Experimental observation of the optical spin–orbit torque

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Electrical and optical control of magnetization are of central importance in the research and applications of spintronics. Current-induced angular momentum transfer or relativistic spin–orbit coupling provide efficient means by which electrical current driven through a ferromagnet can exert a torque on the magnetization. Ferromagnetic semiconductors like (Ga,Mn)As are suitable model systems with which to search for optical counterparts of these phenomena, where photocarriers excited by a laser pulse exert torque upon magnetization. Here, we report the observation of an optical spin–orbit torque (OSOT) in (Ga,Mn)As. The phenomenon originates from spin–orbit coupling of non-equilibrium photocarriers excited by helicity-independent pump laser pulses, which do not impart angular momentum. In our measurements of the time-dependent magnetization trajectories, the signatures of OSOT are clearly distinct from the competing thermal excitation mechanism, and OSOT can even dominate in (Ga,Mn)As materials with appropriately controlled micromagnetic parameters.

The seminal works on current-induced spin-transfer torque (STT) date back several decades and, the effect already plays a key role in commercially developed spintronic technologies. In current-induced STT, spin-polarized carriers are electrically injected into a magnetic object (for example, a ferromagnetic layer or domain wall) from an external polarizer, which is formed by one part of the non-uniform magnetic structure. The origin of STT can be understood from the non-relativistic physics of angular momentum transfer from the external polarizer to the magnetic object. The recently discovered optical spin transfer torque (OSTT) is the optical counterpart of STT, where the direction and magnitude of the non-equilibrium photocarrier spin density are governed again by the properties of an external polarizer. In the OSTT case these properties are the intensity, propagation axis and helicity of the circularly polarized pump laser pulse. In direct analogy to (adiabatic) STT, the coupled dynamics of the magnetization and the non-equilibrium photocarrier spin density can be written as

$$\frac{d\mathbf{M}}{dt} = \frac{j}{\hbar} \mathbf{M} \times \mathbf{s}$$

$$\frac{ds}{dt} = \frac{j}{\hbar} \mathbf{s} \times \mathbf{M} + P\mathbf{n}$$

where $j$ is the carrier–magnetic moment exchange coupling constant and $P$ is the rate at which carrier spins with orientation $\mathbf{n}$ are injected into the ferromagnet from the external polarizer. From the steady-state, non-equilibrium carrier spin density solution ($ds/dt = 0$), we obtain explicitly the direct relation between OSTT and the carrier spin injection rate $P$

$$\frac{d\mathbf{M}}{dt} = \frac{j}{\hbar} \mathbf{M} \times \mathbf{s} = P\mathbf{M} \times (\mathbf{n} \times \mathbf{M})$$

where $\mathbf{M}$ is the magnetization unit vector.

Current-induced spin–orbit torque (SOT) is a distinct relativistic phenomenon in which magnetization dynamics is induced in a uniform spin–orbit coupled ferromagnet in the absence of an external polarizer. In the SOT, the term corresponding to injection from the external polarizer is therefore not present, and the quantum-mechanical dynamics of the carrier spins is given by

$$\frac{d\mathbf{s}}{dt} = \frac{j}{\hbar} \mathbf{s} \times \mathbf{M} + \frac{1}{\hbar} \langle \mathbf{s}, H_{\text{so}} \rangle$$

where $\mathbf{s}$ is the carrier spin operator and $\langle \cdot \rangle$ denotes quantum-mechanical averaging. The carrier Hamiltonian, $H = H_0 + H_{\text{so}} + H_{\text{ex}}$, can be written as a sum of the non-relativistic non-magnetic term $H_0$, the carrier spin–magnetic moment exchange coupling $H_{\text{ex}} = JM\mathbf{s}$, and the spin–orbit term $H_{\text{so}}$. This implies that

$$\frac{ds}{dt} = \frac{j}{\hbar} \mathbf{s} \times \mathbf{M} + \frac{1}{\hbar} \langle \mathbf{s}, H_{\text{so}} \rangle$$

where $s = \langle \mathbf{s} \rangle$. The steady-state carrier spin density solution of equation (2) shows explicitly the spin–orbit coupling origin of SOT:

$$\frac{d\mathbf{M}}{dt} = \frac{j}{\hbar} \mathbf{M} \times \mathbf{s} = \frac{1}{\hbar} \langle \mathbf{s}, H_{\text{so}} \rangle$$

Apart from spin–orbit coupling, the SOT obtained from equation (2) also requires exchange coupling of the carrier spins to the magnetic moments; that is, it requires broken time-reversal symmetry of the exchange-split carrier bands. All these features of the SOT are common to both the previously discussed current-induced SOT and the optical spin–orbit torque (OSOT) reported in this Article.

