EXPERIMENTS PRECEEDING OUR OBSERVATIONS OF OSTT AND OSOT

(Ga,Mn)As and related ferromagnetic semiconductors are potentially ideal test-bed materials for exploring laser induced excitations of magnetization. Their direct-gap band structure allows for strong optical excitations of the electronic system, the photo-carriers can directly interact with magnetic moments via strong exchange coupling, the carrier mediated ferromagnetism produces large and tunable magnetic and magneto-optical effects, and the relatively simple band structure is favorable for identifying microscopic physical origins of the phenomena. Prior to our observations of optical spin transfer torque (OSTT) and optical spin-orbit torque (OSOT) presented in Ref. 1 and in this work, femtosecond laser pulse induced precession of magnetization in ferromagnetic (Ga,Mn)As has been reported by several groups [2–9]. In Ref. 4, magnetization precession was induced by laser pulses of a relatively weak intensity \( \approx 1 - 10 \ \text{µJ cm}^{-2} \) and ascribed to heating effects. On the other hand, in Refs. 2 and 8 the observed precession triggered by comparably weak laser intensities was attributed to an effect of photo-injected holes on the magnetic anisotropy. Apart from these competing views, the photo-carrier based interpretation cannot be reconciled with the theoretical and experimental understanding of static magneto-crystalline anisotropies in (Ga,Mn)As [8]. The interpretation would imply sizable changes of magnetic anisotropy in materials with equilibrium hole densities \( \approx 10^{20} - 10^{21} \ \text{cm}^{-3} \) induced by photo-injected holes of density as low as \( \approx 10^{16} \ \text{cm}^{-3} \). To compare with, e.g., electrical gating experiments [10, 11],
we recall that changes in the magnetic anisotropy are detected for field-induced hole accumulation or depletion of at least $\approx 10^{18} - 10^{19}$ cm$^{-3}$. We also point out that the original interpretation in Ref. 2 in terms of photo-carrier induced changes of in-plane anisotropy was subsequently revised by assuming [8,9] out-of-plane tilts of the easy-axis. Since out-of-plane easy-axis rotation is not consistent with measured trends in static magnetic anisotropies of the considered (Ga,Mn)As/GaAs materials this again illustrates that the non-thermal laser induced magnetization precession due to optically generated carriers in ferromagnetic semiconductors has remained in Refs. 2-9 an attractive yet unproven concept. In Ref. 1 and in this work we provide both the direct experimental evidence for the non-thermal effects and the microscopic interpretation of the physical origin of the observed optical torques.

**SAMPLES**

The time-resolved magneto-optical experiments described in the main text were performed in a large set of optimized (Ga,Mn)As epilayers. Ferromagnetic (III,Mn)V semiconductors, with (Ga,Mn)As as the most thoroughly investigated example, are materials grown under highly non-equilibrium conditions due to the extreme levels of Mn-doping. Therefore, a special care has to be taken when preparing the samples and when generalizing the experimental results obtained in a given sample to the universal behavior of this material system. Very recently we reported a systematic study of optical and magneto-optical properties of an optimized set of (Ga,Mn)As epilayers spanning the wide range of accessible substitutional Mn$_{Ga}$ dopings [12]. The optimization of the materials in the series, which is performed individually for each nominal Mn doping, minimizes the uncertainties in the experimental sample parameters and produces high quality epilayers which are as close as possible to uniform uncompensated (Ga,Mn)As mixed crystals. For each nominal Mn doping $x$, the growth and post-growth annealing conditions were separately optimized in order to achieve the highest Curie temperature $T_c$ attainable at the particular $x$. The highest $T_c$ criterion was found to lead simultaneously to layers with maximized uniformity and minimized compensation by unintentional impurities and defects [12]. All samples are in-plane magnets in which the cubic anisotropy competes with an additional uniaxial anisotropy. At very low dopings, the cubic anisotropy dominates and the easy axis aligns with the main crystal axis [100] or [010]. At intermediate dopings, the uniaxial anisotropy is still weaker but comparable in magnitude to the cubic anisotropy. At very high dopings, the uniaxial anisotropy dominates and the system has one strong easy-axis along the [-110] in-plane diagonal. We confirmed
that the laser pulse-induced precession of magnetization can be observed in all (Ga,Mn)As epilayers except in those with very low and very high doping levels where one of the anisotropies strongly dominates - see Table I.

