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Ultrafast laser-induced dynamics of noncollinear spin structures in amorphous NdFeCo and PrFeCo

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The fanned out, noncollinear spin structure of the Fe(Co) sublattice in amorphous NdFeCo and PrFeCo alloys is shown to strongly affect its ultrafast laser-induced magnetization dynamics. An overshooting effect is discovered at low applied magnetic fields, where the magnetization temporarily increases above its equilibrium value. We explain this phenomenon by considering the dynamics of the noncollinear spin structure. After femtosecond laser excitation the system first reconstructs magnetic order on a time scale of 50–150 ps arriving at a state with a smaller opening angle of the fan. Subsequently, the original opening angle is restored on a time scale of nanoseconds. Increasing the field up to 0.6 T we can fully close the fan and therewith suppress the overshooting behavior.

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Magnetic structures in materials come in much greater variety than the basic collinear ferro- and antiferromagnetic ordering. There is an intriguing diversity of noncollinear spin structures ranging from completely disordered spin glass states to canted structures in frustrated systems and even more exotic constructs, such as magnetic vortices and glass states to canted structures in frustrated systems and spin structures ranging from completely disordered spin ordering. There is an intriguing diversity of noncollinear variety than the basic collinear ferro- and antiferromagnetic noncollinear structures are thought to be key elements in the rise of spintronics. Understanding and controlling the magnetic interactions could possibly lead to yet unexplored magnetic structures, which leads to a sudden increase in magnetization above its equilibrium value in response to a femtosecond laser excitation. This unexpected behavior of the magnetization dynamics can be described by a transient ferromagnetic alignment of the TM moments.

The investigated samples are prepared using magnetron sputtering and have the same general multilayer structure. A 20 nm RE-TM amorphous film is deposited on a glass [AlTi(10 nm) SiN(5 nm)] stack. The AlTi film serves as a heat sink, whereas the dielectric SiN is used as the buffer layer. The samples are capped by 60 nm SiN to prevent oxidation while also being highly transparent in the visible to near infrared, allowing for optical access to the RE-TM layer. The RE-TM layer is of the form REFe100−x+yCo y. In this Rapid Communication, we report results on two representative samples with different light RE elements, Nd30Fe61Co9 and Pr20Fe70Co10 (hereafter referred to as Nd-FeCo and PrFeCo). For comparison, two samples containing the heavy RE elements gadolinium and dysprosium were also investigated. We used conventional static and time-resolved (TR) magneto-optical Kerr effect (MOKE) methods [9] to study the static and dynamic magnetic properties of these samples.

Figure 1(a) shows typical MOKE hysteresis loops at room temperature of the two light RE-TM samples. The samples were probed with light of 630 nm central wavelength corresponding to a photon energy of 1.97 eV. In principle, this photon energy is not sufficient to excite the transitions involving 4f states, which carry most of the RE magnetic moment and lie at energies higher than 3 eV [10,11]. Even though photoemission spectra show splitting of the 4f peaks in certain Pr and Nd-TM alloys [12] to energies below 2 eV, the magneto-optical spectra of our samples between 1 and 3 eV are very similar to those of pure Fe (see Supplemental Material [13]). Therefore we believe our measurements to be mainly sensitive to the TM sublattice but cannot completely rule out a RE contribution to the signal.

The external magnetic field  H was applied along the in-plane easy axis of magnetization [inset Fig. 1(a)]. The results for the PrFeCo [Fig. 1(a) top] and the NdFeCo [Fig. 1(a) bottom] sample show specific similarities. The loops are
This fan structure depends on the relative strengths of the film plane and shows no hysteresis loop but only linear behavior. Magnetization curves with the field applied perpendicularly to our measurements are most sensitive, the TM-TM exchange both magnetic sublattices. For the TM sublattice, to which a fanlike distribution of the magnetic moments in possibly count the sperimagnetic structure of the samples, implying...
applied magnetic fields: After an initial demagnetization, the signal increases to a value greater than at negative probe delay. This indicates a transient higher magnetization than in the unperturbed state before pump arrival. The effect is reduced with increasing magnetic field, showing a strongly field-dependent remagnetization, until it vanishes completely. Interestingly, a complete quenching of this field-dependent behavior occurs with the onset of region III (Fig. 2) for the respective sample. For fields above $H_S$, the magnetization dynamics does not change. This field-dependent remagnetization and overshooting of the signal occurs exclusively in region II of the NdFeCo and PrFeCo samples and was not observed in GdFeCo and DyFeCo.

