Journal of Magnetism and Magnetic Materials

Generation of Terahertz Transients from Co2Fe0.4Mn0.6Si Heusler alloy/ Heavy-Metal Bilayers

--Manuscript Draft--

Manuscript Number:	MAGMA-D-21-01241
Article Type:	VSI: PM21
Keywords:	THz Radiation; Inverse Spin Hall Effect; Spin-to-Charge Conversion; Ferromagnetic Resonance
Corresponding Author:	Sarah Friederike Heidtfeld Forschungszentrum Jülich GmbH: Forschungszentrum Julich GmbH Jülich, GERMANY
First Author:	Sarah Friederike Heidtfeld
Order of Authors:	Sarah Friederike Heidtfeld
	Roman Adam
	Takahide Kubota
	Koki Takanashi
	Derang Cao
	Carolin Schmitz-Antoniak
	Daniel E. Bürgler
	Fangzhou Wang
	Christian Greb
	Genyu Chen
	Ivan Komissarov
	Hilde Hardtdegen
	Martin Mikulics
	Roman Sobolewski
	Shigemasa Suga
	Claus Michael Schneider
Abstract:	We generated pulses of electromagnetic radiation with a frequency content up to three Terahertz by optical excitation of $Co_2Fe_{0.4}Mn_{0.6}Si$ Heusler alloy/heavy metal bilayers (CFMS/HM) using fs-laser pulses. We attribute the generation process to the inverse spin Hall effect. We compared the THz emission efficiency in CFMS/Pt and CFMS/Ta bilayers due to their high spin-orbit coupling of Pt and Ta. Surprisingly, our data reveal that CFMS/Pt shows substantially larger THz amplitudes compared to CFMS/Ta. Both bilayers exhibit a tunability of the THz amplitude by external magnetic field both at 300 K and 100 K. Ferromagnetic resonance measurements demonstrate that CFMS/Ta has a high effective spin mixing conductance, describing the efficiency of interfacial spin transport. We observe that the efficiency of the THz radiation cannot be solely described by the spin-orbit coupling strength and the spin diffusion length in the HM material plays an important role.
Suggested Reviewers:	Tobias Kampfrath tobias.kampfrath@fu-berlin.de
	Tom S. Seifert tom.seifert@fu-berlin.de
	Evangelos Papaioannou evangelos.papaioannou@physik.uni-halle.de

Sarah Heidtfeld, Roman Adam, Derang Cao, Carolin Schmitz-Antoniak, Daniel E. Bürgler, Fangzhou Wang, Christian Greb, Genyu Chen, Ivan Komissarov, Hilde Hardtdegen, Martin Mikulics, Roman Sobolewski, Shigemasa Suga, and Claus M. Schneider*

Dear Editor of Journal of Magnetism and Magnetic Materials:

herewith we would like to submit our manuscript titled "*Generation of Terahertz Transient from $Co_2Fe_{0.4}Mn_{0.6}Si$ Heusler alloy/ Heavy-Metal Bilayers*" for publication in the Journal of Magnetism and Magnetic Materials.

Heusler alloys have been long identified as one of the most interesting class of materials for spin-based electronics. Demonstration of efficient THz generation in these materials may lead to an interesting merge of low power- and high frequency applications.

In our manuscript we show that $Co_2Fe_{0.4}Mn_{0.6}Si$ *Heusler alloy/Heavy Metal (CFMS/HM) nanobilayers generate THz radiation* when excited by femtosecond laser pulses.

We find that the amplitude of the generated THz transients is surprisingly larger in CFMS/Pt compared to CFMS/Ta. We observe that the amplitude in both bilayers flips sign when reversing the direction of an external magnetic field and follows the magnetization in the sample. We show that the THz amplitude is increased when cooling the sample down to 100 K, which is in agreement with increasing magnetization in the sample.

In addition, we employed ferromagnetic resonance experiments in order to gain deeper insight into the conditions affecting efficiency of THz generation. We found that the *spin-orbit coupling cannot completely describe the THz generation process* in the bilayers and that the spin *transmission* efficiency and the *spin diffusion* length in the NM play crucial roles in the THz emission process.

As our detailed research brings new light into the THz generation in Heusler alloy/Hevy Metal bilayers, we feel that our manuscript is very suitable for publication in Journal of Magnetism and Magnetic Materials.

