Quantum oscillations of the thermoelectromotive force of osmium in strong magnetic fields

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The anisotropy and field dependence of the thermoelectromotive force of very pure osmium single crystals with $\rho_{273\,K}/\rho_{4.2\,K} \approx 2000$ were measured at a temperature of 4.2 K and in magnetic fields up to 90 kOe. Above 40 kOe, quantum oscillations of the thermoelectromotive force analogous to the Shubnikov-de Haas effect, which have not been observed earlier in transition metals, were detected.

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Osmium is a transition $5d$-metal with an atomic number $Z = 76$ and a hexagonal close-packed lattice. It is the electron analog of ruthenium and, according to a calculation of the electron energy spectrum,[1] it allows a magnetic breakdown (MB), similar to that in ruthenium. However, strong relativistic effects in osmium, which are difficult to calculate theoretically with an adequate degree of accuracy, can significantly influence its electron spectrum and alter the topology of the Fermi surface in the vicinity of the L point of the Brillouin zone compared to ruthenium (in ruthenium the magnetic breakdown is caused by a small energy gap of a spin-orbital origin between the multiply connected, hole surface KM8h and the lens L7h, centered at the L point). As a result, the possibility of a magnetic breakdown in osmium is nontrivial and interesting.

Therefore, we attempted to search for quantum oscillations of the galvanomagnetic and thermoelectric properties of osmium, primarily the thermo emf oscillations. In the experiments we used samples cut along the principal crystallographic directions from single crystals with $\rho_{273\,K}/\rho_{4.2\,K} \approx 2000$. The sample dimensions were $1 \times 1 \times 15$ mm. The measurements were made in fields up to 90 kOe and at a temperature of 4.2 K.

Figures 1 and 2 show the results of measurements of the magnetothermo emf α for an experimental geometry in which the temperature gradient $\nabla T \| \langle 1\bar{2}10\rangle$, and the magnetic field **H** rotates in a plane perpendicular to this axis. This geometry is based on the following considerations. First, such orientations of ∇T and **H** determine the Fermi surface in the hexagonal direction in the neighborhood of the L point. Secondly, large-amplitude quantum oscillations of the thermoelectromotive force can be observed in the case of such geometry and the existence of magnitude breakdown trajectory rearrangement.

Figure 1 shows the anisotropy of the thermoelectromotive force α and the magnetoresistance $\Delta\rho/\rho$ of an osmium sample with $\nabla T \| \langle 1\bar{2}10\rangle$. It can be seen that there are broad minima at $\mathbf{H} \| \langle 0001\rangle$ whose cause, according to the Fermi surface model,[1,2] is the geometrical decompensation due to the appearance of a layer of electron-type trajectories on the multiply connected KM8h sheet. At $\mathbf{H} \| \langle 10\bar{1}0\rangle$ a maximum in $|\alpha|$

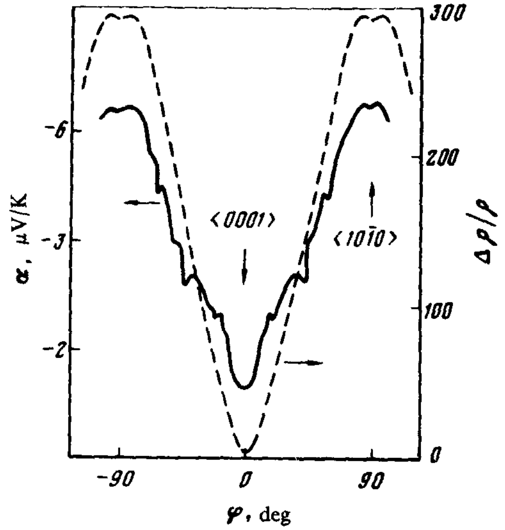

FIG. 1. Anisotropy of the thermoelectromotive force $\alpha(\phi)$ (solid line) and of the magnetoresistance $\Delta\rho/\rho(\phi)$ (dashed line) of an osmium single crystal for ∇T and j along the $\langle 1\bar{2}10\rangle$ axis, $T = 4.2$ K, $H = 86$ kOe.

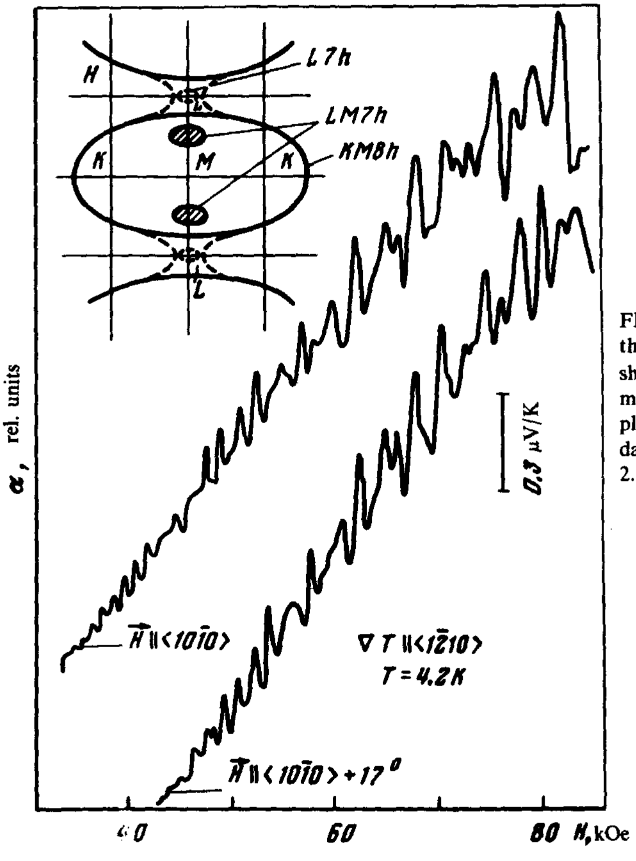

FIG. 2. Typical oscillations of the thermo emf of osmium. The inset shows the cut of the sheets of the Fermi surface of osmium by the LMK plane in the vicinity of the L point: dashed line—Ref. 1, solid line—Ref. 2.

can be observed, and the field dependence $\alpha(H)$ in strong magnetic fields is close to linear; according to Ref. 3, this indicates that there are closed trajectories for the given H direction. Measurements of the magnetoresistance of this sample also showed that for $\mathbf{H} \| \langle 10\bar{1}0 \rangle$ a maximum exists in the anisotropy of $\Delta\rho/\rho$ along with a quadratic dependence of $\Delta\rho/\rho(H)$ corresponding to it. This indicates that there are no closed trajectories along the $\langle 0001 \rangle$ axis, which was pointed out in Ref. 4.

As far as the field dependences $\alpha(H)$ are concerned, an oscillating contribution can be observed in a wide range of magnetic field directions for $H \gtrsim 40$ kOe (Fig. 2). This contribution contains several frequencies in the range of angles up to 70° from the $\langle 10\bar{1}0 \rangle$ axis. A Fourier analysis performed on a computer showed that the observed oscillations contain the fundamental frequencies [for example, for $\mathbf{H} \| \langle 10\bar{1}0 \rangle$ $f_1 = (1.44 \pm 0.03) \times 10^6$ kOe and $f_1 = (2.12 \pm 0.04) \times 10^6$ kOe], which coincide within an accuracy of 3% with the de Haas-van Alphen frequencies[2] from the LM7h hole ellipsoids located between the L and M points of the Brillouin zone (see inset in Fig. 2). These two frequencies agree with the symmetrical arrangement of the ellipsoids.

Generally, the absence of open trajectories (see dashed line in Fig. 2) along the $\langle 0001 \rangle$ direction which were predicted in Ref. 1, can be explained by their magnetic breakdown rearrangement into closed trajectories via the lens L7h. However, the experimental data cast doubt on the existence of magnetic breakdown in osmium for the following reasons: a) the observed oscillation frequencies, rather than coming from the L7h lenses (as is the case in ruthenium), correspond to the extreme cross sections of the LM7h hole ellipsoids, which, according to Ref. 1, are remote from the multiply connected KM8h hole surface; b) in contrast to ruthenium, it has no sharply pronounced anisotropy of the oscillation amplitudes which are an order of magnitude smaller; c) none of the anomalies characteristic of magnetic breakdown, which would indicate magnetic breakdown rearrangement of the electron trajectories, is observed in the field dependences of the thermo emf and magnetoresistance.

Thus, these experimental facts indicate that the observed oscillations of the thermo emf of osmium are apparently due to the Shubnikov-de Haas-type effect, rather than due to magnetic breakdown. This is probably due to the fact that the strong relativistic effects in osmium produce a significant change in the topology of its Fermi surface in the neighborhood of the L point of the Brillouin zone as compared with the calculation.[1]

In conclusion, we note that the thermo emf oscillations in the metals studied to date are caused by magnetic breakdown. So far as we know, osmium is the first transition metal in which quantum oscillations of the thermo emf, analogous to the Shubnikov-de Haas effect in magnetoresistance, have been observed.

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