Effect of Optical Spin Injection on Ferromagnetically Coupled Mn Spins in the III-V Magnetic Alloy Semiconductor (Ga, Mn)As

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We report on the new type of photoinduced magnetization in ferromagnetic (Ga, Mn)As thin films. Optically generated spin-polarized holes change the orientation of ferromagnetically coupled Mn spins and cause a large change in magnetization, being 15% of the saturation magnetization, without the application of a magnetic field. The memorization effect has also been found as a trace after the photoinduced magnetization. The observed results suggest that a small amount of nonequilibrium carrier spins can cause collective rotation of Mn spins presumably through the p-d exchange interaction.

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In Mn-doped III-V–based magnetic alloy semiconductors (III-V–MAS), being conventionally called III-V–based diluted magnetic semiconductors, strong hybridization between the d orbital of incorporated Mn and the p orbital of host III-V semiconductors results in the long-range parallel alignment of Mn spins (ferromagnetism) accompanied by spin-polarized energy bands [1]. This cooperative behavior opens ways to control both magnetic and semiconductor properties by changing the carrier characteristics such as concentrations [2–4] and spin degree of freedom. Indeed, a change between paramagnetism and ferromagnetism as well as alternation of coercive force has been demonstrated successfully by changing carrier concentration in (In, Mn)As/GaSb heterostructures with light illumination [2,3]. The electrical control of Curie temperature has also been accomplished in the (In, Mn)As-based field effect transistor structures [4].

In terms of the direct manipulation of carrier-spin degree of freedom, pioneering works were carried out in the 1980s for II-VI–based paramagnetic diluted magnetic semiconductors (II-VI–DMS) (Hg, Mn)Te [5,6] and (Cd, Mn)Te [7] by using circularly polarized light. These works have shown that nonequilibrium spin-polarized carriers can change the orientation of Mn spins. The observed changes in magnetization have been explained in terms of the effective mean field caused by the spin-polarized electrons for the (Hg, Mn)Te, and the magnetic polaron formation in the (Cd, Mn)Te. In contrast to the *paramagnetic* II-VI–DMS, III-V–MAS inherently has *ferromagnetic* spin exchange coupling between magnetic ions. Thus, optical spin injection in III-V–MAS by the circularly polarized light may cause novel cooperative phenomena, which are inaccessible by the paramagnetic II-VI–DMS. Studies on novel cooperative phenomena are also becoming one of the interesting topics in magnetic oxides [8].

In this Letter, we report on the new type of photoinduced magnetization in ferromagnetic (Ga, Mn)As thin films. Spin-polarized carriers (holes) generated by the circularly polarized light change the orientation of ferromagnetically coupled Mn spins depending on the light ellipticity and induce large perpendicular magnetization without the application of a magnetic field. The memorization effect has also been found as a trace after the photoinduced magnetization. We discuss that spin-flipped Mn spins develop into the collective rotation of Mn spins and yield reversed magnetic domains.

Ferromagnetic p-type (Ga, Mn)As thin films were prepared by molecular beam epitaxy on GaAs/GaAs(001) substrates. (Ga, Mn)As is known as one representative of III-V–MAS [9]. The thickness of the magnetic layer is 200 nm and Mn content x is 0.011. Details of our growth conditions are similar to those described in earlier papers [10,11]. Compressive strain caused by the lattice mismatch between a (Ga, Mn)As epilayer and a GaAs substrate results in the in-plane magnetic anisotropy [11]. Curie temperature, as measured by a dc superconducting quantum interference device (SQUID) magnetometer, was about 30 K. Compared to the electrical transports of typical (Ga, Mn)As with similar Mn content [12] the reduced hole concentrations below 10^{20} cm^{-3} are inferred in the present samples.

Linearly polarized emission from a laser light source was converted into circularly polarized light by using a quarter-wave plate. The range of wavelength was from 685 to 1064 nm. The circularly polarized cw light beam was then impinged normal to the sample plane to generate a spin-polarized carrier in the samples. A magnetic field, when it was necessary, was applied perpendicular to the sample plane (the Faraday configuration). Being similar to a bulk zinc-blende GaAs, the electrons (holes) whose spin polarization is antiparallel (parallel) to the direction of light propagation are generated in the conduction (valence) band, when the σ^+ circularly polarized light is impinged normal to the sample plane [13].

