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Phase-change control of ferromagnetism in GeTe-based phase change magnetic thin-films by pulsed laser deposition

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Phase change control of ferromagnetism in $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ thin film prepared by pulsed laser deposition is investigated. The magnetic property of Fe doped phase change material GeTe is found to vary with phase change between amorphous and crystalline states corresponding to the variation of conductance during phase change. The results indicate that a fast control of ferromagnetism by phase change can be realized. The measurement of temperature dependent magnetization shows a long range ferromagnetic interaction in ordered crystalline phase and a short range ferromagnetic interaction in frustrated amorphous phase, which is consistent with phase change. © 2011 American Institute of Physics. [doi:10.1063/1.3628660]

Phase change material has been widely used in optical and electrical devices and memory.^{1–3} It is known as rapid and reversible control of optical and electrical properties by phase change. However, the magnetic property is not easy to be altered once a certain magnetic material has been prepared.⁴ Extensive studies for controlling magnetic property by external physical methods have been done, including the illumination and electrical field control of ferromagnetism in III–V diluted magnetic semiconductors (DMSs).^{4,5} It is expected that once the spin is effectively injected into the nonmagnetic phase change material, a fast and reversible control of ferromagnetism by phase change could be realized, which will lead new applications in data storage, sensor, logical devices, and multi-functional spintronic devices.^{6,7} Thus, the control of ferromagnetic property will become convenient because many methods inducing phase change can be employed, including light, electricity, heat, and even pressure.⁸

The principle of phase change control of ferromagnetism seems simple due to the alteration of structure during phase change, but the mechanism behind the change of ferromagnetic exchange interaction during phase change is still unknown. In this study, we focus on GeTe-based phase change magnetic material by introducing elemental Fe into GeTe which is known as a promising alternative phase change material to $\text{Ge}_2\text{Sb}_2\text{Te}_5$ due to the reduction of the switching size.⁹ So far, extensive studies have been limited to the materials system of $\text{Ge}_{1-x}\text{Mn}_x\text{Te}$ and $\text{Ge}_{1-x}\text{Cr}_x\text{Te}$.^{10,11} There is scarcely report combining rapid phase change feature with magnetic property in GeTe-based material. Therefore, it is highly desirable to study GeTe-based phase change magnetic material to determine whether its magnetic property is tunable by phase change, similar to the variation of optical and electrical properties with phase change. Moreover, the insight for the change of magnetic property is given in this letter.

$\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ thin films were deposited on Si (001) substrate by pulsed laser deposition. The $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ target was ablated using KrF excimer laser. The background pressure was achieved at 1×10^{-6} Torr during deposition. The structure of the films was analyzed by x-ray diffraction (XRD) with Cu $K\alpha$ radiation and transmission electron microscopy (TEM). The chemical composition was identified by energy dispersive spectroscopy (EDS). The magnetic measurement was performed using a superconducting quantum interference device (SQUID) magnetometer. The electrical property was determined by Hall effect measurement in the van der Pauw four-point configuration.

Figures 1(a) and 1(b) show the cross-sectional TEM image and selected area electron diffraction (SAED) pattern of the $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ film prepared at 300 °C. The image shows a sharp interface between $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ film and Si substrate and a columnar growth of the film. The SAED pattern of the polycrystalline $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ corresponds to a rhombohedral lattice,¹² which is also confirmed by the XRD measurement (lower inset of Fig. 2).

To investigate the control of ferromagnetism by phase change in $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ thin films, we have prepared some samples with different phases. We did not use laser or quenching to induce crystalline–amorphous–crystalline involving

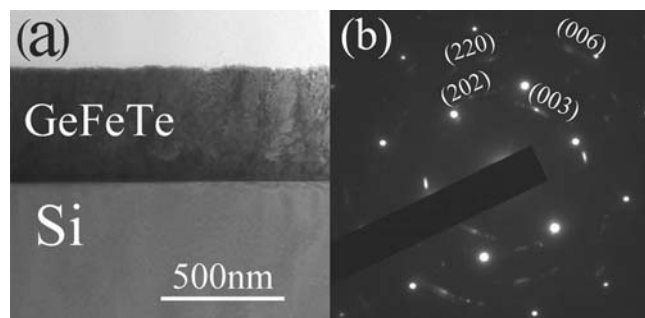


FIG. 1. (a) The cross-sectional bright field TEM of $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ thin film and (b) the SAED pattern of $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ thin film.

