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Electron cyclotron resonance in $In_{1-x}Mn_xAs$

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Abstract

We have made the first observation of cyclotron resonance in $In_{1-x}Mn_xAs$ alloys. Our data unambiguously show that the effective mass of electrons systematically *decreases* with increasing Mn concentration x. Using an 8-band $\boldsymbol{k} \cdot \boldsymbol{p}$ method, we demonstrate that this mass reduction is a combined effect of (1) a decrease of the band gap and (2) an effective reduction of the momentum matrix element P due to the sp–d hybridization. We also observed a significant mass decrease with decreasing temperature, its temperature coefficient slightly depending on x. The temperature dependent sp–d-interaction-enhanced Landau level spin splitting cannot account for this unusual temperature dependence. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

The advent of III–V diluted magnetic semiconductors (DMSs) has created a wide range of new device opportunities that make use of both magnetic and semiconducting properties, but at the same time it has made it possible to explore a new class of many-body phenomena in semiconductors. The sp–d exchange interaction between carriers (electrons or holes) and magnetic ions in these materials has led to a variety of interesting cooperative phenomena, including negative magnetoresistance [1,2] and ferromagnetic phase transitions [3,4]. At the core of these phenomena is the kinetics of interacting electrons that are strongly coupled with localized magnetic moments – a complicated but intriguing physical situation. Such a situation is one of the essential physical bases in high-T_c superconductors and heavy-fermion systems.

Cyclotron resonance (CR) in such strongly correlated systems has attracted much interest [5–7] because the effective mass determined by CR is usually different from the thermodynamic mass obtained by magnetization or magnetoresistance measurements.

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While Kohn's theorem [8] states that CR is sensitive only to the center-of-mass motion of a many-electron system and thus independent of its internal motion (i.e., electron–electron interactions), imperfections breaking the translational symmetry in the system such as disorder and non-parabolicity lead to a breakdown of this theorem and CR can be affected significantly by interactions and correlations [9–11]. If there are carriers of more than one type with different cyclotron frequencies, the center-of-mass motion does not separate from internal motion of the electron gas [12]. Here, we report the first successful CR measurement of the effective masses of conduction electrons in $In_{1-x}Mn_xAs$. The determination of the effective masses in this technologically important DMS alloy system should be extremely useful, but equally important, it should provide new insight into the influence of the exchange interaction on the dynamics of free carriers and the effective mass of quasi-particles like magnetic polarons.

2. Experiment

We studied three n-type $In_{1-x}Mn_xAs$ samples ($x = 0, 0.08$, and 0.12), which were grown on GaAs substrates by molecular beam epitaxy [13]. The room temperature mobility of both the $x = 0.08$ and 0.12 samples was $\sim$450 cm^2/V s and the electron density was 10^{16}–10^{17} cm^{-3}. In order to make CR observable in these low mobility samples, we used very strong pulsed magnetic fields (up to 150 T) produced by a unique single-turn coil technique [14]. We mainly used two wavelengths, 10.6 μm (117 meV) and 5.53 μm (224 meV), provided by CO_2 and CO gas lasers, respectively, to observe CR within our magnetic field range. We also made mid-infrared interband absorption measurements at 300 K using a Fourier-Transform spectrometer to evaluate the band gap in each sample.

3. Results and discussion

We observed pronounced CR absorption peaks in all the samples in the temperature range of $\sim$20 K to room temperature. Representative magnetotransmission spectra for the $x = 0.08$ and 0.12 samples are shown in Fig. 1. The vertical solid lines indicate the CR peak positions. It is very important to note that all the data in the present study were taken in the quantum limit, i.e., under the condition that almost all the electrons were in the lowest Landau level even at room temperature. This is realized above 20 T for the electron density of 10^{16}–10^{17} cm^{-3}. Under such conditions, the effective masses obtained are independent of the electron density. We believe that the broad absorption "band", or the low-field tail, observed in the $x = 0.12$ sample at 224 meV come from either "interband" absorption due to the exchange-field-induced closing-gap effect or impurity-shifted cyclotron resonance [15], but we will not discuss its origin in what follows since it is irrelevant to our determination of free carrier masses.

