Properties of the Shubnikov–de Haas effect in HgMnSe

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(Submitted 24 January 1983; resubmitted 31 March 1983)
Zh. Eksp. Teor. Fiz. **58**, 1638–1646 (November 1983)

The transverse magnetoresistance of HgMnSe crystals in stationary (up to 6 T) and pulsed (up to 33 T) magnetic fields is studied in the temperature range 1.7–77 K. The electron concentration in the samples amounts to $n = 10^{17}$–10^{18} cm^{-3}. The Shubnikov–de Haas (SdH) oscillations reveal certain properties that are accounted for by the effect of the exchange interaction on the energy spectrum of the conduction-band electrons. The value of the exchange integral is obtained.

PACS numbers: 72.20.My, 72.80.Ga

Crystals of $Hg_{1-x}Mn_xSe$ belong to the class of semimagnetic semiconductors and represent solid solutions of II–VI compounds, one of the components of which is the gapless semiconductor HgSe. Replacement of Hg atoms by Mn atoms leads to a smooth rearrangement of the band spectrum. In particular, the energy gap E_g between the bands Γ_8 and Γ_6 changes linearly with the content of manganese x in the following fashion: $E_g(x) = (-270 + 44x)$ meV (Ref. 1), i.e., similar to the case in which other elements of the II group (Zn, Cd) serve as the substitution atoms. However, in contrast to these latter atoms, there is an unfilled Mn shell with an uncompensated spin. The exchange interaction of the conduction electron with the electrons of Mn add to the energy a contribution which obviously depends on the state of the magnetic subsystem and is proportional to the mean value of the magnetic moment of the Mn atoms. At small values $x \lesssim 0.01$, the exchange interaction between the d electrons of the neighboring Mn atoms is insignificant and the system can be considered as paramagnetic with mean value of the spin

$$\langle S_z \rangle = B_{5/2}(\bar{g}\mu B/kT) \quad (1)$$

(μ is the Bohr magneton, $\mu = e\hbar/2m_0c$, $\bar{g} = 2$ for d electrons, $B_{5/2}$ is the Brillouin function). In gapless semiconductors of the HgSe type the effective mass of the electrons is $m \sim 10^{-2}\, m_0$ in order of magnitude and, consequently, the cyclotron energy $\hbar\omega \gg g\mu B$. Therefore the magnetic fields in which ordering of the d electrons and $\langle S_z \rangle$ differ significantly from zero, i.e., $\bar{g}\mu B \sim kT$ are certainly quantizing for the conduction electrons. Thus the role of the exchange interaction should be clearly manifest in the quantum galvanomagnetic effects.

The first measurements of the Shubnikov–de Haas effect (SdH) in HgMnSe in constant magnetic fields up to 7 T were carried out in Ref. 1, and in magnetic fields up to 30 T by the authors of Ref. 2. In these researches, the basic band parameters of HgMnSe were determined. The aim of the present work was to make clear the features of the SdH oscillations that are connected with the presence of uncompensated magnetic moments of the manganese ion.

We investigated the SdH oscillations in samples of $Hg_{1-x}Mn_xSe$ ($x = 1.3\times10^{-2}$) in stationary (up to 5 T) and pulsed (up to 30 T) magnetic fields, over a wide range of temperatures 1.7–77 K. The concentrations of the electrons in the samples studied amounted to $n = 3.5\times10^{17}$–2×10^{18} cm^{-3}. The values of n obtained from measurements of the Hall constant and calculated from the period of the SdH oscillations were practically identical. Typical values of the mobility $u \approx 7\times10^4$ cm^3/W-s. The good quality of the oscillation pictures obtained should be noted; the large number of resolved peaks (more than 20 at low temperatures) and the smooth field dependence of the monotonic part of the resistance. This indicates a high degree of homogeneity of the investigated crystals and small values of the Dingle temperature that characterizes the collision broadening of the Landau levels. Estimates of the Dingle temperature $kT_D = \hbar/\pi\tau$ give $T_d \lesssim 2$K.

In the study of SdH oscillations the following anomalies were observed, particularly for semimagnetic semiconductors, and did not appear in the SdH effect in the usual semiconductors.

