Spin-dependent scattering of conduction electrons in diluted magnetic semiconductors: $Hg_{1-x}Fe_xSe$

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Analysis of the quantum oscillations in the transverse magnetoresistance (Shubnikov—de Haas effect) for $\mathbf{H}||[110]$ in oriented single crystals of the compound semiconductor $Hg_{1-x}Fe_xSe$ with $x\simeq 0.03$ and $x\simeq 0.05$ provide evidence for spin-dependent scattering of the conduction electrons. Line-shape fits to the data allow estimates for the difference Dingle temperature δT_D and the exchange-enhanced effective g factor as a function of temperature and magnetic field. It is concluded that some Fe ions, when incorporated in a HgSe host, exist in a magnetically active state. This result indicates the presence of Fe^{3+} in addition to the Fe^{2+} that might normally be expected in this material.

I. INTRODUCTION

Diluted magnetic semiconductors (DMS's) are semiconducting compound materials whose crystal lattice contains substitutional magnetic ions. These ternary compounds crystalize in the zinc-blende structure with a lattice constant determined by the concentration of magnetic ions. Recently, there has been considerable interest in these materials due to their novel electrical, optical, and magnetic properties.[1–7] The interaction effects between magnetic moments have been studied and there is evidence that in some systems a spin-glass ordering of the magnetic moments results.[8] Other recent work on these materials has included measurements of Shubnikov—de Haas oscillations,[9–12] magnetization measurements,[13,14] specific heat,[14] far-infrared magnetotransmision studies,[15,16] and synchrotron-radiation photoemission measurements.[17]

In general, when a material contains magnetic impurities which possess local magnetic moments, the exchange interaction between these local moments and the conduction electrons produces a shift in the electronic energy levels. Because this interaction is spin dependent, there is reason to expect the scattering lifetimes of the spin-up and spin-down electrons will be unequal. In addition, the exchange interaction may influence the shape of the energy bands at the Fermi level. In order to investigate these spin-dependent effects, it is necessary to use a sensitive probe of the exchange interaction. The quantum oscillatory effects are ideal for this purpose since they provide considerable information about the shape of the band structure at the Fermi energy. Also, careful measurements of the harmonic content of these oscillations are capable of giving information about the effective mass, the cyclotron-averaged electronic g factor, and the difference in the scattering lifetime between the up- and down-spin conduction electrons.

In this study we report measurements of the Shubnikov—de Haas (SdH) effect in single crystals of HgSe ($n=3.82\times10^{18}$ cm^{-3}) and $Hg_{1-x}Fe_xSe$ ($n=5.66\times10^{18}$ cm^{-3} for both $x\simeq0.03$ and $x\simeq0.05$) with $\mathbf{H}||[110]$. The measurements have been performed as a function of temperature ($1.3<T<30$ K) and for magnetic fields of less then 5 T. These materials were chosen for this study because their electron concentrations give rise to oscillatory properties that can readily be analyzed with existing theoretical models applicable at magnetic fields far below the quantum limit. The orientation of $\mathbf{H}||[110]$ was selected because, for this orientation, the resulting quantum oscillatory signal does not exhibit a line shape dominated by beating effects.[12] As a result, a more reliable analysis of the data is possible. The choice of materials was also motivated by the desire to simultaneously conduct a parallel study of the quantum oscillations in both the host material and the diluted compound system with comparable electron concentrations.

These studies are interesting because they provide quantitative information about the behavior of Fe in the zero-gap semiconductor HgSe with zinc-blende structure. This is particularly important since an analysis of the magnetization of Fe^{2+} in a zinc-blende lattice[16] indicates a Van Vleck behavior which leads to a magnetic susceptibility that is independent of temperature.[18] It can thus be expected that the temperature dependence of the quantum oscillatory phenomena in HgSe and $Hg_{1-x}Fe_xSe$ will follow the standard theory for nonmagnetic materials quite closely. As we pointed out previously,[12] and as will be demonstrated below this conclusion is not substantiated by a detailed analysis of the experimental data obtained in this study.

The remainder of this paper is divided into the following sections. In Sec. II we present an outline of the theoretical considerations pertinent to this work. In Sec. III a brief description of the experimental apparatus is given. In Sec. IV we discuss the method of determining the amplitudes and phases of the various harmonics in the quantum oscillatory signal as function of applied magnetic field. In Sec. V we present a detailed analysis of the temperature dependence of the Shubnikov—de Haas oscillations obtained for $\mathbf{H}||[110]$ in HgSe and $Hg_{1-x}Fe_xSe$. In Sec. VI the results of this study are summarized and discussed.

II. THEORY

The derivation of the Shubnikov–de Haas effect is based on the quantization of electron levels in the presence of an applied magnetic field and is discussed in many places.[19–26] The resulting magnetoresistance $\Delta\rho/\rho$ can be conveniently written in the form of a harmonic expansion:

$$\frac{\Delta\rho}{\rho}=2D\sum_{r=1}^{\infty}\frac{\sqrt{H}I_rK_rB_r\cos(\pi rS)}{\sqrt{r}}\times\cos\left[2\pi r\left[\frac{F}{H}-\gamma\right]-\frac{\pi}{4}\right]. \quad (1)$$

If the shape of the Fermi surface is spherical, then F can be directly related to the electron concentration n by

$$F=\frac{\hbar}{2e}(3\pi^2n)^{2/3}. \quad (2)$$

There are several amplitude factors in Eq. (1) which can be further defined. $K_r=e^{-\alpha rT_D/H}$ ($\alpha=2\pi^2m^*k_B/e\hbar$) is an electron-lifetime factor with T_D, the Dingle temperature, providing a measure of the impurity and dislocation scattering in the material. $I_r=X_r/\sinh X_r$ ($X_r=\alpha rT/H$) is the thermal-damping factor that incorporates the temperature T of the sample. The term $\cos(\pi rS)$ (where $S=g_cm^*/2m$) describes the Landau-level splitting in an applied magnetic field and is a function of the cyclotron-averaged g factor (g_c) and effective mass (m^*) of the electrons. Finally, the term B_r represents magnetic breakdown effects which may be required by the Fermi-surface geometry.

