

## Exchange-driven all-optical magnetic switching in compensated 3d ferrimagnets

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Understanding how a single laser pulse can toggle magnetization in a compensated 3d ferrimagnet is a critical problem in ultrafast magnetism. To resolve it, we test single-shot all-optical switching of magnetization in  $\text{Mn}_2\text{Ru}_x\text{Ga}$  at different temperatures using femto- to picosecond pulses in the visible to far-infrared spectral ranges. The switching process is found to be independent of photon energy, but strongly dependent on both the pulse duration and sample temperature. Switching is disabled whenever the starting temperature  $T_0$  is above the compensation point of  $\text{Mn}_2\text{Ru}_x\text{Ga}$ , but as  $T_0$  is lowered below compensation, increasingly longer pulses become capable of toggling the magnetization. We explain the observations in terms of a switching process driven by exchange relaxation of the angular momenta of the manganese sublattices, and propose a common framework to account for the similarities and differences of all-optical switching in  $\text{Mn}_2\text{Ru}_x\text{Ga}$  and  $\text{GdFeCo}$  alloys.

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**Introduction.** Discovering how ultrashort laser pulses can manipulate magnetic materials on nonequilibrium timescales is a key goal of both ultrafast magnetism [1,2] and spintronics [3,4]. All-optical switching (AOS) of magnetization using a single ultrashort laser pulse in the absence of a magnetic field [5] was first identified in amorphous ferrimagnetic  $\text{Gd}_x(\text{FeCo})_{100-x}$  thin films (hereafter referred to as  $\text{GdFeCo}$ ) in 2012 [6]. Great efforts have since been devoted by experimentalists and theoreticians alike to unravel the underlying physical mechanism, with the ultimate aim of designing alternative optically switchable materials with superior functionality [7,8].

Advances in the synthesis of fully compensated half metals [9] have recently resulted in the breakthrough discovery that  $\approx 200$ -fs-long pulses can activate single-shot AOS in thin films of the Heusler alloy  $\text{Mn}_2\text{Ru}_x\text{Ga}$  [10] (hereafter referred to as MRG). In combination with its half metallicity [9], the potential to feature a perpendicular magnetic anisotropy field above 10 T and highly efficient intrinsic spin-orbit torques [11] due to the absence of structural inversion symmetry, MRG's ability to be switched all-optically renders it a compelling candidate for integration in future ultrafast spintronic technology [12–14]. While  $\text{GdFeCo}$  and MRG are both ferrimagnetic metals exhibiting compensation points where the constituent sublattices have equal but opposite magnetization [15], there are important material

differences, which seem to rule out the possibility that the same physics could govern the single-shot AOS process found in both materials.

MRG is a crystalline ferrimagnetic Heusler alloy [9] with two magnetic sublattices, corresponding to manganese on the 4a and 4c sites of the  $XA$  structure (space group  $F\bar{4}3m$ ). Highly spin-polarized electronic states originating from the  $\text{Mn}(4c)$  sites, which form a tetrahedron at the center of the unit cell, dominate near the Fermi level [13]. Consequently, the electrical transport properties (magnetoresistance, anomalous Hall effect) and magneto-optical properties (Kerr effect) are dominated by the 4c sublattice. Based on this alone, we might expect that intense ultrashort optical pulses would initially excite and demagnetize the 4c sublattice and the 4a sublattice would follow later through energy transfer via the lattice [16]. MRG's unusual band structure therefore raises the question of how optical irradiation can simultaneously demagnetize both sublattices [17–19], generating the transient nonequilibrium state from which single-shot AOS emerges [20,21].

