Ultrafast demagnetization of $Co_2MnSi_{1-x}Al_x$ Heusler compounds using terahertz and infrared light

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(Received 5 September 2022; revised 26 May 2023; accepted 30 May 2023; published 9 June 2023)

We investigate the ultrafast demagnetization in $Co_2MnSi_{1-x}Al_x$ quaternary Heusler compounds induced by terahertz (THz) and infrared (IR) pulses. Adjusting the alloy's composition allows us to tailor the spin polarization at the Fermi energy from 97% ± 3% (Co_2MnSi) due to its minority spin gap around 0.6 eV, down to 63% ± 3% (Co_2MnAl) without a spin gap. Here we experimentally compare the cases of ultrafast demagnetization, when the material is excited with the help of laser pulses with photon energies below and above the minority spin gap, respectively. More particularly, the pump-photon energies were tuned from 1.02 eV (IR) down to 4.1 meV (THz). We found that the ultrafast demagnetization time decreases upon substitution of Si by Al and thus destroys the minority spin gap. Moreover, a decrease of the pump-photon energies in the near-infrared spectral range results in a slight increase of the demagnetization time. Nevertheless, further decrease of the photon energy by a factor of 250 hardly changes the characteristic time of ultrafast demagnetization. Both THz and IR pulses cause very similar ultrafast magnetization dynamics in our $Co_2MnSi_{1-x}Al_x$ Heusler compounds.

DOI: 10.1103/PhysRevB.107.224408

I. INTRODUCTION

The seminal discovery of ultrafast laser induced demagnetization of ferromagnetic Ni [1], published nearly 20 years ago, launched the field of ultrafast magnetism. The field aims to understand magnetization dynamics triggered in magnets by ultrashort laser excitation and has become an increasingly interesting subject of experimental [1–5] and theoretical research [6–8]. Despite nearly two decades of studies, understating the actual mechanisms that define the timescale of ultrafast magnetization remains an intricate problem due to the coexistence of different physical mechanisms that could explain the effect. For instance, many models have been proposed considering either a local dissipation of angular momentum which involves different interactions between electrons and other quasiparticles [9–11] or a nonlocal dissipation due to spin transport [12–14]. Usually, two different limits are considered: A Stoner-like description for itinerant electrons, which assumes that the atomic magnetic moment is quenched due to a reduction of the exchange splitting induced by single-particle spin scattering and a localized Heisenberg picture, in which transversal collective excitations, i.e., magnons, lead to a reduction of the net magnetization without altering the atomic moment. Furthermore, using a basic approach based on Fermi's "golden rule," it was proposed that the demagnetization process is directly related to the band structure at equilibrium, especially to the population of minority and majority states at the Fermi level, leading to a dependence of the demagnetization time τ_M on the spin polarization P at the Fermi level with $\tau_M \sim (1–P)^{-1}$ [15,16].

It is nowadays well established that the demagnetization in 3*d* ferromagnets takes place on the timescale of hundreds of femtoseconds (fs). It was recently demonstrated using time-, spin-, and angle-resolved photoemission, that the ultrafast demagnetization of Co was mainly due to collective excitations of magnons [17]. On the other hand, very long demagnetization times, ranging from a few hundred picoseconds to more than 1 ns have been reported for CrO_2, Fe_3O_4, and $La_{0.66}Sr_{0.33}MnO_3$ (LSMO) and were explained to be due to high spin polarizations (larger than 80%) and the existence of a spin band gap in the minority band at the Fermi energy [16]. In an attempt to correlate the effect of the band structure on the spin polarization, the intrinsic Gilbert damping, and the ultrafast demagnetization, we recently studied a series of engineered $Co_2MnSi_{1-x}Al_x$ Heusler compounds [18]. In that paper we controlled the composition of these compounds and thus changed the electronic density at the Fermi energy. In the case of Co_2MnSi ($x = 0$), we reported the existence of a spin band gap in the minority band at the Fermi level [19,20]. Based on photoemission and transport measurements, we can infer that the minority spin band gap is around 0.6 eV, in good agreement with theoretical predictions [21]. By measuring the ultrafast demagnetization induced by laser excitation of these compounds, we verified the $\tau_M \sim (1–P)^{-1}$ law. However, while the longer demagnetization time was obtained for Co_2MnSi with 97% ± 3% spin polarization, it remains

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around 350–400 fs, which is three orders of magnitude smaller than the one observed for magnetic oxides. A similar ultrafast demagnetization process was reported in a series of $Co_2Fe_xMn_{1-x}Si$ alloys by Pan *et al.* [22]. One may argue that in order to achieve a dramatic delay of the ultrafast demagnetization in Heusler alloys as predicted by the rule $\tau_M \sim (1–P)^{-1}$ [15,16] one has to choose pump-photon energy below the minority spin gap. The goal of this paper is to verify this hypothesis.