1. In the quasiclassical region of magnetic fields $\hbar\omega \ll E_F$, where the oscillations are governed by the Landau levels with large numbers N, the harmonic picture of the oscillations is destroyed, beats are observed, and nodes appear at certain fields B_y (Fig. 1). The locations of the nodes B_y shift monotonically toward smaller fields with increase in the temperature.

2. The oscillations are periodic in the reciprocal field both at $B < B_y$ and at $B > B_y$, except for a small region of the

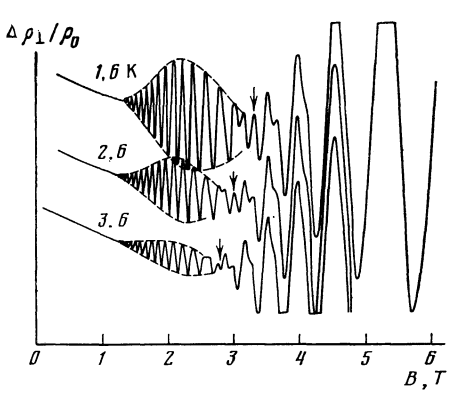

FIG. 1. Formation of nodes of oscillation (shown by arrows) at various temperatures ($n = 1\times10^{18}$ cm^{-3}).

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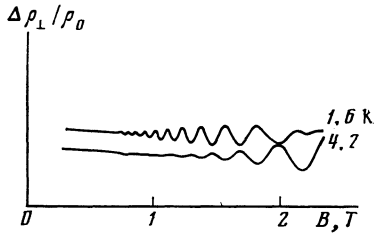

FIG. 2. Effect of the temperature on the phase of the oscillations of the transverse magnetoresistance ($n = 3\times10^{17}$ cm^{-3}).

order of several kG in the neighborhood of B_y; however, the phase of the oscillations is reversed: the maxima in the region $B < B_y$ correspond to the minima in the region $B > B_y$. This can be established if we plot the dependence of the locations of the extrema on integers. This is seen directly from Fig. 2, where the oscillating part of the magnetoresistance is shown for $T = 1.6$ K ($B_y = 2.4$ T) and $T = 4.2$ K ($B_y = 1.2$ T).

3. The amplitudes of the oscillating peaks reveal a strongly nonmonotonic dependence on the temperature. Figure 3 shows an example of such a dependence of the peak in the field $B = 1.8$ T. The detailed temperature dependence of the amplitude of the SdH oscillations were first observed by the authors of Ref. 3 in HgMnTe crystals. The anomalies 2 and 3 noted above are a direct consequence of the presence of the nodes. Obviously, the similar character of the change in the amplitude of the oscillations with temperature makes impossible the traditional method of determination of the effective mass of the electron from the temperature dependence of the amplitude at a fixed magnetic field. Figure 4 graphically illustrates the fact that the ratio of the amplitudes at two temperatures A (1.7 K)/A (4.2 K) can take on values that are both larger and smaller than unity for different oscillating peaks. The latter case corresponds formally to negative effective mass.

4. The first maximum in the resistance from, the side of the quantum limit (usually denoted as the 0^- peak for semiconductors with negative value of the g factor) is shifted significantly toward higher fields with increase in the temperature (Fig. 5). The frequently observed shift of this peak in ordinary semiconductors takes place, on the contrary, always toward the opposite side and is due to the partial lifting of the degeneracy of the electron gas.

5. The third and succeeding peaks split with increase in the temperature, and this splitting increases with increase in the temperature over the entire interval in which it is possi-

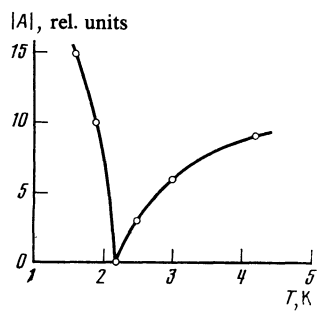

FIG. 3. Temperature dependence of the amplitude of the oscillation peak located in a field $B = 1.8$ T ($n = 3\times10^{17}$ cm^{-3}).

