The Fermi surface of cadmium at an electron-topological phase transition under pressure

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The de Haas–van Alphen (dHvA) effect has been measured in cadmium under a pressure up to 30 kbar. The pressure derivatives (PD) of extremal cross sections and the effective masses have been measured. New frequencies were observed in the dHvA spectrum: the ultralow $F_\delta = 1.2\times 10^5$ G ($p = 21.4$ kbar) and $F_\varepsilon = 2.5\times 10^4$ G ($p = 20$ kbar), corresponding to new parts of the Fermi surface, and $F_\tau \approx 22$ MG ($p = 21.4$ kbar) connected with deformation of the initial Fermi surface under pressure. The pressure derivative of F_δ is 240×10^{-3} kbar^{-1}.

An electron-topological phase transition (ETT), consisting of a change in the majority of a metal's properties when the topology of the Fermi surface (FS) changes, was predicted by Lifshitz[1] and was later found experimentally.[2,3,5] There and in subsequent theoretical and experimental work by many authors, it was shown that this change leads to the appearance of specific features in many electronic, thermodynamic and kinetic properties of the metal.

Itskevich and Voronovskiĭ[5] observed a qualitative change in the form of the angular dependence of magnetoresistance at pressures 10–15 kbar. It was interpreted[5,6] as being the result of the linking of the horizontal arms of the hole part of the FS, the "monster" in the second zone, and the formation of an electron "needle" in the third zone, leading to the appearance of open electron orbits when the magnetic field is directed along [0001]. A pressure of 15 kbar corresponds to such a value of c/a for the hcp structure (which decreases with increasing pressure) and to a change in crystal potential when, according to estimates by Itskevich,[6] its FS takes on a shape similar to that of zinc. Uniaxial stress of zinc whiskers,[7] producing an increase in c/a, led to a form of angular dependence of magnetoresistance, characteristic of cadmium at zero pressure. These works demonstrated the "specularity" of the anisotropy of magnetoresistance and of the properties of the FS of zinc and cadmium which are responsible for it[8] on changing the lattice parameters. An anomaly in the pressure dependence of the resistivity of cadmium at $p = 35$ kbar and room temperature was also ascribed to a qualitative change in the shape of the FS.

In all such work, however, experiment does not provide evidence on the actual changes of the FS at pressures $p > p_{\text{trans}}$ and on the possible manifestation of an ETT in the electronic and kinetic properties of metals. In this work an experiment to observe the de Haas–van Alphen effect (dHvA) is described, allowing a determination of the magnitudes of the areas of extremal sections of the FS; this enables new parts of the FS which arise to be observed directly at pressures above p_{trans}.

METHOD AND SPECIMENS

The solution of the problem posed here became possible when a self-contained piston-and-cylinder type cell was constructed in which hydrostatic pressures of 30–35 kbar could be obtained at helium temperatures.[10] We used a nonmagnetic version of such a cell similar to that used for studies of the Shubnikov–de Haas effect in semiconductors.[11] The external diameter of the chamber was 58 mm, the inner 6 mm. Pressure was applied in a pentane-oil medium (in a ratio 60–40) at room temperature and fixed by a nut, after which the cell was cooled slowly to nitrogen temperature.

The pressure was determined from the temperature of the superconducting transition in indium or tin. In a preliminary article[12] we determined the pressure based on a quadratic $T_c(p)$ dependence obtained for indium.[13] It was found later, however, that the dependence overestimates the value of pressure for $p \gtrsim 15$ kbar and a linear $T_c(p)$ dependence must be used with a value $\partial T_c/\partial p = -3.88\times 10^{-2}$ K kbar^{-1} (Ref. 14). A quadratic $T_c(p)$ dependence was used for tin.

A modulation method[15] was used to record the dHvA effect. The whole modulation and detector coils system (total number of windings ~ 5000) was housed in the high pressure region. The experiment was carried out in the temperature range 4.2–1.7 K, in a magnetic field H of a superconducting solenoid, up to 80 kOe and a modulation field up to 120 Oe. Specimens with dimensions $1.7\times 1.7\times 3$ mm were cut by the electrospark method from a cadmium single crystal with resistance ratio $R_{300\,\text{K}}/R_{4.2\,\text{K}} \approx 20\,000$.

