Precise measurement of effective masses in cadmium by using the de Haas–van Alphen effect

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The effective masses of conduction electrons in cadmium have been measured with high precision on the basis of the temperature dependence of the oscillation amplitudes in the de Haas–van Alphen effect. The results are compared with values of the masses determined by cyclotron resonance. The errors associated both with instrumental effects as well as with the Shoenberg effect are analyzed.

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1. INTRODUCTION

Important characteristics of the conduction electron energy spectrum can be determined from the de Haas–van Alphen effect (dHvA). Among them are the extremal cross sections S_i of all parts of the Fermi surface, the electron effective masses $m_i^* = (2\pi)^{-1}\partial S_i/\partial\varepsilon$ for these orbits, and the effective Dingle temperature T_D which characterizes the broadening of the Landau levels. It is usually considered that accurate values of effective masses are obtained by measurement of cyclotron resonance, while the accuracy of dHvA determinations is around 10%. However, the cyclotron resonance method sets high demands on the quality of the specimen surface and can not thus be used for specimens under pressure. The susceptibility oscillations in the dHvA effect are determined by electron orbits over the whole volume of the specimen and are observed under a pressure p, so that the dHvA effect is the only method for determining dm^*/dp.

It is shown in the present work, using as an example the polyvalent metal cadmium with a complicated Fermi surface (FS), that effective masses can be measured under favorable conditions to an accuracy of $\sim 1\%$, and to ~ 5 to 10% in the most complicated cases. These results are compared with results of cyclotron-resonance measurements and an analysis is given of both instrumental errors and errors due to physical causes, in particular the Shoenberg effect.

The effective mass m^* is determined in the dHvA effect from the temperature dependence of the oscillation amplitude:[1]

$$A = A(H)\,T\,\mathrm{sh}^{-1}\left(\frac{2\pi^2 ckm_0}{e\hbar}\frac{m^*}{m_0}\frac{T}{H}\right).$$

In most cases for normal metals the argument of the hyperbolic sine is several units and sinh can be replaced by an exponential. The error associated with this substitution does not exceed a fraction of 1% and can easily be corrected for by a simple iterative method. m^* is thus determined from the slope of the graph of $\ln(A/T)$ against T at a fixed magnetic field H. To measure to an accuracy of $\sim 1\%$ one must measure T, H and A and maintain T and H during the measurements to an accuracy of $\sim 0.1\%$. The accuracy in determining m^* depends on the magnitudes of H and of the mass itself, and determination of the mass to an accuracy of no worse than $\sim 1\%$ is usually achieved by measuring the amplitude of oscillations to an accuracy of $\sim 1\%$.

2. EXPERIMENTAL TECHNIQUE

The cadmium specimens $0.9\times0.9\times(5$ to $8)$ mm were cut by electrosparking from a large single crystal and were oriented so that the long specimen axis lay in the $(11\bar{2}0)$ plane, while the angle θ between the specimen axis and the [0001] direction took values from 0 to 90° in 10° steps. The orientation of each specimen was fixed accurately by x-rays and also by comparing the measured oscillation frequencies with earlier results.[2] Specimens etched in a 50% mixture of HCl and alcohol were glued with BF-2 adhesive to a kapron thread and were mounted on it inside the receiver coil. Cooling of the apparatus to helium temperature took an hour.

It was established that cycles of warming up to room temperature lead to an increase in Dingle temperature T_D. For example, values of $T_D = 0.7$, 1.1 and 1.2 K were obtained after heating cycles for one of the specimens. In general, therefore, all measurements on a single specimen were carried out without warming up above 4.2 K.

A detailed description of the experimental setup and its separate components has been given earlier.[3–6] Only the main features of the apparatus are given below.

A magnetic field up to 94 kOe was produced in a superconducting solenoid by a source of current up to 200 A with a relative current instability of not more than 0.01%. The field was calibrated by NMR with an absolute error of ~ 0.5 Oe, and the accuracy of field determination from the current through the solenoid was $\sim 1\%$ for $H < 20$ kOe and not worse than 0.06% for $H > 20$ kOe. The minimum rate of field sweep was ~ 0.1 Oe·s^{-1}, which is very important for reducing dynamic errors in amplitude measurement.

The temperature was determined from the helium vapor pressure and was maintained constant to ~ 0.5 mK using a precision temperature controller which had an Allen-Bradley carbon resistor as sensor placed near the specimen.

