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Ultrafast Modulation of Magnetization Dynamics in Ferromagnetic (Ga, Mn)As Thin Films

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Abstract: Magnetization precession induced by linearly polarized optical excitation in ferromagnetic (Ga,Mn)As was studied by time-resolved magneto-optical Kerr effect measurements. The superposition of thermal and non-thermal effects arising from the laser pulses complicates the analysis of magnetization precession in terms of magnetic anisotropy fields. To obtain insight into these processes, we investigated compressively-strained thin (Ga,Mn)As films using ultrafast optical excitation above the band gap as a function of pulse intensity. Data analyses with the gyromagnetic calculation based on Landau-Lifshitz-Gilbert equation combined with two different magneto-optical effects shows the non-equivalent effects of in-plane and out-of-plane magnetic anisotropy fields on both the amplitude and the frequency of magnetization precession, thus providing a handle for separating the effects of non-thermal and thermal processes in this context. Our results show that the effect of photo-generated carriers on magnetic anisotropy constitutes a particularly effective mechanism for controlling both the frequency and amplitude of magnetization precession, thus suggesting the possibility of non-thermal manipulation of spin dynamics through pulsed laser excitations.

1. Introduction

Investigation of multiple functionalities of ferromagnetic semiconductors in the picosecond range lays the ground for future opportunities for high-speed spintronic devices. Time-resolved magneto-optical Kerr effect (MOKE) spectroscopy has been widely used for studying ultrafast photon-induced collective spin dynamics in ferromagnetic semiconductors. Based on this technique, Mn-based III–V ferromagnetic semiconductors have been extensively investigated from the point of view of both fundamental physics and spintronic applications [1–6]. Ultrafast magneto-optical investigation of collective spin excitations in ferromagnetic (Ga,Mn)As thin films has demonstrated the possible multi-functionality of ferromagnetic semiconductors when incorporated in spintronic devices [4,7–9]. Transient modulation involving either thermal or non-thermal mechanisms investigated by laser excitation has been shown to contribute to ultrafast coherent precession of magnetization [10–23]. Modulation of magnetic anisotropy stimulated by transient increases of either local temperature or photocarrier density in (Ga,Mn)As has demonstrated multiple possibilities of modulating the precession of magnetization by linearly polarized pump pulses.

Recently, several types of ultrafast mechanisms for manipulating coherent spin excitations have been discovered in (Ga,Mn)As, such as optical spin transfer torque (OSTT), in which the angular momentum of photo-generated carriers is transferred to the collective magnetization of the system [10]; and optical spin-orbit torque (OSOT), in which torque is exerted on the local magnetic moments of Mn by non-equilibrium spin polarization of photo-excited holes through spin-orbit coupling [19]. Photoionization-like excitation of Mn^{2+} has been recently proposed to account for the photoinduced modulation of perpendicular effective field [23]. Intrinsically, one or more of such mechanisms could simultaneously trigger a collective precession of magnetization [4,7,14,21], and they would compete with each other as to how they contribute to the precession process. On the other hand, they may also compensate each other in their contributions to data storage in the magnetic recording. Previous studies reported that hole cooling after pulse excitation is one of the limiting factors for ultrafast control of carrier-mediated magnetization precession, while thermal dissipation after laser heating is considered as the barrier for the high speed of data storage [11–14,24–27]. In this sense, the interplay between such thermal and non-thermal modulations upon laser pulse excitation is one of key issues in the context of ultrafast manipulation of magnetization.

In this paper, we report a study by means of time-resolved magneto-optical spectroscopy of photo-induced collective spin dynamics triggered by both thermal and non-thermal mechanisms. Using different intensities of linearly polarized optical excitation, we could relate the influence of both laser heating and photo-carrier generation on the precession of magnetization precession, and show how this is connected to modulations of the in-plane and out-of-plane anisotropy fields that result from such excitation. Specifically, for moderately-doped (Ga,Mn)As films with in-plane compressive strain, it is shown that the variation of precession frequency and precession amplitude depend on the superposition of both thermal and non-thermal effects through anisotropy fields of this material. Theoretical calculations show that the out-of-plane anisotropy field via non-thermal mediation mainly contributes to nonlinear variation of magnetization precession, thus pointing to the important role of ultrafast optical spin manipulation in ferromagnetic materials via non-thermal paths.