Note that the previous experiments on ultrafast demagnetization in Heusler compounds were performed using pump-photon energy of 1.5 eV, which is far above the spin band gap. It means that majority electrons from the top valence band will be excited above the spin band gap and therefore will be scattered into the minority conduction band. In this study, we investigate the role of the spin band gap in Heusler compounds in the ultrafast magnetization dynamics using THz photons with energies far less than the spin band gap. To this end, we studied the influence of the photon energy from 1.02 eV down to 4.1 meV. We report a noticeable increase of the demagnetization time when the photon energy becomes smaller than the spin band gap. However, it remains around hundreds of femtoseconds, which is still three orders of magnitude shorter than the time reported for highly spin-polarized materials by the rule $\tau_M \sim (1–P)^{-1}$. Hence, we must conclude that the suggested correlation between spin polarization at the Fermi level and the demagnetization time is not universal.

II. EXPERIMENTAL RESULTS

The Heusler films were epitaxially grown on single-crystalline MgO(001) substrates using molecular beam epitaxy [19]. The different elements (Co, Mn, Si, Al) were codeposited on MgO maintained at a temperature around 450 °C (with a thermocouple located behind the sample holder) and the films were annealed at around 750 °C after the growth, with an accuracy on each flux equal to 1%. The present study was done on a series of 20 nm thick $Co_2MnSi_{1-x}Al_x$ films capped with gold. These films were already used in a previous paper [18] where we reported ultralow magnetic damping coefficients (4×10^{-4} in Co_2MnSi to 1.5×10^{-3} in Co_2MnAl). The variation of the damping was clearly correlated to the spin polarization P (from 97% ± 3% in Co_2MnSi to 63% ± 3% in Co_2MnAl) and the demagnetization time using pump photons with the energy of 1.5 eV. It should be noted that such an ultralow magnetic damping is in agreement with the existence of a spin gap (at least in Co_2MnSi). Indeed, we have shown that even a small deviation on the stoichiometry may disturb the damping constant [19].

Here, we go further by analyzing the demagnetization of these samples using a variable photon energy pump. We first performed time-resolved magneto-optical (TRMOKE) measurements using a standard Ti:sapphire fs laser and a regenerative amplifier system with a wavelength of 800 nm and a repetition rate of 5 kHz. It was combined with a commercial optical parametric amplifier system in order to produce light pulses at the desired wavelength. The probe photon's energy was kept constant at 3.1 eV while the pump photon's energy was varied between 1.02 and 0.55 eV. Autocorrelation characterization was used to monitor the pulse duration for pump and probe pulses when the pump wavelength was changed. In this work, the pulse duration was kept around 80 fs for the pump and the probe. For the penetration depths δ_p, it is comparable ($\delta_p \sim$ 20 nm) for the pump laser pulses with different wavelength, according to the estimation by $\delta_p = \frac{\lambda}{4\pi\kappa}$, where λ is the wavelength and κ is the optical extinction coefficient [23].

Firstly, a reference sample of transition metal Co without any spin band gap was measured carefully via TRMOKE as shown in Fig. 1(a). The magnetization dynamics of Co following laser excitation shows no dependence on the laser pulse energy within the studied range. The magnetization dynamics in $Co_2MnSi_{1-x}Al_x$ compounds were then measured using the same laser conditions. In this study, we kept the amplitude of demagnetization close to 10% for all photon energy excitation by adjusting the laser power. For instance, Fig. 1(b) shows the IR femtosecond laser pulse induced magnetization dynamics in Co_2MnSi as a function of wavelength with a fixed laser fluence of $F = 2.5\,\mathrm{mJ/cm^2}$. Note that this Co_2MnSi epitaxial thin film has a full spin polarization and ultralow magnetic damping. Contrary to the magnetization dynamics obtained from the Co reference sample, a small increase of the maximum demagnetization position is observed when decreasing the photon energy, indicating the presence of a spin blocking effect in Co_2MnSi when the photon energy becomes comparable with the size of the spin band gap. In order to extract the exact ultrafast demagnetization time, the temporal evolution of magnetization was fitted using the following analytic solution based on the three-temperature model (3TM) [24]:

$$-\Delta M = \left\{\left[\frac{A_1}{(t/\tau_0+1)^{0.5}} - \frac{(A_2\tau_E - A_1\tau_M)}{\tau_E-\tau_M}e^{-\frac{t}{\tau_M}} - \frac{\tau_E(A_1-A_2)}{\tau_E-\tau_M}e^{-\frac{t}{\tau_M}}\right]\Theta(t)\right\} * G(t), \quad (1)$$

where $*G(t)$ presents the convolution product with the Gaussian laser pulse profile, $\Theta(t)$ is a step function, and A_1, A_2 are fitting constants. The two critical time parameters τ_M, τ_E are the ultrafast demagnetization time and magnetization recovery time, respectively. The pulse duration used in this study was around 80 fs for all the wavelengths in the near-infrared spectral range, which is the value of τ_G applied in the fitting equation. For τ_E representing the electron-phonon coupling, we assign $\tau_E = 550$ fs according to previous studies [18,24]. τ_0 is the parameter describing the cooling process by heat diffusion, which has to be about one order of magnitude larger than τ_E and τ_M. The value of $\tau_0 = 5$ ps is fixed during the fitting procedure, which is consistent with our previous work where the studied material has the same substrate MgO [25]. τ_M is the parameter we need to compare with various photon energy. We

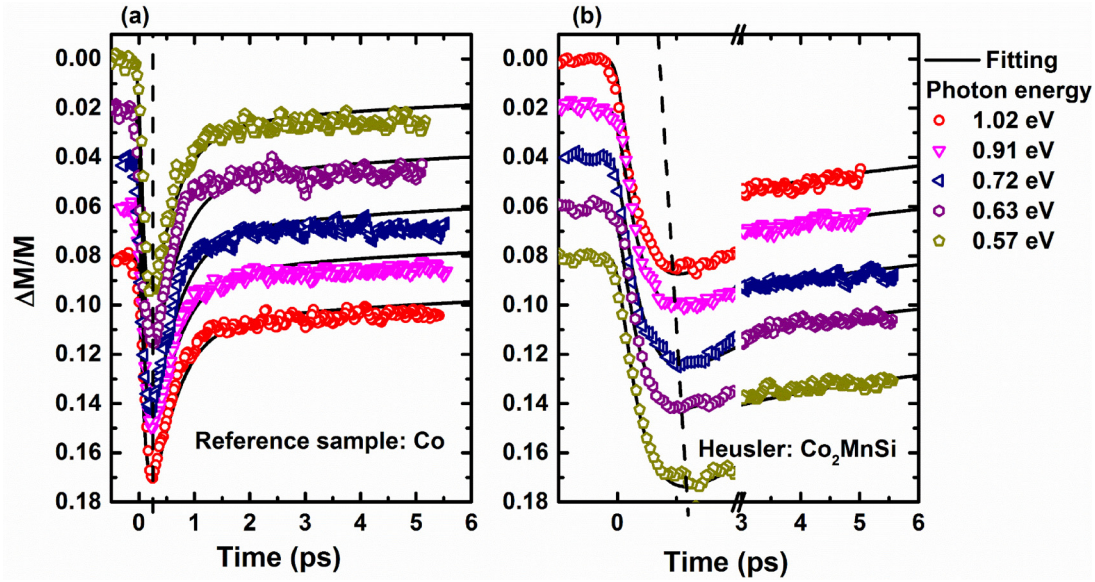

FIG. 1. Dynamics of the magneto-optical Kerr effect induced by the IR pump via TRMOKE for (a) reference sample of Co and (b) Heusler material Co_2MnSi.

manually changed the value of τ_M to fit the experimental demagnetization curves for each wavelength. The errors of τ_M are given by the judgement of the deviation between the fitting curve and the experimental one. According to our previous paper [18,25], an obvious deviation, whether from the demagnetization part or magnetization recovery process, appears if the value of τ_M is further varied up to 30 fs from the best fitted value, so we gave the errors as ± 15 fs which is the maximum variation of demagnetization time to make the fitting and experimental curves almost identical. Meanwhile, in order to obtain a high time resolution, we measured the ultrafast demagnetization with a very fine step of time delay (10 fs). The fitted curves are also shown in Fig. 1 from which the ultrafast demagnetization time τ_M was evaluated.

As illustrated in Fig. 2, τ_M remains constant for the reference sample of Co as a function of the laser wavelength.