<table>
<thead>
<tr>
<th>sample</th>
<th>$x$ (%)</th>
<th>$T_C$ (K)</th>
<th>$M_s$ (emu/cm$^3$)</th>
<th>Laser-induced precession</th>
</tr>
</thead>
<tbody>
<tr>
<td>F010</td>
<td>1.5</td>
<td>29</td>
<td>8.9</td>
<td>No</td>
</tr>
<tr>
<td>F008</td>
<td>2</td>
<td>47</td>
<td>11.6</td>
<td>Yes</td>
</tr>
<tr>
<td>F007</td>
<td>2.5</td>
<td>60</td>
<td>11.5</td>
<td>Yes</td>
</tr>
<tr>
<td>F002</td>
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<td>77</td>
<td>16.2</td>
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</tr>
<tr>
<td>F016</td>
<td>3.8</td>
<td>96</td>
<td>24.7</td>
<td>Yes</td>
</tr>
<tr>
<td>E101</td>
<td>4.5</td>
<td>111</td>
<td>27.8</td>
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</tr>
<tr>
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<td>132</td>
<td>33.3</td>
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</tr>
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</tr>
<tr>
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<td>9</td>
<td>179</td>
<td>63.7</td>
<td>Yes</td>
</tr>
<tr>
<td>F056</td>
<td>14</td>
<td>182</td>
<td>78.1</td>
<td>No</td>
</tr>
</tbody>
</table>

TABLE I: Table summarizing the basic characteristics of selected samples from the series of optimized materials: $x$ is nominal doping, $T_C$ is Curie temperature, $M_s$ is saturated magnetic moment. Last column shows if the impact of ultrafast laser pulse induces a precession of magnetization for the sample temperature of 15 K and the pump pulse fluence of about 30 $\mu$J.cm$^{-2}$.

For simplicity, we show in this Supplementary information only the helicity-independent signals measured in (Ga,Mn)As epilayer with a nominal doping $x = 3\%$, which is the sample that was used for a detailed analysis presented in the main text. The extensive information about the helicity-dependent signals can be found in our previous paper [1].

EXPERIMENTAL DETAILS

We investigated the laser-pulse induced dynamics of magnetization by a pump-and-probe magneto-optical technique. A schematic diagram of the experimental set-up is shown in Fig. 1. The output of a femtosecond laser is divided into a strong pump pulse and a weak probe pulse that are focused to the same spot on the measured sample. Laser pulses, with the time width of 200 fs and the repetition rate of 82 MHz, were tuned to 1.64 eV, i.e. above the semiconductor band gap, in order to excite magnetization dynamics by photon absorption. The pump pulses were usually circularly polarized (with a helicity controlled by a wave plate)
and the probe pulses were linearly polarized. The measured magneto-optical signals correspond to the probe polarization rotation induced by the pump pulses (see Fig. 1). The experiment was performed close to the normal incidence geometry ($\theta_i = 2^\circ$ and $8^\circ$ for pump and probe pulses, respectively) with a sample placed in a cryostat, which was placed between the poles of an electromagnet. The external magnetic field $H_{ext}$ was applied in the sample plane at an angle $\varphi_H$ with respect to the [100] crystallographic direction in the sample plane (see Fig. 1).

![Diagram](image)

**Fig. 1.** Schematic diagram of the experimental set-up for a detection of the magnetization precession induced in (Ga,Mn)As by an impact of the circularly polarized femtosecond laser pump pulse. Rotation of the polarization plane of reflected linearly polarized probe pulses is measured as a function of the time delay $\Delta t$ between pump and probe pulses. The orientation of magnetization in the sample is described by the polar angle $\varphi$ and azimuthal angle $\theta$. The external magnetic field $H_{ext}$ is applied in the sample plane at an angle $\varphi_H$.

**DETERMINATION OF THREE-DIMENSIONAL MAGNETIZATION VECTOR TRAJECTORY FROM DYNAMICAL MAGNETO-OPTICAL SIGNALS**

In this part we describe in detail how we are able to deduce the magnetization movement in the sample plane, $\delta \varphi(\Delta t)$, out of the sample plane, $\delta \theta(\Delta t)$, and the change of the magnetization magnitude, $\delta M_s(\Delta t)/M_0$, from the measured dynamical MO signals *without any numerical modeling* [13].

