Terahertz charge and spin transport in metallic ferromagnets: the role of crystalline and magnetic order

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We study the charge and spin dependent scattering in a set of CoFeB thin films whose crystalline order is systematically enhanced and controlled by annealing at increasingly higher temperatures. Terahertz conductivity measurements reveal that charge transport closely follows the development of the crystalline phase, with increasing structural order leading to higher conductivity. The terahertz-induced ultrafast demagnetization, driven by spin-flip scattering mediated by the spin-orbit interaction, is measurable in the pristine amorphous sample and much reduced in the sample with highest crystalline order. Surprisingly, the largest demagnetization is observed at intermediate annealing temperatures, where the enhancement in spin-flip probability is not associated with an increased charge scattering. We are able to correlate the demagnetization amplitude with the magnitude of the in-plane magnetic anisotropy, which we characterize independently, suggesting a magnetoresistance-like description of the phenomenon.

In this Letter, we design a study to investigate in greater detail how terahertz-driven demagnetization and crystalline structure are related. We choose a similar CoFeB amorphous system studied in Ref. [14], but now we systematically change its lattice structure from amorphous to crystalline under controlled annealing, as shown in Ref. [20]. This study shows that annealing increases the ordering of the lattice, monitored with X-ray diffraction, spectroscopic ellipsometry, and magneto-optical Kerr effect spectroscopy. In turn, the different lattice structure affects the optical and magneto-optical properties of the sample. Here, by comparing the ultrafast demagnetization driven by intense THz fields, and the THz electronic conductivity, we find indication of a more complex interplay between magnetic and structural order, where enhanced spin-dependent scattering is observed without an associated enhancement of the charge scattering at an intermediate state where the magnetic anisotropy is enhanced.

Pristine CoFeB films were deposited via magnetron sputtering. The stacking structure of our samples is Co_{60}Fe_{30}B_{20} (100 nm)/Pt (5 nm), grown on a 500 μm thick silicon substrate with a native oxide layer on top. The samples were oven-annealed at different temperature ranging from 300 °C to 600 °C in steps of 50 °C for 60 minutes. The samples were mounted in a THz-pump / NIR-probe setup, in the geometry shown in Fig. 1a, on top of a permanent magnet producing a magnetic field μ₀H ≈ 400 mT. This magnetic field causes the easy plane magnetization vector M to tilt out of plane, while preserving a sizable in-plane component. An intense THz magnetic field H_{THz} with polarization normal to the direction of M excites the system which maximizes the M × H_{THz} torque on the magnetization [14,18,19]. The dynamical response of the magnetization is measured in the polar

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magneto-optical Kerr effect (MOKE) geometry using a nominally 40 fs near-infrared probe pulse and a balanced detection scheme\textsuperscript{18}.

The intense THz fields are generated by optical rectification of a 1500 nm pulse (generated by optical parametric conversion from the 800 nm laser fundamental) in an organic DSTMS crystal\textsuperscript{25,26}. By performing an electro-optic sampling of the THz pulse in a 50 µm thick GaP crystal\textsuperscript{27}, we reconstruct the temporal variation field of the single-cycle THz pulse shown in Fig. 1(b). We estimate a free-space peak electric field of approximately 800 kV/cm, corresponding to a peak magnetic field of 270 mT, which lasts for less than a picosecond. In Fig. 1(b) we also show that we can monitor and control the sign of the THz field (both electric and magnetic components) by rotating the THz-generating organic crystal by 180 degrees, producing the fields that we label $+H_{THz}$ and $-H_{THz}$ throughout this work. The signals detected at approximately 2.5 ps and 4 ps are the echos of the terahertz pulse traveling through the GaP crystal, reflected at each surface. They are shown for completeness, but they are not relevant for the actual experiment, since the much thicker silicon substrate separates these echos by more than 10 ps, beyond the measurement range of interest. For both the electro-optical sampling and the actual measurements, the time-delay between pump and probe beams is accurately controlled by a delay stage in one of the two beams’ paths, given that both pulses originate from the same laser and are hence intrinsically synchronized.

Fig. 1(c) shows the polar MOKE signal as a function of delay between pump and probe, mapping the magnetization dynamics initiated by terahertz pulses with opposite polarity. From these plots, we first notice the two distinct dynamics already observed in Refs.\textsuperscript{14,18}: a coherent response of the magnetization, which is associated with the torque exerted by the magnetic component single-cycle THz field and which depends on its sign, and an incoherent response that is independent on the field polarity. The latter is understood in terms of an ultrafast demagnetization which is recovered within a few picoseconds. Furthermore, considering that all subplots were plotted on the same scale, and that the measurements were performed in the same geometry, we observe a variation of the maximum amplitude of the MOKE signal between samples that were annealed at different temperatures. In particular, the MOKE response has a relatively low peak-
to-peak amplitude (50 μrad) in the pristine sample, which reaches a maximum of 100 μrad at an annealing temperature of 500 °C, and then decreases at even larger annealing temperatures. In the Supplementary Material, we calculate this variation more precisely by taking the maximum difference, i.e. max[θ_K(+HTHz) − θ_K(−HTHz)] between the MOKE responses to THz fields with opposite polarity, hence removing the one-sided offset caused by the demagnetization. With this analysis, we find that the amplitude variation of the coherent response is consistent with what was observed in the previous studies on similar samples. This observation is very important, because it provides us with a normalizing method within the data themselves, allowing us to separate magneto-optical effects from intrinsic magnetic ones, which are the ones of interest in our work.