We here offer a phenomenological explanation by not only taking into account the longitudinal magnetization dynamics of the magnetic moments but also a dynamic response of the fan structure. If we assume that the pump stimulus causes the opening angle of the fan to first close, then relax back to its original equilibrium angle, we can retrace the observed overshooting behavior as a combination of the fan dynamics and longitudinal dynamics [Fig. 3(a)]. The following approach is used in order to single out and quantify the dynamics of the magnetic fan structure of the TM sublattice.

We first fit the remagnetization occurring in region III [blue curve, Fig. 3(a)] using a biexponential function, which is then subtracted from the measurements at fields in region II. Assuming that in a first approximation, the longitudinal dynamics of the magnetic moments—independently of the fan dynamics—does not change throughout region II and region III, this procedure reveals the fan dynamics only. As an example, Fig. 3(a) shows the separate and combined contributions of the fan (red dashes) and longitudinal dynamics (blue curve) for a low-field measurement on the PrFeCo sample. Using this procedure, one can extract the amplitude [green A in Fig. 3(a)] and delay position of the peak in the fan dynamics signal for all fields. The peak amplitudes shown in Fig. 3(b) clearly illustrate the disappearance of the overshooting effect towards higher applied fields. Furthermore, it is possible to deduce the change in the fan opening angle $\Delta\alpha$ using Eq. (1). It is assumed that the equilibrium opening angle before pump arrival for a measurement at a certain field corresponds to the value derived from the static measurements at the same field. For the lowest fields measured in region II we calculate a change in opening angle of $8^\circ$ at 0.11 T for PrFeCo and $20^\circ$ at 0.08 T for NdFeCo.

The peak position, on the other hand, lies at a delay of $\sim50$ ps for PrFeCo and $\sim150$ ps for NdFeCo and changes little with field. However, first preliminary data suggest that it strongly depends on temperature (not shown) and pump fluence (see Supplemental Material [13]). A more rigorous study has yet to be performed.
In the following, we give a tentative description of the mechanism leading to the overshooting phenomenon. Upon pump arrival, the electron system is heated up which leads to a partial demagnetization within the first couple of picoseconds. Electron-phonon interaction heats up the lattice until thermal equilibrium between electrons and lattice is reached. Taylor et al. [15], investigating sperimagnetic amorphous GdFeCo and GdFeNi alloys, showed evidence that increasing the temperature will in turn increase ferromagnetic alignment in the TM magnetic sublattice. This agrees with our measurement of magneto-optical hysteresis loops at different temperatures (see Supplemental Material [13]) which show a shrinking of region II upon increasing the temperature. As competing antiferromagnetic and ferromagnetic exchange interactions are the main force behind the fanning of the Fe(Co) magnetic moments, they are now (more) free to align collinearly. This effectively reduces the opening angle of the magnetization cone, leading temporarily to a higher net magnetization.

Depending on how fast this process occurs and on how much the longitudinal magnetization has recovered at that point, the measured signal can be above the initial value. Towards longer delay times the energy is dissipated and the local magnetic frustration, and with it the opening angle of the magnetic fan structure, recovers until it reaches its initial value. Following this mechanism one can explain the field-dependent behavior. The higher the applied field, the smaller the initial opening angle. Accordingly, the overshooting effect decreases along with the difference in net magnetization between the aligned state and the fan state and finally vanishes at \( H_S \).

In conclusion, we provided direct experimental evidence through static and dynamic magneto-optical measurements that the TM sublattice in Nd- and PrFeCo alloys possesses a fanlike magnetic structure. We showed that if stimulated by an ultrafast laser pulse, this fan structure undergoes its own dynamics, the results of which mix with the longitudinal dynamics of the magnetic moments. Upon applying an external magnetic field of sufficient strength to align all TM magnetic moments, the fan dynamics is suppressed and only the longitudinal response of the magnetic system remains. By increasing the magnetic field it is thus possible to switch between a regime determined by magnetic frustration to a regime purely governed by ferromagnetic exchange. This behavior might not only be limited to light RE-TM alloys but could potentially be a general feature of noncollinear spin structures.

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