Besides the electronic structure, the magnetic properties of MRG and  $\text{GdFeCo}$  are different. Mean-field analysis [22], x-ray-based measurements [23], and density functional theory [24] show the atomic magnetic moments  $\mu_{4a}$  and  $\mu_{4c}$  of Mn on the two sublattices in MRG are similar (ratio  $\approx 1 : 1.2$  at absolute zero), whereas the ratio of  $\mu_{\text{Gd}}$  to  $\mu_{\text{Fe}}$  in  $\text{GdFeCo}$  is  $\approx 4 : 1$  [25–27]. The inter- and intrasublattice exchange interactions which are in the ratio  $J_{4a-4a} : J_{4a-4c} : J_{4c-4c} \approx 10 : -4.2 : 3.5$  [22] for MRG are much closer to each other than for  $\text{GdFeCo}$  where the ratio is  $J_{\text{Fe-Fe}} : J_{\text{Fe-Gd}} : J_{\text{Gd-Gd}} \approx 10 : -1 : 0.1$  [5,28]; the intersublattice exchange in MRG is up to three times stronger than in  $\text{GdFeCo}$ . These interactions not only accelerate the sublattice-specific demagnetization rates [27], but more importantly, the intersublattice exchange interaction facilitates exchange relaxation [29,30], whereby angular

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momentum  $S_i$  is exchanged directly between the two sublattices. These structural, electronic, and magnetic differences raise the question [10] of whether the same mechanisms underpin single-shot AOS in MRG and GdFeCo.

Here, we address the question of how single-shot AOS occurs in MRG, primarily by testing the limits where it fails. We demonstrate single-shot AOS for photon energies varying by a factor 30, from 1.55 down to 0.05 eV, and we show that pulse durations ranging from tens to several thousands of femtoseconds are able to achieve switching. As in GdFeCo [21,31], we identify a critical pulse-duration threshold  $\tau_c$  below which AOS is enabled that depends strongly on the sample temperature but, unlike GdFeCo, we find that switching vanishes completely when the initial temperature is above the compensation point. Finally, we propose a common framework for single-shot AOS that allows us to account for the similarities and differences between the two ferrimagnetic systems.

**Experimental results.** We have studied two films of  $\text{Mn}_2\text{Ru}_x\text{Ga}$  with  $x = 0.75$  and  $0.80$  of thickness 32 and 18 nm, respectively. Both films were deposited on MgO substrates by dc-magnetron sputtering using a ‘‘Shamrock’’ system, and capped with a 3-nm layer of aluminum oxide. Biaxial substrate-induced strain induces a slight tetragonal distortion of the structure that leads to perpendicular magnetic anisotropy [9,14]. The ferrimagnetic compensation temperatures  $T_{\text{comp}}$  of  $\text{Mn}_2\text{Ru}_{0.8}\text{Ga}$  and  $\text{Mn}_2\text{Ru}_{0.75}\text{Ga}$  were determined by magnetometry to be 345 and 370 K, respectively. These values depend on  $x$ , the film thickness, and the degree of atomic order determined principally by the substrate temperature. In equilibrium at temperatures above  $T_{\text{comp}}$ , the absolute angular momentum of the  $4a$  sublattice  $|S_{4a,0}|$  exceeds that of the  $4c$  sublattice  $|S_{4c,0}|$ , whereas below  $T_{\text{comp}}$ ,  $4c$  is the dominant one. As a point of comparison, we also studied a GdFeCo thin film structure  $\text{Si}_3\text{N}_4(60)/\text{Gd}_{25}\text{Fe}_{65.6}\text{Co}_{9.4}(20)/\text{Si}_3\text{N}_4(5)/\text{glass}$  (numbers in parentheses indicate layer thickness in nanometers), which compensates at 320 K [32], and is known to exhibit single-shot AOS [6].