The field modulation signal was provided by a GZ-35 oscillator having a $\sim 0.2\%$ harmonic coefficient, with a relative frequency drift $\sim 0.1\%$ over an 8 hour period of operation. To maintain constant the argument of the Bessel function that determines the amplitude of the emf in the receiver

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coil, the modulation amplitude h was kept proportional to H^2 to an accuracy no worse than 1.5% and the coefficient of nonlinear distortion was $\sim 0.1\%$. The signal reached the modulation coil after passing through a power amplifier rated up to 50 W, which had a transfer coefficient stable to 0.01% with a null drift not more than 1 mV and a small coefficient of non-linear distortion.

The pickup channel consisted of a step-up transformer with a turns ratio 40, a twin-T bridge with 80 dB suppression of the modulation frequency, a U2–8 selective amplifier, and a phase-sensitive detector. At frequencies from 10 Hz to 15 kHz the phase-sensitive detector ensured conversion linearity no worse than 0.1% over the range of output voltages from 30 mV to 2.6 V, the permissible ratio of out-of-phase noise to useful signal was 50 dB, the suppression of even harmonics was not less than 40 dB and the null drift was not more than $250 \mu V \cdot K^{-1}$. Detection was in all even harmonics of the modulation frequency from the second to the fourteenth.

To measure a signal proportional to the magnetic susceptibility, besides detection of the harmonics of the modulation frequency, measurements were made at the fundamental frequency, which was in this case chosen to be 19 Hz. A variometer was used for total compensation of the signal picked off the measuring coil. The useful signal in such a measuring regime was much less than in measuring the harmonics, and the system itself was more sensitive to mechanical vibrations. Measurements at the fundamental frequency were therefore carried out only in special cases (the Shoenberg effect, verification of the absence of the influence of the skin effect), while the main measurements of effective masses were carried out at harmonics of the modulation frequency.

3. EXPERIMENTAL RESULTS

Measurements of effective masses were made on electrons moving over the pocket α, monster γ and lens λ orbits of the Fermi surface of cadmium,[7] lying respectively in the first, second and third Brillouin zones. These results are shown in Fig. 1.

Strong low-frequency α oscillations were, in general, easily measured and only in orientations close to the [0001] axis did oscillations with frequencies F_β and F_χ (β and γ monster orbits[7] close to the pocket α orbit) mix in with the F_α oscillations, thereby slightly lowering the accuracy in determining the mass.

The γ and λ high-frequency oscillations could only be determined reliably in certain angular intervals. For example, the mass of the λ orbit could only be obtained with satisfactory accuracy for orientations θ from 50 to 90°. Oscillations with frequency F_λ were strongly modulated at small angles by low-frequency oscillations mainly with frequency $F_{\gamma/3}$, although traces of longer-period modulation, possibly at a frequency F_α, were noticeable. Oscillations connected with the lens were unreliably observed at orientations $\theta = 20$ to 40°, and it was impossible to measure the effective mass in these directions.

The effective mass of electrons connected with the γ orbit was measured in orientations from 30 to 60°; it was also modulated at the frequency F_α. The oscillations were extremely weak for $\theta > 60°$, while for $\theta < 20°$ and fields from 20 to 80 kOe used in the experiment the γ orbit undergoes a magnetic breakdown.[8] Masses connected with the magnetic-breakdown orbits $\gamma/3$ and $2\gamma/3$ were also not measured.

The observed picture of signal modulation cannot be related to beating of neighboring frequencies and can only be explained by the Shoenberg effect, since the modulation frequency coincided in all cases with the frequency of some or other low-frequency oscillations. The trace of oscillations with frequencies F_α and F_γ ($\theta = 50°$), made on the sixth harmonic of the modulation frequency for different values of modulation amplitude h, for one and the same field interval, is shown in Fig. 2. It can be seen that the high-frequency F_γ oscillations are modulated according to the amplitude of the low-frequency F_α with a phase shift of $\pi/2$ characteristic of the Shoenberg effect[9] (a term $\propto dM_\alpha/dH$ enters into the oscillation amplitude).