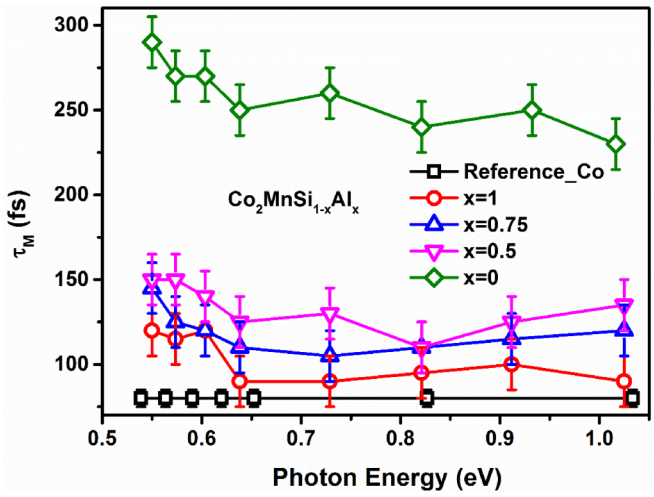

FIG. 2. The extracted ultrafast demagnetization time τ_M as a function of photon energy in the range of IR for Co and Heusler compounds with various concentrations of Si.

Similar results were obtained for Ni [26], where the excitation photon energy was chosen as 1.55 and 3.1 eV. Recently, similar results were found in Ni using a larger range of photon energy [27]. Moreover, an almost identical demagnetization time was reported very recently in Fe [28], by using optical (3.1 eV) and THz pulses (4.1 meV). Regarding the present measurements on the $Co_2MnSi_{1-x}Al_x$ thin films series, three points have to be noted:

(i) The ultrafast demagnetization time τ_M is longer for all the Heusler samples than for Co.

(ii) Focusing on the variation of τ_M with x for a fixed wavelength, the ultrafast demagnetization slows down with decreasing x, i.e., when decreasing the substitution of Si by Al. This was already observed in our previous study for $E = 1.5$ eV/ $\lambda = 800$ nm [18] and was the consequence of the band gap vanishing with increasing x. This variation is still observed here for all the laser wavelengths we used in this study.

(iii) Focusing on the dependence of τ_M variation with the wavelength for a fixed composition x, we found that τ_M shows a weak dependence on the laser photon energy in the Heusler samples. For instance, in the Co_2MnSi sample, the demagnetization time increases from 230 ± 10 fs to 290 ± 10 fs when the photon energy is reduced from 1.02 to 0.55 eV.

To go further, THz measurements were performed using short terahertz pulses with 4.1 meV photon energy. We studied the spin dynamics triggered by nearly single-cycle short terahertz pulses (photon energy 4.1 meV) with a duration of $\sim$1 ps. The THz electric field peak is up to 800 kV/cm with a spectrum ranging from 0 to 3 THz, as shown in Fig. 3(a). In order to trace THz-induced magnetic changes, we detected ellipticity acquired otherwise by a linearly polarized probe pulse at the wavelength of 800 nm upon transmission through the sample. Both pump and probe pulses were at normal incidence. And the ellipticity acquired by the probe pulse is due to the Faraday effect and thus a measure of the out of plane magnetization component. An external magnetic field

