

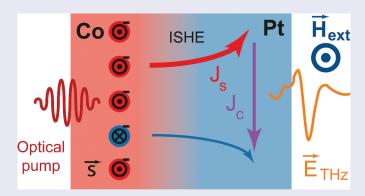


# Enhanced laser-induced single-cycle terahertz generation in a spintronic emitter with a gradient interface

Leonid A. Shelukhin [p], Anna V. Kuzikova [p], Andrey V. Telegin [p], Vladimir D. Bessonov [p], Alexey V. Ognev och, Alexander S. Samardak och, Junho Park och, Young Keun Kim och and Alexandra M. Kalashnikova (Da

### **ABSTRACT**

The development of spintronic emitters of broadband terahertz (THz) pulses relies on designing heterostructures in which the processes of laser-driven spin current generation and subsequent spin-to-charge current conversion are the most efficient. The interface between the ferromagnetic and nonmagnetic layers in an emitter is a critical element. In this study, we experimentally examined single-cycle THz pulse generation from a laser-pulse-excited Pt/Co emitter with a 1.2-nm-thick composition-gradient interface between the Pt and Co and compared it with the emission from a conventional Pt/Co structure with an abrupt interface. We found that the gradient interface improved the efficiency of the optics-to-THz conversion by a factor of two in a wide range of optical fluences up to 3 mJ·cm<sup>-2</sup>. This enhancement was caused by a pronounced increase in the transmittance of the laser-driven spin-polarized current through the gradient interface compared with the abrupt interface. Moreover, it was evident that such transmission deteriorated with the laser fluence owing to the spin accumulation effect.



### **IMPACT STATEMENT**

Pt/Co spintronic emitter with a crystalline composition gradient interface demonstrates twofold increase of the optical-to-THz fluence conversion in comparison with a conventional Pt/Co structure with an abrupt interface. This is caused by an enhanced transmittivity of the laserdriven spin-polarized current through the gradient interface.

# Introduction

The demand for broadband THz emitters for various applications [1] has led to considerable progress in the field, with emitters based on various materials and operating under different principles [2-11]. Multilayer spintronic structures that

convert laser-driven spin dynamics into picosecond charge currents [12] make promising single-cycle THz sources [5,13-19]. The foremost spintronic emitters based on nonmagnetic metal/ferromagnetic metal (NM/FM) heterostructures rely on spin/charge conversion by the inverse spin Hall

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aloffe Institute, St. Petersburg, Russia;

bM. N. Mikheev Institute of Metal Physics, Ural Branch of Russian Academy of Science, Yekaterinburg, Russia;

Institute of High Technologies and Advanced Materials, Far Eastern Federal University, Vladivostok, Russia;

dSakhalin State University, Yuzhno-Sakhalinsk, Russia;

<sup>&</sup>lt;sup>e</sup>Department of Materials Science and Engineering, Korea University, Seoul, Republic of Korea

effect (ISHE) occurring in the bulk of the NM layer, which possesses strong spin-orbit coupling [20,21]. Therefore, optimizing the thicknesses and materials of the NM and FM layers is vital to obtaining a major increase in THz emission in spintronic emitters [5]. Interfaces in such structures have been found to be an alternative source of THz emission, enabled by symmetry breaking [22,23], the Rashba – Edelstein effect [24], and skew scattering by impurities [25].

Nevertheless, interfaces in NM/FM heterostructures can have a strong impact - even on the 'bulk' spin-to-charge conversion - because of their influence on the spin and spin-mixing conductance [26-29] and spin memory loss [30-33]. The manipulation of THz emission by adding nonmagnetic interfacial layers [26] or intermixing [25,34,35] at the NM/FM interface of a spintronic emitter has also been reported. It has been established that the THz emission in these modified structures correlates with changes in interfacerelated spin phenomena like spin pumping [26]. Consequently, examining the THz emission properties of structures with designs that enable the enhancement of particular interface-related spintronic phenomena could be a promising research direction.