Hall voltage was measured in van der Pauw configuration (see the inset in Fig. 2). In magnetic semiconductors, carriers are asymmetrically scattered by local magnetic

moments, causing anomalous (extraordinary) Hall effect (AHE) [14] in addition to the ordinary Lorentz-force Hall effect. For (Ga, Mn)As at low magnetic fields, Hall resistance divided by sheet resistance $R_{\mathrm{Hall}}/R_{\mathrm{sheet}}$ is proportional to the magnetization component that is perpendicular to the sample plane [11]. Photoconductivity was also measured to monitor the reproducibility of the amount of photogenerated carriers from run to run. Samples were immersed in liquid He to minimize the unfavorable heating of sample by light illumination so that all the data discussed in this Letter were taken at 4.2 K. Magneto-optical effects, in particular, magnetic circular dichroism (MCD) with backscattered configuration was also measured to study optically an induced-magnetization change. Both electrical and optical methods have shown qualitatively similar dependence on excitation polarization.

Figures 1(a) and 1(b) show, respectively, raw Hall resistance R_{Hall} and sheet resistance R_{sheet} data taken under light illumination with a rotation of the quarter-wave plate. No external magnetic field was applied to the sample. Wavelength and power of excitation light was $\lambda = 785$ nm and $P = 600\ \mathrm{mW/cm^2}$, respectively. The inset shows the magnetization curve deduced from magnetotransport data. Reflecting the in-plane magnetic anisotropy, there is no noticeable remanent component along the direction normal to the sample plane at the zero magnetic field. As far as the raw Hall resistance values are concerned, there is an offset component ($\sim -180\ \Omega$) due to the misalignment of voltage probes at the corners of the square sample. With a rotation of the quarter-wave plate from 0° to 360°, 180° periodicity is clearly visible in Hall resistance. The change in R_{Hall} is either positive or negative, respectively, for σ^+ and σ^- polarized light excitation. In this particular experiment, the complete circular polarization condition is established at the rotation angles of 45°, 135°, 225°, and 315°. In contrast, the R_{sheet} does not change significantly with the ellipticity of the light. Consequently, the observed results indicate that perpendicular magnetization is induced by the illumination with circularly polarized light that is impinged normal to the sample plane.

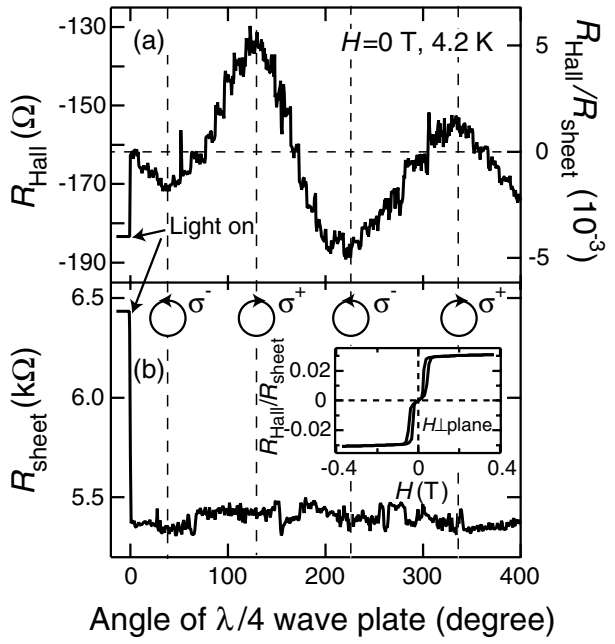

FIG. 1. Changes in (a) Hall resistance R_{Hall} and (b) sheet resistance R_{sheet} at 4.2 K for the $Ga_{0.989}Mn_{0.011}As/GaAs$ sample with in-plane magnetic anisotropy under the light illumination with rotation of the quarter-wave plate from 0° to 360°. No magnetic field is applied. Wavelength and power of excitation light were 785 nm and 600 $\mathrm{mW/cm^2}$, respectively. The inset shows the magnetization curve extracted from the anomalous Hall effect measurements.

The direction of induced magnetization $\Delta M_\perp$ ($\approx R_{\mathrm{Hall}}/R_{\mathrm{sheet}}$) is parallel to the incident σ^+ light. This strongly suggests that it is a spin-polarized nonequilibrium *hole,* and not an electron, that changes the orientation of Mn magnetic moments, as far as *negative p-d* exchange coupling is assumed [15]. A similar increase/decrease in MCD signal has also been observed with excitation and monitoring light wavelengths of 790 and 850 nm, respectively. This optically confirms the inducement of perpendicular magnetization. Note that the observed Hall voltage is not due to the circular photogalvanic effect reported earlier for *p*-doped (001)-oriented GaAs multiple quantum well structures. The effect appears only along $[1\bar{1}0]$ in zinc-blende-type crystals [16], whereas in our experiments, the electromotive force is observed along the ⟨100⟩ crystallographic direction (see the inset in Fig. 2).