We now turn to the incoherent response of the sample to the terahertz field, i.e. to the ultrafast demagnetization driven by the spin-polarized current flowing uniformly in the sample, which dissipates the energy deposited by the pump pulse. In order to better separate the demagnetization from the coherent response, we follow the procedure explained in Ref.14 in Fig. 2, we plot the sum of the MOKE signal originating from THz fields with opposite polarities, i.e. θ_K(+HTHz) + θ_K(−HTHz). This procedure cancels out most of the coherent dynamics and allows us to perform a fit to the data using the approach of Ref.14. Since the MOKE signal varies with annealing temperature, we also normalize the data by the maximum of the coherent MOKE response, as discussed above. Looking at the data in Fig. 2, we can evince that the qualitative shape of the demagnetization is the same for all samples. However, we see an evident trend in the demagnetization amplitude: the effect is relatively small for the pristine sample and for those annealed at 300 °C and 350 °C, it then starts to grow and reaches a maximum at 450 °C, to almost disappear at the highest annealing temperatures. We analyze the results of the fitting procedure in detail later in the text.

Before moving to discuss the results so far obtained, we also estimate the resistivity variation between the different films using both conventional sheet resistance methods, as well as terahertz time-domain spectroscopy. The resistivity is a measure to quantify the charge transport, with the terahertz data accessing the relevant demagnetization time scales. In metals the charge transport is strongly connected with the spin transport, and it is therefore an important parameter to estimate. We show in the Supplementary Material the details of the terahertz setup. In short, since the substrate is the same for all samples, the modulus of the relative THz transmission |T| is proportional to the resistivity. In fact, from the Tinkham formula, it follows that ρ = [(n_r + 1)/(Z_0)]d(1/|T| − 1) ~ 1/|T|, where n_r is the refractive index of the substrate, Z_0 = 377 Ω is the vacuum impedance, and d is the film thickness. Hence, ρ ~ |T|. Fig. 3 shows both the sheet resistance and the THz transmission normalized to the value measured for the pristine samples. We notice that both measurements return values that remain more or less constant and similar to that of the pristine samples for all samples annealed up to 450 °C.
°C, and then suddenly drop to less than half for the samples with the two highest annealing temperatures. In the insets, we plot the magnetic force microscopy (MFM) images for a few selected annealing temperatures, the full data is available in the Supplementary Material. They show that for an annealing temperature of up to 500 °C, the magnetic contrast is rather smooth, with the tip phase shift being less than a degree. The MFM contrast changes abruptly for the 550 °C sample, there is a small remanence which is preserved for all samples by carefully rotating them in the sample plane and overlaying it with the respective demagnetization data, the details on the terahertz time-resolved magneto-optical Kerr effect pump-probe setup, the vibrating sample magnetometer characterization of all samples, and which were then annealed at different temperatures to induce an increasing degree of crystallization. We performed terahertz-induced ultrafast demagnetization and terahertz conductivity experiments to quantify the effect of annealing on both charge and spin scattering. We observed a decrease of the terahertz resistivity as soon as the annealing temperature induced a structural phase transition. The demagnetization experiments revealed a novel enhanced spin-flip process in an intermediate magnetic state characterized by the same electrical conductivity as the pristine samples, but with a much larger in-plane anisotropy. Such anisotropy must be due to an enhanced spin-orbit coupling in the sample plane caused by the annealing process, and is the microscopic mechanism that increases the spin-flip probability at crystalline defects. Our observations, supported by independent experimental data, further deepen our understanding of the fundamentals of spin and charge transport at ultrafast time scales, and their role in ultrafast magnetism of metals.

In conclusion, we performed a systematic study on a series of CoFeB thin film samples which were grown under the same conditions to achieve an amorphous state, and which were then annealed at different temperatures to induce an increasing degree of crystallization. We performed terahertz-induced ultrafast demagnetization and terahertz conductivity experiments to quantify the effect of annealing on both charge and spin scattering. We observed a decrease of the terahertz resistivity as soon as the annealing temperature induced a structural phase transition. The demagnetization experiments revealed a novel enhanced spin-flip process in an intermediate magnetic state characterized by the same electrical conductivity as the pristine samples, but with a much larger in-plane anisotropy. Such anisotropy must be due to an enhanced spin-orbit coupling in the sample plane caused by the annealing process, and is the microscopic mechanism that increases the spin-flip probability at crystalline defects. Our observations, supported by independent experimental data, further deepen our understanding of the fundamentals of spin and charge transport at ultrafast time scales, and their role in ultrafast magnetism of metals.

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FIG. 4. Maximum demagnetization (blue) and relative remanence (orange) for samples annealed at different temperatures. The error bars in the demagnetization are calculated from the function used to fit the delay traces in Fig. 2. Insets: hysteresis loops from the VSM measurements taken along the in-plane hard magnetization axis for the samples for a few selected annealing temperatures.
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DATA AVAILABILITY

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

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