In this study, we explored an approach to boost the THz generation of a spintronic emitter through an advanced Pt/Co interface design. The interfacial layer between the Co and Pt - which comprises a Pt/Co content gradient – makes the interface less abrupt than that of a conventional Pt/Co emitter. Interestingly, such structures with a gradient interface demonstrate an approximately two-fold enhancement of the interfacial spin phenomenon — the Dzyaloshinskii - Moriya interaction (DMI) [36]. We demonstrated that such an interface led to a nearly two-fold increase in the optical-toterahertz fluence conversion efficiency compared to a Pt/Co emitter with the same layer thickness and sharp interface. By excluding the effect of the thickness change and quantifying the spin current generated in the Co layer by ultrafast demagnetization measurements, we concluded that the main role in enhancing THz generation was the increased spin transmittance of the gradient interface to a spin current.

### **Experimental methods**

# Sample preparation and characterization

Two primary samples of spintronic THz emitters were examined; the conventional Pt(3)/Co(1.2)structure with an abrupt interface (numbers in brackets indicate the thicknesses in nm), and the Pt(3)/g-PtCo(1.2)/Co(1.2) structure with a 1.2-nm-thick composition-gradient interface (g-PtCo) between the NM and FM layers having a nominal composition  $Co_{25}Pt_{75}(0.4)/Co_{50}Pt_{50}(0.4)/Co_{75}Pt_{25}(0.4)$ . Three different alloys (Co<sub>25</sub>Pt<sub>75</sub>, Co<sub>50</sub>Pt<sub>50</sub> and Co<sub>75</sub>Pt<sub>25</sub>) were utilized for the deposition of the g-PtCo. The Pt and Co layer thicknesses (Table 1) were chosen to be close to the respective optima for THz emission [15,37]. Single films of the Co(4.6) and Co<sub>75</sub>Pt<sub>25</sub>(4.6) alloys and spintronic emitters Co<sub>75</sub>Pt<sub>25</sub>(4.2)/Co(0.8), Co<sub>50</sub>Pt<sub>50</sub>(4.2)/Co (0.8), and Pt(2)/Co<sub>75</sub>Pt<sub>25</sub>(4.2) were used as references. All samples were fabricated on Si substrates with about 300-nm-thick thermal oxides using UHV DC magnetron sputtering at a base pressure of  $5 \times 10^{-9}$  Torr at room temperature and then post-annealed at 300°C for 1 h in a vacuum of  $1 \times 10^{-9}$  Torr [36]. The bottom 2-nm and top 2-nm Ta layers were deposited as buffer and capping layers, respectively. The Pt/g-PtCo/Co sample structure is schematically shown in Figure 1 (a).

Transmission electron microscopy (TEM) and X-ray diffraction studies showed that the samples possessed an *fcc* (111) structure [36]. TEM study also revealed that the interfacial layer g-PtCo was characterized by a gradual change in the Co-Pt content ratio rather than a step one [36].

Information on the magnetization and magnetic anisotropy of the samples is summarized in Table 1, as obtained from vibrating-sample magnetometry measurements. As expected, changing the composition of the FM layers, adding a Pt layer, and modifying the interface resulted in variations in the strength and sign of the magnetic anisotropy [38,39]. Notably, the addition of a gradient interface to the Pt/Co led to a change in the magnetic anisotropy from out-of-plane to in-plane, which is optimal for spintronic emitters. The magnetization hysteresis loops obtained in the in-plane magnetic field for Pt/Co and Pt/g-PtCo/Co samples are shown in Figure 2 (a). Table 1 lists the DMI parameters of the samples reported in Ref [36].

