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Ab initio study of ultrafast spin dynamics in $Gd_x(FeCo)_{1-x}$ alloys

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ABSTRACT

Using an ultrashort laser pulse, we explore *ab initio* the spin dynamics of $Gd_x(FeCo)_{1-x}$ at femtosecond time scales. Optical excitations are found to drive charges from Fe majority *d*-states to unoccupied Gd *f*-minority states with *f*-electron character excited occupation lagging behind that of the *d*-electron character, leading to substantial demagnetization of both species while leaving the global moment almost unchanged. For $x > 0.33$, this results in the creation of an ultrafast ferromagnetic (FM) transient by the end of the laser pulse with the Gd demagnetization rate slower than that of Fe. For all concentrations, the Gd moments begin to rotate from their ground state orientations developing in-plane moments of between 0.2 and 0.5 μ_B. Thus, the ultrafast spin dynamics of the material captures three important ingredients of all optical switching that occurs at much later (picosecond) times: (i) the development of a FM transient, (ii) the different rates of demagnetization of Fe and Gd, and (iii) the breaking of the collinear symmetry of the ground state. Furthermore, several predictions are made about the behavior of Fe–Gd alloys that can be experimentally tested and can lead to a spin-filtering device.

The field of laser driven spin dynamics on ultrafast time scales (from attoseconds to picoseconds) holds the possibility of profound technological innovation.[1–3] Central to this field in both scientific interest and technological importance is a simple question: how can the magnetic order be controlled by light? Since the pioneering demonstration of ultrafast demagnetization in Ni by light more than 20 years ago,[4] two purely optical approaches to manipulate a long range magnetic order have emerged. At femtosecond time scales, the optical inter-site spin transfer (OISTR) effect[5] predicts that, in multi-sub-lattice magnetic materials, a transient ferromagnetic (FM) state can be created from an uncompensated anti-ferromagnetic ground state. This is a generic early time effect with the long time fate of the transient still uncertain. On the other hand, at comparatively much longer time scales, of the order of picoseconds, the direction of spin (i.e., $+z$ or $-z$) in a broad class of materials can be flipped by laser light, the phenomenon of all optical switching (AOS).[6–8] In contrast to the OISTR effect, the switched state achieved in AOS is permanent and not transient. This represents a broad umbrella of different physical processes, including both helicity-dependent[9–11] AOS (which occurs on slower time scales and only under multiple pulses exposure) and helicity-independent switching,[7,8,12] which is a single-shot excitation that can be switched back and forth multiple times.

The few femtosecond or sub-femtosecond time scales of the OISTR phenomenon ensure that this effect occurs well before either exchange interactions or temperature emerge from the underlying non-equilibrium electronic system as well defined variables. This is exemplified by the theoretically predicted light induced change in the magnetic structure from anti-ferromagnetic to ferromagnetic in Co/Mn multilayers,[5] in which the magnetic moments in one layer reverse direction without rotating from the *z*-axis, a phenomenon that obviously cannot be described within a Heisenberg model picture of the dynamics of the magnetic order.

In Gd based alloys[6,13] and multilayers,[14] AOS can be driven by a single intense laser pulse (the polarization state of which is unimportant) with, however, switching of Gd moments occurring much after the application of the pulse of the order of a few picosecond later. In the case of $Gd_x(CoFe)_{1-x}$ alloys—an uncompensated anti-ferromagnet in the ground state—the laser pulse results first in reorientation of the

Fe moments to create a ferromagnetic transient[8] with the Gd spin direction subsequently switching to return the system to the anti-ferromagnetic structure of the ground state but with all moments now aligned in the opposite direction. Central to this phenomenon are believed to be (i) the distinct time scales on which Fe and Gd demagnetize and (ii) the anti-ferromagnetic coupling between Gd and Fe. It should be emphasized that, in contrast to the OISTR effect, the longer time scales of AOS imply that exchange interactions and local moments provide a valid description of the spin dynamics, and thus, moment rotation may play a role in the long time spin dynamics. In fact, at the experimental stoichiometry simple number of available states, argument shows that the Gd moment cannot flip by spin-flips or optical excitation induced transitions alone.

