Ultrafast nonthermal photo-magnetic recording in a transparent medium

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Discovering ways to control the magnetic state of media with the lowest possible production of heat and at the fastest possible speeds is important in the study of fundamental magnetism¹⁻⁵, with clear practical potential. In metals, it is possible to switch the magnetization between two stable states (and thus to record magnetic bits) using femtosecond circularly polarized laser pulses⁶⁻⁸. However, the switching mechanisms in these materials are directly related to laser-induced heating close to the Curie temperature⁹⁻¹². Although several possible routes for achieving all-optical switching in magnetic dielectrics have been discussed¹³,¹⁴, no recording has hitherto been demonstrated. Here we describe ultrafast all-optical photo-magnetic recording in transparent films of the dielectric cobalt-substituted garnet. A single linearly polarized femtosecond laser pulse resonantly pumps specific d⁻→d transitions in the cobalt ions, breaking the degeneracy between metastable magnetic states. By changing the polarization of the laser pulse, we deterministically steer the net magnetization in the garnet, thus writing ‘0’ and ‘1’ magnetic bits at will. This mechanism outperforms existing alternatives in terms of the speed of the write–read magnetic recording event (less than 20 picoseconds) and the unprecedentedly low heat load (less than 6 joules per cubic centimetre).

To stabilize magnetic states of a single bit of a recording medium at room temperature, a magnetic anisotropy energy barrier of \(60kT \approx 0.25 \text{aJ} \) (where \(k\) is the Boltzman constant and \(T\) is the absolute temperature) is taken as a sufficient value¹⁵. This value would then also correspond to the energy that is ideally required to switch the magnetic state. In practice, however, about eight orders of magnitude more energy is used¹⁶⁻¹⁸ and much of it is subsequently lost via dissipation. It would be greatly advantageous to realize optical rather than current-induced switching of magnetic states, as light can be transferred with minimum losses and effectively modifies the barrier through opto-magnetic and photo-magnetic interactions¹⁹²⁰.

Cobalt-substituted yttrium iron garnet (YIG:Co) is an optically transparent ferrimagnetic dielectric (see Methods and Extended Data Fig. 1) with a cubic lattice and two antiferromagnetically coupled spin sublattices of Fe³⁺ in both tetrahedral and octahedral sites²¹. The dopant Co²⁺ and Co³⁺ ions replace Fe³⁺ in both types of sites²². These Co ions are responsible for the strong magnetocrystalline²³ and photo-induced magnetic anisotropy²³,²⁰ as well as for the very large Gilbert damping²³,α = 0.2. In an unperturbed state at room temperature, the equilibrium orientation of the magnetization is defined by cubic (\(–8.4 \times 10⁴ \text{erg cm}⁻²\)) and uniaxial (\(–2.5 \times 10⁴ \text{erg cm}⁻²\)) anisotropy, which favour orientation of the magnetization along one of the body diagonals of the cubic cell (the \([111]\) axis) and perpendicular to the \([001]\) axis, respectively. It results in four easy magnetization axes which are slightly inclined from the body diagonals, as shown in Fig. 1. To distinguish between different magnetic domains, we used a garnet film with a miscut of about 4°. In Fig. 1, the large stripe-like domains have magnetizations along \(M(1)⁺\) near the \([111]\) axis and \(M(1)⁻\) near the \([111]\) axis and the small labyrinth-like domains have magnetizations along \(M(S)⁺\) near the \([111]\) axis and \(M(S)⁻\) near the \([111]\) axis.