2. Materials and Methods

Ferromagnetic $Ga_{0.964}Mn_{0.036}As$ films with thickness of 97 nm were grown by means of low-temperature molecular-beam epitaxy (LT-MBE). The layers were grown on [001]-oriented GaAs substrates. The hole concentration p of the as-grown film was estimated to be ~2 × 10^{20} cm^{-3}. After annealing at 250 °C in N_2 for 1 h, p was estimated to increase to ~3 × 10^{20} cm^{-3}. Superconducting quantum interference device (SQUID) measurements showed that the Curie temperatures T_C of the annealed and as-grown samples were ~80 K and ~60 K, respectively. The magnetic anisotropy properties of the investigated (Ga,Mn)As layers were characterized by ferromagnetic resonance (FMR) measurements at different temperatures. Time-resolved pump-probe magneto-optical Kerr effect was measured using a Ti:sapphire laser with repetition rate of 80 MHz and spectral width of 10 nm. Pump-induced magnetization dynamics of Mn spins were studied via time-delayed probe pulses. Both the pump pulse and the probe pulse were linearly polarized with the same photon energy ranging from 1.53 eV (810 nm) to 1.58 eV (785 nm). The pump intensity was varied from 1.8 μJ/cm^2 to 28 μJ/cm^2, and the probe intensity was fixed at 0.6 μJ/cm^2. The samples were mounted in a Janis subcompact cryostat. The experiments were performed at 10 K unless noted otherwise. There is no external magnetic field applied in the experiments.

3. Results

Ultrafast photon excitation of (Ga,Mn)As ferromagnetic film can strongly disturb the equilibrium between holes, localized Mn spins, and the lattice [14]. On the subpicosecond time scale, laser pulse excitation leads to a coherent phase of nonequilibrium photoexcited carriers. However, the well-defined coherent phase of carriers quickly decays due to electron–electron, hole–hole, and electron–hole scattering. In this extremely short time regime, the time evolution of the coherent phase of

nonequilibrium photoexcited carriers is controlled by nonlinear optical excitation, and the carrier energy distribution function cannot be characterized by a temperature. After the carriers transfer their excess energy to the lattice (within a few picoseconds), the system acquires a quasi-equilibrium temperature. The enhancement of both the local lattice temperature and the hole concentration by photoexcitation can then modulate the magnitude of magnetic anisotropy fields, which reorients the direction of the easy axes, thus triggering the magnetization dynamics on a time scale of ~100 ps.

Magnetization precession of the coupled Mn spin system triggered by such magnetic anisotropy modulation is distinct from photomagnetic phenomena such as optical-spin-transfer-torque (OSTT). Specifically, it requires no injection of angular momentum, and can thus be triggered by a linearly-polarized laser pulse. Figure 1 shows a typical magnetization precession response to such a pulse at 10 K for the annealed (Ga,Mn)As sample excited by a linearly-polarized pump beam at 1.58 eV. As shown in Figure 1a, the temporal evolution of the photoexcited magnetization observed by time-resolved magneto-optical measurement in ferromagnetic (Ga,Mn)As can be fitted by an exponentially damped sine function superimposed on a pulse-like function [7,21],