**Table 1.** Main sample composition, magnetization  $M_s$ , effective uniaxial anisotropy constant  $K_{\rm eff}$ , magnetic dead layer  $d_0$ , and DMI parameter [36]. The numbers in brackets are the layer thicknesses in nm. Negative  $K_{\rm eff}$  corresponds to perpendicular magnetic anisotropy.

unisotropy.				
Sample	$M_S$	$K_{ m eff}$	$d_0$	DMI
	10 <sup>5</sup> A m <sup>-1</sup>	$10^4  \mathrm{J \ m}^{-3}$	nm	${\rm mJ~m}^{-2}$
Pt(3)/g-PtCo <sup>a</sup> (1.2)/Co(1.2)	10.6	10.6	0.9	-0.82
Pt(3)/Co(1.2)	10.5	-31.5	0.04	-0.44
Co(4.6)	13.6	102	-	-
Co <sub>75</sub> Pt <sub>25</sub> (4.6)	9.8	-14.7	-	0.4

 $<sup>^{</sup>a}Co_{25}Pt_{75}(0.4)/Co_{50}Pt_{50}(0.4)/Co_{75}Pt_{25}(0.4).$ 

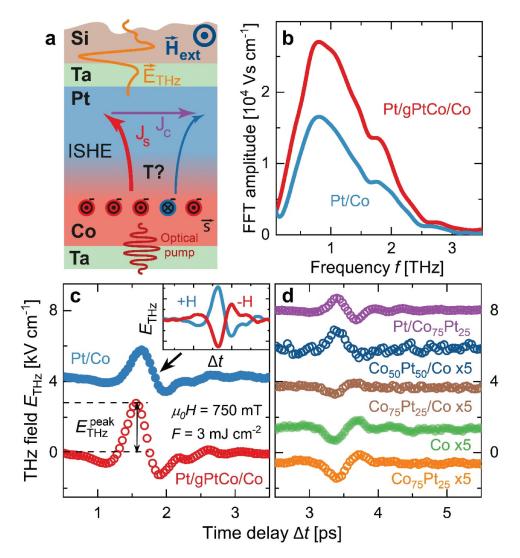


Figure 1. (a) Schematic of the generation of THz emission via the inverse spin Hall effect in a laser-excited Pt/g-PtCo/Co structure with a composition-gradient interface. T denotes the spin transmittance of the interface between the Pt and Co layers. (b) Fourier spectra of the THz pulses generated in Pt/g-PtCo/Co (red line) and Pt/Co (blue line). Electric field temporal profile of the emitted THz pulse (c) for Pt/Co and Pt/g-PtCo/Co, and (d) Pt/Co<sub>75</sub>Pt<sub>25</sub>, Co<sub>50</sub>Pt<sub>50</sub>/Co, Co<sub>75</sub>Pt<sub>25</sub>/Co, Co, and Co<sub>75</sub>Pt<sub>25</sub>. Inset in (c) shows the THz pulse polarity inversion with the change in the external magnetic field sign.

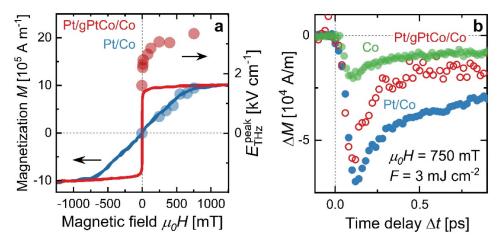


Figure 2. (a) In-plane magnetization (lines) and  $E_{THz}^{peak}$  (symbols) obtained at F=3 mJ·cm<sup>-2</sup> as functions of the external magnetic field applied in the sample plane for Pt/g-PtCo/Co (red) and Pt/Co (blue). (b) Absolute demagnetization value as a function of the time delay (Δt) measured for the pure Co (green circles), Pt/Co (blue circles), and Pt/g-PtCo/Co (red dots) structures.

#### Thz emission

In the THz emission experiment, laser pump pulses with a central wavelength of 800 nm and a repetition rate of 1 kHz were generated using a Ti:sapphire regenerative amplifier (Avesta, Russia). They were collimated into a 1-mm-diameter spot on the sample surface, providing a fluence ranging from 0.01-3 mJ·cm<sup>-2</sup>. Excitation was performed on the side of the structure, and the THz emission was studied after passing through the substrate [Figure 1 (a)]. The resulting THz pulse waveform was detected by electro-optical sampling [40] using a 1-mm-thick ZnTe crystal with (111) orientation. ZnTe was selected because its high electro-optical coefficient enabled the detection of weak THz fields. The electro-optical coefficient of the ZnTe crystal was verified using control measurements with a 200- $\mu$ m (110)-oriented GaP crystal. To set the magnetization state of the sample, an external magnetic field  $\mu_0 H = 50-750$  mT was applied in the sample plane, with  $\mu_0 H = 750 \text{ mT}$ sufficient for the in-plane saturation of all samples, including those with perpendicular magnetic anisotropy [Figure 2 (a)]. All measurements were performed in a dry nitrogen atmosphere with humidity below 10%.