To test the pulse duration and wavelength limits of single-shot AOS, we used the Free Electron Lasers for Infrared eXperiments (FELIX) facility in Nijmegen [33,34]. The photon energy of our Fourier-transform-limited pulses was varied between 50 and 160 meV and their duration [34,35]  $\tau$  ranges from  $\approx 200$  fs to  $\approx 7$  ps. A single optical pulse nominally of energy  $3 \mu\text{J}$  at 50 meV is focused to a Gaussian spot of radius [36]  $110 \mu\text{m}$  at 50 meV on the MRG samples. Other pulse-duration measurements were performed using 800-nm (1550-meV) excitation pulses from a Ti-sapphire laser. The incident optical fluence was  $\approx 8 \text{ mJ}/\text{cm}^2$ , and the pulse duration  $\tau$  was tunable from 48 fs to 6 ps with sub-100-fs resolution. Samples were mounted on a resistive heater allowing the initial temperature  $T_0$  to be set between room temperature and 450 K, thus tuning the equilibrium angular momenta of both sublattices [15,22]. The impact of pulses from either system on the magnetization was imaged via the Faraday effect using a nitrogen-purged magneto-optical microscope. Figure 1 shows typical Faraday images of a sample irradiated with pulses of different durations, giving rise to single-shot AOS or demagnetization.

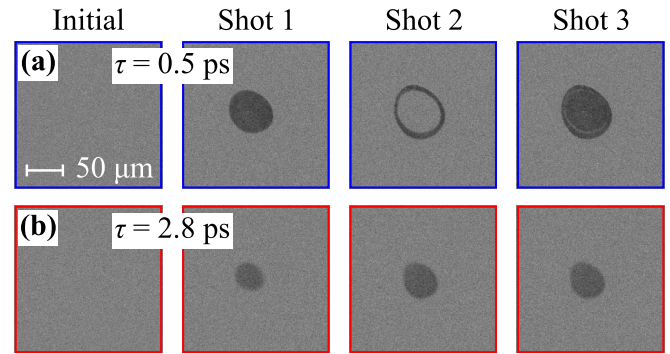


FIG. 1. Typical background-corrected magneto-optical images taken after exposing  $\text{Mn}_2\text{Ru}_{0.75}\text{Ga}$  at room temperature to consecutive optical pulses of photon energy 124 meV and duration  $\tau$  as indicated in rows (a) and (b). The images shown in row (a) demonstrate single-shot AOS whereas those shown in row (b) demonstrate demagnetization.

In Fig. 2 we present the results of testing single-shot AOS in  $\text{Mn}_2\text{Ru}_{0.75}\text{Ga}$  using 800-nm pulses of varying duration  $\tau$  and temperature  $T_0$ . After classifying the resulting images as showing single-shot AOS or demagnetization (as in Fig. 1), we constructed the state map shown.

We consistently identify a threshold pulse duration  $\tau_c$  at each temperature  $T_0$  above or below which the pulses activate either demagnetization or switching. Remarkably,  $\tau_c$  is strongly dependent on  $T_0$ , following a linear relationship

$$\tau_c = 17.6 - 0.048T_0, \quad (1)$$

for  $\text{Mn}_2\text{Ru}_{0.75}\text{Ga}$  with  $\tau_c$  in picoseconds and  $T_0$  in kelvins. It is clear that as  $T_0 \rightarrow T_{\text{comp}}$ ,  $\tau_c \rightarrow 0$ . No pulse is capable