These two examples of the manipulation of the magnetic long range order by light evidently inhabit very different temporal landscapes: the OISTR effect represents an example of profound spin–charge coupling occurring at sub-exchange times, while AOS in $Gd_x(CoFe)_{1-x}$ occurs at picosecond time scales at which not only can the spin dynamics be characterized in terms of exchange interactions but also temperatures reliably assigned to the spin, lattice, and charge sub-systems. The question we wish to address in this work is to what extent these two different pictures of spin dynamics are connected: what role if any does the early time (purely electronic) spin dynamics have in the phenomenon of AOS in Gd_xFe_{1-x}? In particular, could the ferromagnetic transient be partly an early time OISTR phenomenon?

By doing fully *ab initio* calculations, we show (i) that the OISTR effect can create a ferromagnetic transient already at ∼10 fs, (ii) that increasing the concentration of Gd results in larger change in moment of Fe, but concomitantly a reduced demagnetization of Gd, (iii) that the excitation of the Gd *f*-electrons lags behind that of the *d*-electrons mostly likely indicating indirect Fe-*d* to Gd-*f* transitions via Gd-*d* states, (iv) the symmetry (i.e., the collinear magnetic alignment) is broken at very early times with the Gd moments already at 20 fs exhibiting out-of-plane components, and (v) the individual Gd moments behave incoherently, i.e., they all evolve very different in-plane components. Strikingly, therefore, two of the key ingredients of the later time AOS—the existence of a ferromagnetic transient and the breaking of collinearity—already have their footprint at very early times, suggesting the possibility of new routes to control AOS in this material. One should note that the ferromagnetic transient in experiment[8] exists from ∼200 fs to ∼1 ps with these much longer time scales due to the much increased duration and smaller amplitude in experiment as compared to our ultrashort pulse.

In the present work, fully *ab initio* calculations are performed comprising two steps: (a) the ground-state is first calculated using the density functional theory (DFT), the exchange–correlation (xc) functional, the local spin density approximation together with Hubbard U (LSDA+U), and subsequently; (b) the time dependent extension of the density functional theory (TD-DFT)[15,16] is employed as a fully first principles approach to study the dynamics of charges and spins. For this, we use adiabatic LSDA+U as the xc functional with $U = 6.7$ eV applied on the Gd *f*-states with this value held fixed during time propagation. In the recent, it has been debated if the LDA+U method correctly describes the physics of rare-earths.[17] However, for ultrashort time calculations, it is only the localization and position of the *f*-states that are critical, as these determine the transition energies and matrix elements of the optical excitations. While the localization and positioning of the *f*-states are well approximated by the LDA+U and are, thus, sufficient for the present work, exchange effects are less well captured and so at longer times (a few 100s of femtoseconds to picoseconds) at which exchange physics becomes important, the use of the LDA+U may become questionable.

This first principles approach for the ultrafast dynamics of spins and charges has been shown to accurately describe magnetic systems on femtosecond time scales.[5,18–22] A time step of 2.4 attoseconds was used for time propagation (see Ref. 23 for full details of the method). All calculations were performed within the framework of the all-electron full-potential linearized augmented plane wave method[24] as implemented in the Elk code.[23,25]

We have used an ultrashort laser pulse of full width half maximum (FWHM) 3 fs. While the FWHM of this pulse resides within the range of experimentally feasible pulses, the incident fluence (88 mJ/cm^2) and the peak power (4×10^{13} W/cm^2) would appear to be much larger than experimental pulses that have much greater FWHM and significantly lower fluence. Such pulses, due to non-linear effects, result in excitations far above the Fermi level up to 8 eV above the Fermi level as shown in a recent experimental investigation.[26] Such states, however, have very short lifetime and, thus, quickly decay to lower energy states by photon emission or by scattering events. This results in additional transitions to lower energy states (i.e., 1–4 eV above the Fermi level). This decay of high energy states is not included in our theoretical approach. We can, however, determine the energy "locked in" to these high energy states and find it to be of the order of 80% of the energy. Thus, in order to reproduce the energy available for creating low energy excitations in experiment, we require a significantly higher absorbed fluence to compensate for the energy "locked into" high energy states.

We have used the C15 cubic Laves (spacegroup 227) unit cell with a lattice parameter of 7.32 au. This unit cell contains six atoms and, thus, allows for the study of concentration dependence via variation in the number of Gd and Fe atoms (e.g., 33% Gd and 67% Fe unit cell consists of 4 Fe and 2 Gd atoms). Prohibitive computational costs for larger unit-cells limit us to these few concentrations. A $12 \times 12 \times 12$ **k**-point set was used, and conduction states up to 50 eV above the Fermi energy were included.

