Direct Observation of the Magnetic-Breakdown Induced Quantum Interference in the Quasi-Two-Dimensional Organic Metal κ-(BEDT-TTF)$_2$CU(NCS)$_2$

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Magneto-oscillatory behavior of the organic metal κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ has been studied at temperatures above 2 K at which the thermodynamic quantum oscillations are highly suppressed due to the temperature smearing of the Fermi level. Under this condition, we directly observe kinetic oscillations with $F_\beta - 2F_\alpha$ frequency coming from quantum interference of the electron orbits in the magnetic-breakdown regime. The extremely low effective mass of these oscillations causes a very weak temperature dependence of their amplitude, allowing up to $T \simeq 9$ K, the highest temperature reported for the observation of magnetic quantum oscillations in organic metals. [S0031-9007(96)01213-6]

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The interference of conducting electrons moving in a magnetic field is a specific quantum phenomenon which can be realized in coherent magnetic-breakdown (MB) networks. A particular case of the quantum interference (QI), namely, the QI between the electrons passing through open orbits connecting two MB junctions (the so-called Stark interferometer), was investigated in detail by Stark and co-workers [1]. A generalized theory of QI in a common case of a coherent MB network was developed by Kaganov and Slutskin [2], and predicted complicated spectra of kinetic coefficient oscillations including various linear combinations of fundamental harmonics. Recently, Machida *et al.* [3], based on a numerical analysis of linear chain MB networks, proposed that similar combination frequencies also exist in the oscillations of thermodynamic properties; however, the temperature and field dependences of their amplitudes differ from those obtained for the kinetic oscillations [1,2]. From the experimental point of view, QI remains an exotic phenomenon. It has been observed so far in its simplest manifestation (Stark interferometer) in pure Mg and some of its alloys [1] in which the interfering orbits constitute only a small part of a rather complicated Fermi surface. The main reason for the lack of experimental observations is that the MB networks in conventional metals normally include orbits too large to achieve the electron phase coherence on a sufficient part of the Fermi surface including several MB junctions.

Quasi-two-dimensional organic metals are characterized by a relatively small Brillouin zone and, as shown in numerous experiments [4], the phase coherence is often realized in the entire Fermi surface. In particular, the Fermi surface of the organic metal κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ [where BEDT-TTF denotes the bis(ethylenedithio)tetrathiafulvalene molecule] represents a textbook example of a MB linear chain network coherent within at least one whole unit cell. Indeed, both Shubnikov–de Haas (SdH) and de Haas–van Alphen oscillations in this compound show that its Fermi surface consists of a cylinder occupying 16% of the Brillouin zone and a pair of corrugated open sheets separated from the cylinder by a small energy gap at the Brillouin zone boundary (Fig. 1). Oscillations of two main frequencies, $F_\alpha = 600 \pm 5$ T and $F_\beta = 3850 \pm 50$ T, have been observed, corresponding, respectively, to the classical orbit on the cylinder (α) and the magnetic-breakdown orbit (β), which includes both the open sheets and outer arcs of the cylinders, and envelopes the area equal to 100% of the Brillouin zone cross section. Besides the fundamental frequencies, F_α and F_β, and their higher harmonics, additional smaller peaks have been detected in the SdH spectra at frequencies $F_\beta + nF_\alpha$, where $n = \pm 1, \pm 2$ [5,6]. The frequencies with $n < 0$ are

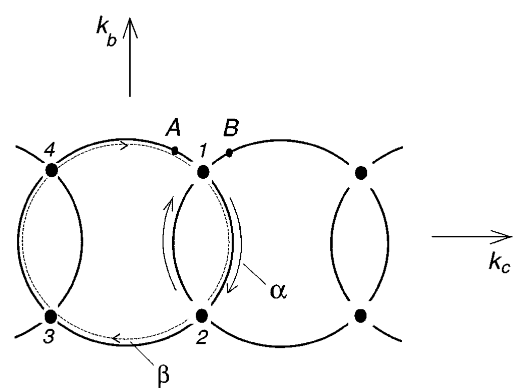

FIG. 1. Schematic view of the Fermi surface of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$. Enumerated filled circles denote the MB junctions. Dashed lines show the classical (α) and MB (β) electron orbits