$$\theta_k = a + be^{-t/t_0} + Ae^{-t/\tau_D}sin(\omega t + \phi), \tag{1}$$

where A, τ_D, ω, and ϕ represent, respectively, the oscillation amplitude, the relaxation time of magnetization, the frequency of oscillation, and the phase of the magnetization oscillation; a is the offset of background; and b and t_0 are the amplitude and decay time parameters of the pulse-like signal, charactering the slow recovery process. Earlier studies reported that the pulse-like signal which persists above the Curie temperature is related to the recovery process of nonequilibrium electron–hole pairs in the GaAs substrate [10,28], and only the oscillatory part is directly related to the ferromagnetic order of (Ga,Mn)As [12]. The numerical fitting result by the Landau-Lifshitz-Gilbert (LLG) equation shown in Figure 1b is focused on the precession part of the observed Kerr rotation, which reflects the uniform precession of magnetization in the annealed (Ga,Mn)As film [21]. The collective spin excitation and modulation of the dynamics of precession discussed below will mainly focus on the characteristics of the oscillatory signal modulated by laser excitation.
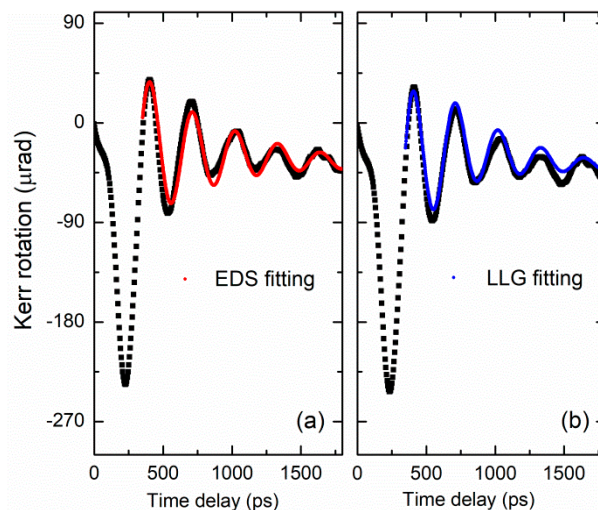

Figure 1. (Color online) Temporal profile of Kerr rotation excited by linearly-polarized pump beam at 1.58 eV with an excitation intensity of 10.9 $\mu J/cm^2$ for the annealed (Ga,Mn)As sample. (**a**) The solid line (red color) shows the best fit by an exponentially-damped sine function. (**b**) The solid line (blue color) shows numerical fitting result to LLG equation. The initial pulse-like-signal persists to above the Curie temperature, and is associated with non-equilibrium electron–hole pairs in the GaAs substrate. The fitting is focused on the oscillatory part of the Kerr rotation, which represents a uniform precession of magnetization in the (Ga,Mn)As film [21].

The observed dependences of the precession frequency and precession amplitude on pump intensity are shown in Figure 2. One can clearly see the nonlinear behavior of precession frequency and precession amplitude as the pump intensity increasing from 1.8 µJ/cm^2 to 28 µJ/cm^2. According to the LLG equation, it is the total effective magnetic field that determines the precession of magnetization. In principle, the total effective magnetic field includes the exchange field, the demagnetization field, the external magnetic field, and magnetic anisotropy fields [4]. Since the precession and relaxation of hole spins are much faster than those of Mn spin, the *p-d* exchange field itself cannot affect the precession frequency [19,29]. Additionally, the signal of magnetization precession has no relationship to the nonequilibrium electron-hole pairs that could also be generated in GaAs substrate [12,30]. Moreover, the ultrafast demagnetization is characterized by the subpicosecond time scale, so it will not influence the magnetization precession on the time scale of hundreds of picoseconds either [27,31]. Therefore, when the external magnetic field is absent, it is the optical excitation induced by magnetic anisotropy field modulation that mainly determines the value of precession frequency. That is, the optical absorption results in a transient variation of magnetic anisotropy fields (and thus of the magnetization) in the (Ga,Mn)As layer due to transient variation of both the lattice temperature and photo-excited carrier density. Thus, the thermal effect arising from laser heating and the non-thermal effect due to photo-excitation of carriers can affect the frequency of magnetization precession by respective changes in magnetic anisotropy fields caused by these two processes [13,14,21,23].
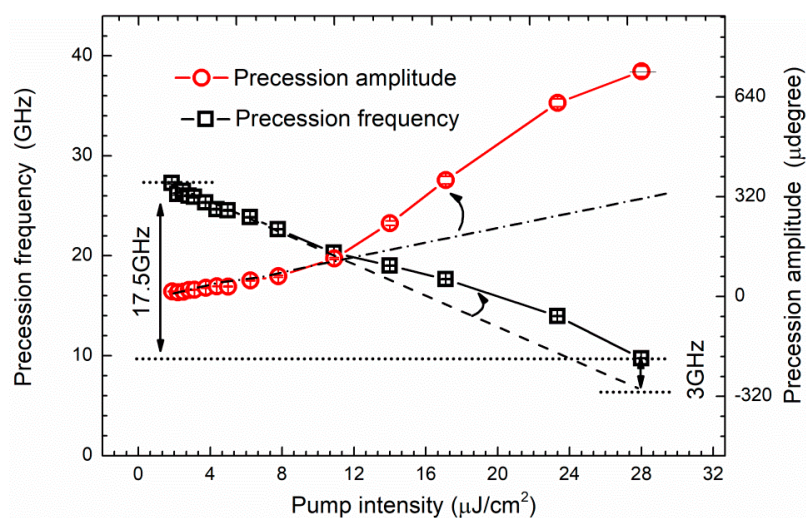