In Fig. 1(a), we present the time dependent moments of Gd and Fe in Gd_xFe_{1-x} for four different values of x under the influence of a pump laser pulse. Note that only the z-component of the magnetization is shown, and in the ground state, all moments are aligned along the z-axis (with the exception of the highest Gd concentration for which the ground state exhibits a small canting of the Gd moments). As can be seen, for all concentrations, the laser pulse results in significant demagnetization of both Gd and Fe with the maximum decrease in Gd moment obtained at the highest Fe concentration and, vice versa, the maximum change in Fe moment occurring for the highest Gd concentration. At the highest Gd concentration ($x = 0.67$), Fe demagnetizes and re-magnetizes in the opposite direction with a moment of $1.6\mu_B$ to create a transient ferromagnetic state. On the other hand, for Gd concentrations close to those at which AOS switching occurs in experiment ($x = 0.33$), the Fe lattice is already fully demagnetized at the end of the pulse.

It should be stressed that due to their opposite alignment, the change in the total moment is comparatively small and within all cases, the change in the global moment is less than 15%. This spin

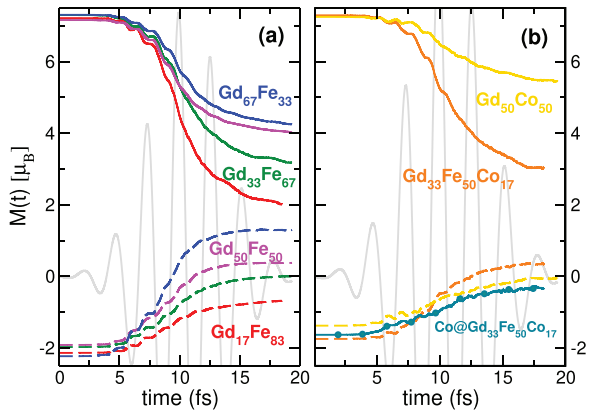

FIG. 1. Moment on Gd (solid lines) and Fe (dashed lines) in (a) Gd_xFe_{1-x} alloys, (b) $Gd_{33}Co_{17}Fe_{50}$ and $Gd_{50}Co_{50}$ alloys. The laser pulse is also shown in grey and has pulse parameters full width half maximum 3 fs, frequency 1.55 eV, peak power 4×10^{13} W/cm^2, and fluence 85 mJ/cm^2. Note that only the z component of the moments is shown here; the laser pulse also induces an out-of-plane component, see Figs. 3 and 4.

dynamics of profound change in local moments alongside a comparatively small change in the global moment rules out spin–orbit coupling as driving the demagnetization and is characteristic of the OISTR effect that dominates early time spin dynamics in multi-component magnets. This reflects the time scales of the key underlying physical processes: the purely optical excitations that drive the OISTR effect occur on a much faster time scale than spin–orbit induced transitions between majority and minority channels, which are yet faster than the characteristic time coupling of spin angular momentum to the lattice degrees of freedom. The demagnetization of Fe and Gd, for the ultrashort laser pulse we consider, would, thus, appear to be dominated by the OISTR effect.

To confirm this in Fig. 2(a), we show the density of states for $Gd_{50}Fe_{50}$ both in the ground state (labeled as $t = 0$) and at $t = 25$ fs after the action of the pulse. The ground state DOS shows, as usual, both occupied and unoccupied states with all states above $E_F = 0$ eV

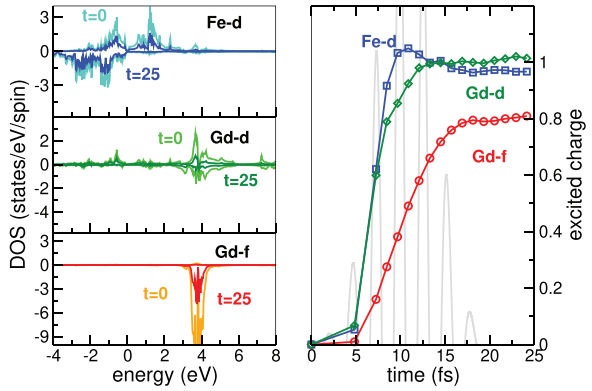

FIG. 2. (a) Species and angular momentum projected, ground-state DOS, and occupied DOS at t = 25 fs (b) excited charge projected on the Gd and Fe states. For both cases, the system shown is a $Gd_{50}Fe_{50}$ alloy. In panel (b), the laser pulse is also shown in grey and has full width half maximum 3 fs, frequency 1.55 eV, peak power 4×10^{13} W/cm^2, and fluence 85 mJ/cm^2.