Sincerely, Sarah Heidtfeld, and Roman Adam on behalf of all authors

*****Author to whom correspondence should be addressed. Electronic address: r.adam@fz-juelich.de

Generation of Terahertz Transients from $Co_2Fe_{0.4}Mn_{0.6}Si$ Heusler alloy/ Heavy-Metal Bilayers

S. Heidtfeld[1,2], R. Adam[1], T. Kubota[3,4], K. Takanashi[3,4,5], D. Cao[1,6], C. Schmitz-Antoniak[1], D. E. Bürgler[1], F. Wang[1,2], C. Greb[1,2], G. Chen[7,8], I. Komissarov[9,10], H. Hardtdegen[11], M. Mikulics[11], R. Sobolewski[7,8,9,12], S. Suga[1,13] and C.M. Schneider[1,2,14]

[1]Research Centre Jülich, Peter Grünberg Institute, 52425 Jülich, Germany

[2]Faculty of Physics, University of Duisburg-Essen, 47048 Duisburg, Germany

[3]Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

[4] Center for Spintronics Research Network, Tohoku University, Sendai 980-8577 Japan

[5]Center for Science and Innovation in Spintronics (Core Research Cluster), Tohoku University, Sendai 980-8577, Japan

[6]College of Physics, National Demonstration Center for Experimental Applied Physics Education, Qingdao University, Qingdao 266071, China

[7]Materials Science Graduate Program, University of Rochester, Rochester, New York 14627-1299, USA

[8]Laboratory for Laser Energetics, University of Rochester, Rochester, New York 14623, USA

[9]Department of Electrical and Computer Engineering, University of Rochester, Rochester, New York 14627-0231, USA

[10]Belarusian State University of Informatics and Radioelectronics, Minsk, 220013 Belarus

[11]Research Centre Jülich, Ernst Ruska Centre for Microscopy and Spectroscopy with Electrons, 52425 Jülich, Germany

[12]Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627-0171, USA

[13]The Institute of Scientific and Industrial Research, Osaka University, Ibaraki, Osaka 567-0047, Japan

[14]Department of Physics, University of California Davis, Davis, California 95616-5270, USA

***Abstract*—We generated pulses of electromagnetic radiation with a frequency content up to three terahertz (THz) by optical excitation of $Co_2Fe_{0.4}Mn_{0.6}Si$ Heusler alloy/heavy metal bilayers (CFMS/HM) using fs-laser pulses. We attribute the generation process to the inverse spin Hall effect. We compared the THz emission efficiency in CFMS/Pt and CFMS/Ta bilayers due to their high spin-orbit coupling of Pt and Ta. Surprisingly, our data reveal that CFMS/Pt shows substantially larger THz amplitudes compared to CFMS/Ta. Both bilayers exhibit a tunability of the THz amplitude by external magnetic field both at 300 K and 100 K. Ferromagnetic resonance measurements demonstrate that CFMS/Ta has a high effective spin mixing conductance, describing the efficiency of interfacial spin transport. We observe that the efficiency of the THz radiation cannot be solely described by the spin-orbit coupling strength and the spin diffusion length in the HM material plays an important role.**

I. INTRODUCTION

The conversion from spin- to charge-currents attracted increasing significance for the future use in spin-based electronic devices operating at high speeds. The inverse spin Hall effect (ISHE) is a process that results in the conversion of a spin current $\vec{J}_s$ into a charge current $\vec{J}_c$, with the spin Hall angle D_{ISHE} describing the conversion efficiency $\vec{J}_c \propto D_{\mathrm{ISHE}}\,(\vec{J}_s \times \overrightarrow{\sigma})$. Here $\vec{\sigma}$ are the spin matrices defining the spin polarization direction of $\vec{J_s}$ [1, 2]. It has been demonstrated earlier that, in laser illuminated ferromagnet (FM)/heavy metal (HM) bilayers, the ISHE can be used for the generation of picosecond electromagnetic transients with a frequency content extending up to the terahertz (THz) frequency range [3].

In the present work we fabricated $Co_2Fe_{0.4}Mn_{0.6}Si$ (CFMS)/HM bilayers with HM standing either for Pt or Ta. The CFMS Heusler alloy is a half-metal with a bandgap in one spin channel that exhibits 100% spin polarization at the Fermi level, in the ideal case. This unique feature is expected to result in a substantial enhancement of the interfacial spin current polarization [4] compared to a standard FM/HM bilayer emitter.