We point out that the electrical and optical SOTs may differ in their specific contributions to $H_{\text{so}}$, which dominate the effect. In the Boltzmann linear-response transport theory of current-induced SOT, $\langle \cdot \rangle$ represents quantum-mechanical averaging constructed from the equilibrium eigenstates of $H$ and with the non-equilibrium steady state entering through an asymmetric redistribution of the occupation numbers of these eigenstates on the
Fermi surface due to applied electrical drift and relaxation. Because of this specific form of the asymmetric non-equilibrium charge redistribution with a conserved total number of carriers, the current-induced SOT requires broken inversion symmetry terms in $H_{\text{so}}$ (refs 10–15). However, OSOT is caused by optical generation and relaxation of photocarriers without applied drift (without a defined direction of carrier flow) and without conserving the equilibrium number of carriers in the dark. Accordingly, broken inversion symmetry in the crystal is not required in OSOT, and inversion-symmetric $H_{\text{so}}$ plus the time-reversal symmetry-breaking exchange-coupling term in the carrier Hamiltonian are sufficient for it to be observed. Note that there is a direct analogy between broken microscopic inversion symmetry being required in current-induced SOT and not required in OSOT and the macroscopic non-uniform magnetic structure being required in current-induced STT but not in OSTT.

**Experimental observation of OSOT**

The ferromagnetic semiconductor (Ga,Mn)As used in our experiments is favoured as a candidate material for observing optical spin-torque phenomena. The direct-gap GaAs host allows the generation of high-density non-equilibrium photocarriers, and the carrier spins interact with ferromagnetic moments on Mn via strong exchange coupling. When the ferromagnetic Mn moments are excited, this can be detected sensitively by probe laser pulses due to the large magneto-optical (MO) signals in (Ga,Mn)As. Several groups have reported MO studies of fast laser-induced magnetization dynamics in (Ga,Mn)As (Supplementary section ‘Experiments preceding our observations of OSOT and OSTT’). However, the direct search for and observation of the optical counterparts of STT and SOT were not the subject of studies until the work described in ref. 7 and in this Article.

The OSTT generated by circularly polarized light requires the injected carriers to have large spin lifetimes, so the weakly spin-orbit coupled photoelectrons in the (Ga,Mn)As conduction band play a key role in this case. OSOT is a fundamentally distinct photomagnetic phenomenon from OSTT, because the helicity-independent excitation does not impart angular momentum. This effect relies on spin–orbit coupling, so the non-equilibrium photoholes generated in (Ga,Mn)As are essential for OSOT. Our physical picture of OSOT in (Ga,Mn)As is based on the SOT formalism of equation (2) and on the following representation of the non-equilibrium steady-state spin polarization of the photoholes. The optically injected photoholes relax towards the hole Fermi energy of the p-type (Ga,Mn)As on a short (∼100 fs) timescale, and the excitation/relaxation processes create a non-equilibrium excess hole density in the spin–orbit coupled, exchange-split valence band. The increased number of occupied hole states (compared to the equilibrium state in the dark) can generate a non-equilibrium hole spin polarization that is misaligned with the equilibrium orientation of Mn moments. This non-equilibrium photohole polarization persists over the timescale of the hole recombination (on the scale of picoseconds), during which it exerts a torque on the Mn local moments.

