Quantum oscillations of resistance and magnetization in the degenerate semiconductor *n*-$HgCr_2Se_4$

A. D. Balaev and V. A. Gavrichkov*)

L. V. Kirenskiĭ Institute of Physics, Siberian Branch of the Russian Academy of Sciences, 660036 Krasnoyarsk, Russia

S. G. Ovchinnikov

L. V. Kirenskiĭ Institute of Physics, Siberian Branch of the Russian Academy of Sciences, 660036 Krasnoyarsk, Russia; Krasnoyarsk State University, 660074 Krasnoyarsk, Russia

V. K. Chernov

Krasnoyarsk State University, 660074 Krasnoyarsk, Russia

T. G. Aminov and G. G. Shabunina

N. S. Kurnakov Institute of General and Inorganic Chemistry, Russian Academy of Sciences, 117907 Moscow, Russia

(Submitted 3 November 1997)
Zh. Éksp. Teor. Fiz. **113**, 1877–1882 (May 1998)

In the magnetic field range $\Delta H = 8-60$ kOe we observed and studied the anomalous oscillations in the magnetic field dependence of the resistance and magnetization of single crystals of *n*-$HgCr_2Se_4$. The absence of periodicity in $1/H$ in the $\Delta H = 8-20$ kOe range can be explained by the non-Fermi-liquid behavior of the electron subsystem and agrees with the theory of the de Haas–van Alphen in systems with intermediate valence. In stronger fields, $\Delta H = 20-60$ kOe, the amplitude of the fundamental harmonic decreases, with the number and amplitude of the higher-order harmonics increasing. As a result, noise is superimposed on the signal as magnetic field strength grows. The temperature dependence of the magnetization is the sum of the monotonic spin-wave contribution and the oscillating part. © *1998 American Institute of Physics.* [S1063-7761(98)02205-7]

1. INTRODUCTION

Landau oscillations, which arise because of quantization of electron orbits in a magnetic field, usually lead to oscillations (periodic in $1/H$) of the thermodynamic and kinetic characteristics of degenerate Fermi systems. If the dependence of the chemical potential μ on temperature and magnetic field differs from that in standard Fermi liquids, e.g., due to strong electron correlations, anomalous quantum oscillations may occur. In an earlier paper[1] we reported the detection of temperature oscillations of magnetization in single crystals of the degenerate magnetic semiconductor *n*-$HgCr_2Se_4$. In the present paper we give the results of observations of weakly periodic and aperiodic (in $1/H$) oscillations of the resistance R and magnetization M of the same samples. While observing temperature oscillations of magnetization required complicated multiparameter processing of experimental data,[1] oscillations in the field dependence of M and R are visible without any theoretical processing.

The ferromagnetic semiconductor *n*-$HgCr_2Se_4$ has a temperature-independent electron concentration n of roughly 10^{18}cm^{-3} in the temperature range $4.2 < T < 77$ K (in which the investigations took place) and a high carrier mobility $\sim 10^3\text{cm}^2\ \text{V}^{-1}\text{s}$ at 77 K, which makes it possible to observe Landau oscillations in fields up to 10 kOe. Non-Fermi-liquid

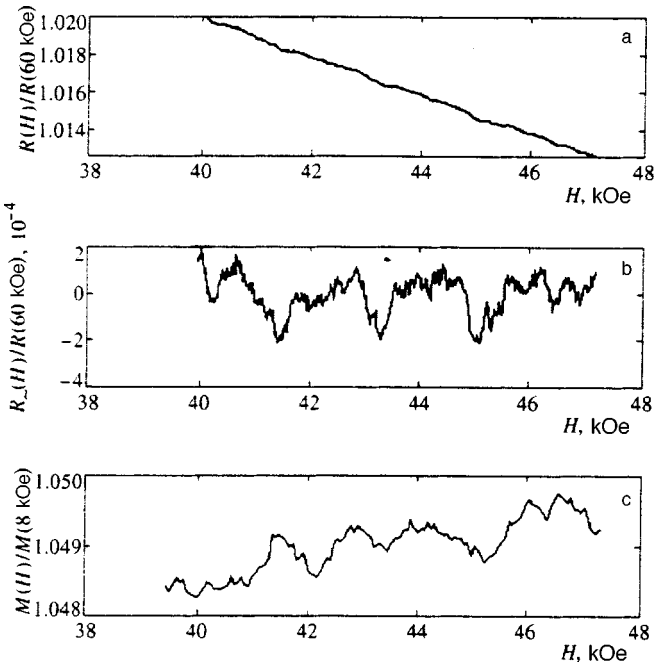

FIG. 1. Magnetic field dependence of the total resistance (a), the oscillating part of the resistance (b), and the relative magnetization (c) of the degenerate semiconductor *n*-$HgCr_2Se_4$ measured at $T = 4.2$ K.