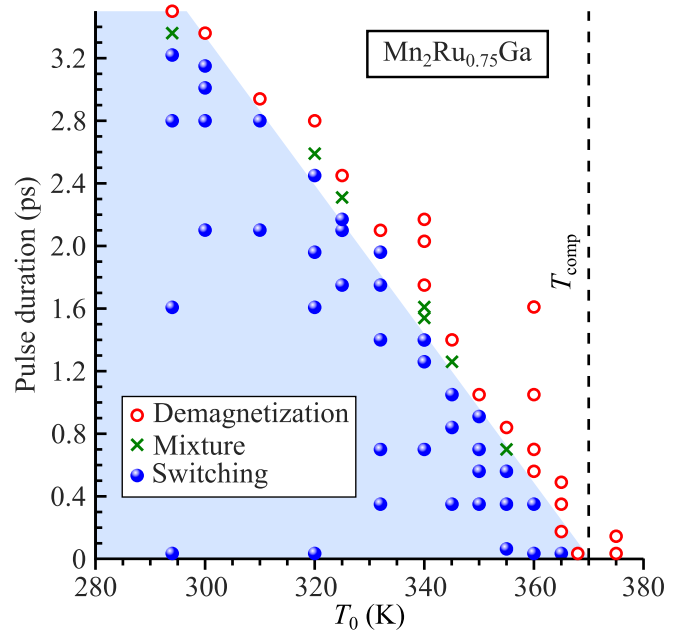


FIG. 2. State map recorded for  $\text{Mn}_2\text{Ru}_{0.75}\text{Ga}$  indicating whether single-shot AOS, demagnetization or a mixture of the two effects is achieved by an 800-nm pulse with the indicated duration at the starting temperature  $T_0$ .

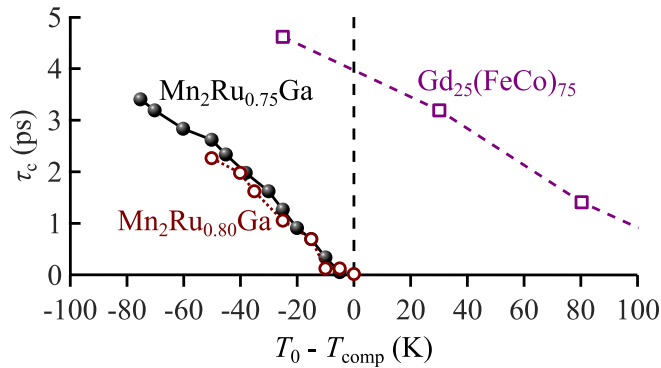


FIG. 3. Threshold pulse duration  $\tau_c$  for the two MRG and the GdFeCo films measured as a function of the difference between the compensation temperature and the measurement base temperature  $T_0$ . The compensation temperatures  $T_{\text{comp}}$  of  $\text{Gd}_{25}(\text{FeCo})_{75}$ ,  $\text{Mn}_2\text{Ru}_{0.8}\text{Ga}$ , and  $\text{Mn}_2\text{Ru}_{0.75}\text{Ga}$  are 320, 345, and 370 K, respectively.

of achieving single-shot AOS when  $T_0 > T_{\text{comp}}$ . The thermal dependencies of  $\tau_c$  in the two MRG films and in the GdFeCo sample are shown in Fig. 3. The underlying state diagrams upon which these trends are based are provided in Supplemental Material Note 1 [37]. In agreement with the composition-based observations in Ref. [10], neither of the two MRG alloys we tested could be magnetically switched by any laser pulse when  $T_0 > T_{\text{comp}}$ . In marked contrast, the magnetization of GdFeCo could be switched [32] when  $T_0$  is below  $T_{\text{comp}}$ , or even as much as 100 K above.

In Fig. 4 we present the results of testing single-shot AOS in  $\text{Mn}_2\text{Ru}_{0.75}\text{Ga}$  using pulses of different photon energies and durations produced by FELIX. Images taken after exposing the sample to several single-shot pulses are classified as evidence of single-shot AOS if the pulse switches magnetization uniformly across the irradiated surface, or demagnetization if the pulse switches the entire irradiated region into a multidomain state (Fig. 1). While there is about 30% fluctuation in the energy of the single pulses obtained from FELIX, we compensate for excessive incident fluence by examining whether the demagnetized region has a switched outer perimeter, where the local incident fluence is lower. Further details of this methodology are provided in Supplemental Material Note 2 [37].

We make two distinct observations. First, single-shot AOS was achieved in  $\text{Mn}_2\text{Ru}_{0.75}\text{Ga}$  across a very wide spectral range, with photon energies as low as 50 meV, and second, there is a critical pulse duration  $\tau_c \approx 2.5$  ps whereby any single pulse longer or shorter than this threshold induces either demagnetization or single-shot AOS. This latter feature is independent of the photon energy, and has recently also been observed for GdFeCo [21]. On repeating these measurements for the other alloy,  $\text{Mn}_2\text{Ru}_{0.80}\text{Ga}$ , we observe the same behavior except for a downward shift in the duration threshold to  $\tau_c \approx 1$  ps (see Supplemental Material Note 3) [37].