(indicated by the vertical line) being unoccupied, while the transient DOS shows only occupied states. As can be seen, there is substantial depletion of both spin channels in Fe; however, the minority channel is largely inactive in the dynamics of the magnetic moment: optical excitations merely promote minority spin charge from below to above E_F without changing the moment. The behavior of the Fe majority channel, however, is dramatically different: optical excitations create transitions from majority Fe d-states to minority Gd f-states. This transition from majority to minority within the same spin channel is a consequence of the anti-ferromagnetic alignment of the Fe and Gd moments, and the net result is to reduce the moment on both Gd and Fe, exactly as seen in the dynamics of the moment presented in Fig. 1(a). Note that while there is some excitation from Fe into Gd d-states [see Fig. 2(b)]—as transitions from Fe to Gd occur in both majority and minority channels, this is less important for the local Gd moment but does contribute to the reduction in the local Fe moment. Most interestingly, as can be seen in Fig. 2(b), the Gd f-state charge excitation discernibly lags behind that of Fe d and Gd d charge excitation, while at the pulse peak, the Fe and Gd d-charges have reached close to their final values, but the Gd f character charge is less that ~50% of its final value. This may indicate that the charge in Gd-f states is excited via Gd-d states due to hybridization; this process of indirect excitation is likely driven by the comparatively small optical matrix element between Fe-d and Gd-f states.

The OISTR effect depends crucially on the number of states available to be excited and the number of empty states available to be excited into. Increasing the Gd concentration creates a greater number of unoccupied minority states, thus leading to a greater change in the Fe moment upon laser excitation but concomitantly less reduction of Gd. Correspondingly, at high Fe concentrations, the number of occupied majority Fe d-states increases, leading both to increased Gd demagnetization on laser excitation and less loss of the average Fe moment. These "state counting" arguments (i.e., the number of available states) evidently can also be extended to changing the alloy species. Substitution of Co for Fe in this way, due to the reduced exchange splitting in Co, is expected to lead to less moment loss on laser excitation. This is shown in Fig. 1(b), where in comparison with $Gd_{50}Fe_{50}$, it is seen that, as expected, in $Gd_{50}Co_{50}$ both species show less change in their magnetic moment. The dependence of the early time spin dynamics on the Fe concentration and Co substitution is theoretical prediction that can directly checked in experiments.

We now consider the question of the time evolution of the in-plane components of the magnetic moments. As shown in Fig. 3, moment in the xy plane is averaged over all the Gd atoms in the cell, i.e., $\overline{m_{xy}} = \frac{1}{N}\sum_{i=1}^{N}\sqrt{m_{i,x}^2 + m_{i,y}^2}$ with N being the number of Gd atoms and $m_{i,x}$, $m_{i,y}$ being the x and y components of the ith atom. As can be seen, except for the case of the highest Gd concentration, $Gd_{67}Fe_{33}$, a clear in-plane moment develops starting at approximately the time of peak of the pulse (shown in grey) at 10 fs. For $Gd_{67}Fe_{33}$, the Gd moments are already slightly canted from the z axis with a finite in-plane moment observed at $t = 0$. Such a non-collinear character ground state is expected for high Gd concentrations as the Gd inter-site exchange interactions are both weak in magnitude and frustrated due to their long range RKKY character. In this case, the average in-plane moment starts to rotate at the same time of ~10 fs seen for the other concentrations. In contrast, the Fe local moments we find,

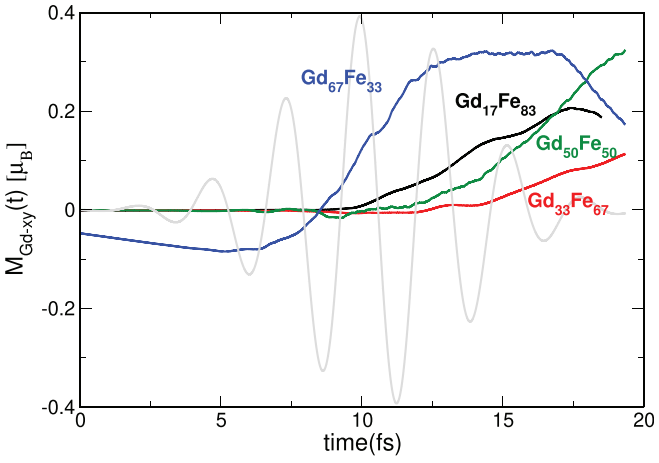

FIG. 3. Average moment for Gd atoms in the *x*–*y* plane for four different concentrations of the Gd_xFe_{1-x} ordered alloy. For the largest Gd concentration, the ground state already exhibits non-collinearity of the Gd moments due to the long range RKKY character of the Gd–Gd coupling. For all concentrations, the Fe local moments are aligned along the *z* axis both in the ground state and after the laser pulse.

for all alloy concentrations, to be always aligned along the *z*-axis, both in the ground state and during the spin and charge dynamics.