Figure 2. (Color online) Precession frequency and precession amplitude as a function of pump intensity for the annealed sample, obtained with linearly-polarized 1.58 eV pump beam at 10 K. No external magnetic field was applied. Pump intensity was varied from 1.8 µJ/cm^2 to 28 µJ/cm^2. The extended grey dashed line and grey dash-dotted are drawn to clearly show the nonlinear dependence of both the precession frequency and the amplitude on pump intensity.

According to ferromagnetic resonance (FMR) measurements, magnetic anisotropy fields are a function of temperature and carrier density [14,32]. For the investigated (Ga,Mn)As thin films, our previous ferromagnetic resonance (FMR) measurement has revealed that: the increase of both temperature and hole density leads to a decrease of in-plane anisotropy fields $H_{4||}(T, p)$ and $H_{2||}(T, p)$; The out-of-plane anisotropy field $4\pi M_{eff}$ is also reduced by increasing temperature, but is increased by the increase of hole concentration that results from annealing [21].

For our annealed (Ga, Mn)As thin film, we estimate that a temperature increase could induce average decreases of $4\pi M_{eff}$, $H_{4||}$ and $H_{2||}$ by about 21.51 Oe/K, 11.89 Oe/K and 0.45 Oe/K, respectively. In addition to the temperature influence, about 20% increase in carrier density (~20% × 10^{20} cm^{-3}) caused by annealing results in an average increase of $4\pi M_{eff}(p)$ of about 635.61 Oe, but in decreases of $H_{4||}(p)$ and $H_{2||}(p)$ of about 392.3 Oe and 148.85 Oe, respectively.

Note that the increase of pump intensity from 1.8 μJ/cm² to 28 μJ/cm² at 1.58 eV is observed to cause an increase of phototexcited carrier concentration of about 30×10^{17} cm^{-3}–120×10^{17} cm^{-3}, based on pulse absorption of photon energy. It can then be estimated that this 3–12% increase of photoexcited carrier density due to optical absorption results in an increase of $4\pi M_{eff}$ of about 95.34–381.4 Oe, and in decreases of $H_{4||}$ and $H_{2||}$ of about 58.84–235.38 Oe and 22.33–89.31 Oe, respectively.

It is known that the lattice constant of the zinc blende (Ga,Mn)As is larger than that of GaAs, so that the (Ga,Mn)As film is compressively strained when epitaxily grown on GaAs substrate, as sketched in Figure 3 [33]. For the compressively-strained (Ga,Mn)As film studied in this work, the influence of magnetic anisotropy field modulation on magnetization precession frequency can be directly described by the expression for the FMR frequency. With the magnetization lying in the plane of the sample, the precession frequency can be expressed as [32,34]:

$$\left(\frac{\omega}{\gamma}\right)^2 = H_{4||}\left(4\pi M_{eff} + H_{4||} + \frac{H_{2||}}{2}\right),\tag{2}$$