The oscillation amplitude related to an extremal cross section can be written in the form

$$A_i(H,T)=M_i(H,T)D_i(H)\left(\frac{\partial^2 S_i}{\partial k_z^2}\right)^{-1/2},$$

where $M_i(H,T)$ is the oscillatory part of the magnetic moment,

$$D_i(H)=\exp\left(-k(m_i^*/m_0)T_D/H\right)$$

is the Dingle factor, T_D the Dingle temperature $(\partial^2 S_i/\partial k_z^2)^{-1/2}$ is determined by the curvature of the FS in the region of its ith section.

The crystal perfection of cadmium specimens decreases on cooling-heating cycles, which shows up in some reduction in the oscillation amplitude and an increase in the Dingle temperature T_D. This damage to the crystal shows up more strongly in experiments at high pressure when the

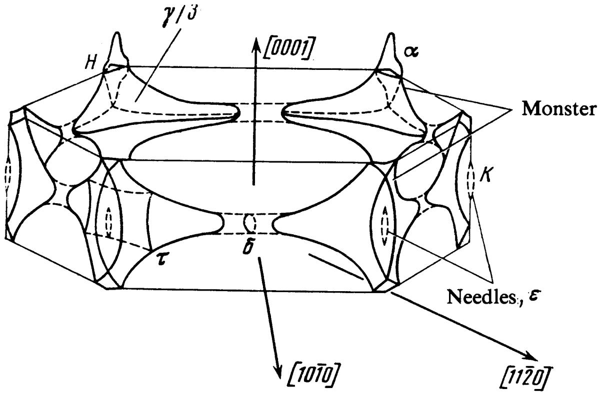

FIG. 1. Fermi surface of cadmium (the electron lens at the center of the Brillouin zone is not shown). The indices indicate the measured extremal sections of the FS.

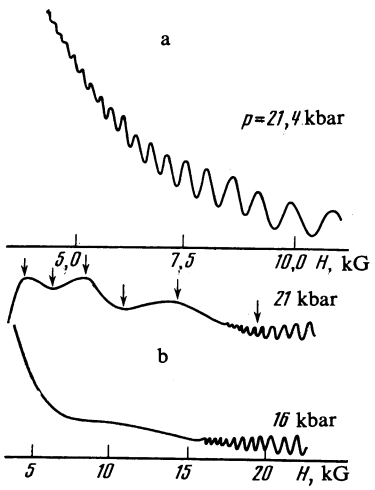

FIG. 2. dHvA oscillations in cadmium, produced by new parts of the FS arising from the ETT: a) frequency δ, $H \parallel [11\bar{2}0]$; b) frequency ε, $H \parallel [0001]$.

transmitting medium freezes. The Dingle factor which appears in the expression for the oscillation amplitude, appreciably decreases the amplitude in small H where infralow frequencies ($F \leqslant 10^5$ G) are usually observed. The amplitudes of these frequencies decrease appreciably on each heating-cooling cycle, while the amplitudes of the average frequencies ($F \approx 10^6$–10^7 G) after decreasing 2–3 fold in the first experiment with pressure, later decrease insignificantly on cycling. In view of this, experiments were usually carried out first at the maximum pressure and this was then gradually decreased.

The oscillations of magnetic susceptibility as a function of the current in the solenoid were recorded on a chart recorder and simultaneously on punched tape which was then analyzed on a computer, using a rapid Fourier transform program.

EXPERIMENTAL RESULTS

a) Magnetic field $H \parallel [11\bar{2}0]$. Two cycles of experiments with maximum pressures 21.5 kbar (specimen 1) and 23 kbar (specimen 2) were carried out for this magnetic field orientation. In the first cycle, measurements were carried out as the pressure was raised to 13.5 and 20 kbar after lowering to 5.4 kbar; the high degree of reproducibility of measurements of medium and high frequencies was then shown.

The following frequencies of the dHvA spectrum with values at zero field coinciding with those known earlier were observed in this orientation.