An important question is whether the out-of-plane moment develops in a coherent manner, i.e., if the canting from the *z*-axis is the same for all Gd atoms. If this were the case, then an angle dependent x-ray magnetic circular dichroism (XMCD) signal would be able to measure this. To better visualize the development of the in-plane components in Fig. 4, we show the evolution of the in-plane components for each Gd atom with the color indicating the size of the moment in the *z* direction. Shown in panel (a)–(d) are, respectively, $Gd_{17}Fe_{83}$, $Gd_{33}Fe_{67}$, $Gd_{50}Fe_{50}$, and $Gd_{67}Fe_{33}$. These have 1, 2, 3, and 4 Gd atoms in the six atom unit cell that we employ. Evidently, these moments behave quite incoherently with each Gd atom developing its own in-plane components. This early time rotation of the Gd moments from the *z*-axis will, therefore, not be measurable in any element-specific magnetic techniques that average over macroscopic laser-excited areas/volumes, e.g., XUV/soft x-rays, XMCD, and MOKE. Large angle canting has also been reported in atomistic spin dynamics simulations;[13] however, these obviously entail much longer time scales than those considered here.

To summarize, using the state-of-the-art *ab initio* time dependent density function theory, we have addressed the early time spin dynamics of ordered $Gd_x(CoFe)_{1-x}$ alloys driven by a short pulse of 3 fs full width half maximum (FWHM), which is a duration comparable to the currently shortest available pulses available in the experiment. We find that the early time spin dynamics is dominated by OISTR physics: optically induced excitations from the Fe-*d* majority to the Gd minority result in a substantial loss of moment from both species, while leaving the global moment almost unchanged. The smaller moment of Fe, nevertheless, results in a much faster demagnetization of this species with for concentrations $x > 0.33$ re-magnetization in the opposite direction creating a ferromagnetic transient. We also find that the Gd *f*-electron character excited charge lags behind *d*-electron excitation of Gd and Fe: at the pulse maximum, the latter are already close to the final values with, on the other hand, the excited charge of Gd *f*-electron character having reached only ∼50% of its final value. This might indicate an indirect transfer of charges from Fe-*d* states to Gd-*f* states via Gd-*d* states, which is a process made more likely by the comparatively small optical matrix element for direct Fe-*d* to Gd-*f* transitions.

The early time spin dynamics also displays clear symmetry breaking of the collinear ground state with small but significant rotation of the Gd moments, occurring within the trailing edge of the pulse (0.2–0.5μ_B in-plane components). Thus, three key aspects of the later time spin dynamics in all optical switching (AOS) in Gd_xFe_{1-x} alloys are, thus, found in the very early time regime: (i) the FM transient, (ii) the slower rate of demagnetization of Gd as compared to Fe, and (iii) the breaking of ground state collinearity.

The magnitude of the OISTR effect depends on both the available empty states as well the states available for optical transitions to excite into this unoccupied band. The moment of the FM transient, as well as the magnitude of Gd demagnetization, can, thus, be tuned both by alloy concentration and alloy species, e.g., by substituting Fe by Co, and our calculations predict that the early time moment loss can be controlled by substitution of Fe by Co. Furthermore, the time scale on which this transient is created will be dictated solely by pulse parameters. We, thus, find that the FM transient in $Gd_x(CoFe)_{1-x}$ is, in principle, highly controllable in the early time regime. The dependence of the early time spin dynamics on the Fe concentration and Co substitution is two key *ab initio* predictions that can be investigated in experiments. Interestingly, while we have only considered ordered alloys, the OISTR effect has previously been demonstrated to hold in an interface geometry.[22,27] In this case, spin currents across the interface result in changes to moments of atoms adjacent to the interface. Recent experiments demonstrating all optical switching in a multilayer geometry are, thus, consistent with the early time picture we study here.

