Ferromagnetic Order Induced by Photogenerated Carriers in Magnetic III-V Semiconductor Heterostructures of (In,Mn)As/GaSb

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We report the inducement of a ferromagnetic order by photogenerated carriers in a novel III-V–based magnetic semiconductor heterostructure p-(In,Mn)As/GaSb grown by molecular beam epitaxy. At low temperatures (<35 K), samples preserve ferromagnetic order even after the light is switched off, whereas they recover their original paramagnetic condition above 35 K. The results are explained in terms of hole transfer from GaSb to InMnAs in the heterostructure, which enhances a ferromagnetic spin exchange among Mn ions in the InMnAs layer. [S0031-9007(97)03396-6]

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Cooperative phenomena based on spin exchange interactions between carriers and local magnetic moments have long been studied in the diluted magnetic semiconductor (DMS). Among various DMS materials, the III-V–based DMS represent a new class of semiconductor alloys that can be grown by molecular beam epitaxy, with a large amount of magnetic ions (concentration of manganese ions: $[\mathrm{Mn}] = 10^{20} \sim 10^{21}\ \mathrm{cm}^{-3}$) [1,2]. It has been shown that n-type (In,Mn)As usually exhibits paramagnetic characteristics, whereas p-type layers and related heterostructures p-(In,Mn)As/(Ga,Al)Sb show various types of ferromagnetic order at low temperatures [3–5]. These facts suggest that holes can change the strength of spin exchange among Mn ions. This phenomenon is reminiscent of the carrier-induced magnetic order found a decade ago in the IV-VI–based DMS (Pb, Sn, Mn)Te [6]. Lately, the light-induced dynamic magnetic effect has been studied in the II-VI–based DMS (CdMnTe) by the micro-SQUID technique, in which circular-polarized light has been utilized to yield spin-polarized carriers and thus magnetic polarons [7]. These works stimulated us to the study of (photo) carrier-induced magnetic order, which is an interesting and challenging subject.

In this paper, we demonstrate, by means of photogenerated carriers, that the strength of ferromagnetic spin exchange can be controlled by changing the hole concentrations in (In, Mn)As/(Ga, Al)Sb heterostructures. The consequence is an enhanced magnetization and, in some cases, a magnetic phase change. The experimental data clearly show that the change in magnetization is purely electronic, and is not accompanied by any photoinduced and/or heat-induced structural or chemical changes.

In Fig. 1, we show schematically the sample structure and expected band edge profile of an (In, Mn)As/GaSb heterostructure. Samples were grown by molecular beam epitaxy. The Mn content x of a top 120 Å (In, Mn)As layer was fixed at 0.06 ± 0.02. Details of growth procedures and general physical properties have been described in other papers [5,8]. The most important point in sample preparation is the use of relatively low substrate temperatures (T_s) for the growth of (In, Mn)As layers ($T_s = 440$ to 470 K). This was necessary to suppress the formation of ferromagnetic MnAs crystallites, and also to control the hole concentration (p) while keeping the Mn content unchanged. The reduction of T_s increases the number of n-type defects, which consequently reduces the number of holes supplied from Mn acceptors [5,8]. Samples studied most extensively in this work are basically paramagnetic. As shown later, however, the samples are in the critical condition in that a small change in carrier numbers significantly influeneces their magnetic and galvanomagnetic properties. Anomalous Hall effect appears to be

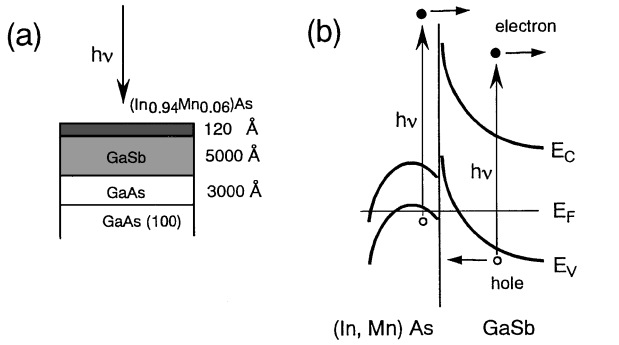

FIG. 1. (a) Structure of the sample. Direction of light irradiation is also shown by an arrow. (b) Band edge profile of (In,Mn)As/GaSb heterostructure. E_c, E_v, and E_F denote band edges of conduction band, valence band, and Fermi level, respectively.

large, making it difficult to determine carrier concentrations unless measured at high fields where magnetization is saturated [9]. Nevertheless, the p values of the samples studied here are certainly lower than those of ferromagnetic heterostructures.