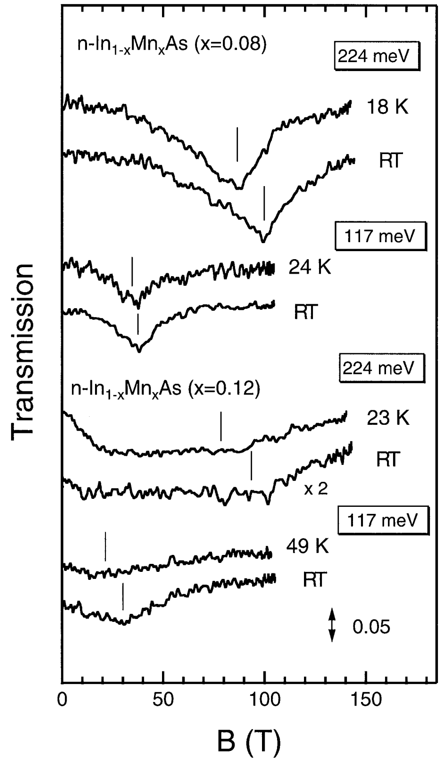

Fig. 1. Magnetotransmission spectra in $In_{1-x}Mn_xAs$ ($x = 0.08$, 0.12). The vertical solid lines indicate the absorption peaks due to the cyclotron resonance.

Our data show that the cyclotron mass (m^*_{CR}) decreases considerably with the Mn concentration, x. For example, from the data taken at room temperature with a photon energy of 224 meV, we obtain $m^*_{CR} = 0.053m_0$, $0.051m_0$ and $0.048m_0$ for $x = 0$, 0.08 and 0.12, respectively. This tendency should be contrasted with our recent study of $Cd_{1-x}Mn_xTe$ [16], where we found the completely opposite behavior, i.e., the cyclotron mass of electrons increased with x. Our mid-infrared Fourier-transform interband absorption spectroscopy showed that the band gap (E_g) decreases with x, which is qualitatively consistent with the observed mass decrease. As a rough estimate, we can expect that the effective mass changes with the band gap as $m_0/m^* \sim 2P^2/(m_0E_g)$, where m^* and m_0 are the band edge mass and the free electron mass, respectively, P is the momentum matrix element $-i\langle \varphi_{CB}|\boldsymbol{p}|\varphi_{VB}\rangle$, and φ_{CB} and φ_{VB} are the s-like and p-like basis functions of the conduction and valence bands near the Γ point. However, we find that the band gap change with x is too large to be consistent with this m^*–E_g relationship. For example, $-\mathrm{d}\ln E_g/\mathrm{d}x$ is found to be ~ 2.4 (taking $E_g = 418$ meV in InAs), while $-\mathrm{d}\ln m^*_{CR}/\mathrm{d}x$ is only ~ 0.69 at 224 meV and room temperature. In brief, the x-dependence of the band gap alone cannot explain the observed mass decrease with x, calling for a better model taking into account the effects of magnetic ions.

A theoretical study by Hui et al. [17] suggests that the effective mass of electrons is affected considerably by the sp–d hybridization in DMSs; it can be calculated by the $\boldsymbol{k}\cdot\boldsymbol{p}$ method whose sp Hamiltonian matrix elements are effectively modified by the sp–d hybridization. They considered 28 basis functions including 10 upper and 10 lower d-levels separated from each other by an on-site Coulomb energy, and two s-levels and six p-levels. They proposed that the effect of the d-levels is put in an (8×8) $\boldsymbol{k}\cdot\boldsymbol{p}$ matrix for the sp band by a perturbation to order k^2. If the influence of the spin–orbit split-off band on the change in the mass due to the sp–d hybridization is small, the modification of P is the main change in the sp Hamiltonian matrix. It is demonstrated in Ref. [17] that the effective masses of electrons and light holes in $Cd_{1-x}Mn_xTe$ increase with x due to this reduction in P. A strong reduction in P with increasing x in $Hg_{1-x}Mn_xSe$ has been indeed observed [18]. In light of these considerations, the large difference between $\mathrm{d}\ln E_g/\mathrm{d}x$ and $\mathrm{d}\ln m^*_{CR}/\mathrm{d}x$ in the present study strongly suggests that the reduction in P due to the sp–d hybridization plays an important role.

