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Cyclotron Resonance of Electrons and Holes in Paramagnetic and Ferromagnetic InMnAs-Based Films and Heterostructures

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We have studied the cyclotron resonance of electrons and holes in various types of InMnAs-based structures at ultrahigh magnetic fields. Our observations, in conjunction with an eight-band effective mass model including the s–d and p–d exchange interactions with Mn d-electrons, unambiguously suggest the existence of s-like and p-like delocalized carriers in all samples studied. The samples studied include Paramagnetic n-type $In_{1-x}Mn_xAs$ films ($x \sim 0.12$) grown on GaAs, ferromagnetic p-type $In_{1-x}Mn_xAs$ films ($x \sim 0.025$) grown on GaAs with Curie temperatures (T_C) > 5 K, paramagnetic n-type $In_{1-x}Mn_xAs$/InAs superlattices, ferromagnetic p-type $In_{1-x}Mn_xAs$/GaSb heterostructures ($x \sim 0.09$) with $T_C = 30 - 60$ K, and ferromagnetic $(In_{0.53}Ga_{0.47})_{1-x}Mn_xAs/In_{0.53}Ga_{0.47}As$ heterostructures ($x \sim 0.05$) grown on InP with T_C up to 120 K.

KEY WORDS: III–V magnetic semiconductors; ferromagnetism; cyclotron resonance; high magnetic fields.

1. INTRODUCTION

Magnetic III–V compound semiconductors such as GaMnAs and InMnAs have provided not only new device opportunities but also novel systems in which to study the physics of itinerant carriers interacting with localized spins. Carriers in the vicinity of a magnetic ion are strongly affected [1], which in turn affect other ions via carrier–carrier interactions, resulting in an indirect exchange interaction between two magnetic ions. The microscopic origin of ferromagnetism as well as the nature of the ground state in these semiconductors is still a matter of debate [2]. Deeper understanding of their band structure, particularly the effective masses and g-factors of carriers, is key to solving these fundamental issues as well as designing ferromagnetic semiconductor structures with high Curie temperatures (T_C).

Here we report results of our cyclotron resonance (CR) studies of InMnAs-based structures. CR is a direct and accurate method for determining the effective masses of carriers (or the curvature of energy vs. momentum relations), and thus can provide significant new insight into the nature of carriers, e.g., d-like or p-like or some sort of admixture. Our measurements combined with detailed calculations clearly show that there are s-like and p-like carriers in these systems that are describable within

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the framework of the Luttinger–Kohn effective mass theory.

2. EXPERIMENTAL TECHNIQUE

Since magnetic semiconductors usually have low mobilities, we performed infrared (IR) CR measurements using ultrahigh magnetic fields in order to satisfy the condition $\omega_c\tau > 1$, where ω_c and τ are the cyclotron frequency and scattering time, respectively. Fields up to 150 T with $\sim$7 μs pulse duration were generated by the single-turn coil technique [3]. The magnetic field was applied along the growth direction and measured by a pick-up coil around the sample inside a continuous flow helium cryostat. We used circularly polarized IR laser beams with wavelengths of 10.6, 9.2 (CO_2 laser), and 5.527 μm (CO laser) and the transmitted radiation through the sample was detected using a fast HgCdTe detector. A multichannel digitizer recorded the signals from the pick-up coil and detector. Although the coil breaks in each shot, the sample survives, making it possible to carry out detailed temperature and wavelength dependence studies on the same specimen. Since the transmission signal is recorded during both the up and down sweeps, a resonance is observed twice in a single pulse, allowing us to check the reproducibility of an absorption peak and to make sure that the spectra are free from any slow heating effects.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1. Electron Cyclotron Resonance

The first n-type samples we studied were $\sim$2-μm-thick $In_{1-x}Mn_xAs$ films ($x = 0, 0.025, 0.05$, and 0.12), grown by molecular beam epitaxy on semi-insulating GaAs substrates at 200°C [4]. The 4.2 K electron densities and mobilities deduced from Hall measurements were $\sim 1.0 \times 10^{16}$ cm^{-3} and 1000 cm^2/Vs, respectively, for the doped samples, and $\sim 1.0 \times 10^{17}$ cm^{-3} and 4000 cm^2/Vs, respectively, for the $x = 0$ sample. The top four traces in Fig. 1 are for these n-type films taken at 30 K. It can bee sen that Mn doping significantly broadens the electron CR, reduces the absorption depth, and shifts the resonance peak to a lower field. We observed a $\sim$25% decrease in CR mass from $x = 0$ to $x = 0.12$ [5]. We performed detailed calculations of conduction band Landau levels