In (Ga,Mn)As there are two magneto-optical (MO) effects that are responsible for the measured signal [13]. In the following we will concentrate on the rotation of the polarization plane $\Delta \beta$ of the reflected linearly polarized light (see Fig. 3(b) for a definition of the probe pulses polarization plane orientation $\beta$) but the same apply also for the change of the light ellipticity (see below). We will also limit the discussion to the case when the incident light is close to the normal incidence. The first of the MO effects is the well-known polar Kerr effect (PKE), which is sometimes called magneto-optical Kerr effect (MOKE), where the rotation of
polarization occurs due to the different index of refraction for $\sigma^+$ and $\sigma^-$ circularly polarized light propagating parallel to the direction of magnetization - see Fig. 2(a). Consequently, $\Delta\beta$ due to PKE is proportional to the projection of magnetization to the direction of light propagation. We also note that this MO effect is linear in magnetization (i.e., the sign of the polarization rotation is changed when the direction of magnetization is reversed) and that $\Delta\beta$ is independent on $\beta$ [13].

![Fig. 2. Schematic illustration of two magneto-optical effects that are responsible for a rotation of the polarization plane $\Delta\beta$ of reflected light at normal incidence in (Ga,Mn)As. (a) Polar Kerr effect (PKE) that is due to the different index of refraction for $\sigma^+$ and $\sigma^-$ circularly polarized light propagating parallel to the direction of magnetization $M$. (b) Magnetic linear dichroism (MLD) that is due to the different absorption (reflection) coefficient for light linearly polarized parallel and perpendicular to $M$ if the light propagates perpendicular to the direction of $M$ [13].](image)

The second MO effect is the magnetic linear dichroism (MLD), which originates from different absorption (reflection) coefficient for light linearly polarized parallel and perpendicular to $M$, that occurs if the light propagates perpendicular to the direction of magnetization $M$ - see Fig. 2(b) [13]. Consequently, $\Delta\beta$ due to MLD is proportional to the projection of magnetization to the direction perpendicular to the direction of light propagation, $\Delta\beta$ is quadratic in magnetization (i.e., the sign of $\Delta\beta$ is not changed when the direction of magnetization is reversed) and it varies as $\sin(2\beta)$ [13].

For normal incidence of light, the simultaneous presence of two MO effects that are sensitive to the out-of-plane orientation of magnetization (PKE) and the in-plane orientation of magnetization (MLD) enables to perform a full three-dimensional stroboscopic reconstruction of magnetization vector movement from the measured dynamical MO signals [13]. In particular, different polarization dependences of PKE and MLD enable us to disentangle the pump-induced out-of-plane and in-plane motions of magnetization, respectively [13]. In equilibrium, the magnetization points to the easy axis direction, which
can be characterized by the polar angle $\varphi_0$ and azimuthal angle $\theta_0$ (see Fig. 3(b) for a definition of the coordinate system). The impact of the pump laser pulse induces a precessional motion of magnetization. The magnetization orientation at any time delay $\Delta t$ is given by $\varphi(\Delta t) = \varphi_0 + \delta \varphi(\Delta t)$ and $\theta(\Delta t) = \theta_0 + \delta \theta(\Delta t)$. The time-dependent position of magnetization leads to a time-dependent MO response of the sample that, moreover, depends on the orientation of polarization plane of probe pulses $\beta$ - i.e., the measured dynamical MO signals depend both on $\Delta t$ and $\beta$: $\delta MO(\Delta t, \beta)$. Consequently, it is possible to determine a time evolution of the magnetization vector orientation - without any numerical modeling - directly from the measured MO data [13]. In particular,

$$\delta \varphi(\Delta t) = \left[\delta MO(\Delta t, \varphi_0) - \delta MO(\Delta t, \varphi_0 - 90^\circ)\right]/\left(4P^{MLD}\right), \quad (1)$$

$$\delta \theta(\Delta t) = -\left[\delta MO(\Delta t, \varphi_0 - 45^\circ) + \delta MO(\Delta t, \varphi_0 - 135^\circ)\right]/\left(2P^{PKE}\right). \quad (2)$$

where $\delta MO(\Delta t, \varphi_0)$ is the pump-induced dynamical MO signal measured by probe pulses with $\beta = \varphi_0$; $\delta MO(\Delta t, \varphi_0 - 90^\circ)$ is the pump-induced dynamical MO signal measured by probe pulses with $\beta = \varphi_0 - 90^\circ$; etc. $P^{PKE}$ and $P^{MLD}$ are MO coefficients that describe the magnitude of PKE and MLD. Similarly, the magnetization magnitude is given by $M_s(\Delta t) = M_0 + \delta M_s(\Delta t)$ and the dynamics of the pump-pulse-induced demagnetization is given by [13]

$$\frac{\delta M_s(\Delta t)}{M_0} = \left[\delta MO(\Delta t, \varphi_0 - 45^\circ) - \delta MO(\Delta t, \varphi_0 - 135^\circ)\right]/\left(4P^{MLD}\right). \quad (3)$$