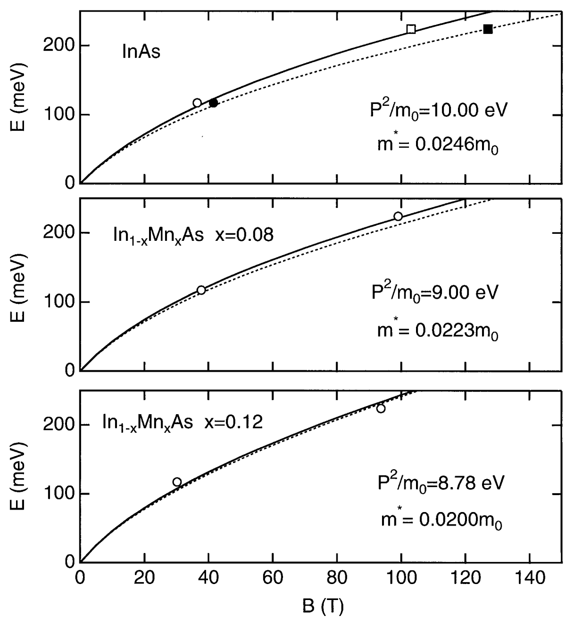

Fig. 2. Plot of the energies of CR peaks as a function of magnetic field at room temperature. Solid and dotted curves are the results of the calculation of the a-set and b-set transitions, respectively, in terms of the $\boldsymbol{k}\cdot\boldsymbol{p}$ method. Open and closed circles denote the experimental results in the present study. Open and closed squares are the results of our previous CR experiment of another bulk InAs sample [21].

We calculated conduction band Landau levels using the standard Pidgeon–Brown model [19] but also taking into account the sp–d exchange interaction [20] in order to understand the experimental results more quantitatively. Here, we take P as an adjustable parameter in order to represent the hybridization effect. Fig. 2 shows the calculated inter-Landau level transition energies ($N = 0$–1) at 300 K along with the experimental results at room temperature for the three samples. We can see an excellent agreement between the calculation and experiment. The solid and dotted curves denote transition energies of the a-set and b-set, respectively, where the a-set and b-set originate from the spin-up and spin-down states. We used the standard set of Luttinger parameters for InAs and took into account the x dependence of E_g. We used 0.22 eV for the s–d exchange constant and -1.2 eV for the p–d exchange constant. Since the exchange constants in $In_{1-x}Mn_xAs$ are not known, we assumed these values to be equal to the s–d exchange constant in one of the

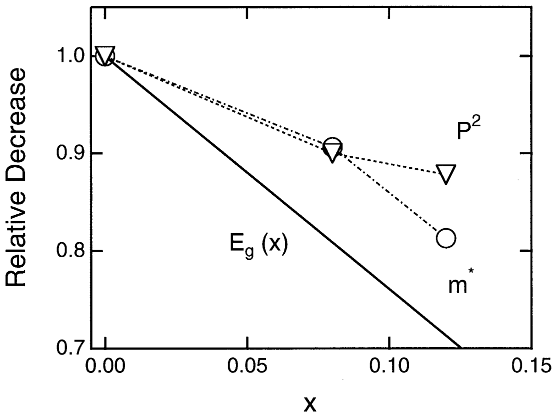

Fig. 3. Relative decrease of the energy gap (E_g), the momentum matrix element (P), and the band edge mass (m^*).

most thoroughly studied DMSs, $Cd_{1-x}Mn_xTe$ [22], and the p–d exchange constant in $Ga_{1-x}Mn_xAs$ [23], which is the only other III–V DMS that can be produced, respectively. The calculated band edge masses and values of P^2/m_0 used are also shown in Fig. 2. The electron mass in InAs obtained ($m^* = 0.0246m_0$) is in good agreement with the electron mass in the literature $m^* = 0.0239m_0$. The relative changes in E_g, m^* and P^2 with increasing x are plotted in Fig. 3. The band edge mass of electrons in $In_{1-x}Mn_xAs$ is approximately given by $(0.0247 - 0.037x \pm 0.0006)\ m_0$ at room temperature.