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not allowed within the standard SdH effect which is caused by the density-of-states oscillations due to the finite electron motion on closed Fermi surface orbits. Caulfield *et al.* [7] suggested that these frequencies may come from the QI in the MB regime. However, the effective mass of the oscillations presented in [7] with $n = -1$, $m^*_{\beta-\alpha} = 4.7m_0$ (where m_0 is the free electron mass) does not agree with the QI theories [1,2] and was not explained satisfactorily by the authors [7]. On the other hand, Sasaki, Sato, and Toyota [6] proposed that the additional peaks arise as an artifact of the fast Fourier transformation procedure. The very low amplitudes of the oscillations with the combination frequencies, which are generally superimposed by much stronger fundamental harmonics, did not allow reliable quantitative information on them to be extracted. Therefore, their origin has not been clear so far.

In this paper, we present the results of high resolution studies of the magnetoresistance quantum oscillations in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ in magnetic fields up to 15 T, at temperatures up to 9 K. Above 2 K, the thermodynamic quantum oscillations are found to be highly suppressed, and we directly observe kinetic oscillations with the frequency of $F_\beta - 2F_\alpha$ coming from QI. The low effective mass of these oscillations causes a very weak temperature dependence of their amplitude. The mass can be tuned to the zero value by applying a moderate quasihydrostatic pressure below 10 kbar; in that case, the damping of the oscillations is determined only by the temperature dependent phase-relaxation time. This allows the $(\beta - 2\alpha)$ oscillations to be observed up to $T \simeq 9$ K, the highest temperature reported so far for the observation of magnetic quantum oscillations in organic metals.

The experiment was carried out on several single crystals of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ synthesized in either the Himeji Institute of Technology or the Institute of Chemical Physics. The low contact resistance, lower than 10 Ω, enabled us to measure the interplane (i.e., normal to the highly conducting b-c plane) resistance of the samples, which was typically ≤ 10 Ω at low temperatures, with the relative resolution of $\leq 10^{-5}$ at the current of 0.1–1 mA. The in-plane resistance was also measured and showed essentially the same behavior as that of the interplane one. However, due to low values of the in-plane resistance ($\sim$10 mΩ), it was not possible to achieve the necessary accuracy without considerable overheating by the measuring current.

Figure 2 shows the oscillatory part of the magnetoresistance in the magnetic field normal to the b-c plane at different temperatures. The slow SdH oscillations, $F_\alpha = 605$ T, corresponding to the classical α orbit, which dominate below 2 K, fade away as the temperature increases and new rapid oscillations become clearly visible. The amplitude of the rapid oscillations is very low, $\sim 10^{-4}$ of the background resistance; nevertheless, they have been found in all the studied samples. The frequency of these

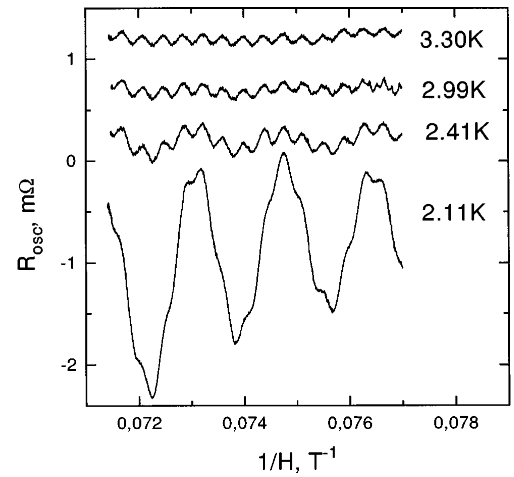

FIG. 2. Oscillatory part of the interplane resistance at different temperatures, at ambient pressure.

oscillations, 2630 T, cannot be represented by a single classical or MB orbit. On the other hand, it perfectly agrees with the difference frequency value, $F_\beta - 2F_\alpha$.

