. Curve 1—$\lambda=6.97$ mm, $T=4.2$ K, $\alpha=0$; 2—6.97, 1.8, 44°; 3—7.86, 4.2, 0; 4—$\lambda=7.86$ mm, $T=1.8$ K, $\alpha=0$. For curve 2, the experimental position of mode C_0 is supplemented with the position (C_0') of this mode, shifted in accordance with a $\cos\alpha$ law. This shifted position corresponds to quantization in the normal component of the magnetic field (as marked by the arrows).

the positions of the features $G_{0,1,2}$ and S are independent of the inclination angle α, so they cannot be interpreted in terms of a cyclotron resonance.

A special study showed that the resonant magnetoabsorption near mode S is complex, consisting of a superposition of two closely spaced lines. The amplitude of one of these lines increases with decreasing temperature. For neither component, however, is there a dependence on the inclination angle. Furthermore, we observe excitation of two components of the mode S and the mode G_0 when the magnetic field is directed parallel to the *ac* conducting plane, and there is no 2D quantization of the orbital motion. These structural features thus cannot be associated with cyclotron resonance.

The most likely interpretation is that the modes of group G correspond to spin waves, while the S mode corresponds to an ESR involving conduction electrons. Our reasoning here is that mode S is greatly broadened (Figs. 1 and 2), as it typically would be in this case. Here the spreading of the ESR results from a strong spin–lattice coupling and a nonuniform distribution of the magnetic field in the sample because of the skin effect.[8] Working from experimental data on mode S, we calculated a g-factor: $g=2.08\pm0.07$. This figure is close to the values characteristic of organic metals.[1-6]

On the other hand, at magnetic fields $H\sim40$–60 kOe we see modes C_0, C_1, and

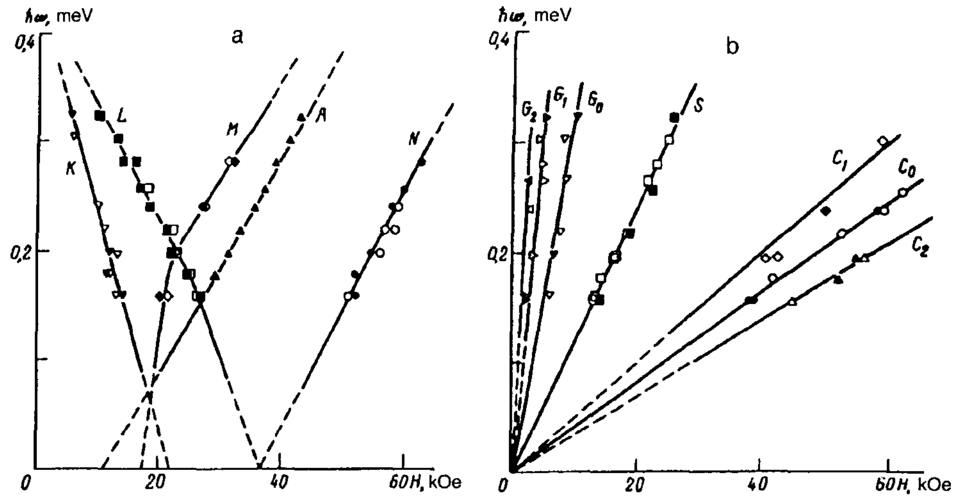

FIG. 3. Mode structure of $(\mathrm{BEDT\text{-}TTF})_2\mathrm{KHg(SCN)}_4$. ●—$T=4.2$ K; ○—$T=1.8$ K. See the text proper for an explanation.

C_2, whose amplitudes increase with decreasing temperature (compare curves 1, 2 and 3, 4 in Fig. 2). The modes of this group shift up the magnetic-field scale by a factor of $(\cos\alpha)^{-1}$ (Fig. 2). In other words, specifically the normal component of the field H is responsible for the resonance in this case. This component determines the quantization of the orbital motion of the 2D carriers. The results show that modes C_1, C_0, and C_2 are probably due to a cyclotron resonance of current carriers with effective masses of $(2.38 \pm 0.09)m_0$, $(2.80 \pm 0.04)m_0$, and $(3.29 \pm 0.08)m_0$, respectively.

From data on the Shubnikov–de Haas effect in $(\mathrm{BEDT\text{-}TTF})_2\mathrm{KHg(SCN)}_4$ we know that effective masses of $1.5m_0$, $2.0m_0$, and $2.4m_0$ correspond to different parts of the 2D Fermi surface.[2,3] It thus appears that a mass renormalization does indeed occur in an organic metal, but the effective mass in $(\mathrm{BEDT\text{-}TTF})_2\mathrm{KHg(SCN)}_4$ increases with the frequency, in contrast with the data of Refs. 2–6. All three masses correspond to approximately the same renormalization factor, $m_{\mathrm{CR}}(\omega)/m_{\mathrm{ShdH}}(0) = 1.4$–$1.6$. This behavior follows from certain Fermi-liquid theories which incorporate the effect of spin and charge fluctuations.[9]

In summary, we have determined the structure of magnetic resonances in the organic metal $(\mathrm{BEDT\text{-}TTF})_2\mathrm{KHg(SCN)}_4$. We have shown that it consists of complex superposition of spin resonances, apparently including an antiferromagnetic resonance and a cyclotron resonance involving 2D charge carriers.

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