According to the pump-probe measurement under the reflection geometry performed in the separate experiments, the photoinduced carrier lifetime is less than 1 ps [17], which presumably is attributed to the nature of

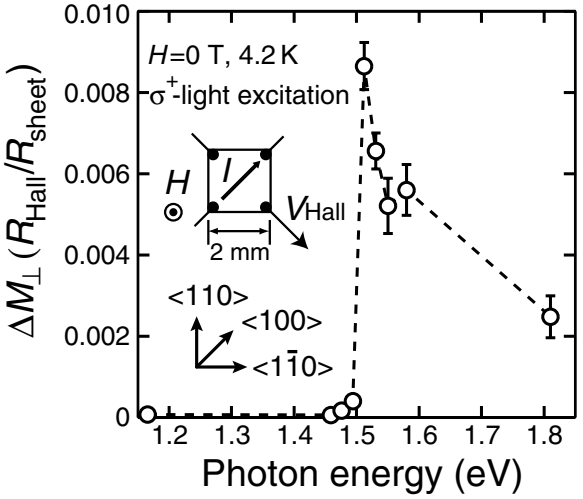

FIG. 2. Dependence of photoinduced magnetization $\Delta M_\perp$ extracted from AHE measurements on excitation photon energy at 4.2 K for the $Ga_{0.989}Mn_{0.011}As/GaAs$ sample with in-plane magnetic anisotropy. Illuminated light is σ^+ polarized. No magnetic field is applied. Excitation light power was about 600 $\mathrm{mW/cm^2}$. The inset shows schematically the sample configuration.

low-temperature GaAs [18]. Based on this observation, the number of photogenerated spin-polarized carriers is estimated to be $10^{10}-10^{11}$ cm^{-3}. This value suggests that the observed change in R_{Hall} is not due solely to the spin polarization of photogenerated carries but is due to the change in the orientation of Mn moments, and thus the magnetization.

Dependence of photoinduced perpendicular magnetization $\Delta M_{\perp}$ on excitation photon energy for σ^{+} polarized light excitation is shown in Fig. 2. The excitation light power was $P = 600$ mW/cm^2. $\Delta M_{\perp}$ is negligibly small when the photon energy is smaller than the band gap energy E_g of (Ga, Mn)As which is eventually very close to that of GaAs. When photon energy is increased and becomes higher than the E_g, $\Delta M_{\perp}$ increases steeply and shows the maximum value at the 1.50–1.51 eV region. In general, a small compressive strain lifts the degeneracy between heavy- and light-hole states at the Γ point. This enhances the polarization of photogenetrated carriers [13]. With a further increase in excitation energy, $\Delta M_{\perp}$ decreases gradually. This is probably attributed to the fact that the spin states in the valence band are mixed by the spin-orbit coupling for $\boldsymbol{k} \neq 0$ [13]. As to the dependence on P, we have observed an almost linear increase in $\Delta M_{\perp}$ with increasing P within the limit of the present study. The data in Fig. 2 indicate that the observed change in magnetization cannot be attributed to the direct excitation of d-state electrons of Mn ions but is attributed to the optically generated spin-polarized carriers.

Surprisingly, the magnitude of the induced magnetization reaches to about 15% (2 mT) of the saturation magnetization at the excitation photon energy of $h\nu = 1.505$ eV with the excitation power of 600 mW/cm^2. This change corresponds to the number of Mn ions $\Delta N_{\mathrm{Mn}} = 4 \times 10^{19}$ cm^{-3} ($S_{\mathrm{Mn}} = 5/2$) and is 10^4-10^5 times larger than that reported in II-VI–DMS [5–7]. First, we can probably rule out the polaron formation because, according to our recent data obtained from time-resolved photoinduced Faraday rotation measurements [17], the development of the Faraday rotation signal, and namely magnetization, occurs quite rapidly within 1 ps. In general, the polaron formation takes much a longer time ($\approx$50–100 ps) [7,19]. The second possibility is a change in Mn spin orientation induced by the magnetic mean field H_{ph} due to the optically generated spin-polarized carriers. This is the mechanism that has been concluded to be responsible for the circularly polarized-light induced magnetization in (Hg, Mn)Te [6]. In view of this mechanism, H_{ph} of 20–40 mT is required to realize the observed change in magnetization (2 mT). Such a large H_{ph} is far from the $H_{\mathrm{ph}} \sim 10^{-5}$ mT estimated with $\Delta p = 10^{11}$ cm^{-3} and $N_0\beta = 1.0$ eV [15] by using Eq. (15) in Ref. [6]. Other candidate mechanisms are those coming from the off-diagonal part of s, p-d exchange interaction [5]. The contribution of this mechanism, as discussed in terms of the dynamic polarization, has been concluded to be not significant when the Mn-Mn interaction is small [6]. In the present case, however, the hole-mediated interaction between Mn ions is strong so that we believe it is worth pursuing the mechanisms associated with the p-d exchange interaction. One possibility is the feedback effect based on the Zener mean-field model [20]. Mn spins that are partially flipped by the optically generated nonequilibrium hole spins promote further flipping of hole spins, and which in turn reflects back to both Mn- and hole-spin subsystems. Another possibility may come from the double exchange [21]/resonance interaction models [22]. Photogenerated holes that occupy the delocalized Mn d or resonant states itinerate and align local magnetic moments successively. In either case, a change in magnetization is presumably a function of spin lifetime and propagation length. It is worth noting that an increase in hole numbers by the light illumination may also change the strain-induced magnetic anisotropy because the magnitude of the anisotropy constant depends on the occupation of the hole subbands [23]. This effect, however, will not cause ellipticity dependence as shown in Fig. 1.

