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[22] L. Passell (private communication) has pointed out to us the difficulty in resolving the narrow energy widths in low-temperature systems like EuS.

Influence of the Electron-Phonon Interaction on the de Haas–van Alphen Effect in Mercury

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Accurate measurements of de Haas–van Alphen effect amplitudes in mercury show, for the first time, departures from quasiparticle behavior. The departures arise from the influence of the electron-phonon interaction.

The controversy relating to the influence of the electron-phonon interaction on the de Haas–van Alphen (dHvA) effect has an interesting history. Wilkins and Woo[1] first demonstrated that the cyclotron effective mass which appears in the thermal damping factor will be renormalized by the electron-phonon interaction. In addition to this, however, Palin[2] anticipated that the Dingle factor,[3] which embraces the effect of electron collisions, might also contain a term proportional to T^3, such as affects the line shape in Azbel-Kaner cyclotron resonance,[4] and that such a term would lead to significant departures from the original theory of Lifshitz and Kosevich[5] (LK) in the amplitude of dHvA-effect oscillations. Surprisingly, however, even for the most favorable metal in this respect, namely mercury with its low-lying phonon mode ($\Theta_D \sim 21$ K) and strong electron-phonon coupling, no such departures were observed.[2] This unexpected result was explained by Engelsberg and Simpson[6] (ES), who showed that the effect of electron-phonon interactions could be incorporated in the oscillatory part of the thermodynamic potential by replacing, at a certain state, the noninteracting single-particle electron energies by the noninteracting energy plus full electron-phonon self-energy. Their result, which is an extension of earlier work by Fowler and Prange,[7] indicates that, with regard to the amplitude of the dHvA effect under accessible laboratory conditions, the departures from the LK theory are expected to be small, even for mercury, and are not inconsistent with the experiments of Palin.[2] This result is sometimes interpreted as being due to a cancellation between changes in the mass-renormalization and electron-phonon scattering rate. However, Engelsberg[8] has shown that this description is not completely valid and has interpreted departures from LK as being the result of a failure of the quasiparticle approximation. The same problem, however, has also been discussed by Gantmakher[9] who claims that Palin's apparently null result follows from a "classical" calculation of electron scattering in a magnetic field in the presence of electron-phonon interactions. The purpose of this Letter is to present new experiments on dHvA amplitudes in mercury, under conditions rather more favorable than those of Palin, in which, for the first time, we have observed departures from quasiparticle behavior. Aside from underpinning the theory of the electron-phonon interaction, these results, we believe, have implications for the experimental investigation of other many-body effects in metals using the dHvA effect.

Prior to discussing the experimental results we shall comment briefly on the theory of ES in order that the optimum experimental conditions for observing the effects might be appreciated and also that, in what follows, we may compare theory and experiment. According to ES, the entire effect of electron-phonon interactions is contained in a term, A_r, for the amplitude of the rth harmonic of the dHvA effect so that

$$A_r = \sum_{n=0}^{\infty} \exp\{(-2\pi r/\hbar\omega_c)[\omega_n + \zeta(\omega_n)]\}, \tag{1}$$

in which ω_c is the cyclotron frequency ($= eB/m_c$),

$\zeta(\omega_n)$ is the full self-energy Σ evaluated at the imaginary frequencies $i(i\omega_n)$ such that $\zeta(\omega_n)=i\Sigma(i\omega_n)$ and where the ples, ω_n, of the Fermi function are defined by $\omega_n=(2n+1)\pi k_BT$. Averaging over the shole Fermi surface,[10] $\zeta(\omega_n,T)$ is then given from the theory of the electron-phonon interaction by[6]

$$\zeta(\omega_n,T)=\pi k_BT\int_0^\infty \frac{d\nu\,2\alpha^2(\nu)F(\nu)}{\nu}\left\{1+2\sum_{l=1}^{n}\left[1+\left(\frac{2\pi l k_BT}{\nu}\right)^2\right]^{-1}\right\}. \tag{2}$$

The behavior of Eq. (1) in various limits has been discussed by both Mueller and Myron[11] and by Engelsberg.[8] At both high temperatures and as $\omega_c\to 0$, only the first pole, ω_0, contributes effectively to the summation. From Eq. (2), $\zeta(\omega_0,T)=\lambda_0\pi k_BT$, with λ_0, the electron-phonon renormalization constant, given by

$$\lambda_0=\frac{m_c^*}{m_c}-1=\int_0^\infty d\nu\,\frac{2\alpha^2(\nu)F(\nu)}{\nu}, \tag{3}$$

so that in this limit Eq. (1) reduces to the LK form[6] with m_c renormalized to m_c^*. The limiting behavior at high fields and low temperatures is less easy to discern. While ζ is not itself field dependent, with increasing field an increasing number of terms become effective in the summation in Eq. (1). If in Eq. (2) $(2\pi l k_BT/\nu)^2\sim 1$ at the highest values of l $(=n)$ which contribute effectively to the summation in Eq. (1), then departures from quasiparticle behavior and hence from the LK theory will ensue. Inspection of Eqs. (1) and (2) shows that larger departures are favored by small effective masses, low characteristic phonon frequencies, and large values of $\alpha^2(\nu)F(\nu)$. The β orbit in mercury[12] is possibly the most favorable case of all.