The existence of such frequency can be readily explained in terms of the generalized theory of the MB kinetic oscillations [2]. This theory predicts the oscillating part of a kinetic coefficient, in particular, electrical conductivity, to be a sum of combination harmonics,

$$\sum_{\lambda,\lambda'} \exp[i(\phi_\lambda - \phi_{\lambda'})] = \sum_{\lambda,\lambda'} \exp\left(\frac{ic\hbar S_{\lambda\lambda'}}{eH}\right), \quad (1)$$

where λ and λ' are a pair of different electron paths in the k space having common starting and ending points, ϕ_λ is the change of the quasiclassical phase of the electron wave packet at its evolution along the λ path under magnetic field H,

$$\phi_\lambda = \frac{\hbar c}{eH} \int_\lambda k_x dk_y \,, \quad (2)$$

and $S_{\lambda\lambda'}$ is the area of the corresponding single- or multiply-connected contour calculated, taking into account the number and direction of traversing the loops included in the λ and λ' paths; the summation is taken over all the possible pairs λ, λ'. For the network shown in Fig. 1, the sum (1) includes, in particular, the paths A-1-2-3-4-1-B and A-1-2-1-2-1-B, giving rise to the oscillating term $\cos[(c\hbar/eH)(S_\beta - 2S_\alpha)]$ in the real part of the conductivity.

The origin of the $(\beta - 2\alpha)$ oscillations can be clearly illustrated in a way similar to that used by Stark and Reifenberger [1], by calculating the probability $\Gamma^{(AB)}$ for an electron to pass from the point A to B in Fig. 1; $\Gamma^{(AB)}$ enters the expression for the open orbit contribution to the magnetoresistance. To do this, we first calculate the probability amplitudes, γ_λ, for different possible trajectories of the electron packet which lead from A to B, restricting ourselves, for simplicity, to those consisting of no more than four elementary arcs connecting the adjacent MB junctions. There are four such trajectories, A-1-B, A-1-2-1-B, A-1-2-1-2-1-B , and A-1-2-3-4-1-B, in the notations of Fig. 1. Denoting the probability amplitude for the transmission through the MB junction as $p = P^{1/2}$ and for the Bragg reflection as $q = i(1 - P)^{1/2}$, where $P = \exp(-H_{\mathrm{MB}}/H)$ is the MB probability and H_{MB} is the MB field [8], we express the probability amplitudes corresponding to the mentioned trajectories as q, $qp^2\exp(i\phi_\alpha)$, $q^3p^2\exp(2i\phi_\alpha)$, and $qp^4\exp(i\phi_\beta)$, respectively, where ϕ_α and ϕ_β are the changes of the quasiclassical phase at traversing the orbits α and β as determined by Eq. (2). Now, taking into account that the probability amplitude of the electron traversing the path λ in time t_λ is damped due to the finite quantum state lifetime τ by the factor $\exp(-t_\lambda/2\tau)$, we obtain, for $T = 0$,

$$\Gamma^{(AB)} = \gamma\gamma^* = \sum_{\lambda,\lambda'} \gamma_\lambda\gamma_{\lambda'}^* = \overline{\Gamma} + \Gamma_\alpha - \Gamma_{2\alpha} + \Gamma_\beta + \Gamma_{\beta-\alpha} - \Gamma_{\beta-2\alpha}\,, \tag{3}$$

