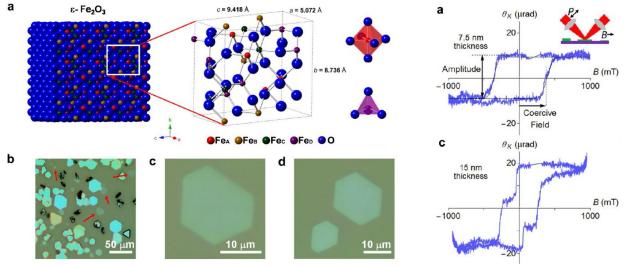


## Room Temperature Magnetic Order in Air-Stable Ultra-Thin Iron Oxide

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## Introduction

Iron oxides are abundant in nature and are present in almost every domain on earth. They are also among the most studied metal oxides, having been applied exhaustively for technological applications such as data storage and catalysis, and biomedical applications such as drug delivery, medical imaging, and cancer treatment. The most common polymorphs of iron oxide are  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> (hematitie),  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> (maghemite), and Fe<sub>3</sub>O<sub>4</sub> (magnetite), which exist in both bulk and nanoscale forms. In contrast,  $\epsilon$ - Fe<sub>2</sub>O<sub>3</sub> is a rare phase with little natural existence and has only been found at the nanoscale. It has received far less research interest than the other polymorphs, due partially to its difficulty in preparation, as it cannot be grown in bulk. However,  $\epsilon$ -Fe<sub>2</sub>O<sub>3</sub> has a variety of interesting characteristics, such as ferrimagnetism, multiferroicity, and a large coercive field, motivating investigation into growth techniques and properties.



**Figure:** Crystal structure and optical images of  $\varepsilon$ -Fe<sub>2</sub>O<sub>3</sub>. a, Non-layered  $\varepsilon$ -Fe<sub>2</sub>O<sub>3</sub> has an orthorhombic structure with a = 5.072 Å, b = 8.736 Å and c = 9.418 Å. There are four independent iron sites, denoted as Fe<sub>A</sub>, Fe<sub>B</sub>, Fe<sub>C</sub> and Fe<sub>D</sub>. Inset: Individual octahedron formed by one center iron atom and six surrounding oxygen atoms, representing the cation coordination. b-d, Optical images of  $\varepsilon$  - Fe<sub>2</sub>O<sub>3</sub> crystals grown on mica by CVD. Ultrathin crystals with a lateral size of many microns can be easily found in each batch of growth, as indicated by the red arrows. [Right plot]: Hysteresis loops acquired by MOKE at room temperature, obtained from crystals with varying thicknesses from 7.5 nm to 50.1 nm, demonstrating magnetic order with symmetric hysteresis and coercive fields  $\approx$  300 mT.

## **Results and Discussion**

Certain 2D materials exhibit intriguing properties such as valley polarization, ferroelectricity, superconductivity and charge-density waves. Many of these materials can be manually assembled into atomic-scale multilayer devices under ambient conditions, owing to their exceptional chemical stability. Efforts have been made to add a magnetic degree of freedom to these 2D materials via defects, but only local magnetism has been achieved. Only with the recent discoveries of 2D materials supporting intrinsic ferromagnetism have stacked spintronic devices become realistic. Assembling 2D multilayer devices with these ferromagnets under ambient conditions remains challenging due to their sensitivity to environmental degradation, and magnetic order at room temperature is rare in van der Waals materials. Here, we report the growth of air-stable ultra-thin epsilon-phase iron oxide crystals that exhibit magnetic order at room temperature. These crystals require no passivation and can be prepared in large quantity by cost-effective chemical vapor deposition (CVD). We find that the epsilon phase, which is energetically unfavorable and does not form in bulk, can be easily made in 2D down to a seven unit-cell thickness. Magneto-optical Kerr effect (MOKE) magnetometry of individual crystals shows that even at this ultrathin limit the epsilon phase exhibits robust magnetism with coercive fields of hundreds of mT. These measurements highlight the advantages of ultrathin iron oxide as a promising candidate towards air-stable 2D magnetism.

## References

[1] Yuan, J. et al., submitted (2018); arXiv:1805.10372