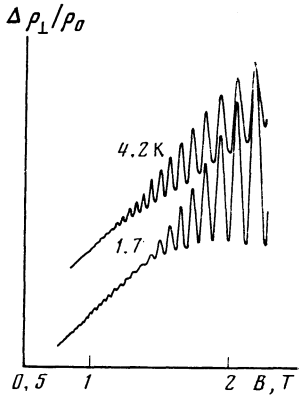

FIG. 4. Curves of the magnetoresistance for samples with $n = 1\times10^{18}$ cm^{-3} at $T = 1.7$ K and $T = 4.2$ K.

ble to resolve the oscillations (Fig. 6). Undoubtedly, the similar doublet structure of the oscillating peaks is connected with the spin splitting of the Landau levels. However, the spin splitting usually observed in other semiconductors becomes smeared out with increase in the temperature: along with a decrease in the amplitude of the oscillations, there takes place an approach toward each other of the peaks of the oscillating doublet and their broadening up to the complete disappearance of the splitting.

The features that have been noted can be understood if we take into account the increment due to exchange interaction, of the spectrum of the conduction electrons. For simplification, we shall neglect the nonparabolic character of the band Γ_8 but shall use the mass of the electrons on the Fermi level for the cyclotron energy $\hbar\omega$. This mass is calculated in the two-band approximation:

$$m = m_n(1+2E_F/|E_g|),$$
$$m_n = 3\hbar^2|E_g|/4P^2$$

is the effective mass at the bottom of the band, and P is the matrix element of the Kane theory. Using the molecular field approximation in the calculation of the exchange contribution to the eneergy of the electron, the authors of Refs. 4 and 5 obtained the following expression for the spin-split Landau levels:

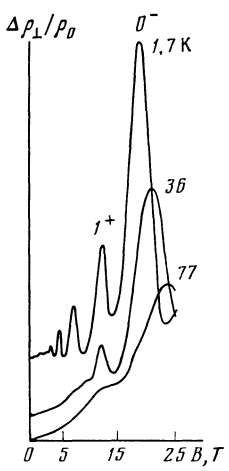

FIG. 5. Shift of the 0^- peak with temperature for a sample with $n = 3\times10^{17}$ cm^{-3}.

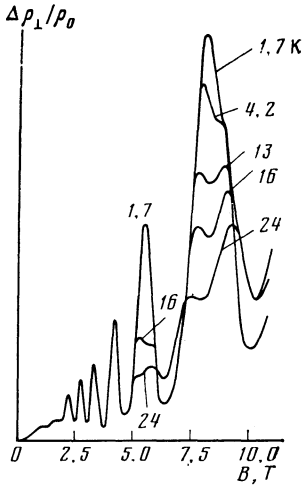

FIG. 6. Effect of the temperature on the spin splitting of the oscillation maxima ($n = 3\times10^{17}$ cm^{-3}).

$$E_N^\sigma=(N+{}^1/_2)\hbar\omega+{}^1/_2\sigma g\mu B+\Delta_N^\sigma,$$
$$N=0, 1, 2, \ldots, \quad \sigma=\pm1, \tag{2}$$
$$\Delta_N^\sigma=J_0x\langle S_z\rangle\begin{cases}\dfrac{8N-1}{8N+2}, & \sigma=+1\\ -\dfrac{9+8N}{8N+6} & \sigma=-1\end{cases}, \tag{3}$$

where J_0 is a parameter characterizing the exchange interaction, g is the g_0-factor of the conduction electrons. At $N = 0$, the sign of the exchange contribution is the same for $\sigma = \pm 1$; at $N\geqslant1$, the signs are opposite. The spin-splitting of the Landau levels increases when account is taken of the exchange if the signs of g and J are the same, and decreases in the opposite case. We recall that according to the Kane model $g<0$ for electrons of the Γ_6 zone. At $N\gg1$, the exchange term Δ_N^σ does not depend on N and leads simply to the renormalization of the g factor, which becomes a function of the magnetic field and the temperature. Equation (2) is conveniently rewritten in the form

$$E_N^\sigma=(N+{}^1/_2+\sigma\nu^*/2), \quad N\gg1, \tag{2a}$$

where we have introduced the parameter

$$\nu^*=\nu+2J_0x\langle S_z\rangle/\hbar\omega, \quad \nu=gm/2m_0, \tag{4}$$

which is equal to the ratio of the value of the energy of the spin splitting to the value of the cyclotron energy. The location of the 0^- peak of the transverse magnetoresistance is connected with the parameter ν^* in the following fashion