where

$$\overline{\Gamma} = Q\left[1 + P^2\exp\left(-\frac{t_\alpha}{\tau}\right) + P^2Q^2\exp\left(-\frac{2t_\alpha}{\tau}\right) + P^4\exp\left(-\frac{t_\beta}{\tau}\right)\right],$$
$$\Gamma_\alpha = \left[QP\exp\left(-\frac{t_\alpha}{2\tau}\right) - Q^2P^2\exp\left(-\frac{3t_\alpha}{2\tau}\right)\right]\cos\varphi_\alpha, \quad \Gamma_{2\alpha} = Q^2P\exp\left(-\frac{t_\alpha}{\tau}\right)\cos(2\varphi\alpha)\,,$$
$$\Gamma_\beta = QP^2\exp\left(-\frac{t_\beta}{2\tau}\right)\cos\varphi_\beta\,,$$
$$\Gamma_{\beta-\alpha} = QP^3\exp\left(-\frac{t_\alpha + t_\beta}{2\tau}\right)\cos(\varphi_\beta - \varphi_\alpha)\,, \quad \Gamma_{\beta-2\alpha} = Q^2P^3\exp\left(-\frac{2t_\alpha + t_\beta}{2\tau}\right)\cos(\varphi_\beta - 2\varphi_\alpha)\,, \tag{4}$$

and $Q = 1 - P$ is the Bragg-reflection probability. Thus, the total probability $\Gamma^{(AB)}$ contains several oscillating terms, including $\Gamma_{\beta-2\alpha} \propto \cos(\phi_\beta - 2\phi_\alpha) = \cos[2\pi(F_\beta - 2F_\alpha)/H]$.

The effect of finite temperature is determined, as in the standard Lifshitz-Kosevich (LK) theory, by the energy dependence of the oscillation phase, in our case, of the difference between the phases of the interfering trajectories λ and λ',

$$\left[\frac{\partial(\phi_\lambda - \phi_{\lambda'})}{\partial\varepsilon}\right]_{k_H} = \left(\frac{\partial\phi_\lambda}{\partial\varepsilon}\right)_{k_H} - \left(\frac{\partial\phi_{\lambda'}}{\partial\varepsilon}\right)_{k_H} = \frac{2\pi c}{e\hbar H}(m_\lambda - m_{\lambda'})\,. \tag{5}$$

Therefore, the corresponding thermal factor contains the effective mass equal to the *difference* between the cyclotron masses on the paths λ and λ'. For the $(\beta - 2\alpha)$ oscillations, the effective mass $m^*_{\beta-2\alpha} = m_\beta - 2m_\alpha$ [9].

Now it becomes clear why the QI oscillations with the frequency 2630 T can be observed at high temperatures at which all the other harmonics are damped by the temperature smearing of the Fermi level. The effective mass determined from the temperature dependence of the oscillation amplitude yields $m^*_{\beta-2\alpha} = (0.9 \pm 0.1)m_0$, which is a factor of 4 smaller than the cyclotron mass of the fundamental α oscillations. Adding the thermal factor with $m = m^*_{\beta-2\alpha}$ to the expression (4) and evaluating the lifetime damping factor through the Dingle temperature determined from the experiment [9], $T_D \approx 0.6$ K, we estimate the term $\Gamma_{\beta-2\alpha} \sim Q^2P^3 \times 10^{-2} \leq 3.5 \times 10^{-4}$, at $T = 2.5$ K, $H = 14$ T. Taking into account that the warping of the Fermi surface, ≤ 1 meV, is sufficiently smaller than the MB gap, $\simeq 6$ meV [10], the probabilities Q and P may be considered as constant over all the Fermi surface. Then $\Gamma_{\beta-2\alpha}$ will represent a rough evaluation of the relative contribution of the $(\beta - 2\alpha)$ oscillations to the total magnetoresistance. On the other hand, the upper limit of the thermodynamic α-oscillation amplitude in the two-dimensional approximation [8] gives $\tilde{R}_\alpha \simeq 4 \times 10^{-4}R_0$ at the same temperature and magnetic field. At higher temperatures, the α oscillations rapidly diminish due to their comparatively high mass, $3.5m_0$. The amplitude of the oscillations shown in Fig. 2 demonstrates a very good agreement of the present estimations with our experimental data. The very low effective mass of the rapid oscillations allows them to be clearly observed at relatively high temperatures and unambiguously proves their QI origin. We note that, in contrast to the kinetic oscillation theory [1,2], the work [3] predicting thermodynamic oscillations with the combination frequencies suggests that the temperature dependence of the amplitude of the oscillations with difference frequency is stronger than that of the fundamental ones (i.e., higher mass may be prescribed to the difference frequency oscillations) and is therefore unlikely to be applied to the observed phenomenon.

