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Formation of Defects in MoS_2 during Data Acquisition of High-resolution Transmission Electron Microscopy

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Understanding electron-sample interactions in High Resolution Transmission Electron Microscopy (HRTEM) is a challenging task due to multiple complex processes occurring simultaneously over a short timescale. Ideally, we want to acquire images of the sample without modifying or destroying it. However, the interaction between the high energy electron beam and the sample necessary for image contrast, also results in high energy electrons transferring part of their energy to the sample through inelastic and elastic scattering. An important consequence of this interaction is knock-on damage, where there is direct momentum transfer from the electron to a target atom. A key quantity in this process is the threshold energy (T_D), which is the energy necessary to remove an atom from its lattice position in a crystalline sample. Here, we investigate the mechanism of knock-on damage, and the effect of electronic excitations on T_D, with a particular focus on molybdenum disulfide (MoS_2).

Recently it has been shown that when electronic excitations are present in a semiconducting material, formation of vacancies through ballistic energy transfer is possible at electron energies much lower than T_D for the ground state [1]. The lifetime of electronic excitations in MoS_2 are long enough to play an important role in the beam damage [2]. In principle, electronic excitations are delocalized, but once a knock-on event cause an atom to be ejected, a pre-existing electronic excitation can localize at the forming defect, as visualized by the electron density distribution in Figure 1. This can significantly lower T_D for a positively charged sample. Such charging is expected in thin films due to secondary electron emission by the electron beam [3], particularly for insulating or semiconducting samples, where charge redistribution is slow (for metallic specimens the electrical neutrality is restored spontaneously in 10^{-15} s). This effect is expected to increase as we increase the electron dose rate since the rate of hole generation becomes greater than hole neutralization. Understanding the role of charging effects gives us a better insight into the mechanisms of beam damage below the so-called knock-on threshold electron voltage (U_{th}) when the electron beam is not able to transfer enough kinetic energy to the recoil atom to remove it without assistance from electronic excitations.

In this work, we studied the defect production dynamics and beam damage in MoS_2 including effects of charging the sample on the T_D for sulfur atoms. A Kinetic Monte Carlo (KMC) algorithm is employed with varying sulfur threshold energies, accounting for their coordination, local geometry and charge. The results of this simulation allow computation of the displacement cross-section and reveal the dynamics and energetics of sulfur vacancy formation, which are compared quantitative measurements derived from experimental HRTEM data.

Obtaining a relationship between the displacement cross-section, beam energy, and electron dose rate is crucial to establishing optimal image acquisition parameters where beam damage is minimized without compromising on signal-to-noise ratio.

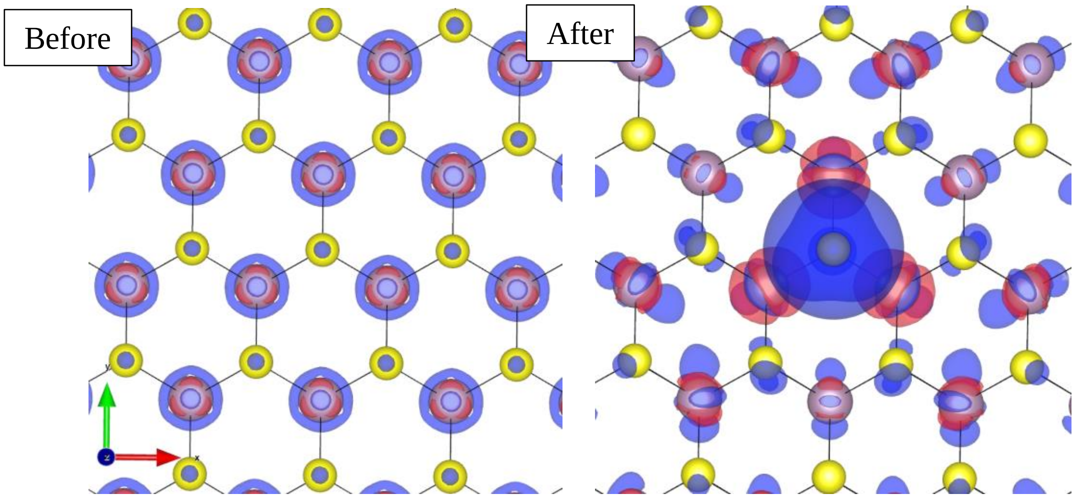

Figure 1. Electron density difference $\Delta\rho = \rho^{+} - \rho^{0}$(positive red, negative blue) between the positive charged and neutral system before and after pulling sulfur atom 7 Å away in the out of plane direction. The iso surface level in both plots is 0.004 e/Å^{3}.

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