We note that for a quantitative evaluation of the magnetization motion from the measured MO signals it is essential to know the magnitude and the sign of the corresponding MO coefficients $P^{PKE}$ and $P^{MLD}$ in the investigated sample [13]. These coefficients can be directly measured if the magnetization is oriented by a strong external magnetic field to the requested position. For a measurement of $P^{PKE}$, the magnetization has to be oriented out-of-plane (i.e., $\theta = 0^\circ$) and any orientation of light polarization plane $\beta$ can be used. For a measurement of $P^{MLD}$, the magnetization has to be oriented in-plane (i.e., $\theta = 90^\circ$) and $\beta = \varphi_0 - 45^\circ$ has to be used [13]. In Fig. 3(a) we show the spectral dependence of $P^{PKE}$ and $P^{MLD}$ measured in a (Ga,Mn)As epilayer with nominal doping $x = 3\%$ that is the sample used for a
detailed study of the magnetization dynamics at different excitation intensities, as described in the main paper.

Fig. 3. (a) Spectral dependence of static PKE and MLD in (Ga,Mn)As epilayer with nominal doping $x = 3\%$, the arrow indicate the spectral position of laser pulses used in the time-resolved experiment. (b) The orientation of magnetization $\mathbf{M}$ in the sample is described by the polar angle $\varphi$ and azimuthal angle $\theta$; the orientation of the polarization plane $\mathbf{E}$ of probe pulses is described by the angle $\beta$.

In Fig. 4 we show MO hysteresis loops that were measured for the in-plane orientation of the external magnetic field (see the inset in Fig. 4). The observed $M$-shaped hysteresis loop is a signature of the existence of four energetically equivalent easy axis positions of magnetization in a epilayer. Prior to the time-resolved experiment, we prepared the magnetization in a state close to the [010] crystallographic direction (i.e., in an easy axis position labeled “1” in Fig. 4).

Fig. 4. Magneto-optical hysteresis loops measured in a (Ga,Mn)As epilayer with nominal doping $x = 3 \%$ at 15 K; the four energetically equivalent easy axis positions of magnetization are schematically labeled “1” to “4” in the inset.

The impact of a laser pulse on (Ga,Mn)As induces a precession of magnetization by two distinct mechanisms – the helicity-dependent and the helicity-independent [1].
separate these effects, we computed from the dynamical MO signals measured after excitation by \( \sigma^+ \) and \( \sigma^- \) circularly polarized pump pulses the helicity-independent \( [(\sigma^+ + \sigma^-)/2] \) and helicity-dependent \( [(\sigma^+ - \sigma^-)/2] \) signals. In Fig. 5(a) we show the as-measured data for \( \sigma^+ \) and \( \sigma^- \) excitation. The helicity-independent part of the signal, which is the main focus of this paper, is shown in Fig. 5(b). We note that the helicity-independent signal is identical to that measured with linearly polarized pump pulses [with any orientation of the polarization plane – see Fig. 5(b)]. We stress that the independence of the measured MO signal on the pump-light polarization excludes the possibility that the observed signal is due to inverse magneto-optical effects (e.g., the well known inverse Faraday effect). Moreover, we in our recent paper on OSTT [1] we clearly showed that inverse magneto-optical effects can be excluded as the origin of the observed optical torques in our experiments based on the comparison of measurements with below and above band-gap excitation energies.

![Figure 5](image_url)

**Fig. 5.** (a) Dynamical magneto-optical (MO) data measured for circularly \((\sigma^+, \sigma^-)\) polarized pump pulses. (b) Comparison of the helicity-insensitive \( [(\sigma^+ + \sigma^-)/2] \) part of the signals shown in (a) with the MO data measured with linearly polarized \((s, p)\) pump pulses. Excitation intensity \( I_0 = 7 \, \mu J/cm^2 \), sample temperature \( T = 15 \, K \), probe polarization orientation \( \beta = 0^\circ \), no external magnetic field applied.