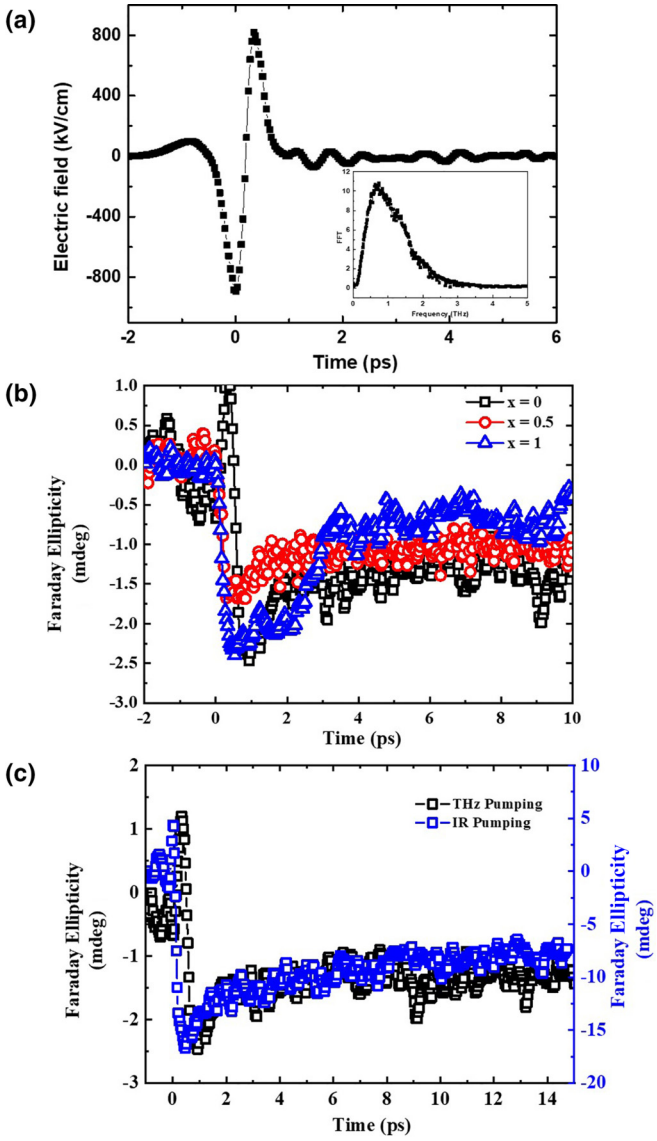

FIG. 3. Experimental results obtained by ultrafast terahertz (THz) pump-infrared (IR) probe. (a) The incident THz electric field in a time domain and its Fourier transform (inset). (b) Terahertz-induced magnetization dynamics in $Co_2MnSi_{1-x}Al_x$ with $x = 0, 0.5, 1$. (c) Comparison of ultrafast demagnetization in Co_2MnSi between THz pump and IR pump.

$H \approx 1.5$ kOe along the sample normal was applied to force the magnetization to get aligned out of plane. Detailed descriptions of the THz setup have been reported elsewhere [29,30].

Figure 3(b) shows the dynamics of the magneto-optical Faraday effect excited by the THz electric field for $Co_2MnSi_{1-x}Al_x$ ($x = 1$, 0.5, 0). It is clearly seen that the magneto-optical signal induced by THz excitation rapidly decreases for $Co_2MnSi_{1-x}Al_x$ with $x = 1$, 0.5 and 0, within less than 1 ps. A pronounced recovery of magnetization is followed closely. Regarding the oscillation observed for $x = 0$ at around zero time delay, it might come from the THz modification of magnetic birefringence. Indeed, Barsaume *et al.* reported that the magneto-optical signal can be substantially changed under the action of the THz pulse [31]. Moreover, we found that the ultrafast magnetization dynamics obtained by THz and IR pumping is quite similar in Co_2MnSi [Fig. 3(c)], indicating that electronic excitations in the THz and IR spectral ranges have similar effects on the demagnetization rate. Our results reveal that both intra- and interband electronic excitations lead to ultrafast demagnetization of the Heusler compounds via similar mechanisms evolving at a subpicosecond timescale.

III. SUMMARY AND CONCLUSIONS

Summarizing the results, we must conclude that the demagnetization time only slightly depends on the pump-photon energy and a change of the latter by a factor of 200 did not result in any substantial changes of the demagnetization time. This finding has far reaching consequences for understanding the physics of ultrafast demagnetization of half-metals. Earlier suggested correlation between spin polarization at the Fermi level and the demagnetization time is obviously not a universal rule. We found the demagnetization is always very fast at the order of hundreds of femtoseconds. In fact, this ultrafast demagnetization that is nearly independent of pump-photon energy suggests that the conventional three-temperature model (3TM) with good precision can still be valid for half-metals even if the pump-photon energy is below the spin band gap. In particular, in the process of ultrafast demagnetization of half-metals one can assume that light interacts exclusively with free electrons. It means that photon energy is spent to increase either potential or kinetic energy of electrons or both. In magnetic metals, light excites intraband electronic transitions, suggesting that light increases only the kinetic energy of electrons. Due to electron-electron interaction (the characteristic time of it in metals is 1–10 fs), the excited electrons exchange the energy with other electrons, electron distribution reaches a statistical Fermi-Dirac distribution, and thus all electrons can be further described by a single parameter—thermodynamic temperature. The time required to reach the statistical distribution is called electron thermalization time and in metals it is about 100 fs. In half-metals, however, the electronic structure is different and different spin subbands are expected to be excited differently. For one of the spin subbands, the excitation will proceed as in metals, while in another one, a part of the photon energy will be spent to increase the potential energy of the electrons. If the photon energy is too small to overcome the band gap in the second subband, as in the case of the THz pump of the samples studied in this work, it is expected that only the electrons in one of the subbands will be excited.

