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Measuring Antiferromagnetism at the Angstrom Scale using 4D-STEM

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Antiferromagnets are a category of magnetic materials with near-zero net magnetization. New magnetic devices involving antiferromagnets may rely on manipulating patterns of magnetic moments such as domain walls or skyrmions, which have been imaged using techniques such as x-ray photoemission electron microscopy or differential phase contrast scanning transmission electron microscopy (DPC STEM) [1, 2]. DPC STEM with an atomically sharp probe and unit-cell averaging has been used to map the magnetic structure of an antiferromagnet in real space [3]. However, electron microscopy studies of antiferromagnets need to isolate and detect the relatively weak signal due to scattering from the magnetic structure and remain limited in signal-to-noise ratio. Here, we propose using antiferromagnetic reflections to perform phase-contrast imaging of magnetic structure at few-angstrom resolution. Such reflections are commonly measured in neutron scattering [4] and, more recently, have been detected in transmission electron diffraction, where they are around 10^4 times less intense than structural reflections [5]. In the regions of convergent beam electron diffraction (CBED) patterns where two disks overlap, interference between the two beams causes the intensity to vary sinusoidally with probe position [6]. Thus, a periodic magnetic structure can be visualized as lattice fringes.

We demonstrate our approach using the metallic antiferromagnet Fe_2As, which has a magnetic unit cell with dimensions $a \times a \times 2c$ (Fig. 1(a)). We view Fe_2As along the [100] direction, with the magnetic moments pointing in-plane. The effect of in-plane magnetic fields on the electron beam is described by the component of magnetic vector potential along the beam direction [7]. Thus, we calculate the magnetic vector potential of Fe_2As from density-functional theory and show that the field imparts a weak phase shift on the order of 10^{-4} radians per unit cell to the electron beam (Fig. 1(b)). Consistent with this periodic magnetic phase shift, in selected area electron diffraction patterns of [100] Fe_2As, we measure a reflection at (001/2). We thus collect a 4D-STEM data set from Fe_2As, choosing the convergence angle such that the (000) disk overlaps substantially with the magnetic {001/2} disks and slightly with the {001} disks (Fig. 2(a)), thus enabling the simultaneous detection of (001/2) and (001) lattice fringes. We integrate over a circular region within the (000) disk to produce a center of mass (COM) image (Fig. 2(b)) which shows (001/2) lattice fringes, consistent with the magnetic structure of Fe_2As. Integrating over an annular virtual aperture, we detect (001) lattice fringes (Fig. 2(c)).

Next, we verify our results with conventional multislice simulations modified to include the magnetic phase shift of Fe_2As in the transmission function. For thicknesses of 10-30 nm, multislice simulations predict (001/2) and (001) lattice fringes with similar amplitudes as in experiment. The positions of (001/2) lattice fringes and the direction of their COM shifts can be intuitively interpreted in terms of the positions of the magnetic moments in Fe_2As and the Lorentz force acting on the electron beam. We will also discuss the feasibility of this method of magnetic imaging for other antiferromagnets with magnetic Bragg reflections [8].

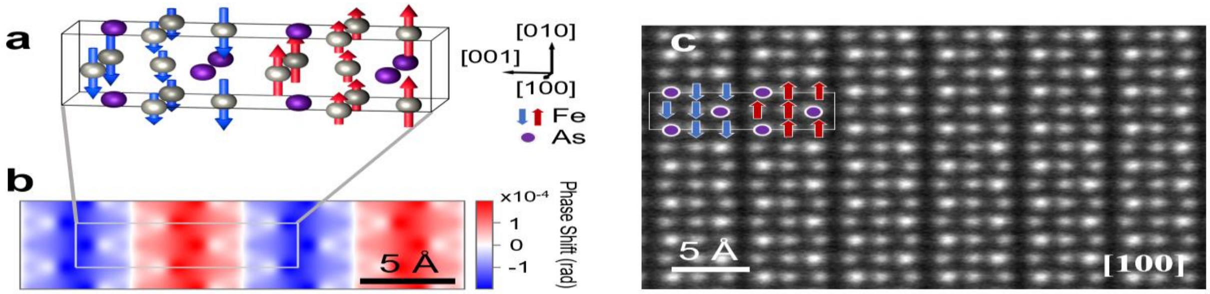

Figure 1. Structure of Fe_2As. (a) Magnetic unit cell of Fe_2As, with Fe atoms in gray and As atoms in purple. (b) Phase shift due to sample magnetic fields accumulated by an electron plane wave traveling through Fe_2As in the [100] direction for a thickness of one unit cell. The white box indicates the extent of the magnetic unit cell in (a). (c) ADF-STEM image of Fe_2As.

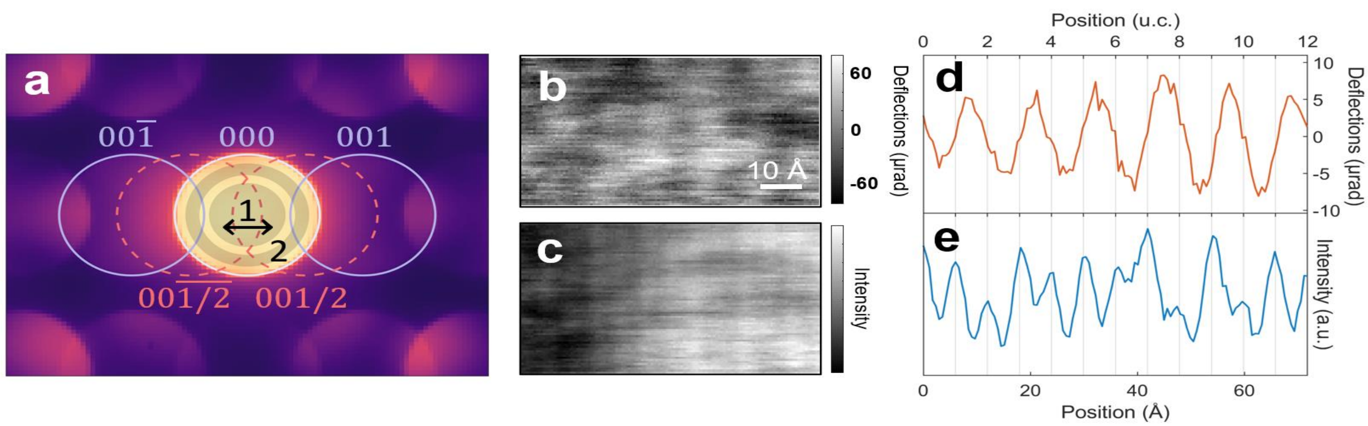

Figure 2. Simultaneously imaging atomic and magnetic structure of Fe_2As using 4D-STEM. (a) Experimental PACBED image of Fe_2As. Positions of {001} and {001/2} disks are labeled in blue and orange, respectively. A circular region (1) and an annular region (2) are indicated by shading. (b-c) Images of the same region of Fe_2As. (b) Center of mass image integrated over region 1, using the component in the [001] direction (arrow in (a)). (001/2) lattice fringes are visible. (c) Image integrated over region 2, showing (001) lattice fringes. (d-e) Background-subtracted line profiles of (b-c).

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