The dependence of helicity-independent dynamical MO signal on the probe polarization orientation \( \beta \) is shown in Fig. 6(a). The measured data can be fitted well by the phenomenological equation

\[
\delta \text{MO}(\Delta t) = A \cos(f_{\text{Mn}} \Delta t + \delta)e^{-\Delta t/\tau_G} + Ce^{-\Delta t/\tau_p},
\]

where \( A \) and \( C \) are the amplitudes of the oscillatory and pulse function, respectively, \( f_{\text{Mn}} \) is the ferromagnetic moment precession frequency, \( \delta \) is the phase factor, \( \tau_G \) is the Gilbert damping time, and \( \tau_p \) is the pulse function decay time. All the measured data in Fig. 6(a) can be fitted
well by Eq. (4) with a one set of parameters $f_{Mn}$, $\tau_G$ and $\tau_p$. The dependence $A(\beta)$ obtained by this fitting procedure is displayed in Fig. 6(b). The position of the maximum in the dependence $A(\beta)$ at $\beta \approx 100^\circ$ corresponds to the equilibrium position of magnetization in the sample $\phi_0$ [13]. We recall that prior to this measurement we prepared the magnetization in the easy axis labeled “1” in Fig. 4, which close to [010] crystallographic direction.

Fig. 6 (a) Dynamics of the helicity-independent MO signal induced by an impact of pump pulse on the sample that was measured by probe pulses with different probe polarization orientations $\beta$ (points). (b) Probe-polarization dependence of the amplitude of the oscillatory part $A$ that was obtained by fitting the measured MO dynamics by Eq. (4) [lines in part (a)] with a fixed values $f_{Mn} = 5.4$ GHz, $\tau_G = 165$ ps, and $\tau_p = 880$ ps (points). The line is a fit (see Ref. 13 for details). The vertical arrow depicts the derived easy axis position in the sample. Excitation intensity $I_0 = 7 \mu$J.cm$^{-2}$, sample temperature $T = 15$ K, no external magnetic field applied.

In Fig. 7 we show the dynamics of magnetization that was deduced from the data depicted in Fig. 6(a) using Eqs. (1), (2) and (3) – in part (a) and (b) the time evolutions, and the corresponding polar plot are depicted, respectively.

Fig. 7 Magnetization real-space trajectory deduced from the MO data shown in Fig. 6. (a) Time evolution of $\delta\phi$, $\delta\theta$ and $\delta M_s/M_0$. (b) Orientation of magnetization at different times after the impact of the pump pulse; the equilibrium position of the easy axis is depicted by the grey spot.
In principle, the measured dynamical MO signals can contain information not only about the pump-induced magnetization change but also about the pump-induced change of the complex index of refraction (the so-called “optical part” of the MO signal) [14]. Consequently, the dynamics of both the rotation and ellipticity has to be measured and compared before the obtained magneto-optical signal is attributed to the magnetization dynamics [14]. In Fig. 8 we show the corresponding data measured at pump intensity $18I_0 = 126 \, \mu\text{J.cm}^{-2}$, which is in the middle of the investigated intensity range (see the main paper). The measured dynamics of the rotation and ellipticity are identical that confirms that the procedure described above can be meaningfully used for a reconstruction of the magnetization movement. In particular, we would like to note that the displayed data which were measured for $\beta = 105^\circ \approx \varphi_0$ provide direct information about the in-plane, $\delta\varphi$, movement of the magnetization [cf. Eq. (1)]. At low excitation intensities, the MO data measured for this value of $\beta$ show a monotonous rise of the (negative) dynamic MO signal for time delays $0 < \Delta t < 50$ ps - see Fig. 6(a) for intensity $I_0 = 7 \, \mu\text{J.cm}^{-2}$. Taking into account that $P_{\text{MLD}} < 0$ in the used spectral range [see Fig. 3(a)], this leads to $\delta\varphi > 0$ – i. e., the magnetization starts its movement towards the [-110] crystallographic direction in the sample (see Fig. 7). On the contrary, at high excitation intensities the measured MO signal has a opposite sign for a time delay $0 < \Delta t < 50$ ps - see Fig. 8 for intensity $18I_0 = 126 \, \mu\text{J.cm}^{-2}$ - and, therefore, $\delta\varphi < 0$ in the initial stages of the magnetization movements – i. e., the magnetization starts its movement towards the [010] crystallographic direction in the sample (see also Fig. 2 in the main paper).