# **Ultrafast demagnetization**

Ultrafast laser-induced demagnetization was measured using the time-resolved longitudinal magnetooptical Kerr effect scheme [41]. The Yb<sup>3+</sup>:KGd(WO<sub>4</sub>)<sub>2</sub> regenerative amplifier (Light Conversion, Lithuania) was the source of pump and probe pulses with a duration of 170 fs emitted at a repetition rate of 5 kHz. The central wavelength of the pump was converted to 800 nm using an optical parametric amplifier to reproduce laser excitation in the THz emission experiment. The pump pulses were focused normally onto the sample surface in an area of with a diameter of 40  $\mu$ m, and their fluence was varied between 0 and 3 mJ·cm<sup>-2</sup> (to match the conditions of the THz experiments) and up to 12.5 mJ·cm<sup>-2</sup>. The central wavelength of the probe was converted to 515 nm using a beta-barium borate (BBO) crystal, enabling filtering of the pump pulses in front of the detector. The probe pulses had a fluence 50 times less than those of the pump pulses and were focused onto a spot of diameter 30  $\mu$ m at an incidence angle of 45°. An external magnetic field of  $\mu_0 H = 750$  mT was applied in the sample plane. Rotation of the probe polarization plane was measured as a function of the time delay ( $\Delta t$ ) between the pump and probe pulses using a balanced photodetector (Newport Corporation, USA) in combination with a Wollaston prism. The rotation of the probe polarization was proportional to the pumpinduced change in magnetization saturation along the external magnetic field. The data were normalized using static magneto-optical Kerr rotation at saturation to determine the relative demagnetization magnitudes. Absolute values of the magnetization change  $\Delta M$  were obtained from the normalized pump-probe data using static  $M_S$  and assuming that the signal was dominated by Co demagnetization.

### **Results**

Typical THz waveforms from the Pt/Co and Pt/g-PtCo/Co measured in the in-plane external field  $\mu_0 H = 750$  mT under the optical fluence  $F = 3 \text{ mJ} \cdot \text{cm}^{-2}$  are shown in Figure 1 (c). The generated THz waveforms were independent of the laser pulse polarization, the THz emission being linearly polarized orthogonally to the applied field direction. The spectra obtained using fast Fourier transforms of the waveforms were similar for both samples [Figure 1 (b)]. The spectra were limited by the sensitivity of the ZnTe crystal, and the small dip at 1.6 THz could be attributed to the features of this crystal [42]. To quantify the THz signals, we designated the largest deviation of the electric field from zero as the peak THz field,  $E_{THz}^{peak}$ [Figure 1 (a)]. In both the Pt/g-PtCo/Co and Pt/Co structures,  $E_{THz}^{peak}$  reached a maximum value and saturated as the magnetization was saturated in the sample plane, as is evident from the field dependences of M and  $E_{THz}^{peak}$ , as shown in Figure 2 (a). Notably, the temporal profiles of the THz pulses remained unchanged under different experimental conditions. Thus, the discussion holds true for the peak-to-peak values of the THz electric field that are often used in the literature.