Equation (1) may not be appropriate for systems containing magnetic impurities which possess local magnetic moments. Under these conditions the exchange interaction of conduction electrons with the local moments produces a spin-dependent shift of the electronic energy levels. The net result is a modification of the Zeeman splitting by introducing an effective g factor $g_{\rm eff}$ and an exchange field $H_{\rm ex}$ such that

$$g_{\rm eff}=g_c+\frac{H_{\rm ex}}{H}. \quad (3)$$

In Eq. (3), $\mu_BH_{\rm ex}=-C_mJ\langle S_z\rangle$, where J is the exchange coupling constant ($J>0$ ferromagnetic exchange, $J<0$ antiferromagnetic exchange), C_m is the concentration of local moments, and $\langle S_z\rangle$ is the expectation value of spin for the magnetic ions, which can be a function of both temperature and magnetic field. As a result the parameter $S=g_cm^*/2m$ in Eq. (1) must be replaced by $S'=g_{\rm eff}m^*/2m$, where S' now is a function of both temperature and magnetic field.

However, Eq. (1) was derived assuming that both up- and down-spin electrons contribute equally to the resultant signal. Because the exchange interaction between the local moment and conduction electrons can be spin dependent, it is expected that the scattering rates for spin-up and spin-down electrons will be unequal. Equation (1) must therefore be generalized to allow for a spin-dependent Dingle temperature.[25] Under these circumstances, Eq. (1) becomes

$$\frac{\Delta\rho}{\rho}=D\sum_{r=1}^{\infty}\sum_{\sigma}\frac{\sqrt{H}I_rB_rK_rE^{\sigma r}}{\sqrt{r}}\times\cos\left[2\pi r\left[\frac{F}{H}-\gamma\right]-\frac{\pi}{4}-\sigma\pi rS'\right], \quad (4)$$

with

$$K_r=e^{-\alpha r\bar{T}_D/H}\ \text{and}\ E^r=e^{-\alpha r\delta T_D/H}, \quad (5)$$

$$\bar{T}_D=\frac{T_D^{\uparrow}+T_D^{\downarrow}}{2}\ \text{and}\ \delta T_D=\frac{T_D^{\uparrow}-T_D^{\downarrow}}{2}, \quad (6)$$

where $\sigma=+1$ is for spin-up electrons and $\sigma=-1$ is for spin-down electrons. In order to analyze data it is more convenient to represent the terms in Eq. (4) with the aid of a phasor diagram.[25] As a consequence, the resultant amplitude A_r for a given harmonic r is

$$A_r=\frac{\sqrt{H}I_rK_r}{\sqrt{r}}[E^{2r}+E^{-2r}+2\cos(2\pi rS')]^{1/2}. \quad (7)$$

Due to the periodicity of the cosine term in Eq. (7), the factor $2\pi rS'$ is uncertain to within an additive constant of $2\pi n$, where $n=0,\pm1,\pm2,\ldots$. In what follows, we arbitrarily set $n=0$. In addition, a shift $\Delta\theta_r$ in the phase of the oscillations due to spin-dependent scattering results and is given by

$$\Delta\theta_r=\tan^{-1}\left[\tan(\pi rS')\left[\frac{1-E^{2r}}{1+E^{2r}}\right]\right]. \quad (8)$$

An important conclusion evident from Eq. (7) is that for nonzero δT_D the amplitude factor A_r does not equal zero even when $2\pi rS'$ equals $m\pi/2$, $m=\pm1,\pm3,\ldots$.

III. EXPERIMENTAL CONSIDERATIONS

The data acquired in this study were obtained using a computer-controlled superconducting-magnet system that allowed digitization of the oscillatory signal at 1000 closely spaced values of the applied magnetic field. A dc current of 50 mA was passed through the sample, and the resulting voltage drop across the sample was measured by a Keithly model-148 nanovoltmeter. Using this technique, no modulation of the magnetic field is required and the data will not be distorted by any Bessel-function multiplicative factors.[27] A small nonoscillatory background magnetoresistance was fitted by a second-order polynomial function and could be subtracted from the measured signal to yield the oscillatory signal of interest.

To employ the method of data analysis discussed below, it is necessary to have a well-characterized oscillatory signal. In this study the typical size of the oscillatory signal at 4.2 K and at a magnetic field of 2.5 T is about 3 μV. A noise signal, typically <0.2 μV, is also present in the signal. The computer-controlled data-acquisition system measures the voltage from the nanovoltmeter ten times at each value of the magnetic field. Thus a signal-to-noise ratio of greater than 47:1 is expected. The signal to noise deteriorates at lower values of magnetic field and is considerably better than the above figure at fields above 2.5

T. In practice, the digitization of such data is sufficiently noise-free to use the data-analysis procedure described in Sec. IV below.

The magnetic field was set at predetermined values by a computer algorithm and was held at the appropriate value while the magnetoresistance voltage was digitized. The computer algorithm that set the value of the magnetic field was independently checked for accuracy with a calibrated axial Hall probe. The difference between the measured magnetic field and the computer-set magnetic field was found to be less than $\pm\frac{1}{2}\%$ over the magnetic field range of interest.