As shown in Fig. 1(b), the (In, Mn)As/GaSb heterostructure most likely consists of a broken-gap type-II band alignment, being similar to the InAs/GaSb [10]. We infer that a large built-in electronic field exists across the p-(In, Mn)As/GaSb heterostructure due to the position of the surface Fermi level which is pinned inside the conduction band [11]. On the basis of these considerations, we expect that photogenerated holes can be accumulated in the (In,Mn)As layer and its vicinity. We discuss this point in later paragraphs to explain our experimental results.

Light-induced magnetization experiments were carried out using a SQUID magnetometer and magneto-optical cryostat, which were equipped with optical fiber and light pipe systems, respectively. Using a 100 W halogen incandescent lamp, samples were irradiated with white light of wavelengths ranging from 0.8 (1.55 eV) to 1.4 μm (0.88 eV). The area of irradiation was about 14 mm^2 and the excitation light intensity was about 5×10^{14} photons $cm^{-2} s^{-1}$. As discussed later, most of the light penetrates through an ultrathin (In, Mn)As layer and is absorbed in a relatively thick GaSb layer whose band gap is 0.8 eV at 5 K. The magnetic field was always applied normal to the sample plane. The demagnetization factor was assumed to be 1 on the basis of the planar geometry of the samples. The diamagnetic contribution from the GaSb/GaAs (100) substrate was measured separately, and subtracted from the overall magnetization signal.

When samples were irradiated with light, an increase in magnetization occurred gradually. It usually took about 20 min to establish a steady state condition. Surprisingly, the enhanced magnetization persisted at low temperatures ($T < 35$ K) even after the light was switched off. This behavior allowed us to ignore any bolometric effects during the light irradiation. On the other hand, it led us to suspect that samples might undergo some irreversible structural/chemical change, such as the development of ferromagnetic MnAs and/or MnSb second phases. This point was examined by repeating the experiments under heat cycling, as described below.

Figure 2 shows the temperature dependence of the magnetization under a magnetic field of 0.02 T during cooldown in the dark (open circles) and warmup after light irradiation (solid circles). Before the irradiation, the sample exhibits paramagnetic behavior throughout the entire temperature range studied in this work (3 to 50 K). A Curie-Weiss plot yields a paramagnetic Curie temperature and Curie constant of 1.1 K and 2.5×10^{-1} K, respectively (inset of Fig. 2). Light irradiation is then carried out at 5 K for 30 min under a zero magnetic field, followed by a 20 min interval in the dark before magnetization experiments. When a magnetic field of 0.02 T is applied, we observe an enhancement in magnetization, as shown by the arrow in the figure. The enhanced magnetization decreases with increasing sample temperature, and almost vanishes at about 35 K. When the sample is cooled down again and irradiated with light at 5 K, the behavior shown in Fig. 2 is reproduced. From these observations, we can conclude that the enhancement in magnetization is not due to the formation of ferromagnetic second phases such as MnAs and MnSb.