$$B_0^-=\frac{\hbar c}{e}\left(\frac{2\pi^4n^2}{|\nu^*|}\right)^{1/3}.$$

In this equation, we have omitted the temperature increment, due to the incomplete degeneracy of the electron gas, because it turns out to be sufficiently small over the entire range of temperatures $T\lesssim60$ K. The experimentally observed shift of the 0^- peak in the region of large fields is connected only with the decrease in the parameter $|\nu^*|$ since the concentration of the electrons n does not change with the temperature. Since $\langle S_z\rangle$ falls off with increase in the temperature, we can state that $J_0<0$, since only in this case will $|\nu^*|$ be a monotonically decreasing function of T.

We shall now analyze the oscillation picture in the region of small fields, where no spin-split peaks are observed. In the limit $N\gg1$, the expression for the oscillating part of the magnetoresistance has the form

$$\frac{\Delta\rho}{\rho_0}=\sum_{r=1}^{\infty}A_r\cos\left(2\pi r\frac{E_F}{\hbar\omega}-\frac{\pi}{4}\right), \tag{5}$$

where

$$A_r=\left(\frac{\hbar\omega}{2E_F}\right)^{1/2}\frac{(-1)^rx}{r^{1/2}\,\mathrm{sh}\,(xr)}\exp\left(-\frac{2\pi^2rkT_D}{\hbar\omega}\right)\cos(\pi\nu^*r), \tag{6}$$
$$x=2\pi^2kT/\hbar\omega.$$

The appearance of the factor $\cos(\pi\nu^*r)$ in (6) can be made clear in the following way. The electrons of both systems of spin sublevels $\sigma = \pm 1$ make an additive contribution to $\Delta\rho$; this contribution can be represented in the form of a sum of harmonics with the same period but shifted in phase: $\cos(2\pi rE_F/\hbar\omega \pm \pi\nu^*r - \pi/4)$. If ν^* does not depend on the field, as in ordinary semiconductors at $J_0 = 0$, the superposition of each pair of harmonics with a given value of r leads to a decrease in the total amplitude of the oscillations A_r. In particular, at $\nu^* = 1/2$, the amplitudes of the fundamental and all the odd harmonics are identically equal to zero, which manifests itself in the doubling of the period of the oscillations (the so-called "spin damping").

In the case in which ν^* changes slightly with field over the interval of the periodicity of the function $\cos(2\pi E_F/\hbar\omega)$, the factor $\cos(\pi\nu^*)$ modulates the amplitude of the oscillations and describes the beats of the fundamental with $r = 1$. In fields B_y satisfying the condition

$$\nu^*=(2l+1)/2, \quad l=0, 1, 2, \ldots, \tag{7}$$

the amplitude of the fundamental vanishes. However, in this case the amplitude of the second harmonic ($r = 2$) is a maximum. Therefore, strictly speaking, we do not observe a distinct node of the oscillations, while in a certain vicinity of the point $B = B_y$ the oscillation pictures vanishes, as is in the decrease of the amplitude, in the loss of periodicity and in the appearance of additional peaks.

The parameter ν^* decreases monotonically both with the magnetic field and with the temperature. At a fixed temperature, in weak fields, when $\bar g\mu B\ll kT$, the parameter ν^* does not depend on the field and reaches its maximum value corresponding to the given temperature:

$$\nu^*=\nu+\frac{35}{12}\frac{J_0x}{kT}\frac{m}{m_0}. \tag{8}$$

In the limit $\bar g\mu B\ll kT$ the Brillouin function is close to its maximum value $B_{5/2}(\infty) = 2.5$ and ν^* falls off as B^{-1}, i.e.,

$$\nu^*=\nu+5J_0x/\hbar\omega. \tag{9}$$

It follows from the properties of the Brillouin function that the field B_y falls off monotonically with the temperature and vanishes at some temperature that can be determined from (7) and (8).