The samples used in this study were oriented by x-ray backreflection techniques and cut into parallelopipeds with dimensions of $1.2\times1.2\times9$ mm^3. The Fe content was determined by electron-microprobe techniques. The oriented samples were mounted inside a variable-temperature Dewar to allow data runs at different temperatures below 77 K. Four platinum pressure-point contacts were attached to the sample in such a way as to allow both the transverse magnetoresistance and Hall voltage to be measured. The electron concentration determined from the frequency of the SdH oscillations through Eq. (2) was always found to be in excellent agreement with the electron concentration deduced from the Hall-voltage measurements. The essential features of the data presented below have been reproduced on four different samples.

IV. METHOD OF DATA ANALYSIS

In a quantitative study of the Shubnikov—de Haas effect, it is important to measure not only the frequency of the oscillations but also the dependence of the amplitude and phase of the various harmonics with magnetic field. A problem with this analysis occurs when oscillatory data from semiconductors are processed because of the low electron concentration. As a consequence, the number of oscillations in a field region is small (typically 30 oscillations or less occur between 2 and 5 T), and the standard approach of Fourier-transform analysis used in the study of quantum oscillations in metals will give inaccurate results. Because of this difficulty, we use a least-squares-fitting technique to determine the amplitudes and phases of the harmonics as a function of magnetic field.

Specifically, a data window spanning one oscillation is defined. The width of this window is based on an estimate of the frequency which is obtained in a straightforward way by measuring the positions in H of the peaks over several oscillations. The data points contained in this window are fitted to a function $f(H)$ given by

$$f(H)=\sum_{r=1}^{2}\tilde{A}_r\cos\left[\frac{2\pi rF}{H}+\tilde{\Phi}_r\right]. \tag{9}$$

In the least-squares-fitting technique implemented in this study, the parameters $\tilde{A}_r$ and $\tilde{\Phi}_r$ given in Eq. (9) are treated as constants independent of the magnetic field over a region spanning one window width. The validity of this approximation is examined below.

In order to justify the use of this technique to analyze our data, considerable effort was expended to show that the fits are reliable. Figure 1(a) shows a computer-generated data trace obtained from a numerical evaluation of Eq. (4) using reasonable estimates for the relevant parameters. The results of a least-squares fit to the amplitudes $\tilde{A}_r$ and phases $\tilde{\Phi}_r$ of the fundamental ($r=1$) and second harmonic ($r=2$) are shown in Figs. 1(b) and 1(c), respectively. The solid lines in these two figures are the expected results based on the direct evaluation of the $r=1$ and $r=2$ terms from Eqs. (7) and (8). As is clear from Fig. 1, there is good agreement between the amplitudes and phases estimated from the least-squares-fitting procedure and the calculated quantities. It can be concluded that for the purposes of quantitatively determining the variation of the various harmonics over a wide range of magnetic field, the least-squares-fitting technique outlined above can give very satisfactory results.

However, for the purpose of data analysis in this paper, the method of phase measurement [Fig. 1(c)] is susceptible to a number of errors for the following reasons. It is convenient to rewrite Eq. (9) as

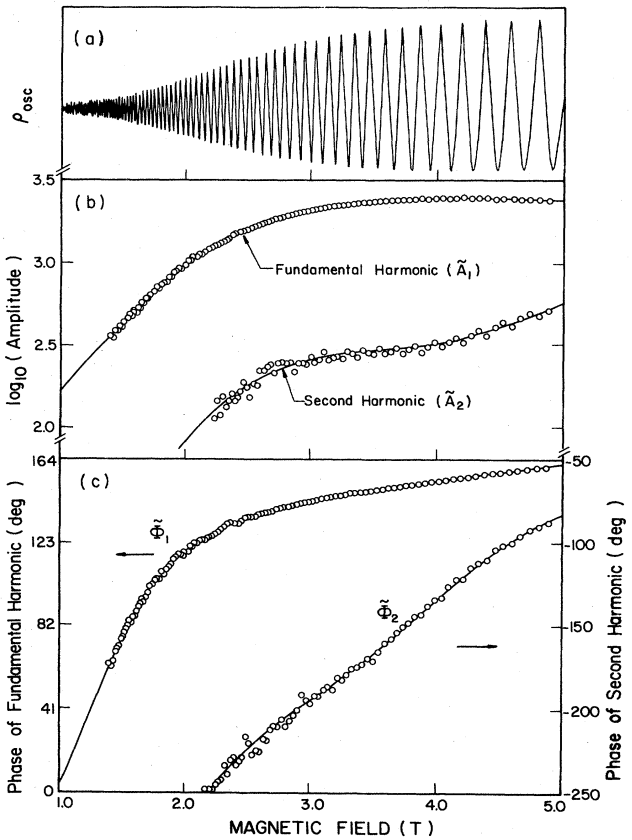

FIG. 1. Least-squares fits to a computer-generated data trace based on Eq. (4). Panel 1(a) shows the behavior of Eq. (4) with $F=100$ T, $T=1.3$ K, $m^*=0.06m_0$, $\overline{T}_D=4.0$ K, $\delta T_D=1.5$ K, $g_0=-10.0$, and $g_1=-60.0$. The parameter g_1 is further defined in Sec. V. The open circles in panels (b) and (c) show the amplitudes and phases for the fundamental and second harmonics determined from the least-squares-fitting procedure. The solid lines in these two figures are the expected variation of these two quantities based on the explicit evaluation of Eqs. (7) and (8).

$$f(H)=\sum_{r=1}^{2}\widetilde{A}_r\cos\left[\frac{2\pi rF}{H}-2\pi r\gamma-\frac{\pi}{4}+\Delta\widetilde{\theta}_r\right]. \quad (10)$$

If the frequency of the oscillatory signal is not constant with changing magnetic field,[12] phase shifts, in addition to the magnetic-field-dependent phase $\Delta\widetilde{\theta}_r$ defined by Eq. (10), will occur. These spurious shifts cannot be readily separated from the $\pi rS'$ term in Eq. (8). Also, any uncertainty in the magnetic field measurment complicates the result of the phase measurement. For example, if the frequency of oscillation is $F=100$ T and the uncertainty in the field measurment is about 1%, a rough estimate gives a 40° error in phase.[26] It is also difficult to separate this unwanted effect from the least-squares-fitted phase $\Delta\widetilde{\theta}_r$.

