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Nanoscale Engineering of Magnetic Textures in the Layered Magnet CrSBr Using Electrons and Helium Ions

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The experimental observation of long-range magnetic order in van der Waals (vdW) layered materials in the single atomic layer limit has sparked great interest for the study of low-dimensional (2D) magnetism. [1] The addition of a spin in the 2D limit offers many exciting opportunities for atomistic manipulation of spin-related phenomena. This is of particular interest due to the strong correlation of vibrational, optical, delectronic and magnetic properties. Recently, CrSBr [2] has been rediscovered: it is a vdW layered magnet that exhibits air stability and therefore offers exciting opportunities for the controlled modification of its structure down to the atomic limit. CrSBr is also highly interesting since the material is a semiconductor with a bandgap of approximately 1.6 eV, a high Néel temperature (145 K) and offers strong light-matter interaction. The combination of all of these exciting aspects makes this material the ideal candidate for the study of magneto-transport and magneto-optical excitations. [3, 4]

Here, we use electrons in a scanning transmission electron microscope (STEM) and helium ions in a helium ion microscope (HIM) for the controlled structural modification of the crystal lattice of few-layer CrSBr. The motivation of these approaches is to artificially create novel magnetic textures in a low-dimensional magnetic material. For the exposure with electrons, we find that controlled irradiation at 200 keV induces a local phase transformation. To our surprise, the transformed material is also a 2D layered structure, but its layers are perpendicular to those in the original material. [5] This finding is corroborated by analyzing the intensity variation of Cr atom columns and local strain as the phase transformation occurs which manifests in the opening of a new vdW gap in the plane-view. Moreover, we theoretically investigate the electronic and magnetic properties of the phase transformed CrSBr using ab-initio and spin-wave theory. Our calculations suggest that the individual layers in the new phase are magnetic, and the material has an energy gap and a fully spin-polarized band structure. In contrast, to electrons, controlled exposure with helium ions creates defects with densities tunable by the ion dose. The disorder from ion irradiation in the CrSBr is further corroborated with Raman measurements that reveal phonon confinement effects.

From our work, we anticipate that the electron and ion beam induced manipulation of CrSBr can create atomically defined spin textures with specific properties that could be tailored towards applications for memory devices and artificially designed atomic scale many-body systems for quantum simulation. This further suggests that CrSBr and other air-stable magnets that can show similar engineerability of their crystal structures have the potential to become versatile quantum material platforms for the design, measurement, and application of nanoscale magnetic textures [6].

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