The accuracy in determining effective masses decreased in all cases when the Shoenberg effect appeared (distortion of the shape of the oscillations, the appearance of anomalies in the magnetic field dependence of the oscillation amplitude), and attempts were made to avoid harmful influences connected with the Shoenberg effect by increasing the temperature or reducing the magnetic field. However, these attempts were not always successful since the conditions for observing the oscillations then deteriorated sharply.

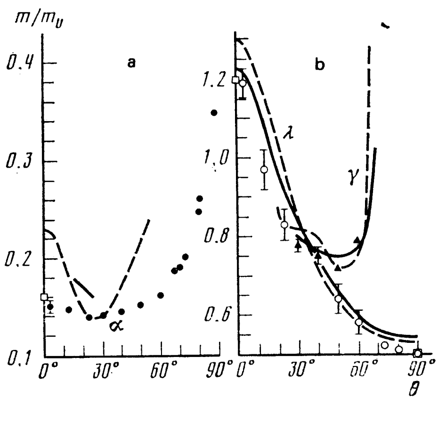

FIG. 1. Angular dependence of effective masses in cadmium: a—for a pocket α orbit; b—for lens λ and monster γ orbits. Dashed lines—results of Naberezhnykh,[10] full lines—Shaw *et al.*,[11] □—Carlin *et al.*[12], ●, ▲, ○—present work.

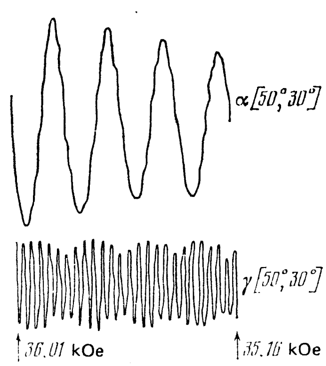

FIG. 2. Trace of oscillations associated with the pocket α and monster γ orbits in one and the same magnetic field interval.

Measurements of the influence on the oscillation with frequency F_α by quasihydrostatic pressure up to 7 kbar were carried out on a specimen with $\theta = 40°$. The measurements were made in a fixed-pressure bomb; a mixture of pentane and transformer oil was used as pressure transmitting medium. The oscillation frequency decreased by 6.2% under pressure while the magnitude of the effective mass stayed unchanged within the limits of $\pm 2\%$.

4. DISCUSSION OF THE RESULTS

The experimental values of m^*/m are shown in Fig. 1 together with those obtained by the cyclotron resonance method.[10–12] It can be seen from Fig. 1, *a* that our values are in good agreement with those of Carin *et al.*[12] for $\theta = 0°$ and with Naberezhnykh[10] for $\theta = 30°$ at the minimum of the curve; at all other angles the disagreement is rather large and reaches 30% at the edges of the curve,[10] which is much greater than the experimental errors.

The ratio m^*/m_0 for orbits λ and γ is shown in Fig. 1, *b*. Our values for the lens near $\theta = 0°$ agree well with earlier results[11,12] and are slightly smaller than the values given by Naberezhnykh[10]; good agreement is observed near $\theta = 90°$ with the results of Carin *et al.*[12] with a small departure from the other results.[10,11] Our results for the γ monster orbit for $30° < \theta < 60°$ agree with results of Shaw *et al.*[11] for all angles except 50°, at which agreement is observed with values given by Naberezhnykh.[10]

It can thus be considered that within the scatter of results obtained by the cyclotron resonance method, our results are in satisfactory agreement with Refs. 10 to 12, the best agreement being with Carin *et al.*[12] It would be interesting to analyze the errors of each of the methods, but we have only analyzed those peculiar to the dHvA effect.

Random errors are connected with industrial interference, inaccuracy in reading the instruments, etc. Errors due to drift in the measuring system are in the same category. Repeated measurements of one and the same mass in different field intervals were made to determine the real random errors. Oscillations with the F_α frequency were chosen, having a large amplitude and monochromatic oscillation structure. The scatter in the values of m_α^* was then within the limits 0.2 to 0.4%, which shows that random errors lead to a real inaccuracy in measuring m^* better than 1%. The random error increased when recording weak high-frequency oscillations, but was not above 2 to 3%.