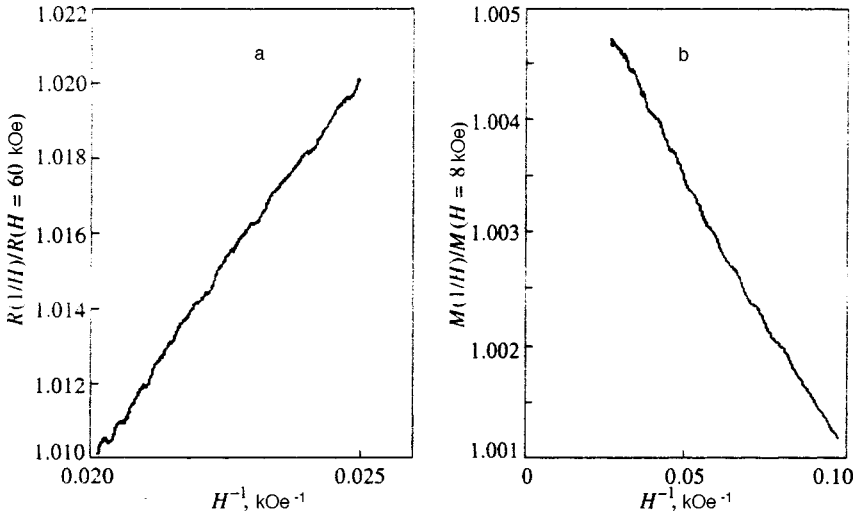

FIG. 2. Dependence of resistance (a) and magnetization (b) on the reciprocal magnetic field for n-$HgCr_2Se_4$ at $T=4.2$ K.

effects in $HgCr_2Se_4$ are due to the presence of a localized d-level of chromium Ω near the bottom of the conduction band.[2] In a degenerate n-type semiconductor, the chemical potential μ is pinned near the localized level and is weakly dependent on T and H, while the bottom of the conduction band is shifted in proportion to the magnetization $M(T,H)$. As a result the function $\mu(T,H)$ acquires non-Fermi-liquid corrections. Temperature oscillations of magnetization in such a multielectron model were predicted in Ref. 3, and after the necessary experimental work was done (see Ref. 1), a more comprehensive theory of the de Haas–van Alphen effect was developed in a recent paper by Val'kov and Dzebisashvili.[4] In the temperature interval $\mu_B \ll T \ll \hbar\omega_c$, realized because of the small effective carrier mass $m \sim 0.01 m_e$, the new theory yields a temperature and magnetic dependence of magnetization such that the chemical potential measured from the bottom of the conduction band can be written as[4]

$$\mu(T,H)=\mu(0)-\frac{J}{2}Z\left(\frac{3}{2}\right)t^{3/2}+Jt\sqrt{h}-\frac{35}{96\pi}J\sqrt{th}, \qquad (1)$$

where $Z(3/2)=2.612$, J is the $3d$-exchange integral, $t=T/4\pi IS$, and $h=\mu_B H/IS$, with I the parameter of exchange between the neighboring spins of a d-ion, and S the spin of the d-ion. Since the oscillating part of the thermodynamic potential and its derivatives is determined by the factor

$$\sin\left(2\pi k\frac{\tilde{\mu}}{\hbar\omega_c}\right)=\sin\left\{\frac{2\pi k}{\hbar\omega_c}\left[\mu(0)-\frac{J}{2}Z\left(\frac{3}{2}\right)t^{3/2}+Jt\sqrt{h}-\frac{35}{96\pi}J\sqrt{th}\right]\right\}, \qquad (2)$$

it is clear that the field dependence of the oscillation phase assumes the form

$$\varphi\sim\frac{a}{H}+\frac{b}{\sqrt{H}},$$

i.e., the periodicity of oscillations in $1/H$ is violated, although the oscillations remain.

2. FIELD DEPENDENCE OF MAGNETIZATION AND RESISTANCE

The magnetization of samples was measured by an automatic vibrating-reed magnetometer with a superconducting solenoid in fields up to 60 kOe (Ref. 5) at $T=4.2$ K. The magnetic field was applied along the $\langle 100\rangle$ axis of the crystal. Longitudinal magnetoresistance was measured by the four-contact method.