Because of the above reasons we use the result of the phase measurements primarily to mark the position of zeros (or minima) in the amplitude of the various harmonics. As can be seen from Eqs. (7) and (8), a sudden phase shift ($\simeq$180°) is expected in $\Delta\theta_r$ whenever A_r goes through a minimum. This phase shift is useful for defining the positions of zeros (or minima) in the data and can be used as a guide to the correct interpretation of the amplitude measurements derived from Eq. (10). It is also important to note that the errors involved in a measurement of $\Delta\widetilde{\theta}_r$ do not seriously effect the determination of the amplitude $\widetilde{A}_r$.

V. EXPERIMENTAL RESULTS AND DISCUSSION

The main results obtained in this study are shown in Figs. 2–4, which illustrate the variation of the Shubnikov–de Haas signal as a function of magnetic field for a variety of different temperatures. In order to quantitatively analyze these data, it is necessary to extract the amplitude variation of the individual harmonics. The magnetic field variation of the fundamental harmonic as determined by the least-squares-fitting procedure described above is shown in Figs. 5–7 for HgSe and $Hg_{1-x}Fe_xSe$ ($x=0.03$ and 0.05) with $\mathbf{H}\|[110]$. An important conclusion drawn by comparing these sets of data is that the signal from HgSe and $Hg_{1-x}Fe_xSe$ samples evolve in a completely different way as the temperature is lowered.

The data for HgSe ($\mathbf{H}\|[110]$, Fig. 5) show that as the temperature is lowered the amplitude of the fundamental harmonic is observed to continuously increase in magnitude. By analyzing these data at various magnetic fields between 2.5 and 4 T an effective mass could be determined in the usual way. The value of m^* was estimated to be 0.061 ± 0.007, where the quoted uncertainty reflects the variation in m^* found at different magnetic fields. Also, an analysis of the frequency of oscillations for $T<4.2$ K gives 77 T and, using Eq. (2), an electron concentration of $n=3.82\times10^{18}$ cm^{-3} is obtained. Our results for m^* are in good agreement with Fig. 1 of Ref. 28. It is important to point out here that no measurable amount of second harmonic was observed in the line shape of these oscillations.

As is clear from Fig. 6 (Fig. 7) the temperature dependence of the fundamental harmonic in $Hg_{1-x}Fe_xSe$ with $x=0.05$ ($x=0.03$) shows a completely different behavior.

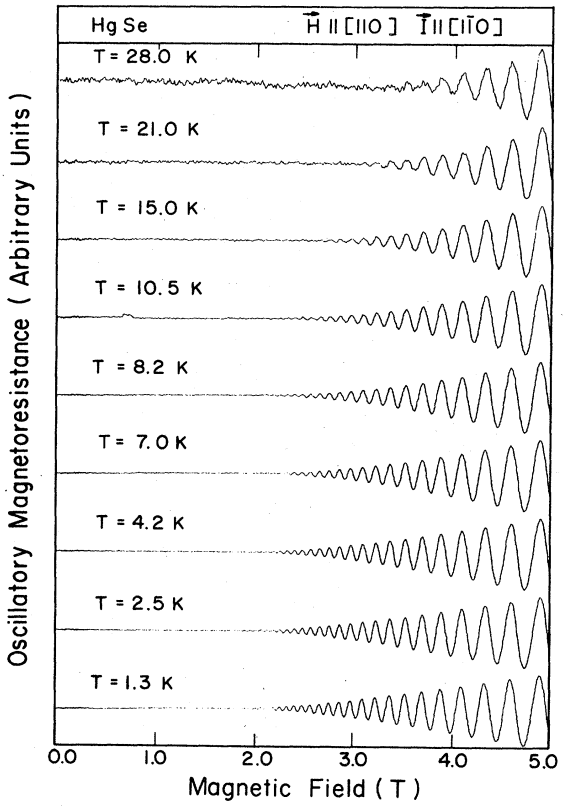

FIG. 2. The temperature dependence of the transverse magnetoresistance oscillations in HgSe for $\mathbf{H}\|[110]$. The scale factors which were used to multiply each data trace before plotting are (from top to bottom) 18.1, 7.1, 3.2, 1.9, 1.5, 1.5, 1.3, 1.1, 1.0[3], and 1.0.

As the temperature is lowered below 15 K (12 K), the amplitude of the fundamental harmonic decreases until a strong second-harmonic component at $T=8$ K (6.5 K) is observed. At this temperature the observed signal is found to contain more second harmonic than fundamental for magnetic fields above 3.5 T (3.0 T). At temperatures below $T=8$ K (6.5 K), the fundamental harmonic is observed to again increase in amplitude. In addition, detailed analysis of the data (for $x=0.05$) reveals the presence of a minimum in the amplitude of the second harmonic for $T<4.2$ K. Studies show that the position of this minimum moves to higher fields by lowering the temperature. For a fixed temperature, the position in H of this feature also depends on the Fe concentration. For example, data on the sample with $x=0.03$, shows that this minimum was only observed for $T=1.3$ K because its position was shifted to lower magnetic fields at higher temperatures. A general result obtained from this study of the $Hg_{1-x}Fe_xSe$ system is that for $x\geq0.03$, zeros (or minima) move toward higher field as the temperature is lowered. In contrast to this result, for samples with $x\simeq0.003$ ($n=4.7\times10^{18}$ cm^{-3}), the position of these zeros are no longer observed to be a function of temperature.