The frequency F_α corresponding to a longitudinal section of the hole pocket in the first zone by the $(11\bar{2}0)$ plane. Pocket α is centered on the point H in Fig. 1. The $F_\alpha(p)$ relation is linear within the limits of error. The value obtained for the pressure derivative (PD) $d \ln F / dp = (-14.2 \pm 0.7) \times 10^{-3}$ kbar^{-1} agrees with known values, for example $(-13.5 \pm 0.5) \times 10^{-3}$ kbar^{-1} (Refs. 16, 17). Measurement of the effective mass at a pressure of 21.4 kbar gave a value $m^*/m_0 = 0.26 \pm 0.05$, which agrees, within the error limits, with the value known at zero pressure.[18]

The frequency F_λ corresponds to a section of the electron lens in the third zone. The value of F_λ obtained by us at zero pressure and the PD are 62.7 MG and $(-2.7 \pm 0.3) \times 10^{-3}$ kbar^{-1} and agree within the error limits with earlier values.[16]

A new frequency appears in the dHvA spectrum at $p \gtrsim 8$ kbar, which we denote by F_τ. Its amplitude grows strongly with increasing pressure and dominates in the field range $65 \geqslant H \geqslant 35$ kG. This frequency decreases as the pressure is increased, its PD being $(-4.0 \pm 0.5) \times 10^{-3}$ kbar^{-1}; at 23.3 kbar $F_\tau = 22.5$ MG. The effective mass at the maximum pressure is $m^*/m_0 = 0.44 \pm 0.1$.

The most interesting result obtained in the $H \parallel [11\bar{2}0]$ orientation is the observation of an unusual infralow frequency with a value of 1.2×10^5 G at 21.4 kbar. The frequency, which we denote by F_δ, was recorded for both specimens in this orientation for pressures above 20 kbar. An example of the oscillation picture at $p = 21.4$ kbar is shown in Fig. 2a. The pressure dependence of the oscillation frequency is shown in Fig. 3; the value $d \ln F_\delta / dp = (240 \pm 30) \times 10^{-3}$ kbar^{-1} is obtained from this. The change in effective mass with pressure was not recorded and its mean value is $m^*/m_0 = 0.07 \pm 0.02$.

b) Magnetic field $H \parallel [10\bar{1}0]$. Two frequencies known before were measured over the whole pressure range for this field orientation: $F_\alpha \approx 11.5$ MG (at $p = 0$ bar) and $F_\lambda \approx 62$ MG ($p = 0$ bar), their PD's were determined and are equal

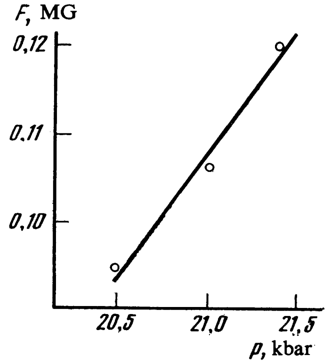

FIG. 3. Pressure dependence of the frequency δ.

to, respectively, $(-8.3 \pm 3.4)\times 10^{-3}$ kbar^{-1} and $(-1.8 \pm 0.5)\times 10^{-3}$ kbar^{-1}. The PD of the lens agrees, within the error limits, with that measured earlier,[16] while the PD of the pocket differs, $(-15 \pm 1)\times 10^{-3}$ kbar^{-1}. This may be related to the specimen misorientation which we noticed in this experiment.

A new frequency was recorded which we suggest is of the same nature as frequency F_τ, as will be justified in the discussion. Its value at the maximum pressure of 24.1 kbar is 27.2 MG. Its amplitude is low and becomes lower than the amplitude levels of the α and λ on lowering the pressure, but was well visible on the Fouriergrams down to pressures ~ 20 kbar. Its PD $= (-5.1 \pm 1.8)\times 10^{-3}$ kbar^{-1}.

c) Magnetic field $H \parallel [0001]$. Three cycles of raising the pressure with different specimens were carried out at this field orientation: up to 18.2 kbar (specimen 1), 23 kbar (specimen 2) and 30 kbar (specimen 3). Specimen 2 had the largest oscillation amplitude of all frequencies. The pressure derivatives of frequencies α and $\gamma/3$, the section of the monster by the (0001) plane in one Brillouin zone, were measured in all three cycles. The values of PD's were, respectively, $(-10.5 \pm 0.5)\times 10^{-3}$ kbar^{-1} and $(2.2 \pm 0.7)\times 10^{-3}$ kbar^{-1} and agree satisfactorily with values measured earlier.[16,17]

An ultralow oscillation frequency denoted by F_ε by us ($F_\varepsilon = (2.5 \pm 0.5)\times 10^4$ G) was recorded in the first measurement with specimen 2 at a pressure of 20.9 kbar. Samples of the experimental traces for two pressures are shown in Fig. 2b. In a subsequent experiment the pressure was raised to 23 kbar, but due to the worsening of the crystal perfection the amplitude of these oscillations decreased to the noise level, so that we were unable to determine the PD and m^*/m_0.