![Fig. 8. Comparison of the laser pulse-induced change of the rotation and ellipticity; the data are normalized. Excitation intensity $I = 18I_0 = 126 \, \mu\text{J.cm}^{-2}$, sample temperature $T = 15$ K, probe polarization orientation $\beta = 105^\circ$, no external magnetic field applied.](image)
CALIBRATION PROCEDURE USED FOR EVALUATION OF THE PUMP-INDUCED TEMPERATURE AND HOLE CONCENTRATION INCREASE

Absorption of pump laser photons leads to a photo-injection of electrons and holes with identical concentrations. However, because the density of states is considerably larger for holes (due to their larger mass), the pump-induced change of the distribution function is much larger for electrons and, consequently, the measured change of complex index of refraction is dominated by electrons [15, 16]. Therefore, the dynamics of the reflectivity change $\Delta R/R$ — that is shown in Fig. 9(a) — provide information about the lifetime of photo-injected electrons in the investigated sample. From the measured data we can conclude that the population of photo-injected free electrons decays with a characteristic time constant (defined as a decrease to $1/e$) of $\approx 10$ ps. This rather short lifetime of free electrons is similar to that reported for the low temperature grown GaAs (LT-GaAs), which is generally interpreted as a result of a high concentration of nonradiative recombination centers induced by the low temperature growth mode of the MBE [17-19]. The nonradiative recombination of photo-injected electrons is accompanied by an emission of phonons (i.e., by a heating of the sample). Consequently, the measured decay time of $\Delta R/R$ corresponds to a rise time of the laser-induced transient change of the sample temperature $\delta T$. We note that the lifetime of the photo-injected holes can be expected to be longer than that of electrons because (Ga,Mn)As is a heavily $p$-doped material and, therefore, the traps for holes are at least partially saturated by the background holes.

We applied the following analysis for an evaluation of the laser-induced change of the hole concentration $\Delta \tilde{p}$. The initial value of $\Delta R/R$ can be regarded as a measure of the relative number of the photo-generated electrons (and thus also of the holes). It is apparent from the inset in Fig. 9(a) that the pump-induced change of the complex index of refraction is linear in a pump-pulse intensity up to $I \approx 25I_0 = 180 \, \mu J \cdot cm^{-2}$ after which the initial values of $\Delta R/R$ start to saturate. The thickness of the investigated (Ga,Mn)As film (20 nm) is considerably smaller than the penetration depth of the laser at the corresponding photon energy ($\approx 600$ nm). Consequently, the absorption of photons leads to a rather constant concentration of photo-injected carriers in (Ga,Mn)As along the growth direction, which can be estimated from the laser spot size on the sample, photon energy, and absorption and reflection coefficients of GaAs. On the other hand, also the carriers photo-injected in the substrate could, in principle, contribute to the measured signals but we have verified that the measured signals in (Ga,Mn)As samples on GaAs substrate are markedly different from that in a bare GaAs...
substrate. In Fig. 10 we show the estimated intensity dependence of the initial concentration of the excess holes $\delta p$.

Fig. 9 (a) Dynamics of pump pulse-induced reflectivity change $\Delta R/R$ at intensity $6I_0 = 42 \, \mu J.cm^{-2}$; Inset: Intensity dependence of the initial value of the reflectivity change at 15 K (points), line depicts the linear dependence. (b) Frequency of precessing Mn moments measured (without external magnetic field) as a function of base temperature at low excitation intensity $I_0 = 7 \, \mu J.cm^{-2}$ and as a function of the laser intensity at low base temperature of 15 K. This figure is re-plotted from the main paper for convenience.

Fig. 10 The pump-pulse induced concentration of the excess holes $\delta p$ and the transient temperature increase $\delta T$ as a function of the laser intensity, $I_0 = 7 \, \mu J.cm^{-2}$.