A schematic illustration and our experimental observation of OSOT are shown in Fig. 1. The experimental identification of OSOT requires separating this non-thermal photomagnetic effect from the competing thermal excitation mechanism of magnetization dynamics. Absorption of the pump laser pulse leads to photo-injection of electron–hole pairs. Non-radiative recombination of photoelectrons produces a transient increase in the lattice temperature that builds up on the timescale of ∼10 ps and persists over ∼1,000 ps. This results in a quasi-equilibrium easy-axis (EA)}
orientation that is tilted from the equilibrium EA. Consequently, Mn moments in (Ga,Mn)As will precess around the quasi-equilibrium EA, as illustrated in Fig. 1a, with a typical precession time of \(~100\) ps determined by the magnetic anisotropy fields in (Ga,Mn)As. As discussed in detail in the following, the EA stays in-plane and the sense of rotation within the plane of the (Ga,Mn)As film with increasing temperature is uniquely defined. In the notation introduced in Fig. 1c, the change of the in-plane angle \(\delta\theta\) of the magnetization during the thermally excited precession can only be positive. OSOT, illustrated in Fig. 1b, acts during the laser pulse (with a duration of \(200\) fs) and fades away within the hole recombination time (on the scale of picoseconds). It causes an impulse tilt of the magnetization that allows us to clearly distinguish the OSOT from the considerably slower thermal excitation mechanism. Moreover, the initial OSOT-induced tilt of the magnetization can yield precession angles that are inaccessible in the thermally induced magnetization dynamics. This provides further evidence for OSOT.

Examples of the direct observation of thermally governed excitation of magnetization at a lower pulse intensity of \(6I_0\) \((I_0 = 7 \mu\text{cm}^{-2})\), and of excitation at a higher intensity of \(12I_0\) with a strong contribution from OSOT, are shown in Fig. 1d for a 3% doped (Ga,Mn)As sample (with a Curie temperature of \(T_C = 77\) K). Note that these dynamical MO signals are independent of the polarization of pump pulses; that is, they are the same for any orientation of the polarization plane of linearly polarized pump laser pulses and they also correspond to the polarization-independent part of the MO signals extracted by summing the laser pulses and they also correspond to the polarization-orientation of the polarization plane of linearly polarized pump pulses; that is, they are the same for any orientation of the polarization plane of linearly polarized pump laser pulses and they also correspond to the polarization-independent part of the MO signals extracted by summing the signals induced by \(\sigma^{+}\) and \(\sigma^{-}\) circularly polarized pump pulses (Supplementary Fig. S5). The distinct features of OSOT, described in the previous paragraph, are clearly visible when comparing the two measured trajectories of magnetization angles. This key demonstration has been enabled by the technique we developed in ref. 30, and which translates the measured dynamical MO signals in our pump-and-probe experiments to the time-dependent magnetization vector trajectory. This is done without assuming any theoretical model for the magnetization dynamics and without using any fitting parameter. Our experimental method uses different dependences of the polar Kerr effect (PKE) and magnetic linear dichroism (MLD) on the orientation of linear polarization of the probe laser pulses to disentangle the contributions to the MO signal from the out-of-plane and in-plane components of the magnetization motion, respectively. The magnitudes of PKE and MLD coefficients in a particular (Ga,Mn)As sample are determined from static MO experiments in which an external magnetic field is used to align the magnetization in a defined orientation (Supplementary Fig. S3). To obtain each point on the trajectory of the magnetization vector excited in the pump-and-probe experiment, we performed a set of measurements of the MO signal as a function of the orientation of the polarization plane of the linearly polarized probe pulse. The dynamical MO measurements in Fig. 1 and also in Fig. 2 were performed at zero magnetic field; before the experiment, the magnetization was aligned with the EA. (For more details on the experimental technique see refs 7,30 and Supplementary section ‘Determination of three-dimensional magnetization vector trajectory from dynamical magneto-optical signals’.)