There is some evidence that suggests that the spin-flipped Mn moments develop into the collective rotation of Mn moments and cause the magnetic domain reversal as well as the change in domain structure. Figures 3(a) and 3(b) show the changes in Hall resistance R_{Hall} and sheet resistance R_{sheet} with the illumination of σ^{+}/σ^{-} polarized lights. As seen in the figures with magnified vertical scales, R_{Hall} does not return to its original value

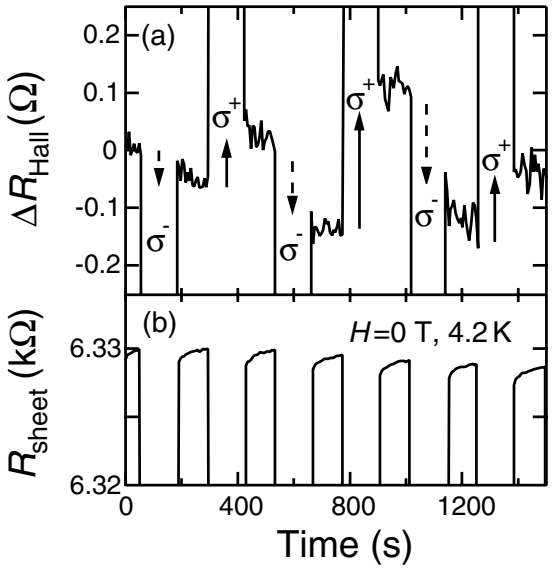

FIG. 3. Changes shown by the magnified vertical scale for (a) the Hall resistance ΔR_{Hall} and (b) sheet resistance R_{sheet} at 4.2 K under the illumination with σ^{+} or σ^{-} light on a $Ga_{0.989}Mn_{0.011}As/GaAs$ sample with in-plane magnetic anisotropy. Wavelength and power of excitation light were 785 nm and 600 mW/cm^2, respectively. Arrows indicate the alternate illumination of σ^{+} and σ^{-} polarized light. The memorization effect is noticeable by the remanent R_{Hall} component after the light illumination. Note that there is no memorization effect in R_{sheet}.

after the excitation light is turned off and appears to leave a small residual component with its relative sign depending on the circular polarization σ^+ or σ^- [Fig. 3(a)]. The magnitude of the residual component is estimated to be less than 0.1% ($\Delta N_{\mathrm{Mn}} = 2 \times 10^{17}$ cm^{-3}) of the saturation magnetization. This suggests that the collective rotation of Mn moments is partially memorized as a reversed magnetic domain. This inference is strongly supported by the fact that the memorization effect disappears when the experiment is done under the application of a sufficiently strong perpendicular magnetic field at which magnetization almost saturates. Under this condition, the sample is magnetically nearly homogeneous in the dark so that the collectively flipped Mn moments are forced to return back to the original homogeneous state when the light is turned off. Therefore, the memorization effect is the remanent of a photoinduced perpendicular magnetization component that is stabilized presumably in the form of small perpendicular ferromagnetic domains imbed in the in-plane ferromagnetic domains.

Finally, it is worth mentioning that changing the magnetization orientation by *electrical* spin-polarized carrier injection can be expected in III-V–MAS. Recently, current-induced switching of magnetization has been proposed theoretically [24] and has been demonstrated experimentally in the magnetic metal multilayer structures [25]. In these particular experiments, a large number of spin-polarized carriers, namely, high current density (10^8 A/cm^2), was required for the magnetization switching. Our present findings suggest that, by using III-V–MAS, the electrical control of magnetization orientation can be achieved by injecting a relatively smaller number of spin-polarized carriers than that required in the magnetic multilayers.

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