As already described above, the fast nonradiative recombination of photo-injected electrons induces a transient pump pulse-induced increase of the lattice temperature $\delta T$ within tens of picoseconds after the impact of the pump pulse and this temperature increase persists over hundreds of picoseconds. An estimate of $\delta T$ can be obtained from the measured precession frequencies because they correspond to magneto-crystalline anisotropy fields in the sample [1], which are rather strongly temperature dependent. In Fig. 9(b) we plot the
dependence of the precession frequency on the base sample temperature at low excitation intensity $I_0$ and on the laser intensity at low base temperature of 15 K. From the comparison of these two measurements we infer the magnitude of the transient temperature change $\delta T$ as a function of the laser intensity, which is shown in Fig. 10. (We note that consistent temperature vs. intensity calibration is obtained from the comparison of the intensity dependence of the pump-induced demagnetization and the temperature dependence of the remanent magnetization measured by SQUID.) Remarkably, the temperature increase saturates at a considerably lower pump intensity than the hole concentration increase does which enables us to identify clearly the influence of the latter in the magnetization dynamics (see the main paper). The physical reason for this saturation can be explained by a closer inspection of the measured intensity dependence of the dynamics of the reflectivity change $\Delta R/R$, which is shown in Fig. 11, and by its comparison with similar experiments performed in LT-GaAs [17-19]. When the laser intensity is increased not only the initial value of $\Delta R/R$ increases [see inset in Fig. 9(a)] but also the speed of the reflectivity change recovery is modified – below $\approx 10I_0$ the dynamics slows down considerably with the excitation intensity while above this value the dependence is much weaker. Similarly, in LT-GaAs the dynamics of the pump-induced change of reflectivity [17] and transmission [18, 19] were reported to be slower at higher excitation intensities, which was interpreted as a saturation of the corresponding trap centers [17-19]. Moreover, at high excitation intensities (above $\approx 10I_0$) the measured slopes of the reflectivity transients are not linear on the logarithmic scale but they are convex (see Fig. 11) that corresponds to an increase of the electron lifetime during the recombination process, which again can be ascribed to the saturation of the electron traps [17]. The exact microscopical origin of the trap centers in currently not clear. In LT-GaAs the carrier traps

Fig. 11 Intensity dependence of the pump pulse-induced reflectivity change dynamics at 15 K, $I_0 = 7 \mu$J.cm$^{-2}$. 
were assigned both to point defects (to single positive charge state of As anisite ($As_{Ga}^+$) and to Ga vacancy ($V_{Ga}$) in particular) and to arsenic precipitates [17-19] but also Mn interstitials ($Mn_I$) can play a role in the case of (Ga,Mn)As [20].

**ELIMINATION OF THERMAL EXCITATION MECHANISM BY EXTERNAL MAGNETIC FIELD**

![Graph](image)

Fig. 12 Suppression of thermal excitation mechanism and of precession damping by external magnetic field applied along the sample easy axis. (a) Time evolution of $\delta \varphi$ and (b) time evolution of $\delta \theta$; note that there are different x-scales in parts (a) and (b). Excitation intensity $24I_0 = 168 \mu J.cm^{-2}$, sample temperature $T = 15$ K,

The influence of an external magnetic field $H_{ext}$ applied along the easy axis in the sample is illustrated in Fig. 12. The initial value of the out-of-plane tilt $\delta \theta$ due to OSOT does not depend on $H_{ext}$ – see Fig. 12(a). On the contrary, the quasi-equilibrium in-plane tilt $\delta \varphi$ of the easy axis, which is induced by the transient temperature increase, is strongly suppressed by $H_{ext}$ – see Fig. 12(b) for time delays larger than $\approx 300$ ps. Moreover, the applied magnetic field suppresses also the precession damping. This behavior, which we observed also in the systematic MO study of micromagnetic parameters of (Ga,Mn)As [21], is quite typical for various magnetic materials. In our case, the most probable explanation seems to be the one that was used by Walowski et al. to explain the experimental results obtained in thin films of nickel [22]. They argued that in the low field range small magnetization inhomogeneities can be build – the magnetization does not align parallel in a weak externally applied field, but forms ripples [22]. Consequently, the measured MO signal, which detects sample properties averaged over the laser spot size, experiences an apparent oscillation damping because the magnetic properties (i.e., the precession frequencies [21]) are slightly differing within the spot size (see Fig. 6 and 7. in Ref. 22). On the other hand, for stronger external fields the sample is fully homogeneous and, therefore, the precession damping is not dependent on the applied...
field as expected for the intrinsic Gilbert damping coefficient [21]. This explanation is also fully in accord with the precession damping enhancement that we observed without $H_{\text{ext}}$ in the sample at high excitation intensity (see Fig. 2 in the main paper) where the pump-induced demagnetization can be expected to lead to an enhancement of the local magnetic inhomogeneities.