To investigate the feasibility of switching with linearly polarized light in YIG:Co, we employed the technique of femtosecond magneto-optical imaging using a pump laser pulse with duration of 50 fs (see Methods and Extended Data Fig. 2). The images of magnetic domains were taken before and after the excitation with a single pump
laser pulse (see Fig. 2a). Taking the difference between the images emphasizes the photo-magnetic changes and is used for detailed analysis. Light can lift the degeneracy between the domains by generating photo-induced magnetic anisotropy\(^\text{22}\). In our case, pumping the initial pattern of magnetic domains with a single laser pulse polarized along the [100] axis (\(\phi = 0^\circ\)) turns large white domains (\(M^{(L)+}\)) into large black ones (\(M^{(L)-}\)). Simultaneously, small black domains (\(M^{(S)-}\)) turn into small white ones (\(M^{(S)+}\)) (see Fig. 2a). The domain pattern stays remarkably unperturbed; only the contrast reverses. The initial state can be restored by pumping with a single laser pulse polarized along the [010] axis (\(\phi = 90^\circ\)). The recorded domains are stable for several days owing to the non-zero coercivity of the garnet film at room temperature (see Fig. 2b). The initial domain pattern can be also restored by a brief application of an in-plane magnetic field of the order of 80 mT. The symmetry of the observed all-optical switching suggests that the point group of the crystal is 4 (see Methods). Although it is expected that the point group of the garnet surface is 4\(\text{mm}\), the actual symmetry can be lowered either by the magnetization component along the [001] axis or simply by distortions during its growth\(^\text{25}\).

The minimum pump fluence required for magnetic recording in YIG:Co is very sensitive to the wavelength of the pump pulse. The switched area estimated from the magneto-optical images is plotted as a function of the pump fluence for different pump wavelengths (see Fig. 3). The wavelength was varied within the range 1,150–1,450 nm (1.08–0.86 eV), where the light resonantly excites electronic \(d\leftrightarrow d\) transitions in Co ions\(^\text{26}\). In the studied YIG:Co film, a resonant pumping of the transitions in Co\(^{3+}\) and Co\(^{2+}\) ions at the tetrahedral sites at 1,305 nm (\(h\omega = 0.95\text{ eV}\))\(^\text{27}\) is accompanied by absorption \(a\) of about 12% of the light energy (Extended Data Fig. 1). The spectral dependence in Fig. 3 reveals a pronounced resonant behaviour around this energy. It can be seen that the minimum pump fluence required to form a domain is about \(I_{\text{min}} = 34\text{ mJ cm}^{-2}\). This means that the magnetic recording is a result of absorption of about \(aI_{\text{min}}/h\omega \approx 3 \times 10^{16}\) photons per cm\(^2\). Given that the film is \(d = 7.5\mu\text{m}\) thick, the absorbed photons required for the switching of the magnetization in a given volume would be about \(10^{19}\text{ cm}^{-3}\), corresponding to depositing \(aI_{\text{min}}/d \approx 6\) cm\(^2\) of heat. For instance, recording a bit with size 20 nm \(\times 20\text{ nm} \times 10\text{ nm}\) would be accompanied by dissipations of just 22 aJ (about 5,300 kT). To the best of our knowledge, this is much lower than for all-optical switching of metals (10 fJ)\(^\text{28}\), existing hard-disk drives (10–100 nJ)\(^\text{16}\), flash memory (10 nJ)\(^\text{17}\) or spin-transfer torque random-access memory (450 pJ–100 fJ)\(^\text{18}\).

Finally, we studied ultrafast dynamics of the magnetization switching, employing time-resolved single-shot magneto-optical imaging (see Extended Data Fig. 2). The magnetic domains were recorded with a single pump pulse and imaged with a single 40-fs unfocused probe pulse with a central wavelength of 800 nm. After each write–read event the recorded domains were erased by application of an external magnetic field of 80 mT in the [1–10] direction. Similarly to static magneto-optical imaging, reference images were taken before each pumping (that is, at negative time delay). This image was subtracted from that obtained at a given pump–probe delay. Varying the time delay \(\Delta t\), a series of the magneto-optical images were obtained (see top inset in Fig. 4). To quantify the dynamics of the laser-induced changes we took an integral over the pumped area, normalized the data and plotted the result as a function of time delay between the pump and probe pulses. It can be seen that the recorded domain emerges with a characteristic time \(\tau\) of about 20 ps and stabilizes after about 60 ps. For the recording and reading out, we used just two femtosecond laser pulses; to the best of our knowledge this experiment is the fastest-ever write–read magnetic recording event\(^\text{9}\). Unlike all-optical magnetic switching in metals\(^\text{6–11}\), the recording in transparent dielectrics does not require any destruction of magnetic order and operates without ultrafast heating of the medium up to the Curie point.