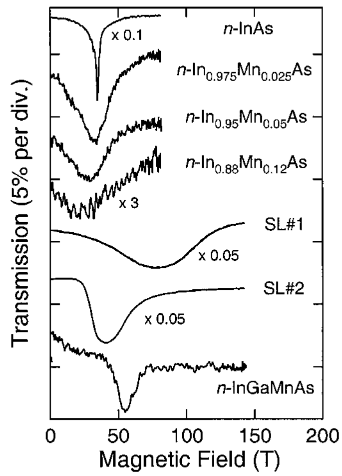

Fig. 1. Ultrahigh field electron cyclotron resonance absorption spectra for InMnAs-based magnetic semiconductor structures. The transmission of electron active circularly polarized 10.6 μm radiation is plotted as a function of magnetic field. The top four traces were taken at 30 K, the fifth and sixth traces were taken at room temperature, and the last trace was taken at 50 K. Different traces are multiplied by the indicated numbers and offset for clarity. SL#1 and SL#2 are InMnAs/InAs superlattices.

based on an 8×8 $\mathbf{k}\cdot\mathbf{p}$ model [6], which successfully reproduced this observation with a proper choice of the exchange parameters, α and β, extremely important parameters characterizing the carrier–Mn ion exchange interaction [1]. We obtained $\alpha = 0.5$ eV and $\beta = -1.0$ eV for InMnAs as the values that best represent the observed trends. In addition, our calculations showed that at low temperatures and high Mn contents the effective g-factor has the opposite sign to InAs and is extremely large ($\sim$100).

We also studied two superlattice samples consisting of alternating repetitions of 5-nm-thick InMnAs and 5-nm-thick InAs layers. One of them (labeled SL#1 in Fig. 1) had 85 periods and was grown at 200°C while the other (labeled SL#2 in Fig. 1) had 101 periods and was grown at 300°C. Hall measurements suggested that both SL#2 and SL#1 were n-type with room temperature electron densities of 1.9×10^{17} and 2.1×10^{18} cm^{-3}, respectively. Their low temperature Hall mobilities were 117 and 1159 cm^2/Vs, respectively. The fifth and sixth CR traces shown in Fig. 1 are for these two samples taken with electron-active circularly polarized 10.6 μm radiation at room temperature. As can be seen, both samples show a pronounced peak due to electron CR, consistent with the Hall measurements. However, the peak positions are very different for the two samples, corresponding to CR masses of $0.079m_0$ and $0.040m_0$, respectively, where m_0 is the mass of free electrons in vacuum. This dramatic difference in CR mass suggests that different types of electrons are responsible for the observed resonances. Detailed calculations of the band structure of these superlattice structures are in progress to elucidate this difference.

Furthermore, we studied a new type of lattice-matched ferromagnetic structure, consisting of a 50-nm-thick $(In_{0.53}Ga_{0.47})_{0.87}Mn_{0.13}As$ magnetic layer on top of a 100 nm $In_{0.53}Ga_{0.47}As$ nonmagnetic buffer layer grown on an InP(001) substrate [7]. This sample had a $T_C \sim 110$ K. Room temperature Hall measurements suggested that the sample had an electron density of 3.5×10^{18} cm^{-3} and an electron mobility of 190 cm^2/Vs. The bottom trace shown in Fig. 1 is the CR spectrum for this sample obtained with electron-active circular polarization at 10.6 μm at 50 K. We clearly observed a pronounced resonance, having an electron CR mass of $0.054m_0$. This value is too high for pure InAs and too low for pure GaAs. Furthermore, similar measurements on a control sample that had only a 100-nm-thick InGaAs buffer layer on InP did not show any sign of CR absorption. These facts lead us to believe that we are observing the CR of electrons in the magnetic (In,Ga,Mn) As layer. More measurements on similar structures with different T_Cs, combined with detailed comparison with theoretical calculations, should provide further information on the free carrier states in these novel high-T_C quaternary ferromagnetic structures.

3.2. Hole Cyclotron Resonance

The first type of p-type samples we studied were ~2-μm-thick $In_{1-x}Mn_xAs$ films with $x \sim 0.025$ grown on GaAs substrates with growth temperatures ~300°C [8]. Their typical room temperature Hall mobilities were less than 100 cm^2/Vs and their room temperature hole densities were between 0.39×10^{19} and 2.3×10^{19} cm^{-3}. Although these films show ferromagnetism at low temperatures ($T_C \sim 5$ K), all of our CR measurements were carried out in the paramagnetic phase ($T = 15$–300 K). In Fig. 2, the upper panel shows experimental hole-active CR absorption traces for the $x = 0.025$ sample taken with 5.53 μm radiation ($\hbar\omega = 0.224$ eV) as a function of magnetic field for three different temperatures. The corresponding theoretical absorption spectra are shown in the bottom panel [6]. In the theoretical simulations we assumed a narrow linewidth of 4 meV and plotted CR absorption spectra for hole concentrations of 3×10^{18}

