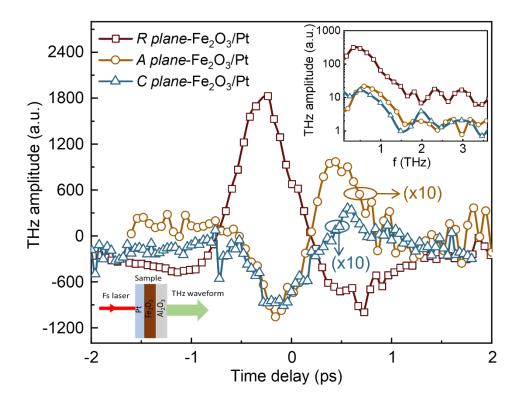
## Controlling Ultrafast Terahertz Dynamics through Crystalline Orientation in Antiferromagnetic Hematite

(in-depth report)

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We report on the generation of ultrafast terahertz (THz) spin currents through femtosecond laser excitation of epitaxial bilayer thin films comprising  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> (an insulating antiferromagnet) and Pt (a heavy metal). The epitaxial thin films of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> were grown in three distinct orientations, namely C, A, and R planes, employing pulsed laser deposition. We show that the magnitude of THz emission from the  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> /Pt system can be controlled by altering the crystalline orientation of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> as shown in Fig. 1. By using the R-plane  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>, we demonstrate a substantial enhancement of the THz signal, nearly one order of magnitude greater compared to the C-plane and A-plane  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> /Pt. We perform a detailed investigation into the azimuthal and polarization dependence of the THz emission. Our investigations establish that the large amplitude of THz emission observed in the R-plane  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> /Pt system is intricately linked to the spin domain distribution, which, in turn, is determined by the crystalline symmetry specific to the R-plane  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>. These results contribute significantly to the understanding of the ultrafast dynamics of spin currents in antiferromagnets.

**Fig. 1.** THz emission amplitudes from R, A and C planes  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>/Pt heterostructure grown on Al<sub>2</sub>O<sub>3</sub> substrates. The inset displays the Fourier spectra of the THz signal in the R, A, and C plane of the  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>/Pt heterostructure.



# Equal contribution as first author, \*Equal contribution as corresponding author