DISCUSSION OF THE RESULTS

The results given above show that the FS of cadmium undergoes qualitative changes in the pressure range 15–20 kbar. We ascribe the frequency F_δ, observed for $H \parallel [11\bar{2}0]$ to the transverse section of the joined horizontal arms of the monster. We calculated the critical pressure at which connection of the monster takes place from the value obtained for $d \ln F/dp$: $p_{\text{crit}} \approx 17$ kbar. Corresponding to this pressure, $c/a = 1.822$ (0 K).

Confirmation of such an interpretation is provided by the observation of large oscillations F_τ for $H \parallel [11\bar{2}0]$, which (as we interpret it) arise on deformation of the body of the monster and lengthening of its arms, which would lead to the appearance of extremal sections such as exist in zinc under normal conditions (orbit σ).[19] The magnitude of the area of the section S_σ in zinc is 7×10^{-2} a.u. and in cadmium $S_\tau = 6\times 10^{-2}$ a.u. corresponding to oscillation frequencies 26.5 and 22.9 MG (at $p = 17$ kbar).

In our view, the frequency of 27 MG observed in Cd at pressures above 20 kbar for $H \parallel [10\bar{1}0]$ corresponds to an oblique section of just this type. The smallness of the amplitude of these oscillations is explained by the fact that for such oblique sections the extremal of the orbit occurs at the boundary. In zinc the oscillation frequency from such an oblique section of the monster is 33 MG; there is thus appreciable geometrical similarity between sections σ in zinc and τ in cadmium. We do not see any alternative plausible interpretation for frequency τ.

According to the present model of the FS of cadmium, the frequency F_ε, recorded for $H \parallel [0001]$, can only be ascribed to the minimum transverse section arising at point K of the electron "needle" in the third zone. With the appearance of this part, the FS of cadmium becomes exactly similar to that of zinc. The question may then arise as to whether F_σ corresponds to the longitudinal section of this same ellipsoid. Such an interpretation cannot be considered plausible for the following reason. The amplitude A for the longitudinal section S_1 of the thin ellipsoid-needle should be about two orders of magnitude less than for the transverse S_c, since $A \sim (\partial^2 S_i/\partial k_z^2)^{-1/2}$. Related to this is the fact that the longitudinal section of the needle in zinc was not recorded by the modulation method but only by the torsional method.[20] In our experiment the amplitudes of oscillations δ were greater than for ε (see Fig. 2).

Proposals about the type and order of appearance of new parts of the FS of cadmium at an ETT have also been made elsewhere.[8,21] Grechnev and Svechkarev[21] studied the nonoscillatory part of the magnetic susceptibility χ of cadmium alloys. The observed anomalies in χ were treated as a consequence of topological transitions, including the joining of the arms of the monster at 5 kbar and the appearance of a needle at point K at $p \approx 30$ kbar. Makarov *et al.*,[8] who analyzed results on the dependence of T_c on the extension of cadmium whiskers,[22] concluded that the observed form of the departure of T_c from linearity is produced by the appearance of an electron needle in the FS. The results of the present work contradict the conclusions[8] on the change in FS for cadmium. We believe that this indicates that the accuracy of quantitative estimates[8] is insufficient to describe adequately the nature of the readjustment of the FS of cadmium on changing the crystal lattice parameters, or that the anomaly in T_c for cadmium[22] is not connected with an ETT.

CONCLUSIONS

Three new frequencies in the dHvA spectrum have been found on studying the FS of cadmium under pressure, two of them being ultralow: $\leqslant 10^5$ G. The latter arise at pressures above 17–18 kbar and correspond to sections of new parts of the FS of cadmium formed by the joining of the arms of the hole monster in the second zone and by the appearance of an electron needle in the third zone. An electron topological transition in metals has thus been found for the first time by a direct method and the FS of cadmium so formed has been determined.

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