Single crystals of high-purity[13] mercury were grown in the cryostat by slow cooling (0.2 K min^{-1}) through the melting point. Subsequently they were rotated to bring the direction 24° from $\{100\}$ in the trigonal-bisectrix plane parallel to the magnetic field, at which orientation the β frequency, which derives from a hole orbit situated at the $\{110\}$ Brillioun-zone faces, has a minimum value of both dHvA frequency ($F\simeq 74$ T) and cyclotron effective mass ($m_c^*\simeq 0.15m_0$). Experiments were performed using the low-frequency field modulation technique. Dictated partly by use of a torque magnetometer, Palin's experiments were performed at a rather less favorable orientation for which $F\simeq 88.5$ T and $m_c^*\simeq 0.18m_0$; with hindsight the effects sought would have been substantially diminished because of this. Finally, using a minicomputer coupled to the experiment, the field and temperature dependences of the amlitudes and phases of the harmonic components of the dHvA effect have been accurately determined.

The experimental results are given in Fig. 1 in the conventional form of a Dingle plot for both the fundamental ($r=1$) and second harmonic ($r=2$) of the dHvA effect. In this case the experiments were performed at $T=2.1$ K over the field range $\sim$1–5 T. At the lower fields ($B\lesssim 2$ T) the results

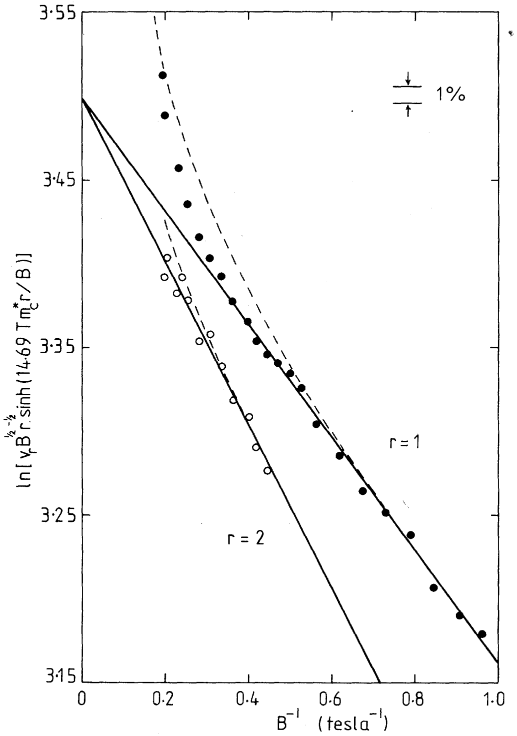

FIG. 1. Dingle plots showing the field dependence of the fundamental (●) ($r=1$) and second-harmonic (○) ($r=2$) components of the dHvA-effect amplitude for the β orbit in mercury. In the ordinate, V_r is the appropriately normalized signal voltage. The full lines correspond to the LK theory, and have a common intercept in view of the fact that the spin factor, $\cos(r\pi g m_c^*/2m_0)$, in this case takes the value $(-1)^r$ (see Ref. 14). The broken lines refer to the predictions of the ES theory as explained in the text. A 1% change in the dHvA-effect amplitude is also indicated.

for the fundamental term show quasiparticle (linear) behavior, the solid line corresponding to the LK theory for a Dingle temperature of 0.15 K and with $m_c^* = 0.152m_0$, as determined from the temperature dependence of the dHvA effect at the lowest fields. At higher fields, however, the observed dHvA amplitude is seen to diverge from the predictions of the LK theory. At 5 T the measured amplitude exceeds the LK value by 8%. For the second-harmonic component, however, the experimental results show no departures from quasiparticle behavior within the experimental uncertainty of ~ 1%.

It is interesting to compare these results with the predictions of the ES theory which we have computed using, for $\alpha^2(\nu)F(\nu)$, the data of McMillan and Rowell.[15] The results is shown in Fig. 1 by the broken lines and is seen to be in very satisfactory agreement with experiment. The small discrepancy may well in part be due the form of $\alpha^2(\nu)F(\nu)$ which we have used in the analysis, which corresponds to an "average" over the Fermi surface and may not therefore be quite appropriate for the β orbit. It is evident from an inspection of Eqs. (1) and (2) and from Fig. 1 that departures from the LK theory will be less pronounced with increasing harmonic index, r. That the experimental results reproduce this feature is convincing evidence of their validity and indicates that the present observations cannot be attributed to crystalline defects or to phase smearing arising from sample strains.

An interesting observation is that the gradients of the Dingle plots for the first three harmonics of the dHvA effect are not in the LK ratio of 1:2:3 but rather $1:(1.44\pm0.09):(1.43\pm0.23)$. (For $r=1$, only observations made for $B<2$ T are included.) Using for m_c^* the value of, $0.152m_0$, determined from the temperature dependence of the dHvA-effect amplitude of the fundamental over the temperature range 1.2–3.8 K at 1.5 T, this yields, for $r=1$, 2, and 3, apparent Dingle temperatures of 0.151 ± 0.004, 0.109 ± 0.007, and 0.072 ± 0.015 K, respectively. We note that magnetic interaction effects, which could in principle influence harmonic amplitudes, are quite negligible under the present experimental conditions. This behavior is not understood, but further experiments on the temperature dependence of dHvA effect amplitudes will clarify this point and allow a closer comparison with the ES theory.

The present experimental results have thus shown, for the first time, that the influence of the electron-phonon interaction leads to departures from the LK theory in the field dependence of dHvA-effect amplitudes. The theory of ES is in very satisfactory agreement with these departures from quasiparticle behavior. This work suggests that careful attention to dHvA amplitudes may well prove to be a useful tool in the experimental investigation of other many-body effects in metals, such as, for example, the electron-paramagnon interaction.

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