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Atomistic Understanding of Damage and Beam-driven Dynamics in 2D Materials

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Transmission electron microscopy is a powerful probe of the chemical structure of materials. Due to efficient aberration correction, the capabilities of modern instruments have advanced so far that the information which can be collected is now mainly limited by irradiation effects. Knock-on damage due to electron backscattering affects all materials, but it is especially important for metals such as graphene, while ionization damage affects other kinds of materials [1]. Recent studies have also established the manipulation of covalently bound impurity atoms in graphene [2] and single-walled carbon nanotubes [3] via beam-induced out-of-plane dynamics as a novel application for focused electron irradiation.

From a theoretical point of view, molecular dynamics simulations have been vital for understanding the atomistic mechanisms of such beam-driven dynamics. However, while approximate models were successful for pure carbon materials, first principles simulations based on density functional theory molecular dynamics (DFT/MD) were found to be necessary to describe knock-on damage even qualitatively correctly in systems with charge transfer such as nitrogen-doped graphene [4] or hexagonal boron nitride [5]. Such modeling was also instrumental in revealing the mechanism and deterministic control of impurity movement in graphene triggered by electron impacts on one of its less strongly bound carbon neighbors [6].

Precision measurements of knock-on damage have established that at lower electron energies below the static displacement threshold, the motion of the atoms due to lattice vibrations determine the cross section [7, 8]. This "Doppler enhancement" mechanism activates processes at a specific electron energy that would otherwise be suppressed due to momentum conservation. Up to date, vibration-assisted knock-on damage has theoretically only been treated in the out-of-plane direction where the momentum transfer is most efficient. However, vibrational velocities and ejections into all directions need to be included for a complete physical description.

We have recently developed a fully three-dimensional theory of electron knock-on damage for non-static target atoms. This formalism provides a correction to the cross section values obtained from the earlier approximate theory that bring experiment and theory into a better agreement, and allows more complicated dynamics to be finally quantified and understood. We explore the implications of this formalism for displacements from pristine graphene [8] as well as beam-induced dynamics at previously intractable graphene impurity sites [9]. Although we here only demonstrate our model for this specific system where the most high-quality quantitative data exists, the theoretical framework is of general validity for the quantitative description of elastic electron irradiation effects in any material.

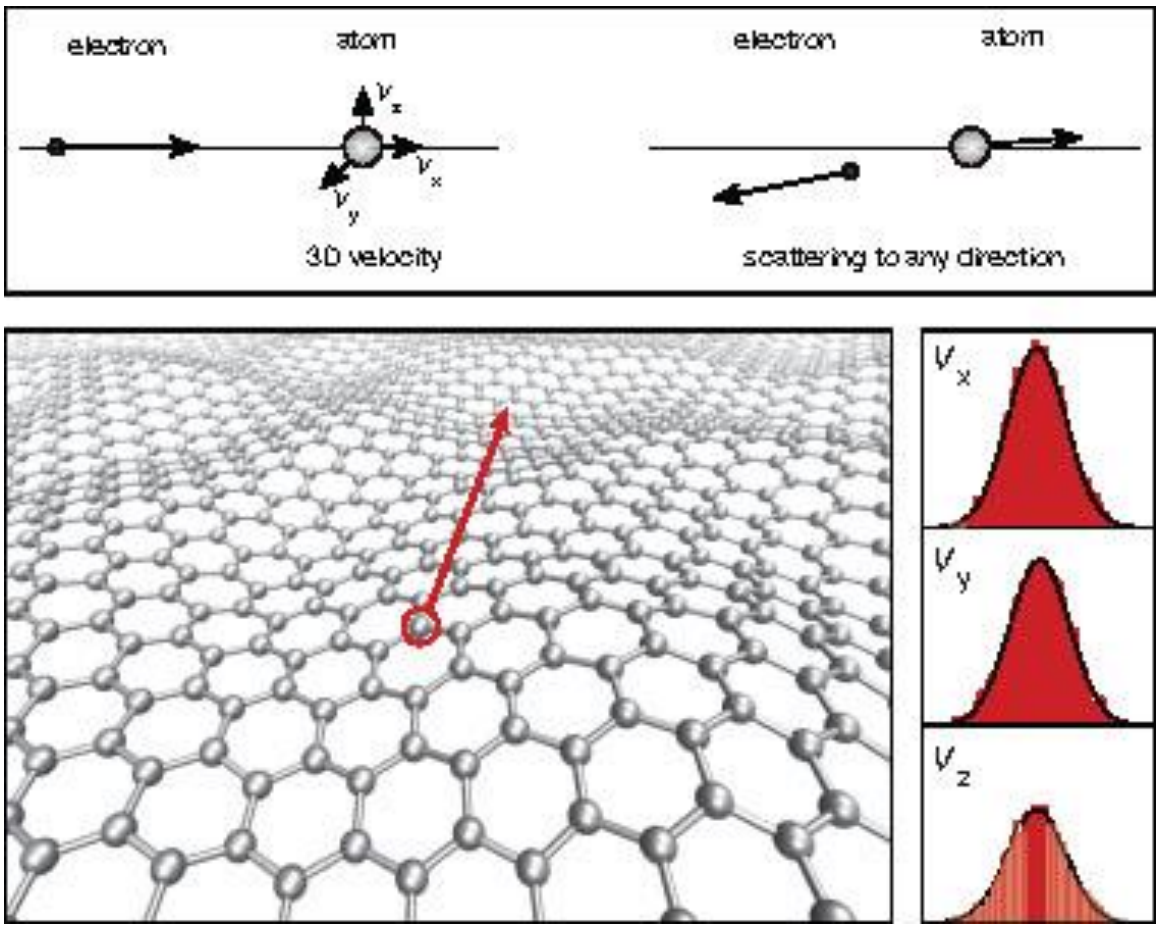

Figure 1. Atoms in real materials are in constant motion due to thermal and quantum zero-point vibrations. The velocity of an atom at the moment a probe electron scatters from its nucleus influences how much energy the atom can receive in a momentum-conserving collision. We have now treated this scattering problem in its full three-dimensional complexity, accounting for the velocity components of the atom via a statistical treatment and describing its displacement into any direction.

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