In order to account for these observations, it is neces-

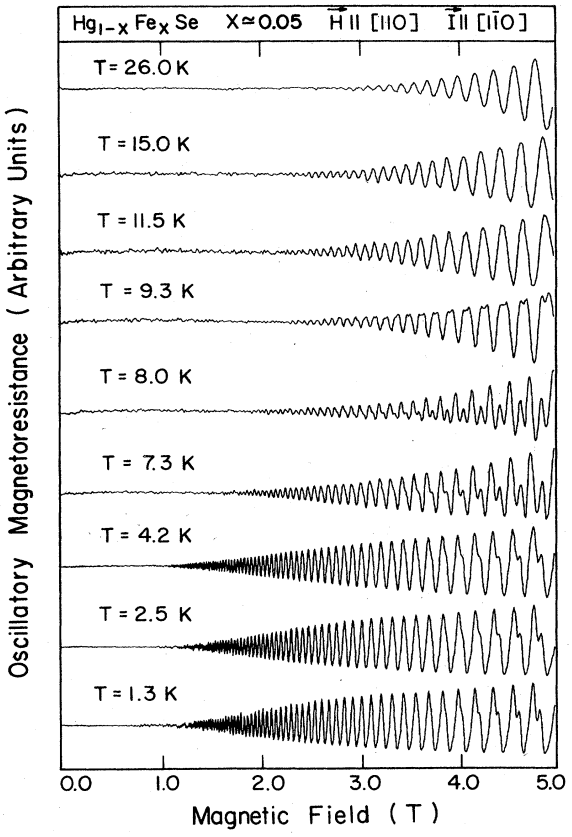

FIG. 3. The temperature dependence of the transverse magnetoresistance oscillations in $Hg_{1-x}Fe_xSe$ ($x=0.05$) for $\mathbf{H}||[110]$. The scale factors which were used to multiply each data trace before plotting are (from top to bottom) 10.1, 3.6, 3.9, 5.8, 5.9, 5.0, 1.4, 1.1, and 1.0. Data are from Ref. 12.

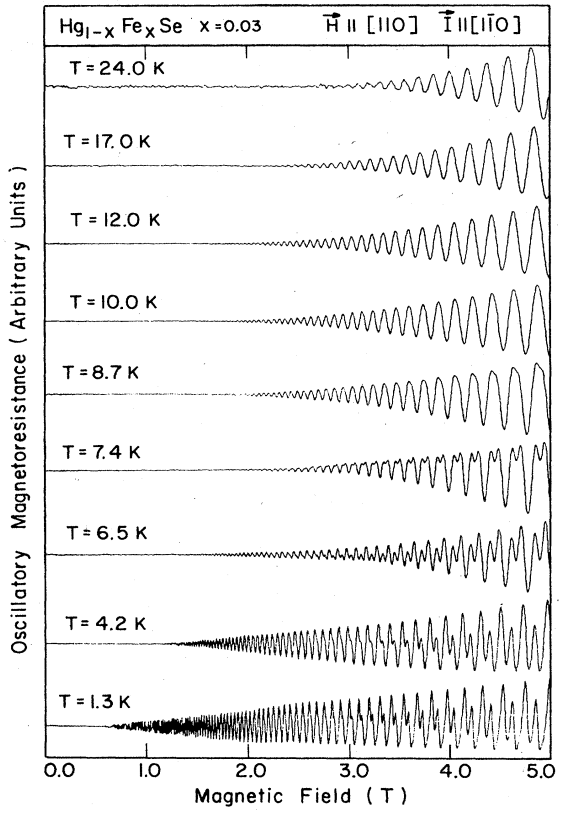

FIG. 4. The temperature dependence of the transverse magnetoresistance oscillations in $Hg_{1-x}Fe_xSe$ ($x=0.03$) for $\mathbf{H}||[110]$. The scale factors which were used to multiply each data trace before plotting are (from top to bottom) 10.1, 3.2, 1.9, 2, 2.3, 2.6, 2.3, 1.6, and 1.0.

sary to include effects due to the temperature and magnetic field dependence of $\langle S_z \rangle$. This information can be obtained from the temperature and magnetic field dependence of magnetization measurements. Because no such information is available at the present time for $Hg_{1-x}Fe_xSe$, we assume that $\langle S_z \rangle$ is proportional to the Brillouin function, $B_j(\eta)$, where $\eta = g\mu_B H/k_B T$. In what follows, the magnetic-ion g factor appearing in the argument of B_j is assumed to equal 2, and j is taken as $\frac{5}{2}$, which is consistent with the $+3$ valence assumed for the magnetically active Fe ions. As a result, the exchange field H_{ex} defined by Eq. (3) can be written as $H_{ex} = g_1 B_{5/2}(T,H)$.[29] The parameter g_1 is expected to be a function of the exchange coupling constant J, the effective magnetic moment per Fe ion, and the concentration of local moments, C_m.

The data in Figs. 6 and 7 can be analyzed in some detail by adjusting m^*, $\overline{T}_D$, δT_D, g_c, and g_1. However, some of these parameters can be fixed in a reasonably straightforward way. For instance, by analyzing several data traces in the high-temperature range ($12\ \mathrm{K} < T < 30\ \mathrm{K}$) for $x=0.05$, an effective mass $m^* = 0.067 \pm 0.001$ ($m^* = 0.064 \pm 0.001$ for $x=0.03$) for three different magnetic fields (3.8, 4.2, and 4.6 T) was obtained. Using this effective mass for the data trace at $T=15$ K (17 K for $x=0.03$), an average Dingle temperature $\overline{T}_D = 3.3 \pm 0.2$ ($\overline{T}_D = 3.2 \pm 0.2$ for $x=0.03$) was estimated from the con-

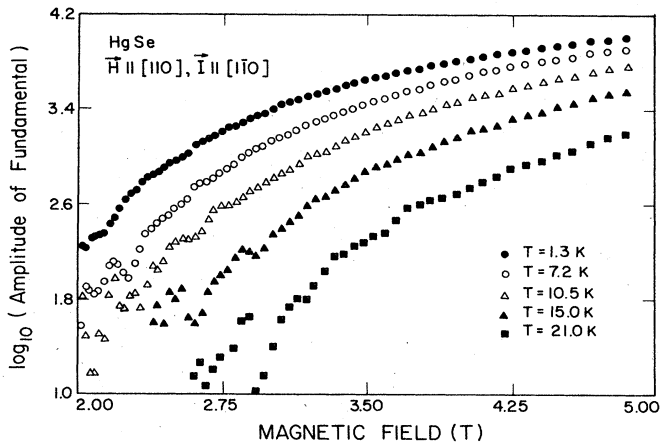

FIG. 5. The variation of the fundamental harmonic amplitude of the Shubnikov–de Haas oscillations in HgSe as a function of magnetic field for a variety of different temperatures.

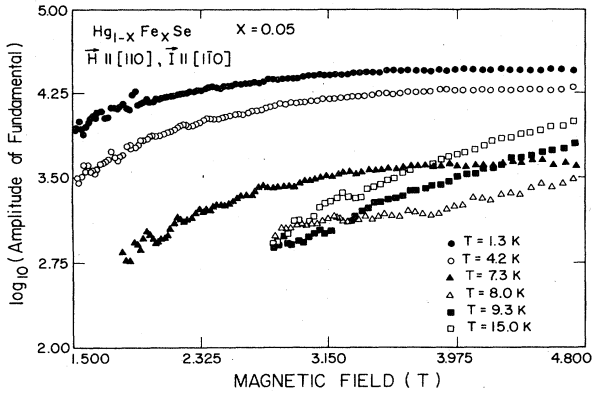

FIG. 6. The variation of the fundamental harmonic amplitude of the Shubnikov–de Haas oscillations in $Hg_{1-x}Fe_xSe$ ($x=0.05$) as a function of magnetic field for a variety of different temperatures.

ventional Dingle plot [$\ln(K_r)$ vs $1/H$]. Since the values of m^* and $\bar{T}_D$ are known, in order to fit the data to Eq. (7) it is necessary to use g_c, g_1, and δT_D as fitting parameters. It is also reasonable to assume that g_c is close to the electronic g factor of HgSe containing no Fe impurities. As a consequence, g_c is independent of temperature and its magnitude can be roughly estimated from Ref. 30 to be $g_c=-10$.[31] Therefore, in fitting the amplitude of the fundamental and second harmonic to Eq. (7), only two adjustable parameters, δT_D and g_1, are required.

Not all data traces display equal sensitivity to these two fitting parameters. From Eq. (7) it can be shown that δT_D controls the depth of a minimum in the amplitude of a given harmonic, while g_1 controls the position of the minimum in H. Therefore, data traces in which zeros (or minima) occur in a given harmonic at a particular value of magnetic field are best suited for a careful adjustment of these two parameters. A typical fit to the data ob-

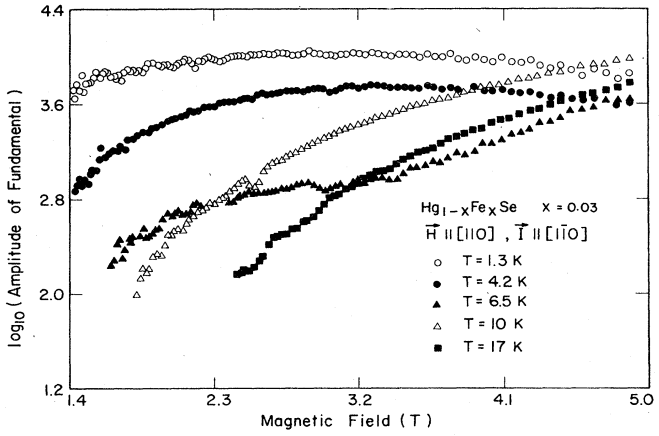

FIG. 7. The variation of the fundamental harmonic amplitude of the Shubnikov–de Haas oscillations in $Hg_{1-x}Fe_xSe$ ($x=0.03$) as a function of magnetic field for a variety of different temperatures.

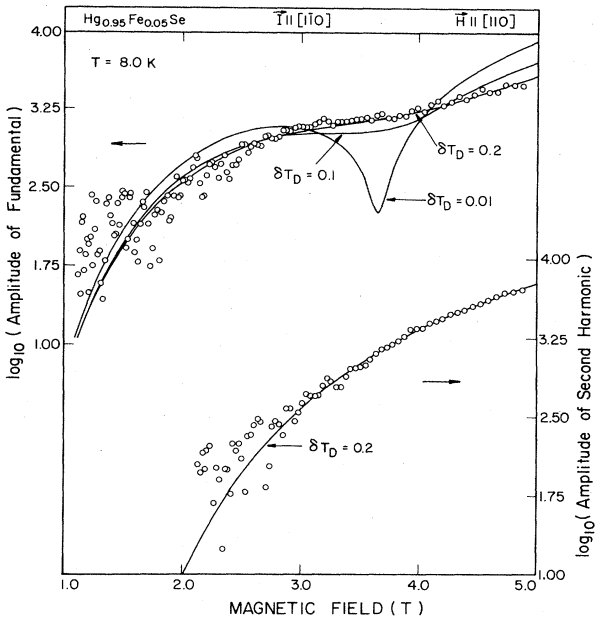

FIG. 8. The determination of the difference Dingle temperature δT_D in $Hg_{1-x}Fe_xSe$ ($x=0.05$) at $T=8.0$ K. Data for both the fundamental and second-harmonic amplitudes are analyzed. The parameters used in generating the theoretical fits are $m^*=0.067$, $\bar{T}_D=3.2$ K, $g_c=-10$, and $g_1=30.5$.