We also found that the cyclotron mass decreases significantly with decreasing temperature as shown in Fig. 1. The resonance fields B_{res} at 117 meV are plotted as a function of temperature in Fig. 4. The calculated lowest two Landau levels in $In_{1-x}Mn_xAs$ ($x = 0.12$) at 20 and 300 K are shown in the inset. It may be expected that the modification of the spin splitting of the Landau levels due to the sp–d exchange interactions can produce temperature-dependent m^*_{CR} since the splitting should be significantly influenced by the magnetization, which is strongly temperature-dependent. However, the temperature dependence expected from this mechanism turns out to be the opposite to what we observed. Since the transitions indicated by arrows (1) and (3) in the inset should be the dominant absorption due to the thermal population of electrons, B_{res} should increase with decreasing temperature from 300 to 20 K, according to the calculation. Although

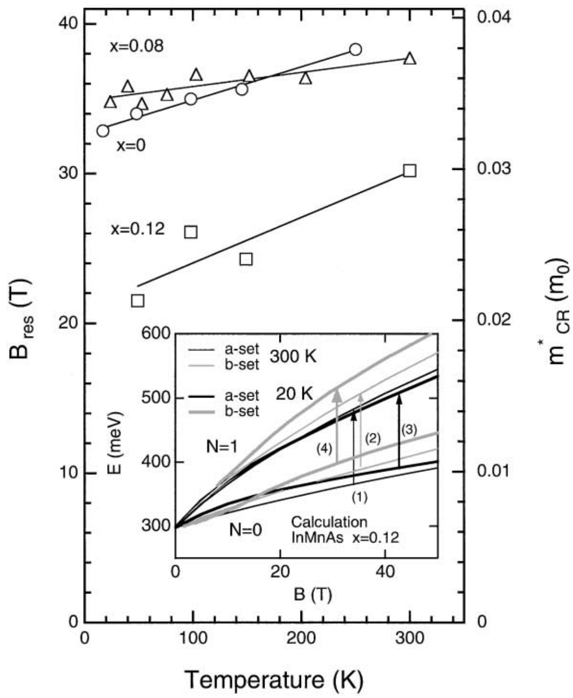

Fig. 4. Temperature dependence of the peak field of CR (B_{res}) in $In_{1-x}Mn_xAs$ ($x = 0, 0.08, 0.12$) at 117 meV. The solid lines are guides for eyes. The calculated Landau levels in $In_{1-x}Mn_xAs$ ($x = 0.12$) at 20 and 300 K are shown in the inset. The arrows denote the possible inter-Landau level transitions.

the temperature dependence of CR is a complicated problem involving the electron–phonon interaction and impurity scattering even in non-magnetic semiconductors, the qualitative disagreement between the experiment and calculation in this work may suggest that we need to consider some other effects related to the exchange interactions. The magnetic polaron effect [24] may be one of the possible mechanisms to explain the decrease of the mass with decreasing temperature. One may envisage that the mass with a spin cloud becomes heavier at high temperatures due to a thermal fluctuation of localized spins. However, in the present work we still have some uncertainties in the exchange constants and the temperature dependence of the band gap. Hence, further experimental and theoretical studies are needed to see if the sp–d exchange interaction leads to some many-body effects on the effective mass beyond the framework of the mean-field exchange theory.

4. Summary

We have successfully determined the effective masses of conduction electrons in $In_{1-x}Mn_xAs$ for the first time by high field cyclotron resonance. A modified Pidgeon–Brown model including the decrease of P and E_g can reproduce the Mn concentration dependence of the electron mass obtained at room temperature. This fact strongly suggests that the sp–d hybridization has a large effect on the effective mass of conduction electrons and its effect can be reduced to the effective change in the matrix element in the $\boldsymbol{k} \cdot \boldsymbol{p}$ Hamiltonian. In contrast to the good agreement between the calculation and experiment done at room temperature, the temperature dependence of the cyclotron mass down to around 20 K shows a large disagreement even qualitatively. This disagreement may come from some uncertainties in the temperature dependence of the band parameters, or can be a manifestation of a many-body effect like the renormalization of the mass due to the magnetic polaron effects.

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