These features, along with reversal of the THz pulse polarity with the magnetic field sign change [see the inset in Figure 1 (c)], are characteristic of the emission originating from the ISHE in the Pt layer, as shown schematically in Figure 1 (a), or ultrafast demagnetization of the Co itself [4,43–48]. To verify that the origin of the THz signal in these two samples was spin-charge conversion by the ISHE, we compared the signals to those from the reference Co and Co<sub>75</sub>Pt<sub>25</sub> samples, where ultrafast demagnetization was the dominant THz source [43]. The signals in the latter samples were 10-15 times lower in agreement with previous findings [28] and were of the opposite polarity compared to the signals in the Pt/g-PtCo/Co and Pt/Co [the orange and green curves shown in Figure 1 (d)]. The fact that spin-to-charge conversion was a leading effect, whereas ultrafast demagnetization itself just corrected the THz field amplitude in the structures with a Pt layer, was further evident from the enhancement of  $E_{THz}^{peak}$  and the reversal of the polarity in Pt/Co<sub>75</sub>Pt<sub>25</sub>, i.e. when the Pt layer was added to a thick Co<sub>75</sub>Pt<sub>25</sub> layer [the violet curve in Figure 1 (d)]. An additional check of the ISHE mechanism involved reversing the

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THz pulse polarity upon reversal of the sample. This experiment was not possible with the studied samples because the Si substrate strongly absorbed the pump pulses.

We also addressed the possible role of Ta capping and buffer layers in all the structures. They could give rise to THz emission via the ISHE effect because the spin Hall angle of Ta was opposite in sign and approximately 10 times smaller than that of Pt. However, in single FM layer structures, the contributions from the two Ta layers cancelled each other because of the opposing directions of the injected spin currents [28]. In the NM/FM structures, the capping Ta layer adjacent to the Co layer provided a small positive correction to  $E_{THz}^{peak}$ . This correction was partly compensated for by the contribution from the buffer Ta layer with a weaker spin current.

### **Discussion**

The above observations bring us to consider the processes responsible for the effective enhancement of different stages of THz generation via the ISHE in the Pt/g-PtCo/Co compared to Pt/Co. At  $\mu_0H=750$  mT both samples were saturated in the plane [Figure 2 (a)], and a quantitative comparison of  $E_{THz}^{peak}$  obtained under such conditions could be made. As is evident from Figure 1 (c),  $E_{THz}^{peak}$  obtained from the Pt/g-PtCo/Co sample was  $\approx 1.7$  times higher than that emitted by Pt/Co at F=3 mJ·cm<sup>-2</sup>.

Three processes potentially lead to the observed enhancement. First, the magnetically ordered Co-Pt alloys at the interface in Pt/g-PtCo/Co were an additional spin current source; second, the Co-Pt interfacial layers contributed to the emission of the THz pulse because of spin-to-charge conversion via the ISHE; finally, the interface in Pt/g-PtCo/Co possess a higher transmittance for the spin current moving from the Co-containing layers to the Pt film.

The fact that the g-PtCo layer added between the Pt and Co could serve as a source of spin current was evident from a comparison of the ultrafast demagnetization data closely related to the spin current [45] for the pairs of Co and Co75Pt25 samples. Ultrafast demagnetization measurements showed that the Co and Co<sub>75</sub>Pt<sub>25</sub> exhibited almost the same absolute demagnetization values [Figure 3 (a)]. Considering that only the fraction of g-PtCo with Pt content above ~ 75% lost its ferromagnetic ordering at room temperature [49], and the estimates of the magnetic dead layer thickness  $d_0$  in the Pt/Co and Pt/g-PtCo/Co (Table 1), we concluded that the thickness of the layer serving as a source of spin current owing to demagnetization increased by  $\approx 0.3$ nm in the Pt/g-PtCo/Co compared to the Pt/Co. The above could lead to an increase in the THz emission as the effective thickness of the FM layer was closer to the optimal one reported to be  $\approx$ 2 nm [15]. However, as shown in Figure 2 (b), the absolute demagnetization value was slightly lower for the Pt/g-PtCo/Co than for Pt/Co, signifying a lower generated spin current, which partly compensated for the effect of increasing the FM layer thickness.