where γ is the gyromagnetic ratio (γ = 1.7588 Hz/Oe for g factor = 2.0023); $H_{4||}$ and $H_{2||}$ are the in-plane cubic and uniaxial anisotropy fields, respectively; and $4\pi M_{eff}$ is the effective perpendicular uniaxial anisotropy field, $4\pi M_{eff} = 4\pi M - H_{2\perp}$, where $H_{2\perp}$ is the out-of-plane perpendicular uniaxial anisotropy field. When the pump intensity increases, the in-plane magnetocrystalline anisotropy fields $H_{4||}$ and $H_{2||}$ are observed to decrease monotonically, presumably due to increasing temperature and increasing carrier density caused by increase of pump intensity. On the other hand, the dependence of $4\pi M_{eff}$ on temperature and carrier density shows variations in opposite directions (as argued above, $4\pi M_{eff}$ decreases with increasing temperature, but increases with increasing carrier density). This indicates that the magnetization precession frequency decreases linearly when the decreasing of in-plane magnetic anisotropy field dominates the behavior of magnetization precession either through thermal or nonthermal effects. This influence of the in-plane magnetic anisotropy field, however, cannot explain the nonlinear dependence of precession frequency on pump intensity shown in Figure 2. Thus, the strong increase of $4\pi M_{eff}$ due to increasing photoexcited carrier density offers the possibility of slowing down the rate of decline of magnetization precession frequency.
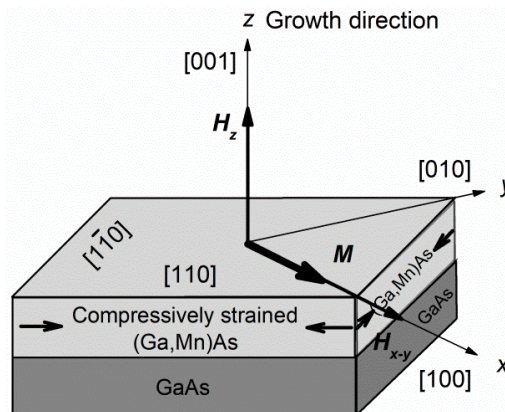

Figure 3. Coordinate system used in this paper. The initial magnetization lies in the plane of the sample along [100] direction. The [100] direction, [010] direction and [001] direction are defined as the directions of *x* axis, *y* axis, and *z* axis, respectively. The magnetic anisotropy field H_{x-y} is in the plane of the film, and the magnetic anisotropy field H_z is out of the plane of the film. The arrows in (Ga,Mn)As layer represent the direction of compressive strain in the plane.

To obtain a quantitative insight into the nonlinear influence of in-plane and out-of-plane magnetic anisotropy fields on magnetization precession, the magnetization precession was numerically simulated by employing LLG equation combined with two different MO effects, polar Kerr rotation

and magnetic birefringence (LLG-2MO) [13,23]. The micromagnetic modeling could be expressed as [14,28,35]

$$\frac{\partial \vec{M}}{\partial t} = -\gamma \vec{M} \times \vec{H}_{eff} + \frac{\alpha}{M} \vec{M} \times \frac{\partial \vec{M}}{\partial t}, \tag{3}$$

where M is the local magnetization contributed from Mn ions, H_{eff} is the effective field (which includes the in-plane and out-of-plane anisotropy fields), and α is the Gilbert damping coefficient. For compressively-strained (Ga,Mn)As films, the initial magnetization before excitation lies along the easy axes direction in the plane of the sample. In the simulation program, the initial state of the magnetization is set to be along the x axis in the *x-y* plane (i.e., in the plane of the film), and z axis is normal to the film plane, as seen in Figure 3. Thus, H_{x-y} and H_z correspond to the effect of $H_{4||}$ and $4\pi M_{eff}$, respectively. δH_{x-y} and δH_z correspond to the modulated value of $H_{4||}$ and $4\pi M_{eff}$, respectively. The total magneto-optical signal is the combined response caused by the out-of-plane component of the magnetization, which is sensed by the polar Kerr effect (PKE), together with the in-plane component of the magnetization, which is sensed mainly by the magnetic birefringence for which the magnetization-induced refractive index change is sufficiently significant to contribute to the observed MO signal in (Ga,Mn)As film [36–39]. To avoid complications arising from the superposition of PKE and MB in the gyromagnetic calculation, we analyze the behavior of M_z and M_{x-y} components separately. For details regarding the gyromagnetic theory and the simulation, we refer the reader to Reference [13].