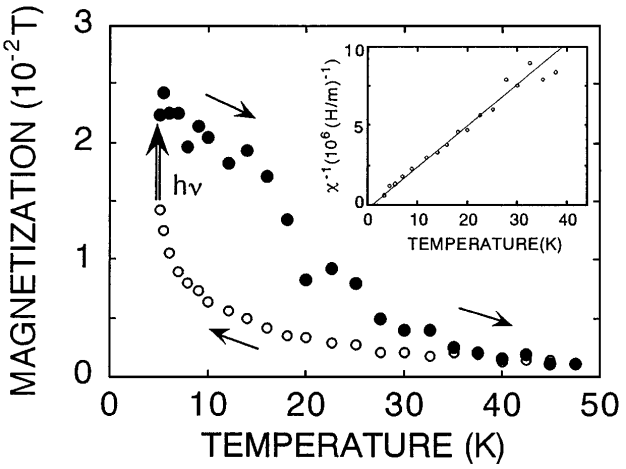

FIG. 2. Temperature dependence of magnetization observed during cooldown in the dark (open circles) and warmup (solid circles) under a fixed magnetic field of 0.02 T. Light irradiation at 5 K is also shown by an arrow. The Curie-Weiss plot for a sample in the dark is shown in the inset.

We now show in Figs. 3(a) and 3(b), respectively, light-induced changes in magnetization and Hall resistivity curves at 5 K. Before the irradiation, the magnetization changes nonlinearly with an external magnetic field with no hysteretic behavior, as shown by the open circles plotted in Fig. 3(a). This curve can be fitted by the Brillouin function (solid line) with $g = 2$, constituent spin $j = 75 \pm 20$, and the effective concentration $\bar{x} = ([\mathrm{Mn}] \times 5/2)/j$ ($[\mathrm{Mn}] = 1.1 \pm 0.3 \times 10^{21}$ cm^{-3} for this sample). This result shows that 30 Mn atoms [$= 75/(5/2)$] form a ferromagnetic cluster, indicating that the sample in the dark is close to the ferromagnetic phase transition. After light irradiation, a hysteresis loop develops, as shown by the solid circles in Fig. 3(a). The magnetization curve saturates at about 0.2 T with a saturation magnetization of about 6×10^{-2} T. This value is reasonably close to the theoretical value of $6 \pm 2 \times 10^{-2}$ T based on the high spin configuration of Mn^{2+} ions ($S = 5/2$). We can therefore conclude that the observed behavior is not due to the changes in the ionic state of Mn ions, as was the case in $FeBO_3$ [12], $CdCr_2Se_4$ [13], and magnetic garnets [14]. In these materials, a change in the ionic state manifests itself in an alternation of the magnetic easy axis and/or a change in the mobility of the magnetic domain walls.

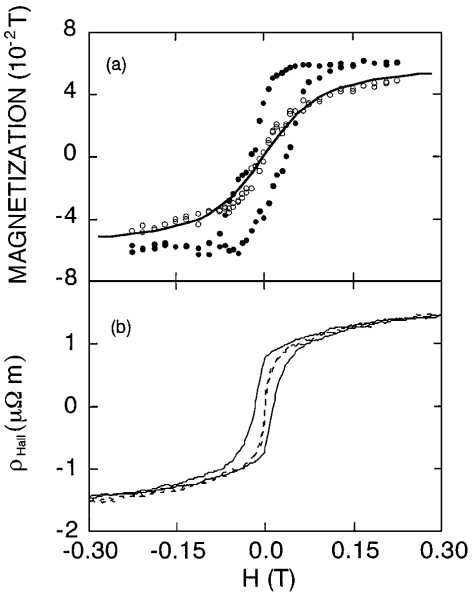

FIG. 3. (a) Magnetization curves at 5 K observed before (open circles) and after (solid circles) light irradiation. Solid line shows a theoretical curve. (b) Hall resistivity (ρ_{Hall}) observed at 5 K before (dashed line) and after (solid line) light irradiation.