In principle, the appearance of a whole series of nodes on the oscillation picture is possible, corresponding to different $l = 0, 1, 2$, and so on in Eq. (7). However, estimates for typical values of the parameter (for example, we have taken

the parameters[1] of HgMnSe) show that the interval between the neighboring nodes amounts to $\Delta B_y \gtrsim 1$ T. We observed the node with $l = 1$ on all the samples studied. The nodes for $l = 2, 3$ lie in the region of such weak fields that the oscillations are practically unresolved. However, for one of the samples with concentration $n = 1\times10^{18}$ cm^{-3}, we succeeded in distinguishing the second node ($l = 2$) in the interval $T = 1.7$–2.4 K. On the other hand, the node with $l = 0$ should be located in the region of very strong fields, close to the quantum limit, where the description of the oscillations with the help of the harmonic formulas loses meaning, because, with increase in the field, the comparable contribution to Eq. (5) is made by harmonics with large r. This is clearly manifest by the appearance of spin splitting of the peaks.

As noted above, the splitting of the peaks has an unusual character: its value increases upon increase in the temperature. Formally, this means that the renormalized g factor or the parameter $|\nu^*|$ increases with temperature. However, as follows from (8), $|\nu^*(T)|$ actually decreases. This apparent contradiction is easily understood if we establish the correct correspondence of the oscillation peaks with the Landau levels. It is simplest to begin the identification with the region of rather high temperatures, when the contribution of the exchange interaction to the parameter ν^* is small. The first peak (from the side of the ultraquantum limit) corresponds to the E_0^- Landau level, the next, to the E_1^+ level, and so forth. By tracking the temperature shift of each peak, we can establish the fact that in the range of temperatures $T < 4$ K, the peak observed at $B = 7.7$ T is due to superposition of the Landau levels E_2^+ and E_1^-. This corresponds to the value $|\nu^*| = 1$, as follows from (2a). In the region of small fields, $B < 7.7$ T, we have $|\nu^*| > 1$, i.e., the E_{N+1}^+ levels are located below the E_N^- level. Obviously, a situation is possible with a superposition of the levels $E_{N+m}^+ = E_{N-}$ with $m = 2, 3, \ldots$, for $|\nu^*| = 2, 3, \ldots$, respectively. With increase in the temperature, such superpositions should be destroyed, which leads to a splitting of the oscillation peaks. However, in the samples investigated by us, in the range of fields in which $|\nu^*| = 2$, the oscillations of the magnetoresistance are described by the fundamental $r = 1$ of the series (5) and therefore it has not been possible for us to observe splitting due to the temperature dependence of $\nu^*(T)$.

Identification of the oscillation peaks enables us to determine correctly the factor $|\nu^*|$ from the location of the maxima of B_1^+ and B_1^- with the help of the usual formulas (see, for example, Ref. 7), taking into account the dependence of the Fermi energy on the magnetic field and the corrections for incomplete degeneracy. The dependence of $|\nu^+(T)|$ thus obtained turns out to be monotonically decreasing, which agrees qualitatively with the behavior of the Brillouin function (1). We note that even at the highest temperatures ($T \approx 30$ K) at which splittings of the peaks $B_1^\pm$ were still observed, the values of $|\nu^*|$ are significantly higher than for the HgSe crystals with about the same concentration of electrons.[7] This shows that the contribution of the exchange interaction to the value of $|\nu^*|$ remains significant even at very high temperatures.

In order to estimate the exchange-interaction parameter J_0, we have used the fact that at $B = 7.7$ T and at low temperatures, $T < 4$ K, superposition of the levels E_2^+ and E_1^- takes place in a sample of $Hg_{1-x}M_xSe$ ($x = 1.3\times10^{-2}$) with concentration of electrons $n = 3\times10^{17}$ cm^{-3}. As is seen from (4), the parameter ν^* depends on the ratio of the exchange energy J_0 to the cyclotron energy $\hbar\omega = \hbar eB/mc$. The experimental determination of the mass of the electrons at the Fermi level, m, from the temperature dependence of the amplitude of the oscillations turns out to be impossible for reasons that were discussed above. Therefore, we have calculated the value of m from the formulas of the two-band Kane model, taking for the matrix element p the value $P = 7.2\times10^{-8}$ eV-cm, as in HgSe,[7] and using the $E_g(x)$ dependence given in Ref. 1. For a small content of Mn, $x \approx 1\%$, the Luttinger parameters that describe the contribution of the remote bands, do not differ significantly in the crystals of HgSe and $Hg_{1-x}Mn_xSe$. Therefore we have taken the same value of the g factor of the electrons as in HgSe with $n = 3\times10^{17}$ cm^{-3}, i.e., $g = -15$. Moreover, we have assumed that the mean value $\langle S_z \rangle$ is determined by Eq. (1), i.e., we have not taken into account the possibility of the formation of any magnetic order or nonuniform distribution in the system of magnetic Mn ions. At a content $x > 1\%$, these effects can be very important, as the investigation of the magnetic susceptibility shows.[8] The value that we obtained amounts to