tained at $T=8.0$ K for $x=0.05$ is shown in Fig. 8. In this figure the theoretical fits to the amplitudes of the fundamental and second harmonic are shown by solid lines. It is clear from Fig. 8 that the data require a value of δT_D different from zero. Similarly, Fig. 9 ($x=0.05$) shows the magnetic field variation of the second-harmonic amplitude at $T=4.2$, 2.5, and 1.3 K. This data is particularly sensitive to the value of g_1, which controls the position of the minimum, and δT_D which controls the depth of the minimum. The dashed line in Fig. 9(c) represents the result of Eq. (7) in the absence of the exchange field ($g_1=0$). This clearly illustrates that the exchange field H_{ex} is required for analyzing the data. In addition, Fig. 10 shows a typical fit to the data obtained at $T=6.5$ K for $x=0.03$. It is also clear from this figure that the best fit to the data requires a nonzero value for δT_D.

The parameters used to fit the amplitude of the fundamental harmonic as a function of temperature for $x=0.03$ and 0.05 are collected in Tables I and II. The range on η (the argument of the Brillouin function) covered by the data in these tables varies from $0\rightarrow 0.45$ at $T=15$ K to $0\rightarrow 5.2$ at $T=1.3$ K. The uncertainties in the temperature measurement at all temperatures $T>1.5$ K are believed to be negligible. A small uncertainty of about ± 0.05 K for the $T=1.3$ K data may be present and this could account for the apparent decrease required in $\bar{T}_D$ for the $x=0.05$ sample at the lowest temperatures. An important result that can be drawn from these tables is that as the temperature is lowered, δT_D and g_1 increase while $\bar{T}_D$ remains essentially constant. In reaching these

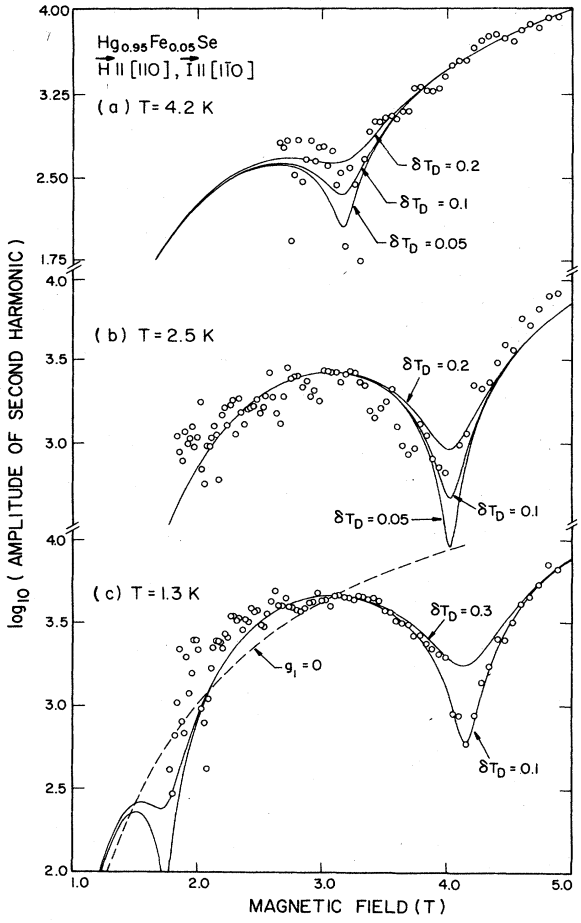

FIG. 9. The sensitivity to the difference Dingle temperature δT_D in the amplitude of the second harmonic as the temperature is lowered from 4.2 to 1.3 K. The presence of a minimum in the data at $T=4.2$ K [panel (a)] is confirmed by a sudden change in the phase $\Delta\tilde{\theta}_2$ by $\simeq 180°$ (not shown). The shift in the position of the minimum as the temperature is lowered is due to the variation of g_{eff} with temperature. The parameters used in generating the theoretical fits are $m^*=0.067$, $\overline{T}_D=3.2$ K, and $g_c=-10$.

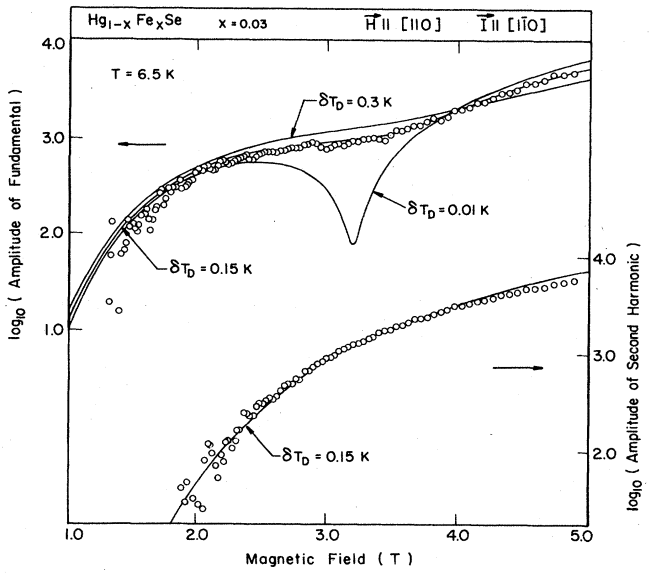

FIG. 10. The determination of the the difference Dingle temperature δT_D by fitting data on the magnetic field dependence of the first-harmonic amplitude from a $Hg_{1-x}Fe_xSe$ ($x=0.03$) crystal at $T=6.5$ K. The parameters used in generating the theoretical fits are $m^*=0.064$, $\overline{T}_D=3.2$ K, $g_c=-10$, and $g_1=-29$.