The role of the gradient layer at the interface as an additional spin-to-charge converter was examined by comparing the THz waveforms of the  $\text{Co}_{50}\text{Pt}_{50}/\text{Co}$  and  $\text{Co}_{75}\text{Pt}_{25}/\text{Co}$ , where the signals had comparable magnitudes and opposite polarities [Figure 1 (d)]. The presence of magnetic ordering at room temperature in the PtCo alloys with Pt content below  $\approx 75\%$  [49] allowed them to emit THz pulses owing to the ultrafast demagnetization, which competed with the contribution of the ISHE owing to the spin current injected from the Co.

The balance between these two effects depends on the composition and leads to a dominant contribution to THz emission from the ISHE in the  $\mathrm{Co_{50}Pt_{50}/Co}$  and from ultrafast demagnetization in the  $\mathrm{Co_{75}Pt_{25}/Co}$ . Consequently, the presence of the interfacial g-PtCo effectively increased the thickness of the layer with the ISHE, which, however, was not expected to produce enhancement of THz emission because the Pt thickness of 3 nm was close to the optimal one [15,17,37,50]. A contribution from skew scattering within g-PtCo [25] was not expected as this layer was crystalline [36].

Therefore, we could conclude that the gradient interface between the Co and Pt layers mediated the delivery of more spin current to the Pt layer. To quantify the corresponding enhancement of the transmittance T of the interface in the Pt/g-PtCo/Co as compared to Pt/Co, a ratio between spin currents  $J_s \simeq T\Delta M$  [26,45] in the Pt in the two samples was evaluated. Based on the formalism in Ref [37],  $J_s$  can be related to  $E_{THz}^{peak}$  as follows:

$$E_{\mathrm{THz}}^{\mathrm{peak}} \propto T \Delta M \mathrm{tanh} \left( \frac{d_{\mathrm{FM}} - d_0}{2 \lambda_{\mathrm{pol}}} \right) \mathrm{tanh} \left( \frac{d_{\mathrm{NM}}}{2 \lambda_{\mathrm{NM}}} \right) \times Z \exp \left( -\frac{d_{\mathrm{FM}} + d_{\mathrm{NM}}}{S_{\mathrm{THz}}} \right),$$
 (1)

where

$$Z = \frac{Z_0}{n_{\rm air} + n_{\rm Si} + Z_0(\sigma_{\rm FM}d_{\rm FM} + \sigma_{\rm NM}d_{\rm NM} + \sigma_{\rm Ta}d_{\rm Ta})}.$$

Here,  $\lambda_{\rm pol}$  denotes the critical thickness of the FM layer above which the generated spin polarization saturates [37],  $\lambda_{\rm NM}$  is the spin diffusion length in the NM layer,  $Z_0$  stands for the free space impedance,  $n_{\rm Si}$  is the optical index of the substrate,  $\sigma_m$  (m = FM, NM, Ta) denotes the conductivity of the layer, and  $d_m$  is its thickness.

In Pt/g-PtCo/Co,  $d_{\rm FM}$  stands for the total thickness of Co and the gradient interface. A typical value of the effective inverse absorption constant,  $S_{\rm THz}$  coming from multiple reflections in metal structures was