*Discussion.* The process of AOS in both MRG and GdFeCo can be understood using a common framework, involving an interplay of relaxation pathways that coexist and dominate on different timescales [5,21]. The strong laser-pulse-induced nonequilibrium state generated in these ferrimagnets results

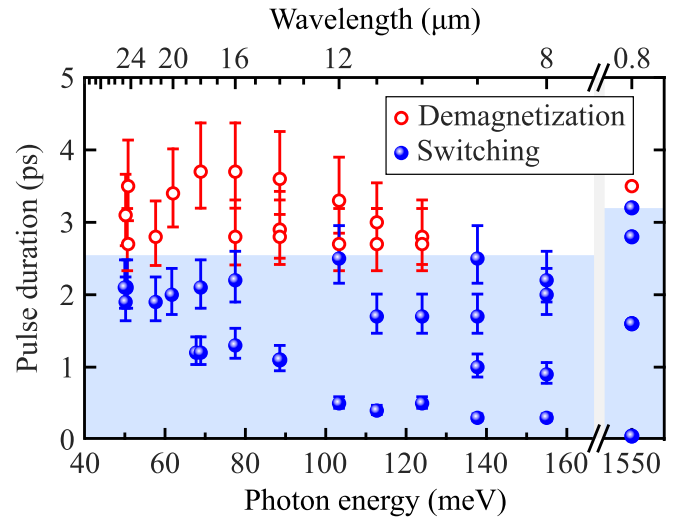


FIG. 4. State map recorded for  $\text{Mn}_2\text{Ru}_{0.75}\text{Ga}$  indicating whether single-shot AOS or demagnetization is achieved by a pulse with the indicated photon energy and pulse duration, measured at room temperature.

in three limiting cases of relaxation: (i) “ultrafast” demagnetization  $\partial S_i/\partial t \propto \alpha_i/\mu_i$ , where the sublattices  $i$  demagnetize independently, irrespective of the exchange interaction between them, at a rate determined by their atomic moment and damping [26]; (ii) demagnetization with conservation of spin angular momentum by exchange between the sublattices, i.e.,  $\partial S_1/\partial t = -\partial S_2/\partial t$ ; and (iii) “slow” magnetization changes, when the sublattices have a common temperature and their magnetizations are at all times in equilibrium with the temperature of the lattice.

We first review the temporal evolution of single-shot AOS in GdFeCo, originally mapped experimentally by Radu *et al.* [26] and later reproduced by more than ten [5,6,18,38–46] distinct models. Electronic states originating from both Gd and Fe sublattices are present at the Fermi level [15], so any suitably short thermal excitation [6,20,21] can efficiently stimulate their ultrafast demagnetization. Subsequent switching is then generally explained in terms of the two fast relaxation processes, (i) and (ii) above, which dominate on different timescales [21]. Prior to the timescale of the electron-lattice equilibration constant ( $\tau_{e-l} \approx 2$  ps [25]), the ultrashort optical pulse demagnetizes Fe four times faster than Gd [case (i)] because of the substantially different atomic magnetic momenta ( $\mu_{\text{Gd}}/\mu_{\text{Fe}} \approx 4$ ) and the relative weakness of the intersublattice exchange coupling [25,27,47,48]. When the magnetization of the Fe sublattice approaches zero, exchange relaxation [29,30,39,49,50] dominates and angular momentum flows between Gd and Fe, conserving the total angular momentum of the system [case (ii)]. This process, persisting for a timescale  $\tau_{s-s}$  connected to the exchange coupling between the two spin systems, drives the magnetization of the Fe sublattice across zero, giving rise to the critical transient “ferromagneticlike” state where Fe and Gd have parallel magnetic polarity. As the magnetization of Fe grows, the magnetization of Gd continues to fall towards zero as total angular momentum is conserved. Finally, antiparallel alignment is reestablished after the magnetic polarity of Gd

switches, and the spins and lattice equilibrate across distinct timescales  $\tau_{s-l}$ . Subsequent equilibrium cooling [case (iii)] finally yields a switched net magnetization [5].