Figure 1 depicts the oscillating part $R_\sim$ of the resistance and the magnetization M as functions of the magnetic field strength, while Fig. 2 depicts the dependence of the resistance R on the reciprocal field strength. From Fig. 1a we see that a magnetoresistance linear in the field strength is superimposed on the oscillating part of the resistance, so we represent $R(H)$ in the form

$$R(H)=R_\sim(H)-cH, \qquad (3)$$

where $c=R(60\text{ kOe})\times 10^{-4}\text{kOe}^{-1}$ is a parameter. The oscillating part $R_\sim(H)$ defined in this manner is depicted in Fig. 1b. A comparison of the $R_\sim$ vs. H curve in Fig. 1b and the M vs. H curve in Fig. 1c shows that the extrema in the two curves $R_\sim(H)$ and $M(H)$ coincide. As expected, the oscillations are periodic neither in H nor in $1/H$. The spectral density of the signal depicted in Fig. 2b has a smeared peak corresponding to an approximate period in $1/H$ in 0.8×10^{-6}Oe. However, it is possible to detect a distinct signal only in the magnetic field range $\Delta H=8$–20 kOe (Fig. 3a). In stronger fields, e.g., in the range $\Delta H=20$–60 kOe, the amplitude of the fundamental harmonic decreases, with the number and amplitude of the higher-order harmonics increasing. As a result, noise is superimposed on the signal (Fig. 3b). Knowing the period, we calculated the area S of the extremal cross section of the Fermi surface and the carrier concentration n. The results were $S=9.3\times 10^{13}\text{cm}^{-2}$ and $n=4.3\times 10^{18}\text{cm}^{-3}$.

The relative amplitudes of the oscillations are moderate, $\sim 10^{-4}$, but they are larger than the magnetization measurement errors by a factor of approximately ten.[5] The smallness of the magnetization oscillation amplitude can be explained by the smallness of the carrier concentration, since at n

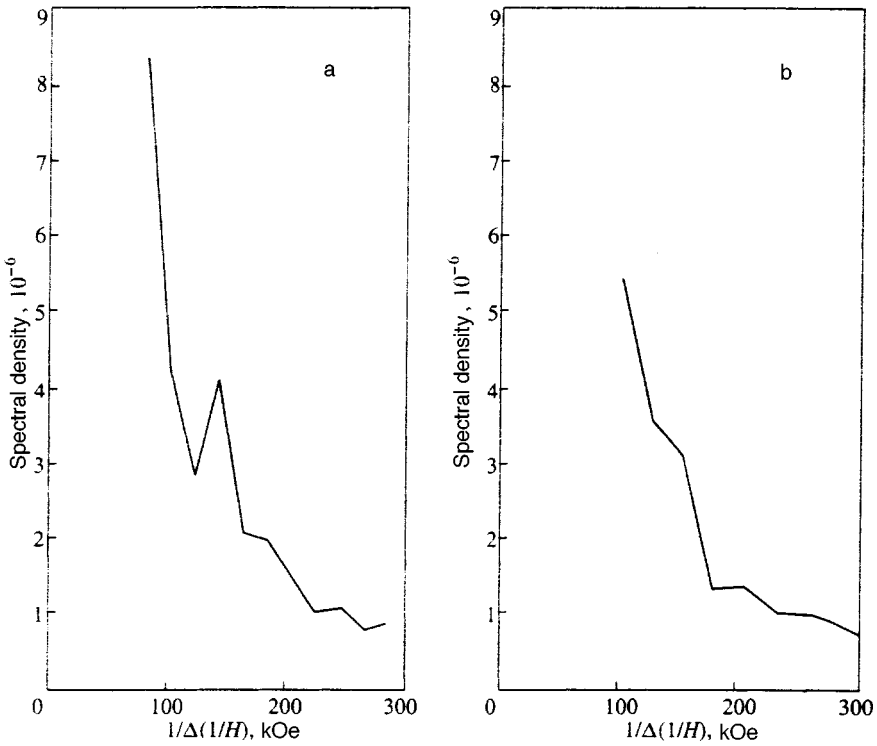

FIG. 3. Spectral density of magnetization $M(1/H)$ in magnetic field ranges $\Delta H = 8-20$ kOe (a) and 20–60 kOe (b).

$\sim 10^{18}\text{cm}^{-3}$ the number of electrons per cell is roughly 10^{-4}. The relative amplitude of resistance oscillations is approximately five times larger than the magnetization oscillation amplitude.

3. TEMPERATURE DEPENDENCE OF MAGNETIZATION AND RESISTANCE

Formula (2) shows that temperature variations cause large shifts in the chemical potential and intersections with the Landau levels, which gives rise to oscillations in the temperature dependence of the magnetization and resistance. Since the oscillation phase is a nonlinear function of temperature, the oscillations are aperiodic in T. Here, however, the damping of the oscillation amplitude with increasing temperature makes observation of a large number of temperature oscillations difficult.