In Figure 4, the LLG-2MO calculation results show that the precession frequency is a linear function of the in-plane magnetic anisotropy field H_{x-y}. These results suggest that, as the pump intensity increases, the reduction of the in-plane magnetic anisotropy field caused either by laser heating or by increasing carrier density only contributes to a linear decrease of precession frequency. However, Figure 5 shows an increase of the out-of-plane magnetic anisotropy field H_z can lead to an exponential increase of the precession frequency. From the FMR results, we note that only the out-of-plane anisotropy field increases due to the non-thermal effect of increasing carrier concentration due to the increase of pump intensity. Thus, the decrease of magnetization precession frequency can be regarded as a slow-down caused by an increase of the out-of-plane magnetic anisotropy field that follows from the increase of carrier density due to optical excitation. In this case, we conclude that the nonlinear variation of the precession frequency shown in Figure 2 results mainly from the non-thermal contribution of photoexcited carriers and their effect on the out-of-plane magnetic anisotropy field. It should be noted that the increase of photoexcited carrier density causes an increase in the value of $4\pi M_{eff}$ of about 380 Oe, but laser heating causes a decrease in $4\pi M_{eff}$ of about 21 Oe/K. If we ignore the saturation for thermal effects, optical excitation at an intensity of 15 μJ/cm^2 leads to a local lattice temperature increase of 10 K based on earlier studies [12].
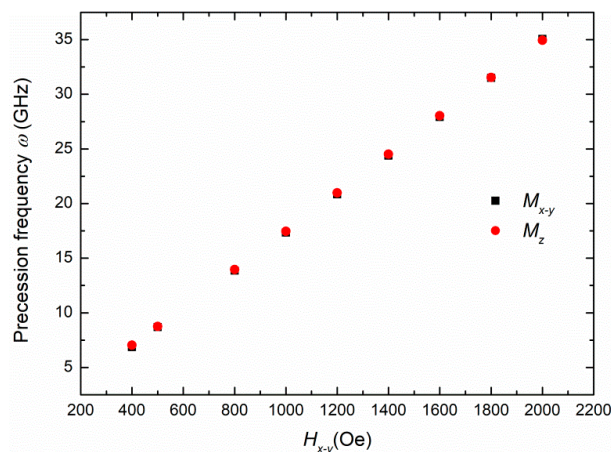

Figure 4. (Color online) Precession frequency simulated by LLG equation as a function the in-plane magnetic anisotropy field H_{x-y} for annealed compressively-strained (Ga,Mn)As film.
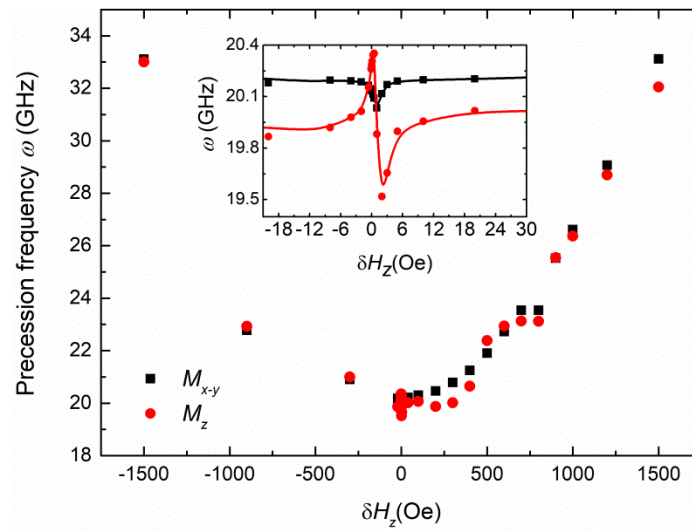
Figure 5. (Color online) LLG simulation of magnetization precession frequency as a function of the modulated value of out-of-plane magnetic anisotropy field (δH_z) for the annealed compressively-strained (Ga,Mn)As film. The inset shows details of precession frequency changes near zero field.

4. Discussion

As shown in Figure 2, with the pump intensity varying from 1.8 μJ/cm^2 to 28 μJ/cm^2, the joint thermal and non-thermal effects associated with the incident pulses cause the precession frequency to decrease by about 17.5 GHz. Although it is difficult to distinguish the accurate proportion of the thermal and nonthermal effects, the 3 GHz slowdown by $4\pi M_{eff}$ could be considered as the nonthermal contribution due to photoexcited carriers. We can therefore estimate that at least 17% of the observed effect on the precession frequency can be ascribed to nonthermal causes.