The four-temperature model, in which the electron, lattice, and the two spin systems are assigned different heat capacities, coupling constants, and temperatures [45], quantitatively reproduces the experimental data for GdFeCo. The possibility of switching is unaffected by the ambient temperature  $T_0$  and the alloy composition [32], with magnetic reversal being achievable in  $\text{Gd}_x(\text{FeCo})_{100-x}$  alloys with  $\approx 22 < x < 28$  and with  $T_0$  a hundred or more degrees above or below the compensation point. With increasing  $x$ , however, the maximum pulse duration capable of activating AOS increases dramatically, with  $\tau_c = 400$  fs for  $x = 22$  [21] shifting to  $\tau_c \approx 15$  ps for  $x \approx 27.5$  [31]. This behavior can be explained [21] in terms of the equilibrium angular momentum reservoir of Gd rapidly growing with  $x$ , combined with the condition that the magnetization of Fe (the sublattice with the stronger intrasublattice exchange coupling) must cross zero before that of Gd for AOS to follow [21,51,52]. This condition can be qualitatively understood by considering the state in which both sublattices are considerably demagnetized, and are starting to recover. The sublattice with the stronger intrasublattice exchange interaction will also be the fastest to recover, and so successful AOS requires the magnetization of this sublattice (Fe) to cross zero before the other.

We now turn to MRG, which displays ultrafast demagnetization [16] similar to the ferromagnetic metal Ni [1] or the ferromagnetic half metal  $\text{Co}_2\text{MnSi}$  [53]. Oxide half metals such as  $\text{CrO}_2$  [53] generally exhibit very slow demagnetization because the Elliot-Yafet spin-flip relaxation channel is blocked and the charge fluctuations necessary for relaxation occur on a nanosecond timescale. Our finding that far-infrared photons can still efficiently demagnetize both sublattices (Fig. 4) suggests that the band structure of MRG promotes exchange scattering [case (ii) at the expense of case (i)] between the strongly exchange-coupled ( $-55$  meV)  $4a$  and  $4c$  sublattices on the nonequilibrium timescale [16], allowing for fast demagnetization at equal but opposite rates despite MRG's high ( $\approx 60\%$ ) spin polarization at the Fermi energy [9].

The combination of the equilibrium thermal dependence of angular momenta ( $S_{4a,0}$  and  $S_{4c,0}$ ) presented in Fig. 5(a) and the sublattice-specific demagnetization rates of case (ii) underpin the process of AOS in MRG. If  $T_0 < T_{\text{comp}}$ , the equilibrium thermal dependence of MRG implies  $|S_{4a,0}| < |S_{4c,0}|$ . Thus the equal demagnetization rates cause equal changes in  $S_{4a}$  and  $S_{4c}$  [ $|\Delta S_{4a}| = |\Delta S_{4c}|$  in Fig. 5(b)] and so  $S_{4a}$  crosses zero first with  $S_{4c}$  soon to follow. The  $4a$  sublattice has stronger intrasublattice exchange coupling compared to that of  $4c$  ( $J_{4a-4a} \approx 3J_{4c-4c}$ ), and so the antiferromagnetic intersublattice exchange coupling drives  $S_{4c}$  across the state with zero magnetization [54]. After the electrons and lattice have equilibrated ( $\tau_{e-l} \approx 1$  ps in MRG [16]), angular momentum not only flows between the spin sublattices but also increasingly leaks from the spins to the lattice, finalizing the switching process during the cooling of the ferrimagnet. It is important during this entire process that both  $S_{4a}$  and  $S_{4c}$  do not simultaneously fall to a level where thermal spin correlations can dominate, leading to loss of magnetic memory and