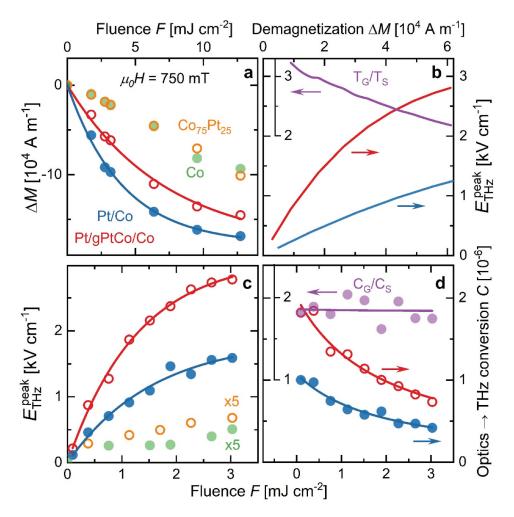


Figure 3. (a) Absolute demagnetization (ΔM) value as a function of the pump fluence. Lines are fit using an exponential function. (b)  $E_{THz}^{peak}$  as a function of the absolute demagnetization (ΔM) (right axis: red and blue lines) plotted using fit functions from panels a, c. The ratio between the interface effective transmittances of Pt/g-PtCo/Co ( $T_G$ ) and Pt/Co ( $T_G$ ) (additional left axis: purple line). (c)  $E_{THz}^{peak}$  as a function of the pump laser fluence measured in the external magnetic field  $\mu_0H = 750$  mT applied in the sample plane. Lines are fit using an exponential function. (d) Conversion of the optical fluence to THz radiation of Pt/g-PtCo/Co ( $T_G$ ) and Pt/Co ( $T_G$ ) (right axis: red and blue symbols). Lines are fit using a reciprocal function.  $T_G/T_G$  as a function of fluence ( $T_G$ ) using a linear fit (additional left axis: purple symbols).

used. All the parameters used in the calculations are listed in Table 2.

The dependences  $E_{THz}^{peak}$  ( $\Delta M$ ) for the Pt/g-PtCo/Co and Pt/Co samples are shown in Figure 3 (b), as obtained from the fluence dependences of these values interpolated by the exponential functions  $1 - A\exp(-F)$  [Figures 3(a), 3(c)]. Using these data, Equation (1), and assuming that the spin Hall angle in the Pt layer was the same in the two samples, we could obtain a ratio  $T_G/T_S$  between the spin transmittance of the gradient interface  $T_G$  in the Pt/g-PtCo/Co and of the abrupt interface  $T_S$  in the Pt/Co at different degrees of demagnetization  $\Delta M$  [as shown by the purple line in Figure 3 (b)].  $T_G/T_S \approx 3$  at

low demagnetization, with the gradient interface allowing a pronounced increase of the injected spin current into the Pt layer. However, as the degree of demagnetization increased,  $T_{\rm G}/T_{\rm S}$  steadily decreased.

The latter could be ascribed to spin accumulation in the NM layer, which limited the growth of the spin current in this layer [28]. Indeed, in heavy-metal layers, the injection of spin-polarized electrons results in spin accumulation because of relatively low electron mobility. In the limiting case of a strong spin current, spin accumulation prevents a further increase in the injected spin current. Thus, the transmittance which connects  $J_s$  and  $\Delta M$  is effectively reduced with increased spin

**Table 2.** Material parameters used in the calculations of  $E_{THz}^{peak}$ .

			1112		
$\lambda_{Pt}$	3.4 nm	[32]	$S_{THz}$	150 nm	[51]
$\lambda_{Co}$	0.7 nm	[52]	n <sub>Si</sub>	3.42	[53]
$\sigma_{Co}$	$3.10^{-3} (\Omega \text{ nm})^{-1}$	[26]	$Z_0$	377 Ω	[37]
$\sigma_{Pt}$	$4.10^{-3} (Ω nm)^{-1}$	[26]			
$\sigma_{Ta}$	$2.4 \cdot 10^{-3} (\Omega \text{ nm})^{-1}$	[54]			

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accumulation. The decrease in  $T_G/T_S$  indicates that spin accumulation was more pronounced in Pt/g-PtCo/Co, which resulted from the higher initial transmittance  $T_G$  of the interface in this structure.

A characteristic signature of the spin accumulation affecting the spin current injected into the Pt layer is the saturation behavior of the optical fluence dependence of  $E_{THz}^{\it peak}$  and THz fluence [28]. Indeed,  $E_{THz}^{\it peak}$  (F) in the Pt/ g-PtCo/Co and Pt/Co exhibited a tendency toward saturation, which was distinct from the dependence of the THz field generated by ultrafast demagnetization in the Co and Co<sub>75</sub>Pt<sub>25</sub> samples [Figure 3 (c)]. Moreover, there was a decreasing trend in the optical-to-THz fluence conversion coefficient C(F), as shown in Figure 3 (d). The THz fluence was calculated by integrating  $E_{THz}^2$ over the THz pulse duration divided by the irradiated area of the electro-optical crystal [47]. In both samples, the conversion decreased with optical fluence, similar to the results reported in Ref [55]. Fitting this dependence using a reciprocal function  $(c_0+d\ F)^{-1}$ , we determined that the maximum conversion achievable at low fluences amounted to  $2 \cdot 10^{-6}$  in Pt/g-PtCo/Co and  $1 \cdot 10^{-6}$  in Pt/Co. The optics-to-THz conversion efficiency ratio  $C_G/C_S$ between the Pt/g-PtCo/Co and Pt/Co was ≈ 2 and was constant in the studied optical fluence range [as shown by the linearly fitted purple symbols in Figure 3 (d)].