II. Results

We used laser 100-fs-wide laser pulses at 800 nm wavelength to illuminate CFMS/HM bilayers from the HM side, with a laser fluence of 6 µJ/cm². The generated THz transients, detected at the MgO substrate side, were first collimated and then focused onto an antenna detector patterned on top of LT-grown GaAs. When comparing the THz transients, we observed that the THz transient amplitude $A^{\mathrm{THz(Ta)}}$ for CFMS/Ta was only $A^{\mathrm{THz(Ta)}} \approx 0.1\ A^{\mathrm{THz(Pt)}}$ [see **Fig. 1(a)**]. Furthermore, we noted a reversed polarity of the THz transient amplitude of Ta compared to Pt - in agreement with spin Hall angles from literature [5]. To investigate the origin of the THz transients we reversed the externally applied magnetic field direction and observed an opposite polarity of the transient amplitude (not shown). We therefore ascribe the generation process of THz transients from CFMS/HM bilayers to the ISHE. The spin Hall angle describing the efficiency of the ISHE depends on the material-specific spin-orbit coupling (SOC), which scales approximately with Z^4, where Z is the atomic number. This means that $SOC^{\mathrm{(Ta)}} \approx 0.77\ SOC^{\mathrm{(Pt)}}$. We note that if the THz transient amplitude was determined solely by SOC, the same ratio should be expected for the THz amplitudes A^{THz}.

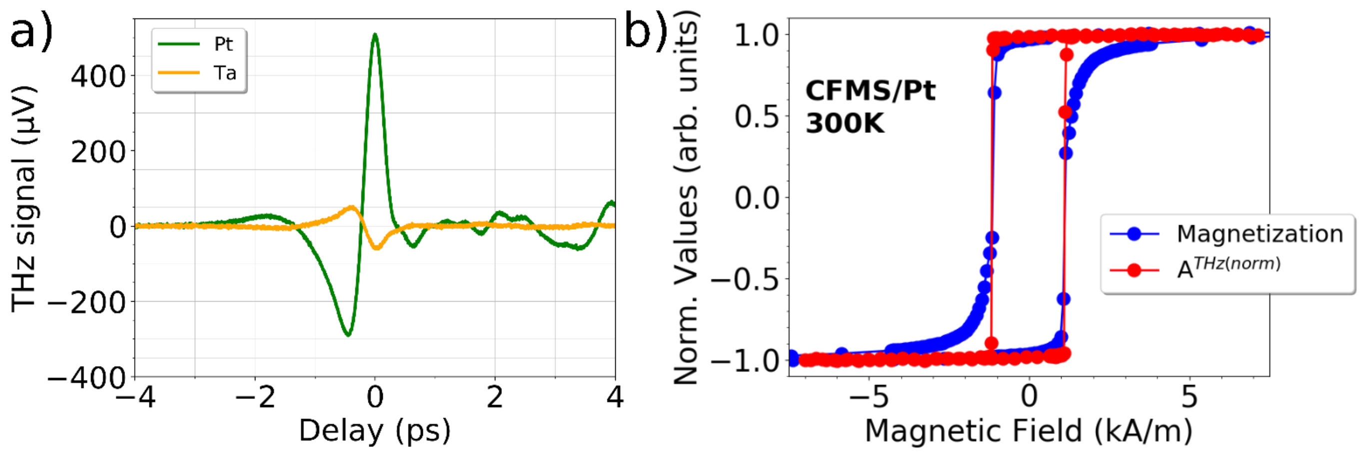


Fig. 1. (a) THz transients generated by CFMS/Pt (green) and CFMS/Ta (yellow) bilayers. (b) Normalized THz amplitude from CFMS/Pt bilayer as a function of the in-plane magnetic field (red) and the normalized magnetization in the sample measured by a VSM (blue).

Figure 1(b) shows the THz transient amplitude $A^{\mathrm{THz(Pt)}}(H)$ (red dots) and the sample magnetization (blue dots) of CFMS/Pt bilayer as a function of the magnetic field. The sample magnetization was measured by a vibrating sample magnetometer (VSM). We observe that the THz amplitudes measured for each transient follow the magnetization of the sample and show a very similar hysteretic behavior, as seen for a Py/Pt bilayer in [6]. The difference in the sharpness of the reversal can be explained by a different sample volume probed by the two measurement techniques.