VI. SUMMARY AND CONCLUSIONS

The data presented in Figs. 2—10 represent a systematic and quantitative study of the quantum oscillatory properties of the ternary diluted magnetic semiconductor $Hg_{1-x}Fe_xSe$ over a wide range of temperature and magnetic field. This study indicates quite clearly the significant differences between the temperature dependence of the Shubnikov—de Haas oscillations in the HgSe and $Hg_{1-x}Fe_xSe$ systems. The different temperature dependence observed for these two materials indicates that a fraction of the Fe ions exists in a magnetically active state (presumably Fe^{3+}), in addition to Fe^{2+}, which apparently does not exhibit a temperature-dependent magnetization

conclusions it is important to realize that we have assumed $g_c=-10$ for both $x=0.03$ and 0.05 samples. If the value of g_c is not the same for both concentrations of Fe ($x=0.03$ and 0.05) or if its value is significantly different from -10, the magnitude of g_1 will still increase as the temperature is lowered, but its value will be different from those given in Tables I and II.

Similar results could be tabulated from an analysis of the second-harmonic data. It was found that the values of $\overline{T}_D$ and g_1 required were essentially the same as for the fundamental harmonic. However, for the $x=0.05$ sample, over the low temperature range for which a sizable second-harmonic component was evident, δT_D was about 5 times smaller than the nominal value of δT_D required to fit the fundamental harmonic. The origin of this discrepancy is not known.

TABLE I. The results of a line-shape analysis for the fundamental harmonic amplitude of the Shubnikov—de Haas oscillations obtained for $\mathbf{H}\|[110]$ in $Hg_{1-x}Fe_xSe$ ($x=0.03$). The uncertainties listed in the various parameters are worst-case values determined by adjusting the parameter until unacceptable agreement between theory and experiment were obtained.

T (K)	$\overline{T}_D$ (K)	δT_D (K)	$\lvert g_1 \rvert$
17.0	3.2±0.2	0	0
12.0	3.2±0.2	0	0
10.0	3.2±0.2	0	15.0±5
8.7	3.2±0.2	0	22.0±3
7.4	3.1±0.2	0.04±0.01	28.0±0.5
6.5	3.1±0.2	0.15±0.02	29.0±0.2
4.2	3.1±0.2	0.3 ±0.1	36.0±1
1.3	3.0±0.2	0.3 ±0.1	36.0±1

TABLE II. The results of a line-shape analysis for the fundamental harmonic amplitude of the Shubnikov–de Haas oscillations obtained for $\mathbf{H}||[110]$ in $Hg_{1-x}Fe_xSe$ ($x=0.05$). The uncertainties listed in the various parameters are worst-case values determined by adjusting the parameter until unacceptable agreement between theory and experiment were obtained.

T (K)	$\overline{T}_D$ (K)	δT_D (K)	$\lvert g_1 \rvert$
15.0	3.3±0.2	0	0
11.5	3.3±0.2	0	20.0±10
9.3	3.3±0.2	0.1 ±0.05	29.0±2
8.0	3.2±0.2	0.25±0.05	30.5±1
7.3	3.2±0.2	0.3 ±0.2	38.0±5
4.2	3.2±0.2	0.5 ±0.4	50.5±1
2.5	3.2±0.2	0.5 ±0.4	51.5±1
1.3	2.7±0.2	0.85±0.15	51.5±1

in crystals with zinc-blende structure.[16] This conclusion is in agreement with the initial results obtained on this system.[12]

Another important conclusion drawn from these data is the necessity for different scattering rates to characterize the up- and down-spin conduction electrons. This result is consistent with the above conclusion that Fe^{3+} is present, since it is probable that the microscopic origin of δT_D lies in the exchange interaction between localized magnetic moments of Fe^{3+} and the conduction electrons. The fact that δT_D increases (and saturates) as the temperature is lowered provides convincing evidence that the origin of δT_D lies in the alignment of the magnetically active Fe ion spins. It should also be pointed out that $\overline{T}_D$ found from this analysis does not decrease as x increases. Measurements of $\overline{T}_D$ over a wider range of x are required to further investigate this unexpected feature of our study.

Finally, another result drawn from Tables I and II is that the magnitude of g_1 increases as the temperature is lowered. This indicates, assuming the underlying validity of Eq. (3), that either the magnetic moment per spin or the exchange coupling constant J, or both, are functions of temperature. From the analysis presented here, this temperature dependence seems to saturate for $T<4.2$ K. Furthermore, the ratio of $g_1(x=0.05)$ to $g_1(x=0.03)$ at low temperatures is very close to the ratio of Fe ions present. Although this result may arise from a fortuitous choice of g_c, it provides good evidence for the proportionality of H_{ex} to C_m, the concentration of local magnetic moments.

Further work on this ternary-compound system will be devoted to studies of the temperature and angular dependence of the oscillations as a function of Fe concentration. In addition, by studying the de Haas–van Alphen effect for $T<1.3$ K at higher magnetic fields, it will be possible to determine the amplitude of the third harmonic in the data. Once the amplitude of the first three harmonics are measured, standard techniques[25] can be used to determine $\overline{T}_D$, δT_D, and g_{eff} independent of any particular assumption for the exchange field H_{ex}.

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