Measurements for several intensities of pump pulses are shown in Fig. 2. The key characteristics of the magnetization dynamics at low intensities \(I_0\) and \(6I_0\) reflect the strong thermal excitation mechanism described in Fig. 1a. In equilibrium, the EA in the 3% Mn-doped sample is tilted by \(\sim 10^\circ\) from the [010] \((\varphi = 90^\circ)\) crystal axis towards the [110] \((\varphi = 135^\circ)\) in-plane diagonal as a result of the competing biaxial and in-plane uniaxial anisotropy fields\(^2\). With increasing temperature, the EA rotates further towards the [110] direction. This is because the uniaxial anisotropy component scales with magnetization as \(\sim M^2\) while the biaxial component scales as \(\sim M^4\), so the uniaxial anisotropy is enhanced relative to the biaxial anisotropy with increasing temperatures. This expected EA rotation is confirmed by independent superconducting quantum interference device (SQUID) measurements and microscopic calculations based on the exchange-split, Kohn–Luttinger representation of the Hamiltonian \(H\) for (Ga,Mn)As (refs 16,32) shown in Fig. 3a,b. (Note that the model Hamiltonian we use is consistent with \textit{ab initio} density functional calculations and X-ray photoemission measurements\(^3\) that show that the spectral weight of Mn \(d\)-states has a maximum around \(3–4\) eV below the Fermi energy, the Mn \(d\)-states are hybridized with the host valence-band orbitals continuously up to the Fermi energy, and no detached impurity band\(^1\) from the valence band is observed.) The amplitude of the precession angles for an intensity of \(6I_0\) (Fig. 2b) is larger than for an intensity of \(I_0\) (Fig. 2a). This is consistent with a larger increase of the transient temperature (and corresponding larger tilt of the quasi-equilibrium EA) for the larger pump pulse intensity. We have deduced the temperature increase due to pump pulses from the measured precession frequencies, which reflect the temperature-dependent magnetocrystalline anisotropy energies. In Fig. 3c we plot the dependence of the precession frequency on base sample temperature at low excitation intensity \(I_0\) and on laser intensity at a low base temperature of 15 K. From comparison of these two measurements we can infer the magnitude of the transient temperature change as a function of laser intensity. (Note that consistent temperature versus intensity calibration is obtained from comparison of the intensity dependence of the pump-induced demagnetization and the temperature dependence of the remanent magnetization measured by SQUID.)

Figure 3c confirms a sizable difference in transient temperatures for intensities \(I_0\) and \(6I_0\). Remarkably, heating of the sample by the laser pulses saturates at approximately \(10I_0\), as seen from Fig. 3c.
Characterization of 3% Mn-doped (Ga,Mn)As. a, Temperature dependence of the equilibrium EA orientation determined from SQUID magnetization measurements. Consistent EA orientations are inferred from the MO experiments. b, Microscopic calculations of the temperature and hole density-dependent EA orientation. c, Frequency of precessing Mn moments measured at zero magnetic field as a function of base temperature at low excitation intensity \(I_0\), and as a function of pump intensity at a low base temperature of 15 K. d, Dynamics of pump-induced reflectivity change at intensity \(6I_0\). Inset: intensity dependence of the initial reflectivity change at a base temperature of 15 K (line depicts a linear dependence). The error bars correspond to standard deviations and depict the uncertainty in the determination of the displayed value from the experimentally measured data.

The connection between OSOT and photocarrier generation is shown in Fig. 3d. Here we show the observed change in measured reflectivity of the (Ga,Mn)As film, which correlates with the number of generated photocarriers (see Supplementary section 'Calibration procedure used for evaluation of the pump-induced temperature and hole concentration increase'). The pump-induced change in the index of refraction is linear in pump-pulse intensity up to \(\approx 25I_0\), after which it starts to saturate. This means that, unlike the transient temperature, the number of generated photocarriers keeps increasing with increasing pump pulse intensity above \(10I_0\). The concentration of photo-injected carriers can be estimated from the laser spot size, photon energy, and absorption and reflection coefficients of the sample. For the higher intensity measurements, the obtained photocarrier density is of the order of \(\approx 1 \times 10^{19} \text{ cm}^{-3}\).

Theoretical modelling
In Fig. 3b we show that the equilibrium EA is sensitive to hole density variations\(^{3,36}\) and that the sense of the tilt of the EA with increasing hole density can be opposite to that due to an increase in temperature. As explained in detail in the following, Fig. 3b provides a clue as to why the instantaneous out-of-plane component of the tilt of the magnetization due to OSOT is opposite to the initial out-of-plane component of the precessing magnetization around the thermally excited quasi-equilibrium EA at lower pump intensities. The relation between OSOT and the magnetocrystalline anisotropy field can be microscopically demonstrated using the OSOT formalism of equation (2) and, similar to the theory for current-induced SOT, assuming in the quantum-mechanical averaging constructed from the eigenstates of \(H\). The non-equilibrium steady state differs from the equilibrium state in the dark in that the distribution function has a shifted Fermi level corresponding to the extra density of the photoholes. In this approximation, the OSOT is determined by the hole density-dependent magnetocrystalline anisotropy field \(H_{\text{an}}\) because\(^{12}\)