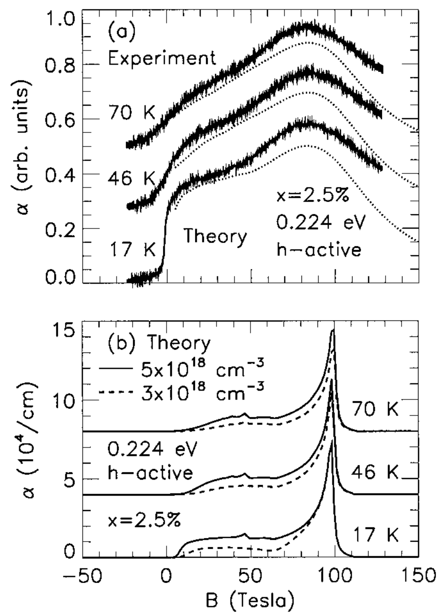

Fig. 2. The upper panel shows experimental and theoretical (hole-active) cyclotron resonance absorptions for 5.53 μm radiation ($\hbar\omega = 0.224$ eV) as a function of magnetic field in p-type $In_{0.975}Mn_{0.025}As$ taken at temperatures of 17, 46, and 70 K. The broadening in the theoretical curves was taken to be 40 meV and the density was 5×10^{18} cm^{-3}. The corresponding theoretical absorption spectra are shown in the bottom panel. We assume a narrow linewidth of 4 meV and plot cyclotron resonance absorption spectra for hole concentrations of 3×10^{18} and 5×10^{18} cm^{-3}.

and 5×10^{18} cm^{-3}. Our calculations show that doping of InAs with Mn strongly modifies the valance band structure, resulting in a camel's back structure near the top of the valence band. This, in combination with carrier population in states with finite wavevectors in the magnetic field direction (k_z), leads to a very asymmetric lineshape, observed both in experiment and theory. In addition, the dramatic sharpening of the low-field tail with decreasing temperature observed in the experiment is successfully reproduced in the theoretical plots. We ascribe this effect to the sharpening of the Fermi edge of the Fermi-Dirac distribution function for the free holes in the valence band.

The second type of p-type structures we studied were ferromagnetic $In_{1-x}Mn_xAs$/GaSb single heterostructures ($x \sim 0.09$) containing high densities of holes [9,10]. The holes were provided by the Mn acceptors. Unlike the p-type InMnAs films described earlier, these heterostructure samples showed ferromagnetism with relatively high Curie temperatures ($T_C =$ 30–60 K). Typical room temperature hole densities and mobilities were 1.0×10^{19} cm^{-3} and 300 cm^2/Vs, respectively, estimated from Hall

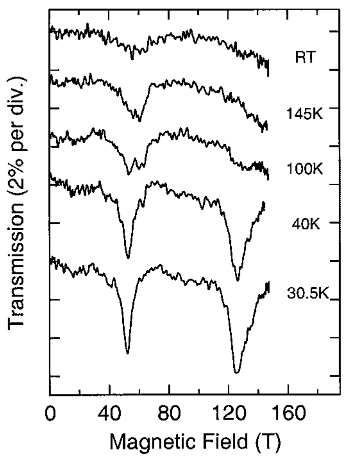

Fig. 3. Hole cyclotron resonance spectra for an InMnAs/GaSb heterostructure with $T_C = 40$ K. The transmission of of hole-active circularly polarized 10.6 μm radiation is plotted as a function of magnetic field at different temperatures. Two strongly-temperature-dependent features are observed.

measurements. The magnetization easy axis of the samples was perpendicular to the epilayer because of the strain-induced structural anisotropy caused by the lattice mismatch between InMnAs and GaSb (InMnAs is under tensile strain). Figure 3 show CR traces for a sample with $T_C = 40$ K taken with hole-active circularly polarized 10.6 μm radiation at different temperatures. From room temperature down to slightly above 100 K, one broad resonance feature (around ~58 T) is observed with almost no change in intensity, position, and width with temperature. However, at lower temperatures we see quite abrupt and dramatic changes in the spectra. First, a significant reduction in linewidth and a sudden shift to a lower magnetic field (~50 T) occur simultaneously. This line rapidly increases in intensity with decreasing temperature. In addition, a second feature appears around 125 T, which also grows rapidly in intensity with decreasing temperature and saturates, similar to the first feature. We are currently developing a model to explain this very intriguing behavior observed only in ferromagnetic InMnAs samples with relatively high Curie temperatures.

4. SUMMARY

We observed cyclotron resonance in InMnAs-based films and heterostructures with different Mn contents at various temperatures to investigate their band structure, carrier states, and the exchange interaction of conduction and valance band carriers with localized Mn d-electrons. Clear observation of cyclotron resonance, in combination with comparison with calculations based on an effective mass theory, leads us to conclude that there exist itinerant carriers in all the structures studied. Furthermore, our determination of the effective masses, effective g-factors, and exchange parameters for these InMnAs-based systems should be very useful for designing new spin-based semiconductor devices.

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