$$|J_0| = (0{,}28\pm0{.}03)\ \text{eV}. \tag{10}$$

The accuracy of the estimate of the parameter J_0 under the reservations made relative to the values of m and g used is limited principally by the accuracy of the determination of the field B at which a strict superposition of the Landau levels E_2^+ and E_1^- takes place, i.e., by the halfwidth ΔB of the oscillation peak, which at low temperatures is equal to $\Delta B = 0.7$ T.

The oscillation peak, which is located at $T = 1.7$ K in a field $B = 5.6$ T, is split only in the range of temperatures $T > 15$ K (Fig. 6). It is due to the intersection of the Landau levels with energies E_2^- and E_3^+ and the Fermi level $E_F(B)$. Calculation shows that at a value $|J_0| = 0.28$ eV, strict superposition of the levels $E_2^- = E_3^+$ in a field $B = 5.6$ T is achieved at a temperature $T = 8.5$ K. At lower temperatures, inversion of the levels occurs: $E_3^+ < E_2^-$. However, even at the very low temperature $T = 1.7$ K, when the exchange contribution to the energy reaches its maximum value, the values of the fields calculated from the condition $E_N^\pm = E_F(B)$ differ little: $B_2^- = 5.3$ T and $B_3^+ = 5.8$ T. The difference $|B_2^- - B_3^+|$ turns out to be less than or of the order of magnitude of the halfwidth of the oscillation peak in the range of temperatures $1.7 < T < 15$ K.

Attempts to determine the parameter of exchange interaction were undertaken earlier in Refs. 1 and 2. The authors, using the procedure of matching of parameters, calculated the dependence of the Landau levels $E_N^\pm$ on B and T, and then determined the locations of the oscillation maxima from the condition $E_N^\pm = E_F$. The dependence of the Fermi level on the magnetic field was not taken into account here

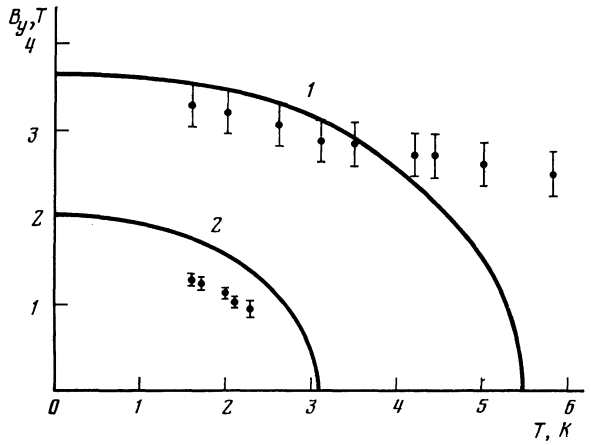

FIG. 7. Temperature dependence of the position of the nodes of oscillation of B_y. The solid curves are the result of calculation (1—for $l = 1$; 2—for $l = 2$), the points are the experimental values for a sample with $n = 1 \times 10^{18}$ cm^{-3}.

and the value of E_F at $B = 0$ was used. As a criterion for the correct choice of the parameters, they used the coincidence of the calculated and the experimentally observed positions of the peaks. Such a procedure seems unsatisfactory to us. We first note that in the region of large quantum numbers $N \gg 1$, where $E_F = \text{const}$, the oscillation peaks are not split, while the locations of the maxima, as also the period of the oscillations, are actually determined only by the concentrations of the electrons and are not sensitive to the band parameters (the effective mass m and the g factor). On the other hand, in the region of fields that are close to the quantum limit, account of the dependence of $E_F(B)$ is quite necessary: the $E_F(B)$ dependence, without significantly distorting the periodicity, has a significant effect on the location of the oscillation peaks.