\[
H_{\text{an}} = -\frac{\partial}{\partial M} \sum_{a} \int d\epsilon_{a,k} f_{a,k} = -\sum_{a} \int d\epsilon_{a,k} |f_{a,k}| \frac{\partial H}{\partial M} |a, k| f_{a,k} = -\sum_{a} \int d\epsilon_{a,k} f_{a,k} \frac{|\epsilon_{a,k}|}{\epsilon_{a,k}} f_{a,k} = -\frac{f_{a,k}}{\epsilon_{a,k}}
\]

Here \(\epsilon_{a,k}\) and \(f_{a,k}\) are the eigenenergy of \(H\) and the Fermi distribution function, respectively, labelled by the band and wave vector index. Consistent with the in-plane orientation of the EA,
we find that the out-of-plane transverse component of the anisotropy field is given by $H_m,\theta = -J_\theta = 0$ for any in-plane $M$ and any considered hole density. The in-plane transverse component, $H_m,\psi = -J_\psi$, is zero when $M$ is aligned with the EA at a given hole density and non-zero for other orientations of $M$ at the same hole density. Because EA orientation is sensitive to hole density, as shown in Fig. 3b, $H_m,\psi$ for a given orientation of $M$ can change when the hole density is increased by the photoexcitation (Supplementary Fig. S13). The sign of the calculated $H_m,\psi$ is consistent with the sense of the initial magnetization tilt observed in experiments governed by the OSOT. The amplitudes of $J_\theta$, obtained from the calculated hole density-dependent anisotropy fields, are on the scale of $\mu$eV, which is about 10 times smaller than the experimental strength of OSOT fields inferred from the measured out-of-plane tilts of the magnetization. The exchange-split, Kohn–Luttinger model is known to underestimate small lattice-matching strain of 1\% (Ga,Mn)As grown on a GaAs substrate, which has a relatively observe the largest OSOT in our annealed 3\% Mn-doped (Ga,Mn)As sample; however, OSOT decreases with increasing Mn doping, as shown in the following. This confirms that the strain-related broken inversion terms in $H_m,0$ do not play a significant role in OSOT in our (Ga,Mn)As samples.

**Experimental elimination of the thermal mechanism**

In the remaining paragraphs we discuss the effects of controlling the micromagnetic parameters of (Ga,Mn)As in relation to the observed magnetization dynamics. In particular we demonstrate that controlling the micromagnetic parameters allows us to eliminate the thermal mechanism while still detecting OSOT. One of the pronounced features of the measurements in Fig. 2, omitted in the previous discussion, is the enhanced damping of the magnetization precession at higher pump pulse intensities. The higher excitation intensity causes higher demagnetization and, correspondingly, a stronger inhomogeneous ferromagnetic resonance damping. In Fig. 4 we demonstrate that the damping due to magnetic inhomogeneities is suppressed at applied magnetic fields along the EA (at higher precession frequencies), as previously established in the systematic MO study of micromagnetic parameters of (Ga,Mn)As and as is typical for MO ferromagnetic resonance measurements in high-crystal-quality ferromagnets (see Supplementary section ‘Elimination of thermal excitation mechanism by external magnetic field’ for more details). Apart from confirming that weaker damping of the precession is recovered when performing our experiments at non-zero external magnetic fields, the experiments have another direct implication for our OSOT study. Applied fields of the order of tens of millitesla (that are comparable to the in-plane anisotropy fields) not only reduce damping, but also gradually suppress the quasi-equilibrium tilt of the EA induced by the transient temperature increase (Fig. 4a–c). The OSOT is, however, much less affected by the applied field, so at 100 mT we still observe the impulse tilt of the magnetization due to the OSOT but the thermal effect is already diminished (Fig. 4d).