When cooling down the sample system to a temperature of 100 K, we observed that the shape of the THz transient of the CFMS/Pt bilayer remains unchanged, whereas the amplitude is increased compared to the signal at 300 K, see **Figure 2(a)**. We observed an increase in the THz transient amplitude of (8 ± 5) % at 100 K. This agrees with the magnetization increase seen in VSM measurements of (4.4 ±0.1) %.

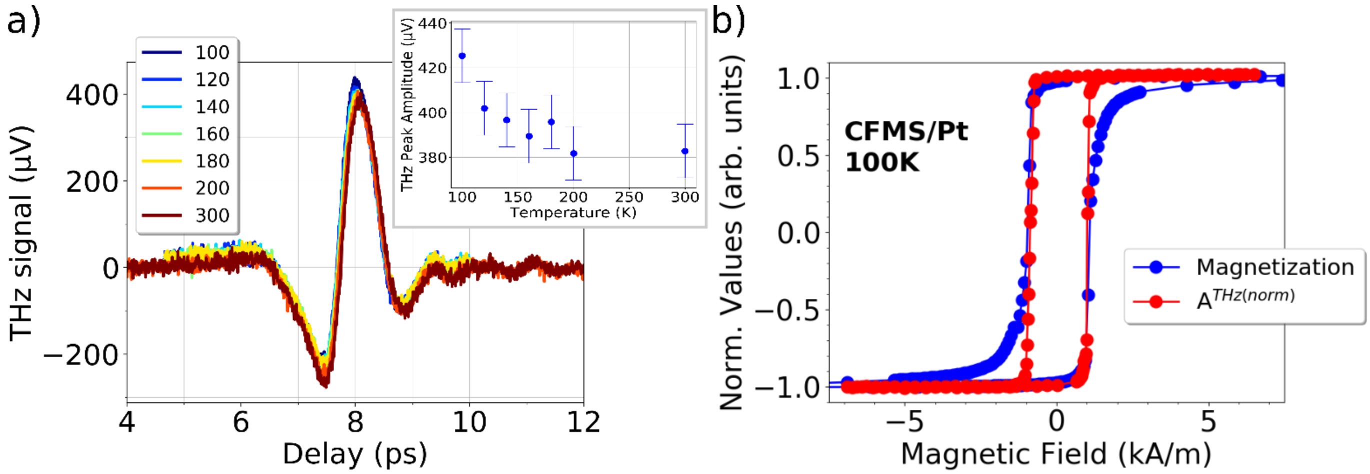


Fig. 2. (a) THz transients for CFMS/Pt at different temperatures between 100 K and 300 K. Peak amplitudes are shown as an inset. (b) Normalized THz amplitude from CFMS/Pt bilayer as a function of the in-plane magnetic field (red) compared to the normalized magnetization in the sample measured by a VSM (blue) at 100 K.

In order to investigate the spin injection from the CFMS into the HM layer we employed ferromagnetic resonance (FMR) to generate spin pumping across the interface. We placed the samples on top of a coplanar waveguide and measured the S_{12} parameter as a function of frequency, between 50 MHz and 20 GHz at different externally in-plane applied magnetic fields, see **Figure 3(a)**. Fitting of the FMR linewidth versus the resonance frequency allows the extraction of the effective spin mixing conductance $g_{\mathrm{eff}}^{\uparrow\downarrow}$, a parameter defining the interfacial spin transport efficiency [7]. **Figure 3(b)** shows the normalized THz transient amplitudes of the two tested sample systems (yellow columns), the normalized SOC values approximated by Z^4 (red columns), and the extracted $g_{\mathrm{eff}}^{\uparrow\downarrow}$ (blue columns). Our data show that CFMS/Ta exhibits both relatively large $g_{\mathrm{eff}}^{\uparrow\downarrow}$ and SOC. The large discrepancy of the THz amplitude ratio between $A^{\mathrm{THz(Ta)}}$ / $A^{\mathrm{THz(Pt)}}$ ~ 0.8 expected solely based on SOC and the measured ratio $A^{\mathrm{THz(Ta)}}$ / $A^{\mathrm{THz(Pt)}}$ ~ 0.1 points to additional mechanisms affecting the THz amplitude. When comparing the spin diffusion length λ_{sf} in Ta and Pt, we find that $\lambda_{\mathrm{sf(Ta)}}$= 1.9 nm is very small compared to $\lambda_{\mathrm{sf(Pt)}}$= 7.3 nm [8] and even smaller than the thickness of the Ta layer. We adjusted the expected values from SOC alone by multiplying with $g_{\mathrm{eff}}^{\uparrow\downarrow}$, which describes the efficiency of spin injection from the CFMS into the HM layer, and with $\lambda_{\mathrm{sf}} \tanh(\frac{t_{Ta}}{2\,\lambda_{\mathrm{sf}}})$, which accounts for the spin diffusion length in the HM, with t_{Ta} being the thickness of the Ta layer [9]. Although, we then observed a closer agreement for the ratio based on $A^{THz\,(theo)} \propto SOC \cdot g_{\text{eff}}^{\uparrow\downarrow} \cdot \lambda_{\text{sf}} \tanh(\frac{t_{NM}}{2\,\lambda_{\text{sf}}})$ with the measured ratio, the amplitude measured for CFMS/Ta still remains lower by a factor of 5. A possible reason for the low $A^{THz\,(meas)}$ in Ta could be the oxidation of the Ta layer, since no capping layer was added on top of Ta. In [10] it was seen that the spin Hall angle can be increased or reduced depending on the level of oxygen content.