In principle, the hot electrons will exchange heat with a colder lattice on a time-scale of electron-lattice interaction ($\sim$2 ps) in metals. If the electrons are hot, an additional channel of heat exchange between spins and electrons opens up. Hot electrons will result in a temperature increase of spins and thus will cause ultrafast demagnetization also on the scale of 1–2 ps. As the spin-flip relaxation channels can be tuned in half-metals, in theory [15], the demagnetization should take a much longer time, whether because the degree of spin polarization is high or only one of the spin subbands is excited. Experimentally, we observed that even in the case of THz excitation the demagnetization of Heusler alloys is

similarly fast with metals. In fact, we have no reason to ignore the 3TM model. The demagnetization in the THz spectral range is almost as fast as demagnetization in the visible spectral range. Such an insensitivity of the demagnetization time to the pump wavelength is consistent with the 3TM model which ignores the electronic structure of magnets. The model assumes that ultrashort pump pulses interact exclusively with free electrons, increase their effective temperature, and, subsequently launching heat exchange between electrons, lattice, and spins, eventually result in a spin temperature increase and the demagnetizations. The demagnetization time in this model depends on the characteristic times of electron-spin and spin-lattice interactions in the magnet and thus does not depend on the pump wavelength [3].

For other Heusler materials such as NiMnSn ($T_C = 319$ K), a much longer demagnetization time was observed by Bonda *et al.* [32] and it is attributed to the low Curie temperature leading to the critical slowdown effect. When the Curie temperature is too close to room temperature, the magnetic heat capacity would diverge. As a result, it becomes harder to heat the magnetic system and leads to a slowdown of the demagnetization. Obviously, this mechanism underlying the slow demagnetization is not relevant to the electronic band structure, either, and such kind of slowdown effect is avoided in the case of our $Co_2MnSi_{1-x}Al_x$ samples, whose Curie temperature is much higher ($T_C \sim 700-985$ K).

To summarize, we studied the influence of the photon energy on the ultrafast demagnetization dynamics of Heusler $Co_2MnSi_{1-x}Al_x$ compounds for a broad range of photon energies from meV (THz) to 1 eV (IR). We found that the ultrafast demagnetization only weakly depends on the photon energy. A very small but noticeable increase of this demagnetization time for photon energies lower than the spin gap is, however, observed in the fully polarized Co_2MnSi film. This effect becomes less pronounced when substituting Si by Al, that is, when the spin gap is destroyed. However, these demagnetization times remain very small, much smaller than what was reported and thus expected for the case of fully spin-polarized oxides. This therefore begs the question of the microscopic mechanism responsible for the slow demagnetization observed in fully spin-polarized oxides. In half-metal magnetic materials like the Heusler compounds we studied, the presence of a spin gap for one spin channel is not sufficient to block the spin scattering even for low photon energy excitations, suggesting that both intra- and interband electronic excitations contribute to the magnetization dynamics of Heuslers via similar mechanisms taking place at the subpicosecond timescale.

ACKNOWLEDGMENTS

W.Z. gratefully acknowledges the National Natural Science Foundation of China (Grant No. 12104030), the China Postdoctoral Science Foundation (Grant No. 2022M710320), and the China Scholarship Council. This work is supported by No. ANR-20-CE09-0013 UFO, by the Institute Carnot ICEEL for the project "CAPMAT" and FASTNESS, by the Région Grand Est, by the Metropole Grand Nancy, for the Chaire PLUS by the impact Project No. LUE- N4S, part of the French PIA project "Lorraine Université d'Excellence" Reference No. ANR-15-IDEX-04-LUE, and by the "FEDERFSE Lorraine et Massif Vosges 2014–2020," a European Union Program by the European Union's Horizon 2020 research and innovation program COMRAD under the Marie Sklodowska-Curie Grant Agreement No. 861300, and by the Academy of Finland (Grant No. 316857), de Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO) and the European Research Council ERC Grant Agreement No. 101054664 273 (SPARTACUS), as well as COST Action No. CA17123 MAGNETOFON, supported by COST (European Cooperation in Science and Technology). The authors would like to thank Michel Hehn for providing the Co sample, K. A. Grishunin for helping to prepare the THz pump-probe setup, and S. V. Semin and C. Berkhout for technical support.

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