The time of 60 ps taken for the switching is in very good agreement with a quarter-period of the laser-induced precession of magnetization in YIG:Co film (see Extended Data Fig. 3). Therefore it is reasonable to suggest that, unlike all-optical switching in metals\(^\text{9}\), the mechanism of the spin switching in garnets proceeds via the precession of the net magnetization. In this scenario, to switch the magnetization from the initial...
The size of the images is 240 μm × 260 μm.

As a result, the magnetization in large domains moves along a trajectory between these states. When started from the initial metastable state, optical excitation should induce magnetic anisotropy, which favours the + and + states, respectively. This means that the magnetization will start precession around a direction somewhere in between these and states (small domains) as well as between the and states (large domains) with the help of the laser pulse polarized along the [100] axis. The pump fluence was 150 mJ cm$^{-2}$, that is, after the first quarter of the precession period equal to 250 ps (see Extended Data Fig. 3a), the magnetization vector will be closer to the − state. If, at this moment, owing to relaxation of the photo-excited electrons, the initial magnetic anisotropy is restored, the magnetization will start to precess around the − direction, eventually arriving at this metastable state on a timescale defined by the damping of the oscillations. In reality, in YIG:Co film, the lifetime of the photo-induced anisotropy at room temperature is also of the order of 60 ps (ref. 13) and the damping is indeed very large (see Extended Data Fig. 3a).

A set area to the magneto-optical signal in the case when the magnetization is aligned along the [001] axis. The dots show the mean values and the errors are no larger than the dot size, being all below 5%. However, the stronger signal noise is caused by the sensitivity of the single-shot imaging to even the slightest illumination drift, which is unavoidable due to the motion of the delay line. The red solid line was fitted using the exponential increase $\exp(-\Delta t/\tau)$ with the characteristic time $\tau = 19.5 \pm 1.6$ ps. The top inset shows the images of domains obtained at different time delays after subtraction of the reference image obtained at negative $\Delta t$. The lower inset shows the schematics of the magnetization trajectory during the switching. The magnetization is switched between the + and − states with the help of the laser pulse polarized along the [100] axis. The pump fluence was 150 mJ cm$^{-2}$ and the central wavelength of the pump was 1,250 nm.

The mechanism operates at room temperature and outperforms existing write–read events, accompanied by an unprecedentedly low heat load. Even faster switching can be anticipated if the photo-induced magnetic anisotropy is enhanced when the temperature is decreased$^{26}$ or when iron is substituted by ions more anisotropic than cobalt ions.

We anticipate that magnetization switching caused by this photo-magnetic phenomenon will open up many opportunities for the design and development of materials and methods in the field of opto-magnetic recording. For instance, using the photo-magnetic garnet as a recording medium has similarities to heat-assisted magnetic recording, but without the need for much heat or for an electromagnet. Furthermore, it is known that magnetic anisotropy in garnet films can also be controlled by electric fields$^{29}$. Tuning the strengths of the magnetocrystalline, photo-magnetic and electrically induced anisotropies such that switching is possible only under simultaneous electric field and light seems to enable faster and less dissipative magnetic random access memory.
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Author Contributions A.S. conceived the project with contributions from A.K. and A.V.K. The imaging and time-resolved magnetization precession were performed by K.S. D.A. developed femtosecond single-shot imaging and performed time-resolved imaging together with K.S. A.K. and A.V.K. co-wrote the manuscript with contributions from A.K., K.S. and D.A. The project was coordinated by A.S.

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Materials. The magnetization switching results were obtained on Co-substituted yttrium iron garnet (YIG:Co) film $d = 7.5 \mu m$ thick with composition Y$_2$CaFe$_5$O$_{12}$. The single-crystal YIG:Co garnet film was grown by liquid-phase epitaxy on gadolinium gallium garnet Gd$_2$Ga$_2$O$_{12}$ (001)-oriented substrates with $4^\circ$ miscut and thickness 400 $\mu m$. The saturation magnetization at room temperature was $M_s = 90$ G and the Néel temperature was 445 K. The Gilbert damping measured using the ferromagnetic resonance technique gives $\alpha = 0.02$. At room temperature the sample has both cubic ($8.4 \times 10^{-3}$ erg cm$^{-3}$) and uniaxial ($-2.5 \times 10^{-3}$) anisotropy, which were measured by means of both ferromagnetic resonance and torque magnetometry.

The cubic anisotropy term is dominant, yielding easy axes of magnetization along $<111>$—type directions. The uniaxial term modifies this by tilting the easy axes slightly towards the sample plane. Thus such a crystal has eight possible magnetization states in a zero applied field, directed close to the body diagonals of a cube. By symmetry, those magnetization states should be energetically equivalent, but owing to substrate miscut this degeneracy is lifted and some of them have slightly lower energy. This is why in the demagnetized state the sample shows an alternating stripe pattern of magnetic phases. The measured absorption coefficient within the spectral range of 1,150–1,450 nm for the studied sample is shown in Extended Data Fig. 1.

Magneto-optical imaging under single pump pulse excitation. The design of the time-resolved pump–probe experimental set-up for investigations of all-optical magnetic recording is shown in Extended Data Fig. 2. The domain structure of the garnet films we studied was visualized using a magneto-optical polarizing microscope with a standard light-emitting diode (LED) source of polarized light as a probe. In this case, the central wavelength of pump pulses with duration of 50 fs was varied within the spectral range 1,150–1,450 nm. Relying on the fact that domains with different orientation of the magnetization will result in different angles of the Faraday rotation, the domains were visualized with the help of an analyser and a charge-coupled device (CCD) camera. The images were acquired about 10 ms after excitation with a single pump pulse.

Time-resolved femtosecond single-shot imaging. For the investigation of magnetization switching dynamics we used laser pulses with duration of 40 fs and with the central wavelength of 800 nm as a probe. The linearly polarized unfocused probe beam passing through the sample was collected by an objective and the magneto-optical contrast was gained with the help of an analyser (see Extended Data Fig. 2). The acquired magneto-optical image was digitized and recorded with the help of the CCD camera. A single-shot pumping was achieved by placing a mechanical shutter in the path of the pump beam. The actuation time of the shutter was set to the minimum possible value of 60 ms. To exclude any possibility of excitation by more than one pump pulse, the repetition rate of the amplifier was brought down to 10 Hz. To improve the signal-to-noise ratio in the detection of the probe pulses, the exposition time of the camera was set at 1 ms. The actuation time of the camera and the mechanical shutter were controlled by an electrical delay generator synchronized with the laser. Adjustment of the electrical delays for the shutter and the camera allowed us to capture the magneto-optical image produced by a single pump pulse. The delay generator was set to the standby mode and was controlled by an external computer. The asynchronous trigger from the computer results in the generation of a single trigger signal synchronized with the laser; this signal activates the camera and the shutter. To erase a long-lived state with the switched magnetization and to reinitialize the magnetic state, a magnetic recording is shown in Extended Data Fig. 2. The domain structure of the magnetic recording is shown in Extended Data Fig. 2. The domain structure of the magnetic recording is shown in Extended Data Fig. 2.

Estimation of temperature increase under laser pump pulse. For the investigation of magnetic recording we take into account the minimum intensity required for the switching $I_{\text{min}} = 34$ mJ cm$^{-2}$, the heat capacity of the garnet $C = 430$ J mol$^{-1}$ K$^{-1}$ at room temperature, the molar mass $m = 706$ g mol$^{-1}$ and the density $\rho = 7.12 g cm^{-3}$. The measured absorption of the pump at the wavelength of 1,300 nm is about 12% (see Extended Data Fig. 1). The temperature increase as a result of absorption is thus $\Delta T = \alpha I_{\text{min}}/C\rho = 1.25 K$. Such a temperature increase is at least two orders of magnitude lower than the one required to reach the Néel temperature.

Laser-induced magnetization precession. To study laser-induced spin oscillations induced by femtosecond laser pulses in YIG:Co film, we also carried out conventional time-resolved measurements using a magneto-optical pulse–probe method. Pump pulses with duration 50 fs arrived at the sample with a repetition rate of 500 Hz. The angle of incidence was set to $10^\circ$ from the sample normal; that is, from the [001] crystallographic axis of the garnet film. Equally short probe pulses had repetition rates twice as high and arrived at normal incidence to the sample. The central wavelength of the pump was set to 1,200 nm. This wavelength corresponds to a large amplitude of the spin oscillations. The central wavelength of the probe was 800 nm. The pump, with fluence below 70 mJ cm$^{-2}$, was focused to a spot about 130 $\mu m$ in diameter. The delay time $\Delta t$ between the pump and the probe pulses could be adjusted within the range from 50 fs to 1 ns. The polarization plane of the linearly polarized pump pulse was set at an angle $\phi$ with respect to the [100] axis. The polarization plane of the probe beam was along the [1–10] axis. Using a balanced photodetector we measured the Faraday rotation of the probe as a function of the delay time $\Delta t$ between the pump and probe pulses (see Extended Data Fig. 2). The Faraday rotation is proportional to the out-of-plane component of the magnetization $M_z$. All measurements were done under zero applied magnetic field and at room temperature. The measurements were performed in a stroboscopic mode and thus reveals the pump-induced dynamics, which is reproducible from pulse to pulse.

The magnetization precession signals show a strong dependence on the pump polarization (see Extended Data Fig. 3a). Moving the pump spot across the boundary between large domains with $M^{[1]}_z$ and $M^{[1]}_z$ — also allows us to excite spin precession with opposite phases. It is also interesting to note that if the probing spot is placed exactly on the wall, the antiphase signals from each domain average to zero. This is only possible if the underlying dynamics is due to a coherent rotation of the magnetization and not due to domain wall motion. The characteristic rise time of the signal is about 20 ps. The time delay at which the signal is at maximum is about 60 ps (see Extended Data Fig. 3a). Orthogonal polarizations along $<100>$—type direction result in the highest amplitude of the spin precession (see Extended Data Fig. 3b). These polarizations correspond to the global symmetry axes of the tetrahedrons in our garnet film hosting the Co ions. The amplitude of the precession follows a cos(2$t$) dependence, which is typical for photo-magnetic effects13,20,24. The amplitude of the magnetization precession increases linearly with the pump fluence owing to the stronger light-induced effective field (see Extended Data Fig. 3c). The period of the precession is nearly constant (about 250 ps) and follows the frequency of the ferromagnetic resonance mode23 in the field of magnetic anisotropy. Note that the central wavelength of the pump corresponding to the most efficient switching does not coincide with the wavelength corresponding to the most effective excitation of the oscillations. This is because the oscillations are detected in a stroboscopic mode, which gives the largest signal when no switching takes place.

The microscopic mechanism of the photo-induced anisotropy has been extensively discussed previously13,20,24. The charge transfer process redistributes electrons among Co ions in non-equivalent crystal sites, changing the valence states of the Co ions and consequently, their contribution to magnetic anisotropy. The latter can be explained in terms of the single ion anisotropy model25. The pump light is used to excite optical transitions in the YIG:Co film in the tetrahedral Co ions. This approach allows us to achieve a large amplitude of the effective field of the photo-induced anisotropy (hundreds of Oe). Another important feature is that even though the number of the excited Co ions does not reach 100%, the single-ion anisotropy from Co is very high, producing a large light-induced field. Thus the photo-magnetism in the YIG:Co film is very efficient.