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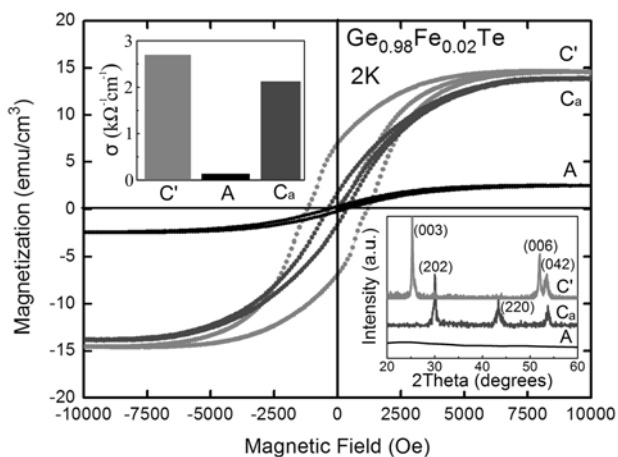


FIG. 2. (Color online). Magnetization curves of $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ films at 2 K for three states: C' , A, and C_a . C' : *in situ* deposited crystalline phase at 300°C , A: deposited amorphous phase at 150°C , C_a : the crystalline phase after annealing at 280°C for 30 min. The upper inset shows the variation of electrical conductivity with phase change. The lower inset is the XRD patterns of different phases.

dynamic phase change process in single sample because neither method could induce complete crystalline nor amorphous for a large area $1\text{ mm} \times 3\text{ mm}$ requested by SQUID measurement. Instead, the crystalline $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ thin film prepared by *in situ* deposited at 300°C was denoted as C' , the deposited amorphous state at 150°C denoted as A, while the obtained crystalline state after annealing the amorphous state at 280°C for 30 min was denoted as C_a .

The lower inset in Figure 2 shows the XRD patterns for different phases of $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ thin films. No obvious peak appears in curve A, indicating that an amorphous phase is formed. The peaks of (202), (220), and (042) appear in the XRD pattern of crystalline phase C_a , which corresponds to a rhombohedral GeTe structure.¹² The lattice planes (202) and (042) also appear in crystalline phase C' though they are rather weak due to a preferred (001) oriented growth. The lattice plane (220) does not appear in the XRD pattern of C' but appears in the SAED pattern shown in Fig. 1(b). These facts provide evidences that the crystalline phases C_a and C' have the same rhombohedral structure. One reason for the discrepancy between the two crystalline states is the crystallinity just like the two crystalline states during a reversible switching period used in phase change memory. Nevertheless, the slight difference in composition of the two crystalline phases measured by EDS could also contribute to the discrepancy.¹³ The upper inset in Figure 2 shows the dependence of electrical conductivity on the phase change between amorphous and crystalline states. The significant change of electrical conductivity between amorphous and crystalline phases is a typical phenomenon going with phase change in GeTe-based phase change material.¹⁴ Note that the electrical conductivity of the amorphous Fe-doped GeTe deposited at 150°C is much higher than that of amorphous GeTe prepared at room temperature. In principle, amorphous thin films deposited at higher temperature may consist of very small crystallites.¹⁵ Thus, it is understandable that the conductivity in our amorphous $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ thin film would be higher than that of the reported amorphous GeTe deposited at room temperature. Additionally, the carrier concentra-

tion of $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ thin film reported here is increased due to Fe doping into GeTe, which can increase electrical conductivity at amorphous state accordingly. This phenomenon is similar to the previous observation in metal doped $\text{Ge}_2\text{Sb}_2\text{Te}_5$.^{7,16}

The magnetization curves for the three states (C' , A, and C_a) at 2 K with a magnetic field parallel to the film surface are shown in Fig. 2. The typical hysteretic behavior of the three states indicates that the interaction between Fe ions is dominated by ferromagnetic exchange. The saturated magnetization M_s of amorphous state A is 2.5 emu/cm^3 . After annealing into crystalline state C_a , the M_s increases to 13.9 emu/cm^3 . The saturated magnetization M_s is quite close to the same rhombohedral crystalline state C' , which is 14.9 emu/cm^3 . The observed changes of M_s during the $C'-A-C_a$ process indicate that the ferromagnetism varies with phase change between amorphous and crystalline states. The discrepancy of saturated magnetization M_s and coercivity H_c between the two crystalline states C' and C_a is due to the crystallinity as shown in the XRD patterns (lower inset of Fig. 2). If this phase change magnetic material is used as the reduced switching area, its ferromagnetism will experience a reversible change during phase transition, just like the conductance. Therefore, not only the electrical property varies with phase change between amorphous and crystalline states but also the magnetic property differs between the two states. Phase change can be used as an effective method to control ferromagnetic property in $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ phase change magnetic material.

The alteration of magnetic property is apparent during phase change in $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ thin films. Another issue is how the ferromagnetic exchange interaction varies with phase change. We performed temperature-dependent magnetization for both amorphous (A) and crystalline (C') $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ films at an in-plane 500 Oe magnetic field, as shown in Fig. 3. The $M(T)$ behaviors are quite different between amorphous and crystalline $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ films below Curie temperature. The $M(T)$ curve is concave and experiences a rapid decrease in low temperature region for

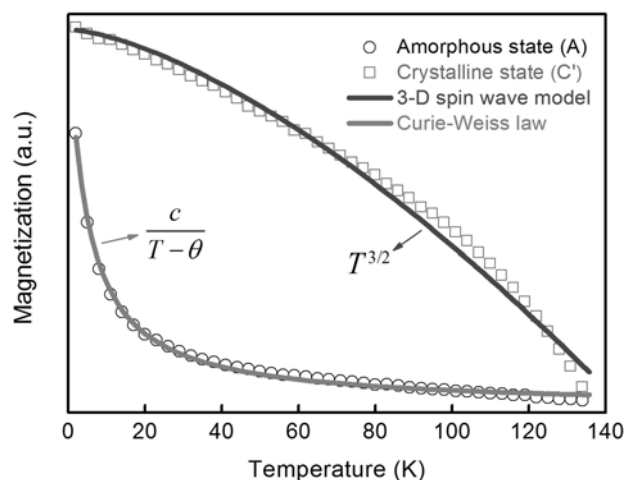


FIG. 3. (Color online) $M-T$ curves of the amorphous (circle) and crystalline (square) $\text{Ge}_{0.98}\text{Fe}_{0.02}\text{Te}$ thin films under an in-plane applied magnetic field 500 Oe. The lines are the theoretical simulation by a mean field theory for the crystalline film and Curie-Weiss model for the amorphous film, respectively.

amorphous Ge_{0.98}Fe_{0.02}Te film. On the other hand, for crystalline Ge_{0.98}Fe_{0.02}Te film, a convex and subdued $M(T)$ behavior is observed. The different $M(T)$ dependences imply different magnetic interaction mechanisms.¹⁷

The $M(T)$ data of amorphous and crystalline Ge_{0.98}Fe_{0.02}Te films were fitted by different models, which reveals the insight magnetic interaction. The fitting results were shown in Fig. 3. For the concave $M(T)$ curve of amorphous phase, a Curie-Weiss model, $\chi = \chi_0 + c/(T - \theta)$, where c is the Curie constant and θ is the Curie-Weiss temperature, can be used to well reproduce it, giving the fitting parameters $c = 11.85 \text{ emu K/cm}^3 \text{ Oe}$ and $\theta = -4.85 \text{ K}$.^{18,19} It implies a short range ferromagnetic order due to a lack of the carrier concentration to mediate the homogeneous ferromagnetic exchange interaction, which is common in poor-conductance DMSs.²⁰ In our amorphous Ge_{0.98}Fe_{0.02}Te film, the conductivity is low as can be seen in the upper inset of Fig. 2. Thus, the short range ferromagnetic interaction is determined by the frustrated amorphous structure. In contrast, the $M(T)$ curve of crystalline Ge_{0.98}Fe_{0.02}Te film can be well fitted by a standard 3-D spin wave model which predicts $M(T) = M_0 - 0.117u_B(k_B T/2S J d^2)^{3/2}$, where M_0 is the zero temperature magnetization, d is the spacing between magnetic ions, and J is the exchange interaction, which is expected for a homogeneous ferromagnet.²¹ The value of exchange integral J is estimated to be between 0.504 eV (for Fe²⁺, $S = 4/2$) and 0.630 eV (for Fe³⁺, $S = 5/2$). It is the typical value for IV-VI DMSs.²² The $M(T) \sim T^{3/2}$ dependence and estimated exchange interaction J demonstrate that the crystalline Ge_{0.98}Fe_{0.02}Te film is a uniform long range ferromagnetic system.²³ Also note the conductivity from the upper inset of Fig. 2, it is large in the crystalline phase (C') and the measured hole concentration reaches up to $1.21 \times 10^{21} \text{ cm}^{-3}$. The ferromagnetism is enhanced by the high carrier concentration via the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction in crystalline state. Therefore, we can conclude that magnetic interaction mechanism of phase change control of ferromagnetism is the change of short range and long range ferromagnetic exchange interaction between amorphous and crystalline phases, which is consistent with the characteristics of amorphous and crystalline structures.

In summary, a GeTe-based phase change magnetic material Ge_{0.98}Fe_{0.02}Te has been presented, which exhibits different magnetic property as well as electrical property

between amorphous and crystalline phases. The reversible control of ferromagnetism by phase change is found to be an alternative change of short range and long range ferromagnetic exchange interactions between amorphous and crystalline structures. The ferromagnetism seems to be consistent with a certain state during phase change. We may expect a new multi-functional spintronic device once this phase change magnetic material is used as the switching area in a phase change random access memory.

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