Hall resistivity (ρ_{Hall}) curves [Fig. 3(b)] also showed a behavior similar to the magnetization curves [Fig. 3(a)], reflecting the fact that the ρ_{Hall} directly correlates with magnetization through the skew scattering [4,8]. As shown by the dashed line, the anomalous Hall effect is noticeable even in the dark, indicating again that the sample is in the critical condition for the ferromagnetic phase transition. Here, the contribution of negative magnetoresistance ($\Delta\rho_{xx}/\rho_{xx} \sim 3\%$ at 0.3 T) is separated from the raw Hall resistance data on the basis of symmetric (antisymmetric) characteristics of ρ_{xx} (ρ_{Hall}) with respect to the polarity of a magnetic field. To extract a change in the carrier concentration, we have carried out Hall resistivity measurements under high magnetic fields (± 15 T), where the contribution of the anomalous Hall effect is expected to be negligible [4,5]. The hole concentrations p thus obtained at 5 K before and after light irradiation are 3.76×10^{19} cm^{-3} and 3.90×10^{19} cm^{-3}, respectively, assuming a homogeneous distribution of holes across the 120 Å thick (In,Mn)As layer. An increase in sample conductivity by about 6% has also been consistently observed. These results suggest that a slight increase in p results in a change in magnetic order, reminiscent of a phase transition. Similar light-induced enhancement in magnetization has also been observed for samples with lower p values [8].

Figure 4 shows the dependence of the persistent photoconductivity (ΔG) on temperature. As can be seen in the figure, ΔG is clearly detectable up to about 45 K. Note that the observed temperature dependence is similar to that for light-induced magnetization (Fig. 2). This coincidence strongly supports the conjecture that the enhanced magnetization is induced by photogenerated carriers. In the inset of Fig. 4, we show the dependence of ΔG on excitation photon energy. This experiment was done at 77 K, and a little change in conductivity was detected by the lock-in technique. As shown in the inset, the ΔG is almost negligible for $h\nu < 0.8$ eV, whereas it increases steeply at about 0.8 eV and higher. This behavior indicates that carriers are generated primarily in a GaSb layer, and raises an interesting question as to how this process results in enhanced magnetization and its persistent characteristics.

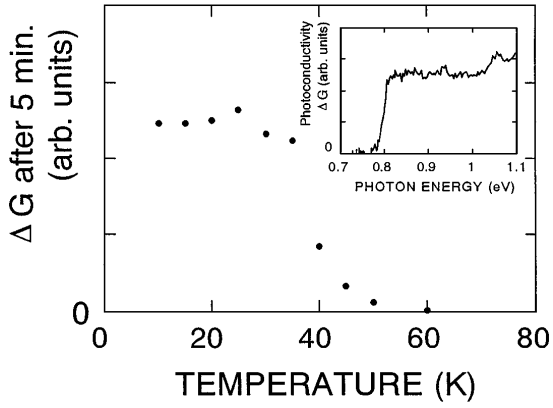

FIG. 4. Temperature dependence of the persistent photoconductivity ΔG. Inset shows the excitation photon energy dependence of ΔG at 77 K.

We strongly believe that the key to explaining the observed results lies in the unique band edge profile of (In,Mn)As/GaSb heterostructures, as shown previously in Fig. 1(b): When holes are generated in a GaSb layer by light irradiation, they are transferred effectively to a thin p-type (In, Mn)As layer due to large band bending across the heterostructure. Electrons will drift towards the opposite direction, so that holes do not recombine immediately with electrons when the light is switched off. This type of photogalvano effect has been invoked earlier to explain the persistent negative photoconductivity in n-InAs/AlSb quantum wells [15,16]. Holes thus transferred to the p-(In, Mn)As layer enhance the ferromagnetic spin exchange among Mn ions and induce the observed enhancement in magnetization and phase change. The gradual increase in both magnetization and conductivity during the light irradiation can probably be explained in terms of the existence of carrier trapping centers in GaSb and (In, Mn)As layers, and/or the (In, Mn)As/GaSb heterointerface.

In conclusion, we have shown experimental evidence for a ferromagnetic order induced by photogenerated holes in p-$(In_{0.94}Mn_{0.06})As/GaSb$ heterostructures. We have established that the ferromagnetic order in the p-(In,Mn)As is induced by the presence of excess holes. Critical hole concentration for the ferromagnetic order in the sample shown in this study is found to be about 3.8×10^{19} cm^{-3}.

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