Systematic errors can arise from a number of causes. One of them is related to replacing the hyperbolic sine by an exponential, as noted above. Another can be connected with a temperature jump on passing the λ point,[13] and measurements were carried out at temperatures between 1.3 and 2.1 K to avoid it. The skin effect can lead to error, but the same result was obtained when detecting at the fundamental modulation frequency of 19 Hz and at the 6th harmonic of the modulation frequency 490 Hz. The main source of errors is evidently the Shoenberg effect,[14] wherein the electrons feel not the external magnetic field **H**, but the induction $\mathbf{B} = \mathbf{H} + 4\pi\mathbf{M}$. The magnetic moment then satisfies the self-consistent equation $\mathbf{M} = f(\mathbf{H} + 4\pi\mathbf{M})$, where $f(B)$ is given by the Lifshitz-Kosevich equation,[1] taking account of all extremal sections of the Fermi surface. In cadmium there is a large collection of extremal sections, different in magnitude, and this can lead to low-frequency modulation of the high-frequency oscillations. Amplitude modulation (AM) can arise both because of dephasing[9] and because of apparatus effects,[15] for example because of field nonuniformity or of features, connected with frequency-amplitude modulation, of the method of measuring the dHvA effect (FM-AM effect[15]). In the case of strong diamagnetism the Shoenberg effect leads to distortion of the shape of the oscillations and to a dependence of the amplitude on the field[9,14] and on the temperature, compared with the Lifshitz-Kosevich formula.

The effect of AM on the determination of the effective mass can be treated qualitatively for the case of limitingly weak AM, when the signal is of the form

$$A = A_0[1 + \xi \sin(2\pi F_\alpha/H + \varphi)] \sin(2\pi F_\lambda/H),$$

where $\xi \ll 1$ and is proportional to $\partial M_\alpha/\partial H$; the frequencies F_α and F_λ are respectively low and high (in our case, corresponding to the pocket α and lens λ orbits), and A_0 is the initial amplitude determined by the Lifshitz–Kosevich formula. Since M_α increases with decreasing temperature, the amplitude of the oscillations of A changes faster than the initial amplitude A_0 at the maximum of the train and more weakly at the minimum. This leads to the value of m^* determined from the maximum amplitude being raised, while on working with the minimum amplitudes it is lowered. The temperature dependence of $\xi \propto \partial M_\alpha/\partial H$ decreases as the parameter $2\pi^2/mckT/e\hbar H$ approaches unity, i.e., the scatter in mass is smaller the higher H. Such behavior was observed experimentally.

If the amplitude averaged over the AM period is used, then the systematic error in determining the effective mass, due to the AM, decreases in the case of limitingly weak AM by about F_λ/F_α compared with the result of using extremal amplitudes. Such averaging also decreases the random error in determining the effective mass.

A situation close to the model case is realized experimentally for $\theta > 60°$; averaging the amplitude over angle gave a value $m^*/m = 0.508$ at $\theta = 80°$ with an error $\pm 0.6\%$. The value obtained by working with the maximum amplitude was greater by $(1 \pm 0.8)\%$, while the value obtained from the minimum amplitude was less by $(2.6 \pm 1.0)\%$.

The results for orbits λ and γ at $\theta < 50°$ were appreciably worse; the difference between the masses determined by averaging the maximum and minimum amplitudes of the train was not of a systematic nature. Many measurements for the lens at $\theta = 13°$, carried out in different but close fields, gave after averaging values differing by up to 10%, while the scatter of points on the plot $\ln(A/T)$ *vs.* T gave an error in determining mass of not more than 2 to 4%. All this points to the fact that it is not possible to carry out a correct treatment of the results either within the framework of the approximate model, or when there is a low-frequency spectrum: the only possibility is an attempt to reduce the Shoenberg effect itself by changing the magnitudes of the magnetic field and the temperature. This applies in full measure to those cases when the Shoenberg effect manifests itself in an enrichment of high harmonic oscillations, i.e., in a distortion of the shape of the oscillations.

5. CONCLUSIONS

Comparison of the values of effective masses obtained in the present work with those obtained in measurements of the dHvA effect in pulsed magnetic fields,[8] without precise determination of T, H, and the amplitude of oscillation, show that the precisely measured masses agree considerably better with values of m^* determined by cyclotron resonance. On considering the internal disagreements of the results of three studies,[10–12] which exceed the mass accuracy obtained in each study, it would be very desirable to carry out a new set of precision measurements of $m^*(\theta)$ by the cyclotron resonance method for comparing the results obtained by this method with those from the dHvA method. Such a comparison would provide faith in the reliability of measurements of m^* under pressure.

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