**THEORETICAL MODELING OF OSOT**

Absorption of pump laser pulse with a photon energy of 1.64 eV leads to photoinjection of charge carriers with a rather high excess energy (the low-temperature band gap energy of GaAs is 1.52 eV). Immediately after photoinjection, the carriers have a non-thermal distribution. The Fermi-Dirac distribution is established on a time scale from tens to hundreds of femtoseconds (mainly due to mutual collisions between the carriers) but the resulting characteristic carrier temperature significantly exceeds that of the lattice. The carrier temperature starts to cool down towards the lattice temperature (by an emission of phonons) on a time scale from hundreds of femtoseconds to several picoseconds and, finally, the non-equilibrium carriers recombine. During this process holes relax through the available states in the spin-split valence band of the ferromagnetic semiconductor. Consequently, a change in the occupation of the hole states, as compared to the equilibrium state in dark, leads – due to the spin-orbit coupling – to a non-equilibrium hole spin polarization which is misaligned with the equilibrium orientation of Mn moments. This non-equilibrium photo-hole polarization persists over the timescale of the hole recombination during which it exerts a torque on the Mn local moments via the kinetic-exchange coupling. The exact theoretical treatment of the mutual interplay of all the involved effects is rather complicated and, therefore, derivation of a full quantitative theory of OSOT is a challenging problem. (We note that in the case of OSTT the non-equilibrium spin-density of weakly spin-orbit coupled photo-electrons, producing the OSTT, is directly determined by the external polarizer - i.e., by the intensity, propagation axis, and helicity of the circularly polarized pump laser beam – which make the corresponding theory much simpler [1]). As a simplified theory of the non-equilibrium photo-hole spin polarization, related to OSOT by Eq. 2 in the main text, we consider the hole density dependent magnetic anisotropy fields (see Eq. 3 in the main text). The simplification is that we consider OSOT produced by fully equilibrated photo-holes acting on the local moments over the recombination time. Within this approximation, the misalignment of the non-equilibrium hole polarization with the equilibrium orientation of Mn moments has the same
physical origin as the dependence of the static magnetic easy-axis orientation on the hole density [23].

Figure 13: Computed in-plane angular dependence of the anisotropy energy $E$ (with respect to the energy along the crystallographic direction $[100]$) (a) and the anisotropy field $H_{an,\varphi} = -\delta E/\delta \varphi$ (b) in (Ga,Mn)As with 3% of Mn and strain $\varepsilon_{xy} = 0.01\%$ at 0 K. Inset: Dependence of the easy axis position on the concentration of holes.

In Fig. 13(a) we show the in-plane angular dependence of the anisotropy energy in (Ga,Mn)As, which was computed using the $k \cdot p$ kinetic-exchange Hamiltonian [23]. The position of the energy minimum (i.e., the easy axis position) depends strongly on the concentration of equilibrium holes [see the inset in Fig. 13(a)]. The femtosecond pump-pulse induced change of hole concentration $\delta p$ (from the sample equilibrium hole concentration at dark $p_0$) leads to an abrupt in-plane movement of the energy minimum and, consequently, to a misalignment of the energy minimum and the equilibrium orientation of magnetization $M$. The resulting anisotropy field, which is aiming at having them aligned again, has a zero out-of-plane component $H_{an,\theta} = -\delta E/\delta \theta$ and a non-zero in-plane component $H_{an,\varphi} = -\delta E/\delta \varphi$ [see Fig. 13(b)]. Consequently, the resulting impulse torque acting on $M$ is oriented in the out-of-plane direction (see Eq. (2) and Fig. 1(b) in the main paper) which explains why we observed the initial out-of-plane magnetization tilt for high excitation intensities [see Fig. 2(c) and (d) in the main paper]. On the other hand, at low excitation intensities the pump-induced temperature increase is considerably slower and the resulting temperature-related movement of the energy minimum (quasi-equilibrium easy axis) is in the opposite direction than in the case of the hole concentration-related movement [see Fig. 3(b) in the main paper]. This explains why we observed at low excitation intensities a much slower rise of the out-of-plane component of the precessing magnetization and towards the opposite direction than in the case of OSOT at high excitation intensity [see Fig. 2(a) and (b) in the main paper].
simplified analysis therefore provides a physically transparent qualitative description of the experimentally observed OSOT.

REFERENCES