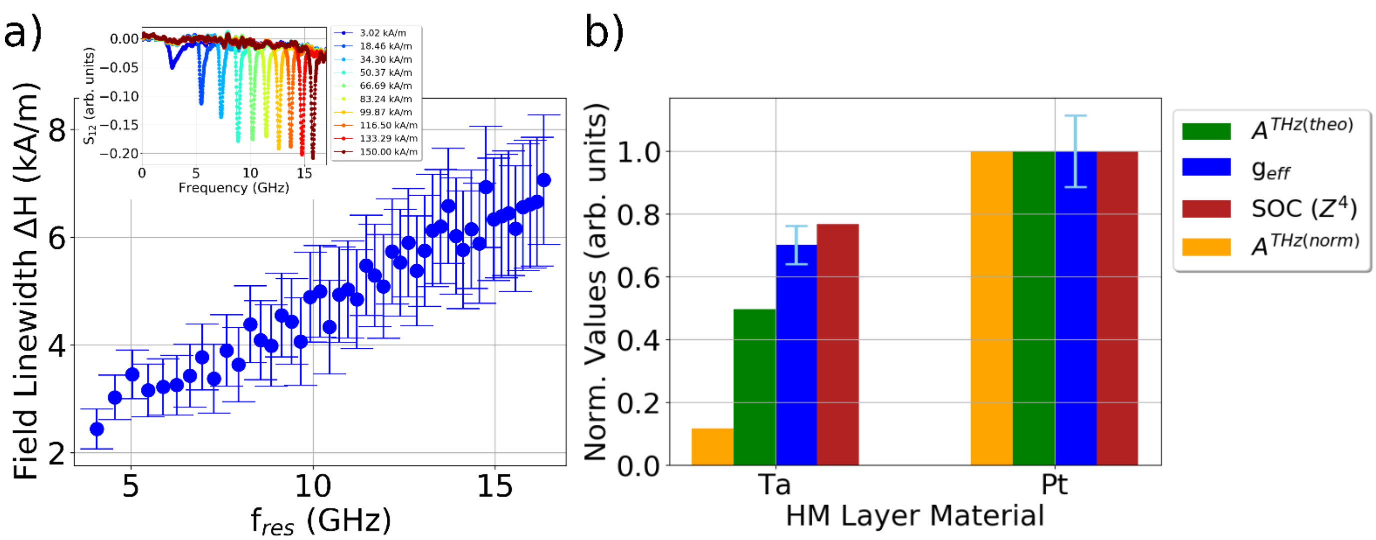


Fig. 3. (a) Field linewidths for CFMS/Ta, extracted from the FMR measurements, versus the resonance frequency. The inset shows absorption dips at the resonance frequencies for the S_{12} parameter at different magnetic fields. (b) Normalized THz amplitude $A^{\text{THz(norm)}}$ (yellow bars), effective spin mixing conductance $g_{\text{eff}}^{\uparrow\downarrow\text{(norm)}}$ and Z^4 (SOC) for CFMS/Ta and CFMS/Pt. The green bar shows the expected value for the THz amplitude compared to Pt via $A^{THz\,(theo)} \propto SOC \cdot g_{\text{eff}}^{\uparrow\downarrow} \cdot \lambda_{\text{sf}} \tanh(\frac{t_{NM}}{2\,\lambda_{\text{sf}}})$. All values are normalized to the corresponding values for Pt.