The other fact that sets temperature oscillations apart from field oscillations is that temperature oscillations are masked by a complicated temperature dependence of both magnetization and resistance. Hence to identify the oscillation contribution one must subtract the monotonic parts. For instance, for the average spin the spin-wave theory yields[6]

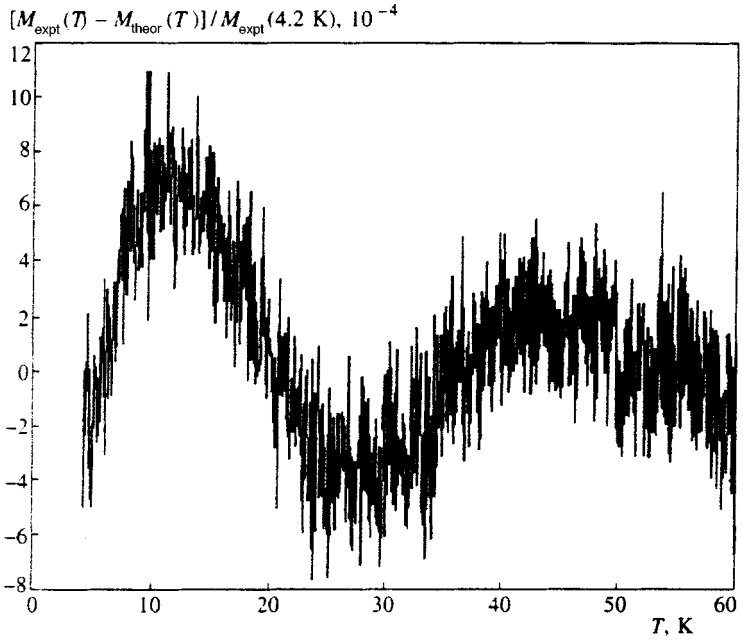

FIG. 4. Difference of the experimental curve $M_{expt}(T)$ for n-$HgCr_2Se_4$ and the theoretical curve $M_{theor}(T)$ expressed in the spin-wave approximation as a function of temperature for a field of 60 kOe.

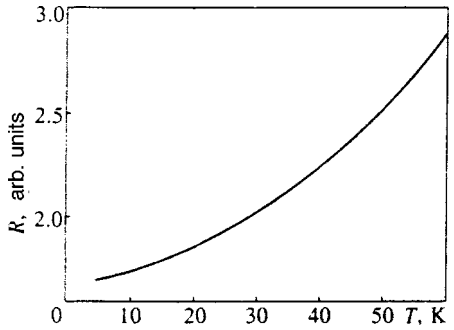

FIG. 5. Temperature dependence of the resistance of n-$HgCr_2Se_4$ in a magnetic field of 60 kOe.

$$\langle S^z\rangle_{SW}(T)=\frac{3}{2}-at^{3/2}Z_{3/2}(x)-bt^{5/2}Z_{5/2}(x)-ct^{7/2}Z_{7/2}(x), \tag{4}$$

where $x=2\mu_B H/kT$, and $Z_p(x)$ is the generalized Riemann zeta function,

$$Z_p(x)=\sum_{n=1}^{\infty}\frac{\exp(-nx)}{n^p}.$$

The parameters a, b, and c were found by fitting the results to the experimental curve $M_{\text{expt}}(T)$ measured in a field $H=60$ kOe via the simplex method. The values are $a=0.8499$, $d=-0.5545$, and $c=0.1294$.

The difference of the measured curve $M_{\text{expt}}(T)$ and the theoretical curve $M_{\text{theor}}(T)$ determined via (4) is depicted in Fig. 4. Thus, the total temperature dependence of the magnetization can be written as the sum of the monotonic curve (4) and the oscillating part.

The temperature dependence of the electrical resistance measured in $H=60$ kOe is depicted in Fig. 5. As in the case of the $M_{\text{expt}}(T)$ curve, to identify the oscillations we must subtract the monotonic temperature dependence, which is not related to Landau quantization. Although the various mechanisms of the temperature dependence of the electrical resistance of magnetic semiconductors have been thoroughly studied,[7] quantitative comparison with experimental results requires special calculations that allow for the behavior of the band structure of n-$HgCr_2Se_3$. Such calculations are outside the scope of the present paper.

5. CONCLUSION

In this paper we have shown that the Shubnikov–de Haas and de Haas–van Alphen effects in the degenerate semiconductor n-$HgCr_2Se_3$ can be observed in magnetic fields of the 8–60 kOe range but that their field dependence is not described by functions periodic in $1/H$, in contrast to the case of an ordinary Fermi liquid. The non-Fermi-liquid nature of the temperature dependence of the chemical potential also gives rise to quantum temperature oscillations of magnetization.

This work was made possible by a grant from the Russian Fund for Fundamental Research (Grant No. 96-02-16075).

*)E-mail: gav@iph.krasnoyarsk.su

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Translated by Eugene Yankovsky