The transient FM state of Fe–Gd also implies that this material is an excellent choice for the so-called OGMR effect (OISTR induced giant magnetic resistance). The material being initially ferri-magnetic with oppositely aligned Gd and Fe moments implies that if a spin polarized current was to pass through such a material, both spin channels would feel a large resistance. However, during the laser induced transient FM state, one of the spin channels (parallel to the direction in which the FM spins are oriented) feels much less resistance. Hence, such a setup can act as a transient spin-filter. This is again a prediction, which can be experimentally directly tested and could lead to a transient spin-filtering device.

For the longer pulses typically employed in the study of AOS, the OISTR physics described here is likely to be of importance, as such physics plays an important role at such time scales in other materials, augmented by lattice and spin-orbit coupling (SOC) effects that become more significant at 100s of femtoseconds. How this highly non-equilibrium early time regime evolves into a regime in which the effective exchange interactions and temperatures emerge as good variables remains perhaps the most significant unanswered question for AOS. Indeed, all present theories of AOS address this later time domain and cannot describe early time electronic effects such as the OISTR physics demonstrated here. At longer times, both inter-atomic exchange and intra-atomic exchange (between the *f*- and *d*-states of the rare earth) have been shown to be important,[28] highlighting the importance of treating the underlying electronic structure realistically,

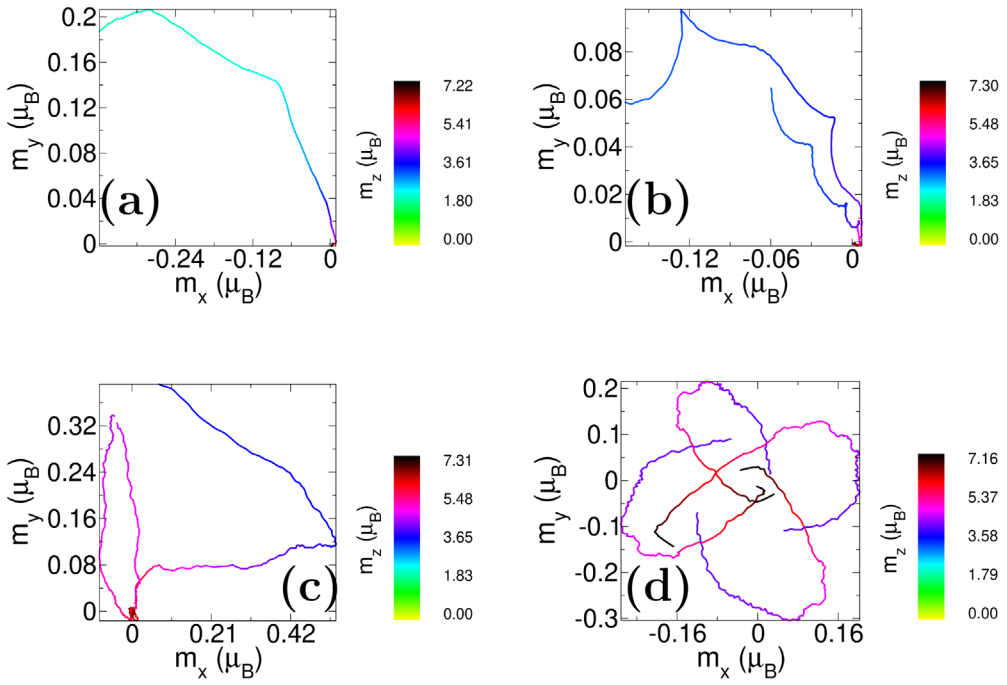

FIG. 4. Moment on various Gd atoms in the xy plane for four different concentrations of Gd_xFe_{1-x} ordered alloy: (a) $Gd_{17}Fe_{83}$, (b) $Gd_{33}Fe_{67}$, (c) $Gd_{50}Fe_{50}$, and (d) $Gd_{67}Fe_{33}$. Note that the Gd local moments all move in different directions under laser excitation, rendering its difficulty to probe them via angle dependent spectroscopy. The Fe local moments, in contrast, are aligned with the *z*-axis both in the ground state and under laser excitation.

as we do here. On the other hand, simply "freezing" the underling electronic system into a set of fixed exchange parameters may not be sufficient even at longer times, as electronic excitations driven by exchange scattering between the delocalized band states and the localized *f*-electrons may play a role.[29] Future work to bridge these two time regimes and their distinct theoretical methodologies, at the simplest level with *ab initio* calculations providing "initial conditions" for models of later time dynamics, might be expected to lift many of the uncertainties surrounding the origins of this remarkable effect.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors declare no conflict of interest.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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