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# Magnetization manipulation in (Ga,Mn)As by subpicosecond optical excitation

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We demonstrate complete reversal of a full magnetic hysteresis loop of the magnetic semiconductor (Ga,Mn)As by ultrashort optical excitation with a *single* subpicosecond light pulse, with obvious implications for ultrafast magneto-optical recording. Our approach utilizes the fourfold magnetic anisotropy of (Ga,Mn)As, in combination with the magnetic linear dichroism of the material. © *2005 American Institute of Physics.* [DOI: 10.1063/1.1899231]

During the last decade (III,Mn)V ferromagnetic semiconductors[1] (FS) such as (Ga,Mn)As have been the subject of intense research. This huge interest is caused by the very promising multifunctional circuits that can be envisioned when FSs are integrated with standard semiconductor devices for example, spin injection,[2] electrical control of ferromagnetism,[3] very large magnetoresistance,[4] tunneling anisotropic magnetoresistance,[5] and current-induced switching[6] have been demonstrated in (III,Mn)V-based devices. In the following we show how the specific magnetic properties of (Ga,Mn)As can be applied to ultrafast magneto-optical recording.

The ferromagnetic materials that are commonly used in magneto-optical memory devices possess uniaxial magnetic anisotropy, i.e., there is an easy axis, along which the magnetization can be either parallel or antiparallel. As a result, the magnetization as a function of external magnetic field exhibits a hysteresis loop resulting from the switching between two metastable magnetization states. In contrast, the ferromagnetic semiconductor (Ga,Mn)As has a biaxial magnetic anisotropy[7] and as a result possesses two equivalent easy axes. This property modifies the hysteresis loop such that a switching of the magnetization between two different pairs of states can be observed.[8–10] In this letter we demonstrate that this switching may be induced by applying a single laser pulse that can be as short as $10^{-13}$ s. Moreover, these four possible orientations of the magnetization can serve for recording of two bits of information at one spot, thus leading to a doubling of the recording density.

We present here results for a 350-nm-thick $Ga_{0.98}Mn_{0.02}As$ epilayer. It was deposited on a (001) GaAs substrate followed by a 3 $\mu$m $Al_{0.5}Ga_{0.5}As$ buffer by low-temperature molecular beam epitaxy.[11] The high structural quality of the sample was confirmed by x-ray diffraction. Magnetic characterization of the sample was done with a superconducting quantum interference device magnetometer. The Curie temperature is about 50 K. For temperature below 20 K the magnetic anisotropy shows two in-plane easy axes along the [100] and [010] crystallographic directions, respectively.

For the ultrafast laser excitation of the sample we used amplified 100 fs pulses from a Ti:sapphire laser at a repetition rate of 1 kHz and a wavelength of 805 nm. A mechanical shutter was used to select *one single pulse*. This pulse was focused on the sample to a spot size of 100 $\mu$m. Pump fluence was up to 175 $mJ/cm^2$. A magnetic field was applied in the plane of the sample at an angle of 41° with the [100] axis. During all experiments the sample was kept at a bath temperature of 10 K.

As a local probe of the magnetization ($M$), we used a linearly polarized continuous-wave laser at the wavelength of 815 nm, which was focused at the sample onto a spot with
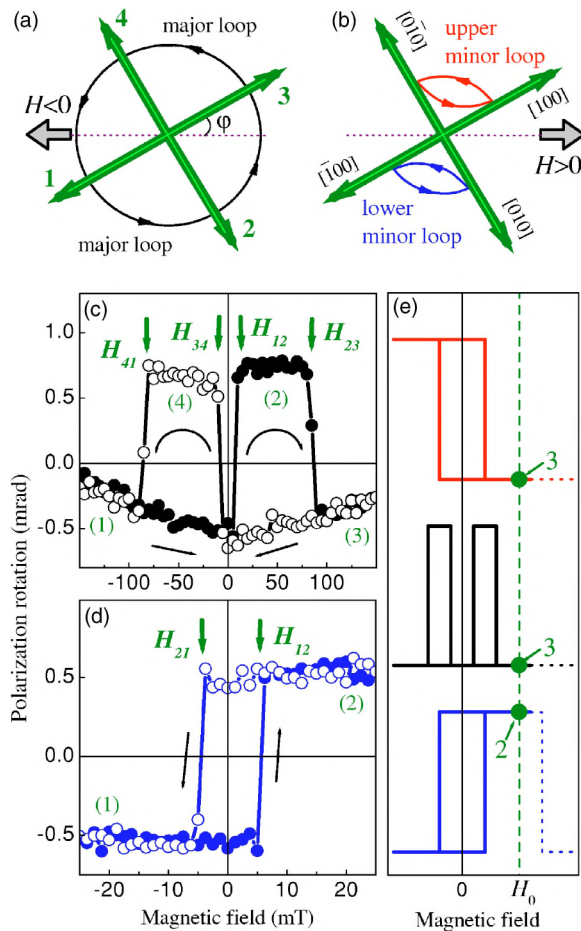
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FIG. 1. (Color online) Hysteresis loops of the $Ga_{0.98}Mn_{0.02}As$ sample with in-plane easy axes along the [100] and [010] crystallographic directions. The applied in-plane magnetic field makes an angle $\varphi = 41°$ with the [100] direction. (a) The major hysteresis loop addresses all four possible magnetization states: $1 \rightarrow 2 \rightarrow 3 \rightarrow 4 \rightarrow 1$. (b) Minor hysteresis loops address only two states: lower $1 \rightarrow 2 \rightarrow 1$ or upper $3 \rightarrow 4 \rightarrow 3$. (c) Experimental major hysteresis loop measured via the linear magnetic dichroism. The vertical arrows indicate the magnetic fields $H_{IJ}$ where switching from magnetization state $I$ to state $J$ occurs. Solid and open symbols correspond to up-sweep and down-sweep of the hysteresis loops. (d) Experimental lower minor hysteresis loop measured via the linear magnetic dichroism. (e) Schematic behavior of the magnetic birefringence signal accompanying a switch from state 2 to state 3 in a constant magnetic field $H_0$ at low temperature (bottom panel), when reducing the coercivity through a laser heat pulse (middle panel), and when returning to low temperature (top panel).

a diameter of 50 $\mu$m. The angle of incidence was about 20° and the plane of incoming linear polarization was set at 45° with respect to the [100] axis. In order to read out the magnetization orientation with respect to the crystallographic axes, the magnetic linear dichroism (MLD) was used. This effect, frequently referred to as the Voigt effect, results from the difference of the optical index of refraction for light polarized parallel and perpendicular to $M$ in the $Ga_{0.98}Mn_{0.02}As$ layer. In our scheme it results in a rotation of the polarization of the reflected light. In absolute value the rotation occurs over the same angle, but clockwise for $M \| [010]$ and counterclockwise for $M \| [100]$.[8] The polarization rotation was detected with the help of a modulation technique described elsewhere.[12]

When an in-plane magnetic field ($H$) is too weak to produce a rotation of $M$ away from the easy axes, only four different orientations of $M$ (two for each of the two [100] and [010] axes) are possible; in Fig. 1 these states are labeled

as (1), (2), (3), and (4). This occurs provided $\mathbf{H} \cdot \mathbf{M}/2 \ll K_c$ ($K_c$ is the cubic anisotropy coefficient),[7,13] which holds for the experiments presented in this letter. In this limit, hysteresis loops exhibit somewhat unusual multiple switching events.[8–10]

The process of magnetization reversal in an applied magnetic field at an angle $\varphi = 41°$ with the [100]-axis of $Ga_{0.98}Mn_{0.02}As$ is shown schematically in Fig. 1(a). The reversal occurs via multiple 90° switches between the states labeled 1 to 4. Consequently, after two jumps the magnetization is reversed with respect to the original configuration. This is due to the perpendicular-biaxial anisotropy, where [100] and [010] are the easy axes. The biaxial anisotropy leads to an M-shaped major hysteresis loop in the magnetic birefringence, as shown in Fig. 1(c). Two jumps, at fields $H_{12} = 6$ mT and $H_{23} = 88$ mT, are clearly seen when the magnetic field is swept from minus to plus. Similar jumps occur at $H_{34} = -H_{12}$ and $H_{41} = -H_{23}$ when the field is swept in the opposite direction.

These magnetization jumps, appearing at magnetic fields $H_{IJ}$, are related to the sudden appearance of a single macroscopic domain with magnetization in the final state ($J$) oriented under 90° with respect to that of the initial state ($I$).[10] Such a behavior is confirmed by the sharp magnetization loops which were mapped using the MLD [Figs. 1(c) and 1(d)] and found to be independent of the spot size. The fields $H_{IJ}$ can be estimated through a characteristic pinning energy of such domains[8] ($\varepsilon$) using the following simple equation:

$$\frac{\mathbf{H}_{IJ} \cdot \mathbf{M}_J - \mathbf{H}_{IJ} \cdot \mathbf{M}_I}{2} = \varepsilon. \tag{1}$$

Based on this equation, we were able to measure precisely the sample orientation with respect to the magnetic field direction:

$$\frac{H_{23}}{H_{12}} = \tan(\varphi + 45°), \tag{2}$$

and obtain in our case $\varphi = 41°$.

When the field sweep is limited to $H_{41} < H < H_{23}$, two *mutually reversed* hysteresis loops appear instead of the major loop [see Fig. 1(d)]. These are referred to as minor hysteresis loops and characterize the magnetization switching between the states 1 and 2 (lower minor loop), as well as between states 3 and 4 (upper minor loop) as shown in Fig. 1(b).

Let us now turn to the single pulse switching behavior that is the main point of the present letter [Fig. 1(e)]. When a small field $H_0 < H_{23}$ is applied, the magnetization is brought in state 2. Any reduction of the coercivity at this stage will result in a decrease of $H_{23}$. As soon as $H_0 > H_{23}$, switching from state 2 to state 3 occurs. This is the switching process we now focus on. The reduction of the coercivity may be induced by heating of the sample, using either a current or laser pulse. The latter has been exploited in this work.

The essential figure demonstrating single pulse magnetization switching is Fig. 2. We measured the minor birefringence hysteresis loop of Fig. 1(d) before and after optical excitation of our $Ga_{0.98}Mn_{0.02}As$ layer with a single ultrashort (100 fs) laser pulse. Before the photoexcitation, the lower minor hysteresis loop (open symbols) is measured. After a single pulse excitation in a magnetic field of 75 mT, the magneto-optical hysteresis has been turned to the upper mi-
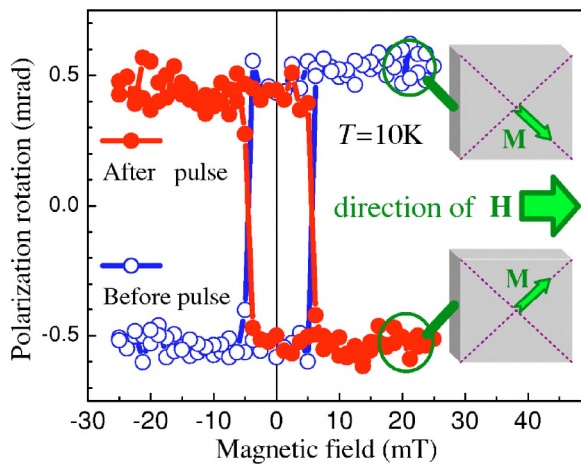
FIG. 2. (Color online) Minor hysteresis loops of the $Ga_{0.98}Mn_{0.02}As$ sample, recorded before (open symbols) and after (solid symbols) single optical pulse with a duration of 100 fs, are shown together with corresponding magnetization orientations (insets).

nor loop (solid symbols). This observation clearly demonstrates that ultrafast laser excitation is able to induce far more drastic changes than just to switch the orientation of the magnetization. In the present experiment, the hysteresis loop as a whole is completely reversed.