These observations can be explained as follows. On one hand, the rate of increase of the demagnetization value  $\Delta M$  grows slower with optical fluence in the Pt/ g-PtCo/Co than in Pt/Co. As a result, the increase in demagnetization became sublinear in Pt/Co at lower fluences than in Pt/g-PtCo/Co, as shown in Figure 3 (a). This affected the generation of spin current. However, the higher transmittance of the gradient interface in the Pt/g-PtCo/Co resulted in an overall larger spin current injected into the Pt layer and a larger spin accumulation effect. The interplay of these two effects resulted in a nearly constant ratio of optical-to-THz fluence conversion of  $C_G/C_S \approx 2$ . Notably, increased optical fluence led to more extensive heating of the Pt layer, which in turn affected the spin Hall angle in the Pt owing to the higher spin-dependent scattering at elevated temperatures [56]. However, this effect was expected to be similar in both samples studied, thus it did not influence the  $C_G/C_S$  and  $T_G/T_S$  ratios.

Finally, we addressed the possible connection between the increased transmittance of the gradient interface for the spin-polarized current revealed in the experiments and the recently reported increase in the interfacial DMI [36]. The transmittance is related to the average spin conductance  $(g^{\uparrow} + g^{\downarrow})/2$ . Ref. [26] suggested a direct correlation between the average spin conductance in an NM/FM structure and the spin-mixing conductance  $g^{\uparrow\downarrow}$  of the NM/FM interface. Further, interfacial DMI and spin-mixing conductance are correlated in NM/FM structures [29]. Thus, the results reported here and

those in Ref. [36] demonstrated that the gradient interface in the Pt/Co structure led to an increase in the THz emission and DMI, supporting the conclusions of these studies.

### **Conclusions**

This study highlighted the crucial role of interface design in spintronic emitters, which dramatically affects their spin current transmittance. Using a crystalline composition-gradient interface between the ferromagnetic and heavy-metal layers led to a two-fold increase in the efficiency of the Pt/Co spintronic emitter compared with conventional heterostructures. These findings paved the way for further optimization of NM/FM emitters by exploring the effect of the gradient steepness and thickness of the interfacial layer on the THz emission. The enhancement of the interface spin transmittance was observed alongside the increase in interfacial DMI, suggesting an intrinsic link between the spin and spin-mixing conductance values and the DMI in Pt/Co structures. The Pt/Co structure with a gradient interface supported the in-plane magnetic anisotropy required for THz emitters in combination with a small Co layer thickness, which was optimal for efficient spin current generation. This contrasts with Pt/Co structures, which were typically characterized by out-of-plane anisotropy.

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# **Disclosure statement**

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### **ORCID**

Leonid A. Shelukhin http://orcid.org/0000-0001-8538-3773

- Anna V. Kuzikova (b) http://orcid.org/0000-0002-6648-8817
- Andrey V. Telegin (b) http://orcid.org/0000-0001-7209-4307
- Vladimir D. Bessonov http://orcid.org/0000-0002-7837-
- Alexey V. Ognev http://orcid.org/0000-0002-1619-3666 Alexander S. Samardak http://orcid.org/0000-0001-5917-4361
- Junho Park http://orcid.org/0000-0003-0294-1288
  Young Keun Kim http://orcid.org/0000-0002-0868-4625
  Alexandra M. Kalashnikova http://orcid.org/0000-0001-5635-6186

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