It is interesting to note that the effective mass of the $(\beta - 2\alpha)$ oscillations can be further reduced and even tuned to zero by applying a moderate quasihydrostatic pressure. Indeed, as shown by Caulfield *et al.* [11], the areas of the α and β orbits have considerably different dependences on pressure. Therefore, one can also expect the relation between their masses to change with pressure. Figure 3 displays the oscillatory part of the magnetoresistance at $P = 8.5$ kbar at different

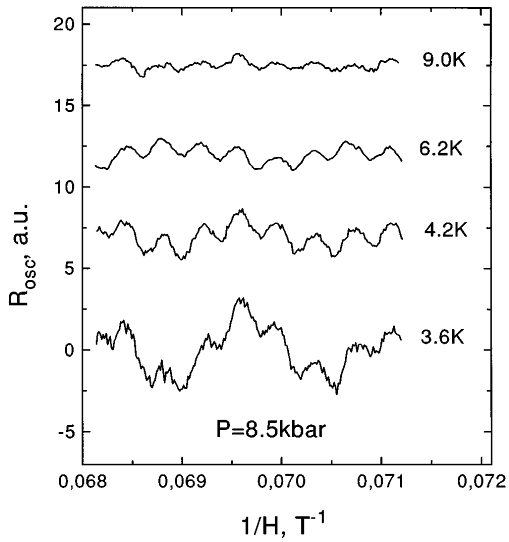

FIG. 3. Oscillatory part of the interplane resistance at different temperatures, at a pressure of 8.5 kbar.

temperatures. The $(\beta - 2\alpha)$ oscillations can be resolved up to 9 K. The standard LK plot for the determination of the effective mass shown in Fig. 4 yields $m^*_{\beta-2\alpha} \approx 0.3m_0$, an order of magnitude smaller than the classical effective mass. This plot does not take into account that the quantum state lifetime τ may change considerably in the relatively wide temperature range. This $\tau(T)$ dependence should cause an additional, independent of the LK thermal factor, damping of the oscillations at heating the sample. Noting that below 4 K the oscillation amplitude is constant within the experimental error, we may suppose that the true effective mass $m^*_{\beta-2\alpha} \approx 0$ at the present pressure, attributing the decrease of the amplitude at higher temperatures to the increasing temperature dependence of τ. Further detailed studies under pressure should check this supposition and provide important information on the quantum state lifetime and its temperature dependence.

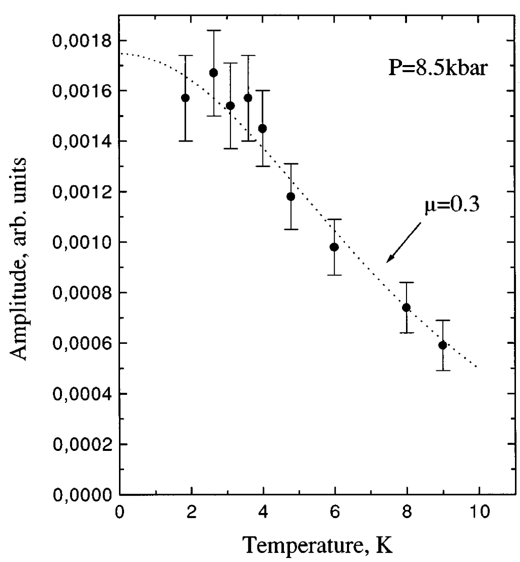

FIG. 4. Temperature dependence of the $(\beta - 2\alpha)$-oscillation amplitude. The dashed line represents the standard LK plot with $\mu = m^*/m_0 = 0.3$.

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