Using the value that we have found for the exchange energy (10), we have calculated the temperature dependence of the locations of the nodes of the oscillations at $|\nu^*| = 3/2$ ($l \neq 1$) and $|\nu^*| = 5/2$ ($l = 2$) for a sample with concentration

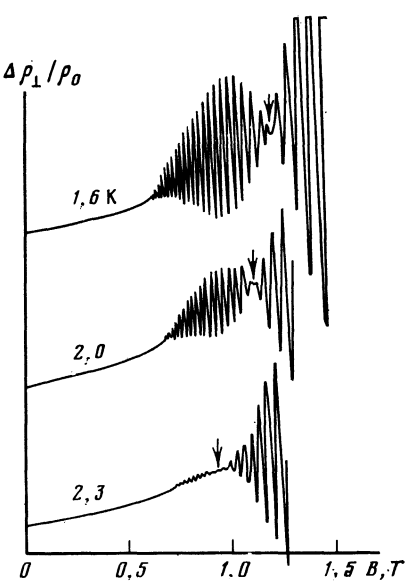

FIG. 8.

$n = 1 \times 10^{18}$ cm^{-3}. These dependences are shown in Fig. 7. Also shown are the segments corresponding to the experimentally observed locations of the nodes. The exact position of a node could not be determined experimentally because of the contribution of the higher harmonics. As is seen from Fig. 7, the values of the fields where the nodes are observed, and the general tendency toward a decrease in B_y with temperature are in agreement with the calculated curve.

At temperatures $T > 2.4$ K, the oscillations in the region of magnetic fields $B < 1$ T (the node with $l = 2$) are not resolved even at maximum sensitivity of our experimental apparatus (Fig. 8). The node corresponding to $l = 1$ is reliably fixed, up to temperatures $T = 6$ K ($B_y = 2.4$ T). Upon further increase in the temperature, the clearly pronounced modulation of the amplitude of the oscillation peaks disappears. However, in the region of fields $B = 2.2$–2.8 T, a destruction is again observed of the harmonic picture of the oscillations (loss of periodicity, distorted shape of the oscillation peaks), which is undoubtedly connected with the anomalously small amplitude of the fundamental in these fields. Thus, the experimentally investigated shift of the node with temperature does not agree with that calculated theoretically on the basis of Eq. (1).

The results of Ref. 8, where the magnetic susceptibility of $Hg_{1-x}Mn_xSe$ crystals were studied, show that the value of $\langle S_z \rangle$ for samples with $x > 0.011$ increases with field and falls off with the temperature; however, it cannot be described by the dependence $\langle S_z \rangle = f(B/T)$. Thus, the value of saturation as $B \to \infty$ is less than the theoretical $B_{5/2}(\infty) = 5/2$, and the law for the approach to the limiting value $\langle S_z \rangle_{\text{sat}} - \langle S_z \rangle$ is much smoother than for the function $B_{5/2}$ (in the latter case this law is an exponential one). Such a behavior of $\langle S_z \rangle$ corresponds qualitatively to the shift of the node with temperature at $\bar{g}\mu B/kT > 2$ observed above; even upon satisfaction of the latter inequality, the location of the node is not stabilized, but changes appreciably with decrease in the temperature.

Analysis of the dependence of the parameter $\nu^*(4)$ on the temperature from the position of the $B_1^{\pm}$ peaks also leads to the conclusion that the Brillouin function $B_{5/2}(\bar{g}\mu B/kT)$ gives a poor description of the behavior of $\langle S_z \rangle$ at large values of the argument. This can be seen immediately from Fig. 6. The superposition of the peaks B_2^- and B_1^+ takes place at $T = 1.7$ K ($\bar{g}\mu B/kT = 6$), but it is clearly destroyed even at $T = 4.2$ K ($\bar{g}\mu B/kT = 2.5$) although the Brillouin function at such values of the argument differs from the limiting value by less than 4%. This shows the necessity of use of a more accurate formula for the description of the magnetic moment of Mn ions in solid solutions $Hg_{1-x}Mn_xSe$ at $x > 0.01$ than is given by Eq. (1).

The authors express their deep gratitude to I. M. Tsidil'kovskiĭ for interest in the research and discussion of its results.

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Translated by R. T. Beyer