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WITHDRAWN-Decoding defect ordering from ADF-STEM images of van der Waals $CrGa_2Te_7$ ferromagnetic crystals using the unsupervised machine learning algorithm

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Two-dimensional (2D) van der Waals (vdW) ferromagnet materials have triggered significant research interest due to their potential applications in spintronic devices.1 $Cr_2Ge_2Te_6$ and Fe_4GeTe_2 are two examples of the successfully synthesized 2D-vdW ferromagnets.1,2 While there has been some investigation on the structure and properties of these novel crystals, little has been done to uncover the defects in this class of materials. Defects can play an important role in the manipulation and control of the macroscale magnetic behavior. In this study, we focused on the characterization of the 2D-vdW ferromagnet $CrGa_2Te_7$ using scanning/transmission electron microscopy (S/TEM). Figure 1 shows the atomic structure of $CrGa_2Te_7$. Using the selected area electron diffraction pattern (SAED), we observe the diffused superlattice spots surrounding the main Bragg peaks. While, the atomic resolution ADF-STEM images of the sample do not show an apparent defect ordering in the crystal, the Fourier transform of the STEM images exhibits similar superlattice spots Fig 1(b). Diffused scattering in electron diffraction patterns usually indicates disorder in the atomic arrangement or presence of short-range order in the crystal. While our preliminary studies indicate point defects exist in the form of Te vacancies in the acquired ADF-STEM, the nature of the disorder or short-range order is unclear in this crystal. To further understand the underlying disorder or short-range order in this crystal, we have used unsupervised machine learning methods, namely the principal component analysis (PCA), to help analyze the nature of the vacancies and their possible ordering from the STEM images. PCA is a mathematical tool for reducing the dimensionality of the multi-dimensional datasets, maximizing interpretability but mitigating the loss of information.3,4

Fig 1(c-d) shows the workflow and results of the PCA analysis on the atomic resolution STEM images. First, the atomic positions in the STEM images are quantified via a 2D-Gaussian fitting routine, and the image is further divided into image patches centered around those atom positions.5 A three-dimensional image stack is constructed through stacking the sub-images, as shown in Fig 1(c). The image stack dataset is then flattened into two dimensions and is fed into the PCA algorithm for dimensionality reduction. The dimensionality reduction results in principal components that contain the most important correlations in the data. Since the input dataset is centered on a single type of atomic column in the image, the principal components can help directly visualize the main features across the sub-images.6 We can directly interpret the first few components as the perfect lattice and the shift of atomic columns in two directions and later components indicate the intensity variations across the sub-images, as shown in Fig 1(d). This analytical method enables direct correlation of the short-range order in point vacancies observed in the 2D-vdW ferromagnet $CrGa_2Te_7$. This presentation further explores the origin of ordering arising from superlattice peaks in the crystal structure.

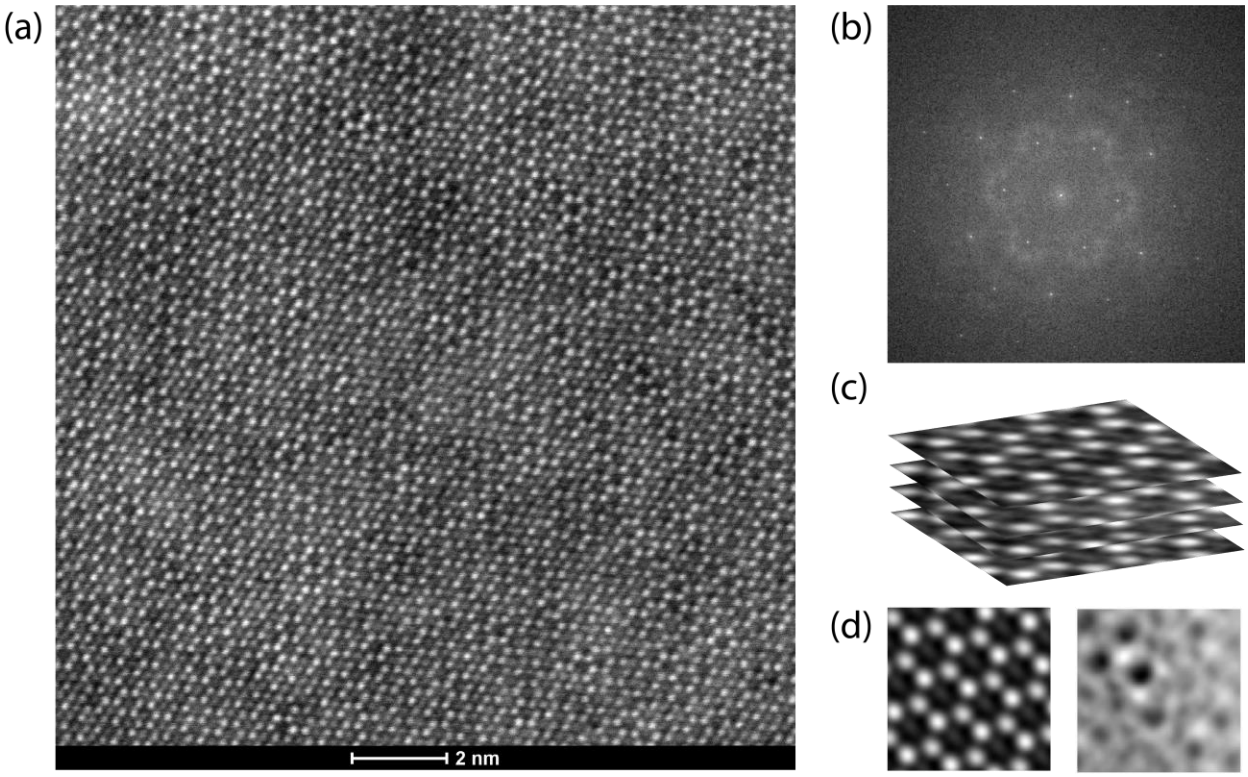

Figure 1. Figure 1: (a) ADF-STEM image of the structure of $CrGa_2Te_7$ taken from [001] zone axis. (b Fourier transform pattern of $CrGa_2Te_7$ ADF-STEM image taken from [001] zone axis. (c) an ADF-STEM image is converted into a three-dimensional image stack based on fitted atom positions for PCA. (d) the mean and one of the principal components extracted from PCA.

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