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Terahertz Emission from Giant Optical Rectification in a van der Waals Material

Exfoliation and stacking of twodimensional (2D) van der Waals (vdW) crystals have created unprecedented opportunities in the discovery of quantum phases. A major obstacle to the advancement of this field is the limited spectroscopic access due to a mismatch in sample sizes $(10^{-6} - 10^{-5} \text{ m})$ and wavelengths $(10^{-4} - 10^{-3} \text{ m})$ of electromagnetic radiation relevant to their low-energy excitations. Here, we introduce ferroelectric semiconductor NbOl₂ as a 2D vdW material capable of operating as a vdW terahertz (THz) emitter. We demonstrate intense and broadband THz generation from NbOl₂ with optical rectification efficiency over one-order-of-magnitude higher than that of ZnTe, the current standard THz emitter. Moreover, this NbOl₂ THz emitter can be integrated into vdW heterostructures to enable on-chip near-field THz spectroscopy of a target vdW material and device. Our approach provides a general spectroscopic tool for 2D vdW materials and quantum matter.

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Fig. 1. Near-field THz spectroscopy exploiting efficient THz emission of 2D vdW ferroelectric NbOl₂. a, Schematic illustration of sub-diffraction limit THz spectroscopy. The inset shows the structure of the vdW layered NbOl₂. **b**, Time-domain waveform of THz emission from 2.5-µm NbOl₂ (red) and 0.2mm ZnTe (blue, multiplied by 0.5). **c**, Frequency-domain spectra for NbOl₂ (red) and ZnTe (blue). **d**, Pump fluence dependences of the emitted field amplitudes, normalized by the thickness. The dotted lines show a linear dependence. The thickness normalized THz emission efficiency from NbOl₂ is 20x to 100x higher than that from ZnTe, the standard THz emitter used today.



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