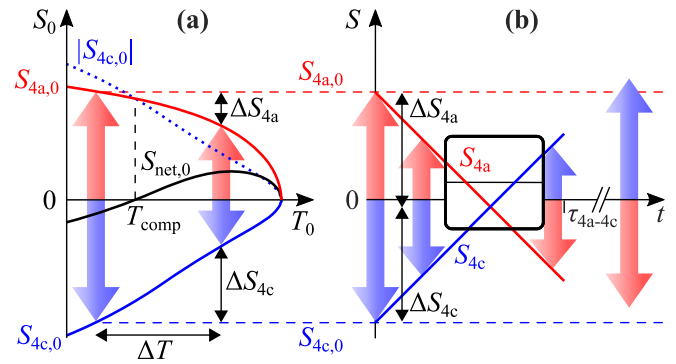


FIG. 5. (a) Representative equilibrium thermal dependence of angular momentum  $S$  in MRG, showing the change in angular momentum of the  $4a$  (red) and  $4c$  (blue) sublattice as the temperature ( $T_0$ ) varies in equilibrium. Equilibrium changes in temperature  $\Delta T$  lead to nonequal changes of angular momenta  $|\Delta S_{4a}| \neq |\Delta S_{4c}|$  via spin-lattice relaxation. (b) The nonequilibrium exchange-driven process of magnetic switching triggered by the 100-fs laser pulse at  $T_0 < T_{\text{comp}}$ . The exchange relaxation results in  $|\Delta S_{4a}| = |\Delta S_{4c}|$  when the temperature changes in nonequilibrium. Since  $|S_{4a,0}| < |S_{4c,0}|$ ,  $S_{4a}$  crosses zero before  $S_{4c}$ , giving rise to successful magnetic switching.

subsequent random behavior. This situation is encountered if the net incident fluence [21,55] is excessive, giving rise to the spatially inhomogeneous pattern shown in Supplemental Fig. S2.

If instead  $T_0 > T_{\text{comp}}$ , we obtain from Fig. 5(a)  $|S_{4a,0}| > |S_{4c,0}|$ . The pulse-induced  $-1:1$  demagnetization rates instead now cause  $S_{4c}$  to cross zero before  $S_{4a}$ , generating a transient “ferromagneticlike” state but with reversed polarity [51,52] compared to that shown in Fig. 5(b). Net switching cannot emerge from this nonequilibrium state because  $J_{4c-4c}$  is three times weaker than  $J_{4a-4a}$ ; the sublattice with weaker intrasublattice coupling ( $S_{4c}$ ) cannot drive the sublattice with a stronger intrasublattice coupling ( $S_{4a}$ ) across the state with zero net magnetization. Switching is therefore impossible to achieve in MRG when  $T_0 > T_{\text{comp}}$ , in agreement with our experimental findings shown in Fig. 3.

The above explanation also accounts for why GdFeCo can still be magnetically switched if  $T_0 > T_{\text{comp}}$ , when  $|S_{\text{Fe},0}| > |S_{\text{Gd},0}|$ , as experimentally shown in Fig. 3. Since  $\mu_{\text{Gd}} \approx 4\mu_{\text{Fe}}$ , an ultrashort pulse demagnetizes the Fe sublattice four times faster than Gd within the first 2 ps of electron-lattice equilibration, i.e.,  $|\partial S_{\text{Fe}}/\partial t| \approx 4|\partial S_{\text{Gd}}/\partial t|$  via case (i). Accordingly, at the time  $t \approx \tau_{e-l}$  when exchange relaxation [case (ii)] begins to dominate,  $|S_{\text{Fe}}| < |S_{\text{Gd}}|$ , and so the  $-1:1$  demagnetization rates promoted by exchange relaxation enable  $S_{\text{Fe}}$  to cross zero before  $S_{\text{Gd}}$ , resulting in magnetic switching in the same manner as in MRG.

