## The role of the magnetic sublattices on the picosecond spin current generation in ferrimagnetic GdCo

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Ultrafast spintronics aims to exploit the electronic spin to develop unprecedently fast and energyefficient data storage and processing technologies. Rare earth-transition metal (RE-TM) ferrimagnets have emerged as one of the most promising types of systems in this research field, since their magnetization can be directly switched with light [1]. Furthermore, these materials are versatile sources of picosecond spin current when they are excited with ultrashort laser pulses. Such spin current has been employed to reverse the magnetization of an adjacent ferromagnet in spin valve structures [2]. In devices containing ferrimagnetic GdFeCo, it has been claimed that the spin current produced by the Gd sublattice plays a crucial role in the switching of the nearby ferromagnet [3]. On the other hand, other studies have argued that Gd cannot produce a spin current at THz frequencies [4].

In this work, we examine the strength and time scales of the RE and TM contributions to the picosecond spin current generated by GdCo via THz emission spectroscopy. We systematically investigate the temperature and alloy composition dependences of the THz signal, which is proportional to the generated spin current in the frequency domain. As shown in Figs. 1(a) and 1(c), we find that the THz emission vanishes at the magnetization compensation point, when an external magnetic field is applied. However, in case there is no external field, the spin current remains across compensation (see Figs. (b) and (b)).

Previously, this vanishing of the THz emission from RE-TM ferrimagnets close to compensation has been attributed to the cancellation of the spin current produced by the two magnetic sublattices [5]. Our experiments indicate that this explanation does not hold for GdCo since the THz emission persist under zero applied magnetic field. Instead, through magneto-optic Kerr effect imaging and the analysis of the external field dependence of the THz signal, we demonstrate that the suppression of the spin current stems from the formation of domains in the GdCo static magnetic configuration.

Subsequently, we investigate why the Gd and Co spin currents do not cancel out. By considering the changes of the THz emission as the Gd atomic concertation (*x*) is varied (see Fig. 2), we determine that, at room temperature, the picosecond spin current is dominated by hot spin-polarized electrons excited from the Co sublattice. However, at low temperature, we detect a substantial contribution to the THz spin current from Gd, with a spectrum containing, on average, lower frequency components. We ascribe this activation of the Gd spin current to the increase of the spin polarization of the Gd 5*d* bands with lowering temperature [6]. Therefore, we show that the Gd and Co spin currents cannot fully offset or compensate each other due to their different time scales.

Lastly, we investigate how the spin current spectrum changes across magnetization compensation. Our experiments reveal a blueshift of the THz emission associated with the multi-domain structure formed close to compensation. This effect, together with previously discussed vanishing of the THz signal, underscores the importance of the equilibrium micromagnetic state in the laser-induced spin current generation.

Our results showcase the tunability of RE-TM ferrimagnets as sources of THz spin current and are relevant for the design of optically controllable spin valve devices.

## References

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## **Figures**



**Figure 1.** THz emission intensity and GdCo static magnetization as a function of temperature. The THz intensity is defined as the integral of the absolute value of the THz pulses. (a) and (b) THz emission intensity measured under in-plane applied magnetic fields of 8.5 kOe and 0 Oe, respectively. The dashed line in (a) indicates the temperature at which the THz emission is the weakest, denoted *T*<sub>THz</sub>. (c) and (d) In-plane magnetization measured by superconducting quantum interference device (SQUID) magnetometry under magnetic fields of 8.5 kOe and 0 Oe, respectively. The dashed lines in (c) and (d) mark the magnetization compensation temperature *TM.*



**Figure 2.** THz emission for different Gd concentrations. (a) and (b) THz signal at 300 K and 6 K, respectively. The multiplicative factors on the left indicate by how much the pulses were rescaled to have the same amplitude as the Co emission  $(x = 0\%)$  at the corresponding temperature. In (b), the black dashed lines are linear combinations of the Gd and Co signals. The purple (green) dashed lines are the corresponding Co (Gd) components in these linear combinations.