As already shown in Figure 2, the increase of precession amplitude is accelerated near the same pump intensity as the accelerated reduction of precession frequency. We simulated the change of magnetization precession under different in-plane and out-of-plane magnetic anisotropy fields. The extracted oscillation amplitude presents the change of amplitude with H_z and H_{x-y} in opposite directions, as shown in Figure 6. When pump intensity increases from 1.8 μJ/cm^2 to 28 μJ/cm^2, the decrease of in-plane magnetic anisotropy field is accompanied by a linear increase in precession amplitude. Thus, the nonlinear variation of precession amplitude seen in Figure 2 cannot be ascribed only to the change of in-plane magnetic anisotropy field. Recall that the increase of pump intensity decreases the out-of-plane magnetic anisotropy field through laser heating, but increases it through the increase of carrier concentration. In addition to the calculated results in Figure 6, only the increase of out-of-plane magnetic anisotropy field can lead to an increase of the oscillation amplitude. Thus, the accelerated increase of precession amplitude shown in Figure 2 can result from the increase of the out-of-plane magnetic anisotropy field. It is therefore suggested that non-thermal effects have the distinct advantage over thermal effects for manipulating the out-of-plane magnetic anisotropy field through pump intensity variation. This conclusion also supports our analysis above, i.e., that non-thermal effects due to photo-carrier excitation could effectively influence magnetization precession by their effect on the out-of-plane magnetic anisotropy field.
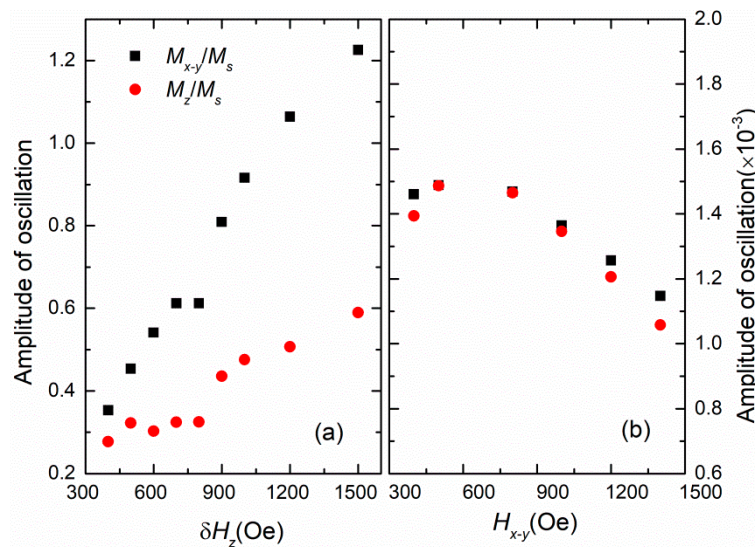
Figure 6. (Color online) Oscillation amplitude of magnetization precession as a function of modulated value of out-of-plane (**a**) and in-plane (**b**) magnetic anisotropy fields for the annealed and compressively strained (Ga, Mn)As thin film, obtained by LLG simulation.

5. Conclusions

We report a study of photoinduced collective spin excitation and their dynamics in ferromagnetic (Ga,Mn)As using time-resolved magneto-optical spectroscopy. Our observations indicate the effectiveness of non-thermal photo-carrier excitation via its effect on magnetization precession. For the (Ga,Mn)As films with in-plane compressive strain, the non-thermal modulation arising through out-of-plane magnetic anisotropy field causes a nonlinear influence on precession frequency and precession amplitude. Our results reveal the existence of a competing path of non-thermal effects occurring via out-of-plane magnetic anisotropy field, providing direct experimental evidence for the possibility of ultrafast nonthermal manipulation of magnetization dynamics in ferromagnetic (Ga,Mn)As by linearly polarized optical pulse excitation.

Author Contributions: H.L., X.Z. and X.L. conceived and designed this work. X.L., M.D. and J.F. prepared and characterized the sample. H.L. performed the experiment and analyzed the results. H.L., X.Z. and X.L. wrote the manuscript. All authors have reviewed and commented on the manuscript.

Conflicts of Interest: The authors declare no conflict of interest.

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