Reducing the equilibrium initial temperature of MRG below compensation results in increasingly longer pulses being capable of activating the switching (Fig. 3). When  $T_0$  falls below  $T_{\text{comp}}$ ,  $|S_{4c,0}|$  grows twice as fast as  $|S_{4a,0}|$  [Fig. 5(a)], while the damping  $\alpha_i$  presumably remains constant [56]. This has important implications for the maximum pulse duration  $\tau_c$  capable of activating AOS. While the two sublattices  $4a$  and  $4c$  transfer angular momentum between each other via exchange relaxation, angular momentum may leak from both

of them to the structural lattice, presumably at similar rates reflecting their similarity. This competition between the spin-spin and spin-lattice equilibration processes [cases (ii) and (iii) above] introduces  $\tau_c$ , since  $\tau$  is directly responsible for triggering the three distinct limiting cases of demagnetization, introduced above.

When  $\tau < \tau_{e-l}$ , the sublattices demagnetize independently of each other. This, however, evidently does not dominate the demagnetization process in MRG (where  $\tau_{e-l} \approx 1$  ps [16]), as can be inferred from the absence of AOS above the compensation point.

Thus, when  $\tau < \tau_{s-s}$ , the sublattices demagnetize with conservation of total angular momentum. The equal changes of angular momentum in Fig. 5(b)  $|\Delta S_{4a}| = |\Delta S_{4c}|$  then correspond to unequal changes of the sublattice-specific temperatures in Fig. 5(a). When  $T_0$  is only just below  $T_{\text{comp}}$ , the demagnetization rates of both sublattices would have to be strictly  $-1 : 1$  for  $S_{4a}$  to successfully cross zero before  $S_{4c}$ . Consequently, only ultrashort pulses can achieve AOS when  $T_0$  is below but very close to  $T_{\text{comp}}$ , since the switching process must have been completed before any leakage whatsoever of angular momenta to the lattice occurs.

If instead  $\tau > \tau_{s-s,l}$ , the sublattices demagnetize persistently in equilibrium according to Fig. 5(a). Both sublattices experience the same change in temperature  $\Delta T$ , which leads to nonequal changes of angular momenta  $|\Delta S_{4a,0}| \neq |\Delta S_{4c,0}|$ . This case cannot activate AOS due to the absence of exchange relaxation.

If  $T_0$  is therefore far below  $T_{\text{comp}}$ , the criterion for AOS ( $S_{4a}$  crossing zero before  $S_{4c}$ ) can still be satisfied with some deviation from  $-1 : 1$  demagnetization rates. This situation is typically triggered by longer pulses (still satisfying  $\tau < \tau_{s-s}$ ) that realistically activate a combination of cases (ii) and (iii). Indeed, by using the experimentally deduced relation Eq. (1) along with the assumption that spin-lattice relaxation consumes all angular momentum remaining after exchange relaxation, we estimate [37] for MRG a characteristic spin-lattice relaxation time  $\tau_{s-l} \approx 10$  ps.

*Conclusion.* In conclusion, we have established how the photon energy, pulse duration, and sample temperature affect the single-shot AOS process in MRG. We found that switching can be achieved using far-infrared pulses, showing that the half metallicity of MRG is not an obstacle to the process, and suggesting that demagnetization of both sublattices evolves via exchange relaxation. This is further supported by our discovery that the pulse-duration threshold is strongly dependent on the sample temperature, with the compensation temperature representing a practical limit. This is not the case for GdFeCo.

We have introduced a framework in which the pulse-duration and temperature thresholds emerge naturally from the process of exchange relaxation, which dominates the demagnetization process at nonequilibrium timescales shorter than the spin-lattice relaxation time of 10 ps. Our work provides an experimentally grounded basis for the development of microscopic *ab initio* models of AOS in MRG [57–59], which benefits from the intrinsic crystallinity of MRG compared to the amorphous nature of GdFeCo alloys. Such microscopic models could resolve the question of how, for example, the transfer of angular momentum between the sublattices is electronically mediated. Our work also represents an important step towards understanding how single-shot AOS can be applied in engineered ferrimagnetic devices for spintronic applications.

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