Symmetry analysis of the all-optical switching. We choose the coordinate system in which the coordinate axes $x$, $y$ and $z$ are aligned along the crystallographic directions [100], [010] and [001], respectively. To explain the effect of light on the magnetic anisotropy, the energy of the light–matter interaction should contain terms $\chi^{\text{eff}}_i E_i E_j^* M_x M_y$ where $\chi^{\text{eff}}_i$ is a polar fourth-rank tensor, $E_i$ is the ith component of the electric field of light, $E_j^*$ is the complex conjugate of the j-component of the electric field of light and $M_x$ is the kth component of the magnetization28. In our experiment, polarized light switches two domains with $M_x > 0$ to two domains with $M_x < 0$ and back; in all four states between which the light switches the magnetic effects13,20,24. The amplitudes for which $k = 0$ do not depend on the sign of the magnetization components and thus cannot be responsible for the sign change of $M_x$. We can also see that the switching does not depend on the sign of $M_x$, because the same pulse has opposite effects on $M_x$ in large and small domains. Hence if light is polarized along the [100] or the [010] direction, the part of the energy of the photo-induced magnetic anisotropy that is responsible for the switching can be written as:

$$W_i(E, M) = \chi_{xyx} E_i E_j^* M_x M_y + \chi_{xxy} E_i E_j^* M_x M_y + \chi_{yxy} E_i E_j^* M_x M_y + \chi_{yyx} E_i E_j^* M_x M_y$$

(1)

This energy must be considered as a thermodynamic potential. Its minimization with respect to $M_x$ allows us to find the potential minimum and define the equilibrium orientation of the magnetization.
The expected point group for the (001) surface of the garnet is 4\,mm, for which $\chi_{yyyx} = \chi_{xxyx} = \chi_{xxxy} = \chi_{yyxy} = 0$. However, a non-zero magnetization along the [001] axis, that is, $M_z$, effectively lowers the symmetry down to 4. The symmetry of the as-grown (with defects) garnet films can also be lowered simply owing to distortions during growth\textsuperscript{25}.

For point group 4 $\chi_{yyyx} = -\chi_{xxyx}$ and $\chi_{xxyx} = -\chi_{yyxy}$ (ref. 31). Assuming that $A = \chi_{xxyx} + \chi_{yyxy}$ equation (1) can be simplified to:

$$W_2(E, M) = A \left( E_x E_y^* M_x M_y - E_y E_x^* M_y M_x \right)$$  \hspace{1cm} (2)

Assume that $A < 0$. In this case, under illumination of the garnet with light linearly polarized along the [100] direction the thermodynamic potential will be minimized if $M_y > 0$. Hence, such an excitation will promote switching of large white domains ($M^{(L)}_y$) into large black ones ($M^{(L)}_y$). Simultaneously, small black domains ($M^{(S)}_y$) will be switched into small white ones ($M^{(S)}_y$). If the light is polarized along the [010] axis, the thermodynamic potential is minimized when $M_y < 0$ and it means that the photoexcitation will promote switching of the magnetization in the large and small domains back into the $M^{(L)}_y$ and $M^{(S)}_y$ states, respectively.

**Data availability.** Source data accompanies Figs 3 and 4 online. The data supporting other findings of this study are available from the corresponding author (A.S.) upon reasonable request.

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Extended Data Figure 1 | Spectral dependence of the absorption coefficient of YIG:Co film.
Extended Data Figure 2 | Schematics of the time-resolved magnetization dynamics and single shot imaging. The inset shows the magneto-optical visualization of the magnetic domains formed by a single laser pulse excitation of YIG:Co.
Extended Data Figure 3 | Time-resolved magnetization precession induced by the femtosecond pump pulses in YIG:Co film. The out-of-plane component of the magnetization $M_z$ is detected with the help of time-resolved magneto-optical Faraday rotation. a, The left axis shows the laser-induced magnetization precession for the case when the light is polarized along the [100] orientation and the magnetization is in either the $M^{(+)}$+ or in the $M^{(-)}$ state. The right axis shows the domain structure and the spots in which the dependences shown on the left panel were measured. b, Dependence of the precession amplitude on the pump polarization $M^{(+)}$. The solid line is a fit to the $\cos(2\phi)$ function. c, The dynamics measured at different pump fluences $I$ in the range 7.4–61 mJ cm$^{-2}$. The pump polarization was in the [100] direction. The inset shows the linear dependence of the precession amplitude on the pump fluence.