III. Summary

We generated THz transients from CFMS/Pt and CFMS/Ta bilayers and could attribute the THz generation mechanism to the ISHE. We compared the THz amplitudes for the two HM materials and observed that Ta showed a substantially lower, by a factor of 8, THz amplitude, as compared to the expected value from the SOC dependence of the spin Hall angle. We observed that SOC and $g_{eff}^{\uparrow\downarrow}$ (value extracted from a FMR experiment) were not solely responsible for the lower THz amplitude. A possible explanation could be the oxidation of the Ta layer. Additionally, we measured the magnetic field dependence of CFMS/Pt and observed a similar hysteretic behavior for the VSM measurement and the THz transient amplitudes at 300 K and at lower temperatures down to 100 K. We measured an increase of the THz amplitude by (8 ± 5) %, which is in agreement with the magnetization increase from VSM of (4.5 ± 0.1) %.

IV. Acknowledgment

The work at the Research Center Jülich was performed within JuSPARC (Jülich Short-pulse Particle Acceleration and Radiation Center), a strategy project funded by the BMBF. Research in Rochester was funded in part by the National Science Foundation grant #1842712.

References

1. Saitoh, E., et al., *Conversion of spin current into charge current at room temperature: Inverse spin-Hall effect.* Applied Physics Letters, 2006. **88**(18): p. 182509.
2. Yu, R., et al., *Determination of spin Hall angle and spin diffusion length in β-phase-dominated tantalum.* Physical Review Materials, 2018. **2**(7): p. 074406.
3. Kampfrath, T., et al., *Terahertz spin current pulses controlled by magnetic heterostructures.* Nature Nanotechnology, 2013. **8**(4): p. 256-260.
4. Kubota, T., et al., *Half-metallicity and Gilbert damping constant in* $Co_2Fe_xMn_{1-x}Si$ *Heusler alloys depending on the film composition.* Applied Physics Letters, 2009. **94**(12): p. 122504.
5. Seifert, T., et al., *Efficient metallic spintronic emitters of ultrabroadband terahertz radiation.* Nature Photonics, 2016. **10**: p. 483.
6. Adam, R., et al., *Magnetically and optically tunable terahertz radiation from Ta/NiFe/Pt spintronic nanolayers generated by femtosecond laser pulses.* Applied Physics Letters, 2019. **114**(21): p. 212405.
7. Tserkovnyak, Y., A. Brataas, and G.E.W. Bauer, *Spin pumping and magnetization dynamics in metallic multilayers.* Physical Review B, 2002. **66**(22): p. 224403.
8. Wang, H., et al., *Scaling of Spin Hall Angle in 3d, 4d, and 5d Metals from* $Y_3Fe_5O_{12}$*/Metal Spin Pumping.* Physical Review Letters, 2014. **112**: p. 197201.
9. Seifert, T.S., et al., *Terahertz spectroscopy for all-optical spintronic characterization of the spin-Hall-effect metals Pt, W and Cu80Ir20.* Journal of Physics D: Applied Physics, 2018. **51**(36): p. 364003.
10. Yang, L., et al., *Maximizing spin–orbit torque efficiency of Ta(O)/Py via modulating oxygen-induced interface orbital hybridization.* Applied Physics Letters, 2021. **118**(3): p. 032405.