The laser-induced switching requires the application of an external magnetic field $H_0$ and an intense optical excitation with pump fluence $P$. We have analyzed the influence of these parameters on the hysteresis reversal, finding as general tendency the larger $P$ the weaker $H_0$. For $P = 150$ mJ/cm$^2$ the switching was found to happen for magnetic fields $H_0 > 50$ mT [Fig. 3(a)]. In a constant magnetic field of $H_0 = 75$ mT the switching occurred only for pump fluences $P > 130$ mJ/cm$^2$ [Fig. 3(b)]. We did not detect any switching for pump fluences below $P < 110$ mJ/cm$^2$, while for $P$ between 110 and 130 mJ/cm$^2$ the switching was not complete. The deviation from ideal step-like behavior could point to an inhomogeneous distribution of Mn ions within the pump spot. In addition, one should note that a strong
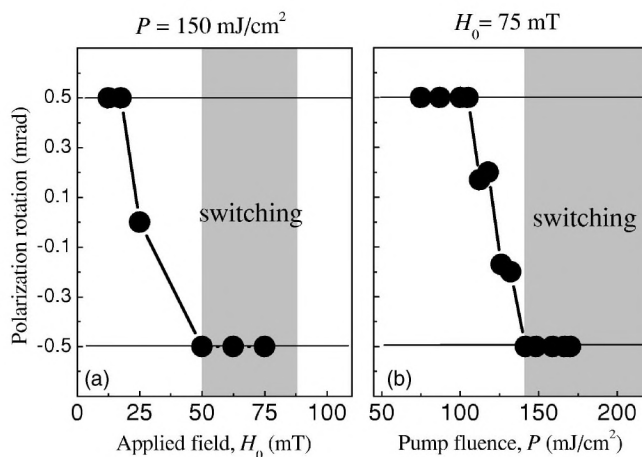
laser pulse may introduce large local stresses that may remain present even after heat relaxation and cannot be wiped out by moderate fields.

These observations conclusively demonstrate that by applying ultrashort optical pulses we can manipulate the magnetization in $Ga_{0.98}Mn_{0.02}As$ layer between its four metastable states, which can be used for application in a magneto-optical memory. In this context, it is important to note that there are two different time scales that characterize the switching process. The first one is the excitation time, or in our case the duration of the laser pulse, $\tau = 100$ fs. The other, $\tau_R$, is the relaxation time the magnetization needs to relax into its new equilibrium state after the laser pulse is turned off. The latter may vary in a wide range, depending on the specific relaxation mechanism, and usually $\tau_R > \tau$. The rate at which one can rewrite a bit of information is given by $R_{write} = 1/(\tau + \tau_R) \lesssim \tau_R^{-1}$, and one must know $\tau_R$ for a reliable estimate of this rate. In order to write $N$ different bits in parallel at once, however, a total writing time $T_{tot} = N\tau + \tau_R$ is sufficient. For a massively parallel memory ($N \rightarrow \infty$), the recording rate $R_{write} = N/T_{tot} \rightarrow \tau^{-1}$; i.e., $R_{write}$ is determined by the excitation time only. In this theoretical limit, one may achieve $R_{write} = 10$ THz. However, it should be understood that other factors, e.g. the time required to focus at another spot, can limit memory performance.

In summary, the ferromagnetic semiconductor (Ga,Mn)As possesses a combination of magnetic and optical properties, which enabled us to demonstrate that its magneto-optical hysteresis loop can be reversed with a single 100 fs laser pulse. This hysteresis reversal occurs in an applied magnetic field and only for pump fluences above a well-defined threshold value. These observations may be adopted in ultrafast magneto-optical recording.

FIG. 3. Polarization rotation of the linear magnetic dichroism of our $Ga_{0.98}Mn_{0.02}As$ sample after excitation with a single optical pulse, plotted (a) versus magnetic field $H_0$ at fixed pump fluence $P = 150$ mJ/cm$^2$, and (b) versus pump fluence $P$ at a fixed magnetic field $H_0 = 75$ mT. A polarization rotation of about $-0.5$ mrad corresponds to a switching event, and is indicated by the gray areas. Note, in panel (a), when $H_0 > 88$ mT, a switching event already occurs without optical pulse assistance [see Fig. 1(c)].

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