Controlling the rich doping dependence of the micromagnetic parameters of our high-quality (Ga,Mn)As materials allows us to eliminate the thermal mechanism, even at zero applied field, as demonstrated in Fig. 5. The idea of these measurements is as follows. Increasing the temperature always tilts the EA towards the in-plane diagonal ($\varphi = 135^\circ$ in our notation). Consistent with the systematic measurements of the anisotropy fields in our series of...
GaMnAs materials, we found that the base-temperature EA of the 3% Mn-doped sample (with $T_c = 77$ K) is at 100° and that of a 5% Mn-doped sample (with $T_c = 132$ K) is at 121°, but for a 7% Mn-doped sample (with $T_c = 159$ K) and a 9% Mn-doped sample (with $T_c = 179$ K), the base temperature zero-field EA is already aligned with the $\varphi = 135^\circ$ in-plane diagonal. As shown in Fig. 5a, we indeed again observe a clear thermal effect in the 5% sample. The precession trajectory is less damped and more elliptic compared to the 3% Mn-doped samples (Fig. 2b). This is consistent with the systematic doping trends in the (Ga,Mn)As micromagnetic parameters, namely the reduction of Gilbert damping and enhancement of the out-of-plane anisotropy field in more doped samples. In the 7% and 9% Mn-doped samples the thermal effect is completely suppressed (Fig. 5b–d); because higher temperature can only strengthen the uniaxial anisotropy along the diagonal, the EA does not tilt from the 135° angle when the temperature is increased by the pump pulse. Remarkably, we still observe impulse out-of-plane tilts in the 7% and 9% samples; that is, we observe magnetization excitation that we can ascribe entirely to OSOT. Also consistent with expectations, the amplitude of OSOT increases with increasing pump intensity (Fig. 5c,d) but the thermal mechanism is absent, independent of pump intensity. OSOT in these highly doped materials is significantly weaker than in the low-doped 3% sample.

**Conclusion**

We have reported an experimental study of the magnetization dynamics in (Ga,Mn)As induced by helicity-independent laser excitations. We have performed several complementary experiments that identified the presence of a non-thermal mechanism of the photocarriers exerting a torque on the magnetization. Because the helicity-independent excitations do not impart angular momentum and thermal effects have been separated out, the observed non-thermal photomagnetic torque originates from spin–orbit coupling effects of the photocarriers. This makes OSOT fundamentally different from the previously reported non-relativistic SSTT, in which angular momentum of the circularly polarized light is transferred via photocarrier spins to the magnetization. Our work demonstrates the possibility of studying SOTs on the short timescales achievable using the optical pump-and-probe experimental technique. The relativistic OSOT effects should be observable in other systems, including antiferromagnetic semiconductors, which, unlike their ferromagnetic counterparts, can have magnetic transition temperatures well above room temperature. It is well established that magnetocrystalline anisotropies are equally as present in spin–orbit coupled antiferromagnets as in ferromagnets, and it has been demonstrated recently that spin–orbit coupling induced anisotropic magneto-transport effects can also be strong in antiferromagnets. OSOT belongs to this family of relativistic effects and its exploration in antiferromagnets may open another novel avenue of optical spin torque research beyond (Ga,Mn)As.

Received 20 September 2012; accepted 28 February 2013; published online 21 April 2013

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Acknowledgements
The authors acknowledge discussions with A.H. MacDonald, J. Sinova and J. Wunderlich, and support from the European Research Council (advanced grant no. 268066), Framework Programme 7 (grant no. 215368), the Engineering and Physical Sciences Research Council (grant no. EP/H029257/1), the Ministry of Education of the Czech Republic (grant no. LM2011026), the Grant Agency of the Czech Republic (grant nos. 202/09/H041 and P204/12/0853), the Charles University in Prague (grant nos. SVV-2012-265306 and 443011) and the Academy of Sciences of the Czech Republic Praemium Academiae.

Author contributions
V.N. and K.O. prepared the samples. N.T., P.N., E.R., T.Ja., D.B., P.M., K.O. and T.Ju. conducted the experiments and analysed the measured data. P.N. and F.T. performed data modelling. J.Z. and T.Ju. developed the microscopic theory and performed the calculations. P.N. and T.Ju wrote the manuscript.

Additional information
Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to P.N.

Competing financial interests
The authors declare no competing financial interests.