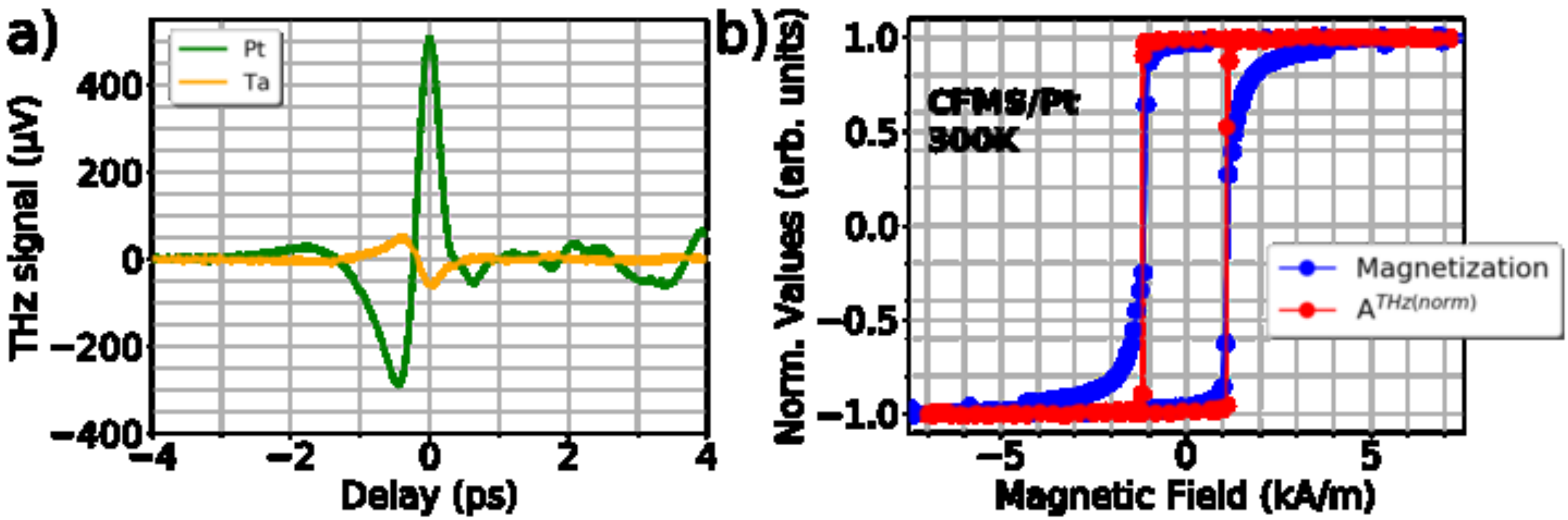
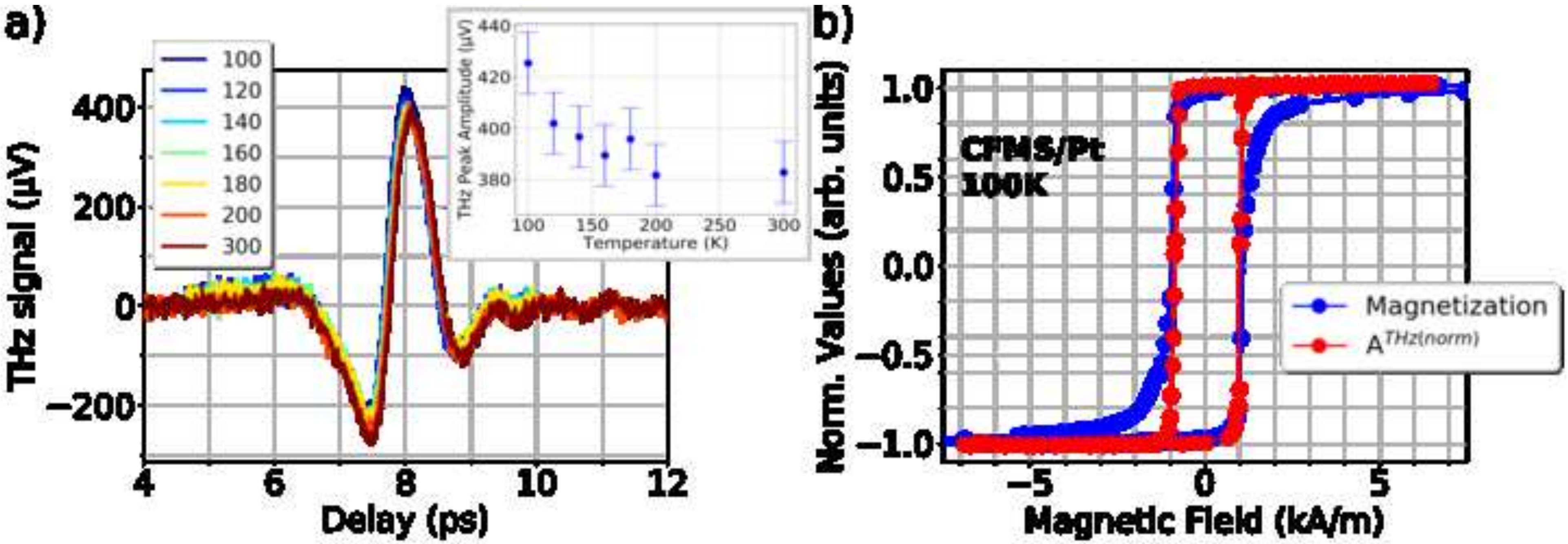
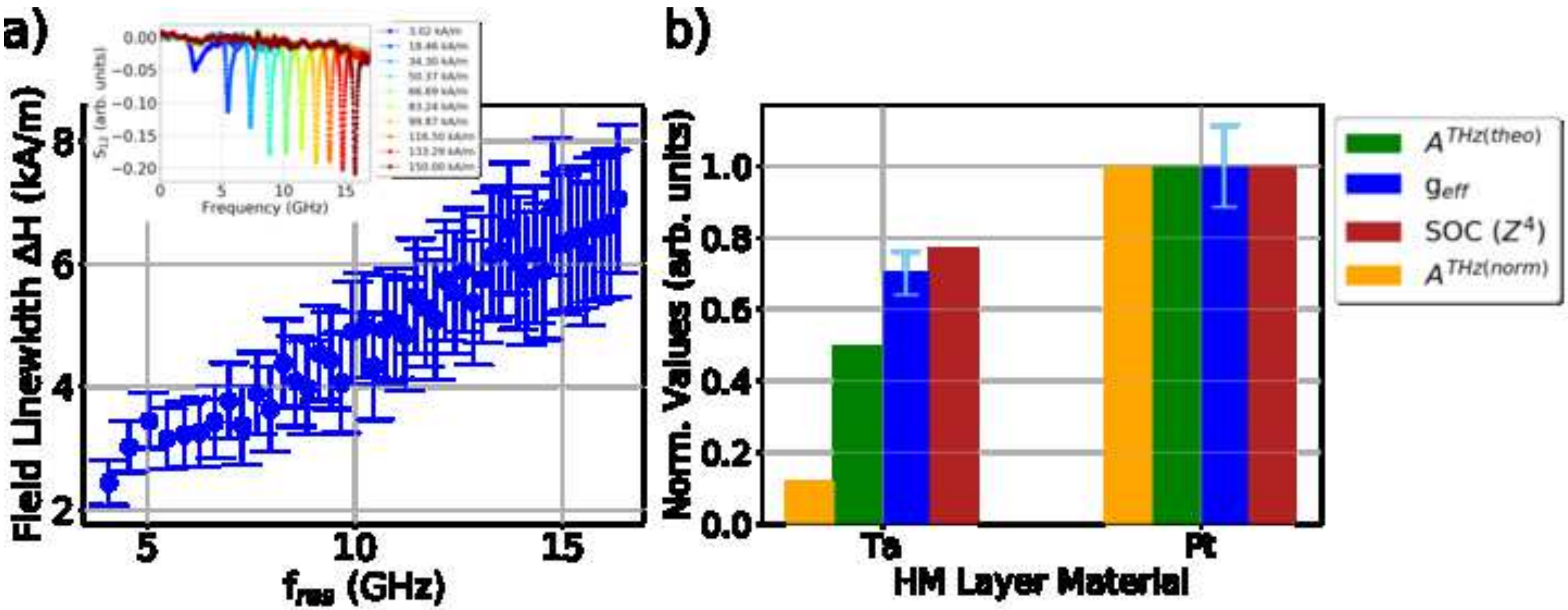

a)
Pt
Ta
THz signal (μV)
400
200
0
−200
−400
−4
−2
0
2
4
Delay (ps)
b)
Norm. Values (arb. units)
1.0
0.5
0.0
−0.5
−1.0
CFMS/Pt
300K
Magnetization
$A^{THz(norm)}$
−5
0
5
Magnetic Field (kA/m)

Figure2

Click here to access/download;Figure;figure2.png




a)
b)
Field Linewidth ΔH (kA/m)
f_{res} (GHz)
S_{11} (arb. units)
Frequency (GHz)
Norm. Values (arb. units)
HM Layer Material
Ta
Pt
$A^{THz(theo)}$
g_{eff}
SOC (Z^4)
$A^{THz(norm)}$

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: