Isotope analysis in the transmission electron microscope

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Recent advances in aberration-corrected scanning transmission electron microscopy (STEM) and electron energy loss spectroscopy have allowed the identity and bonding of individual atoms to be determined in two-dimensional materials such as graphene. However, discerning the isotopes of a particular element has not yet been possible. Isotopes of carbon, in particular, have proven invaluable for archeology through radiocarbon dating, and in materials science, isotope labeling is a powerful tool for understanding chemical vapor deposition.

Here we differentiate between two isotopes of the same element by quantifying how likely the energetic imaging electrons are to eject atoms. Our technique rests on a crucial difference between electrons and photons when used as a microscopy probe: due to their finite mass, electrons can transfer significant amounts of momentum. When the transferred energy is comparable to the energy required to eject an atom from the material when probing pristine or doped single-layer graphene with 60-100 keV electrons, atomic vibrations need to be included for the correct description of the process. We measure the displacement probability in graphene grown from either ^{12}C or ^{13}C and describe the process using a quantum mechanical model of lattice vibrations coupled with density functional theory simulations. We then test our spatial resolution in a mixed sample by ejecting individual atoms from nanoscale areas spanning an interface region that is far from atomically sharp, mappi ng the isotope concentration with a precision better than 20%.

Although these results were achieved with graphene, our technique should work for any low-dimensional material, including hexagonal boron nitride and transition metal dichalcogenides such as MoS_2. This could potentially extend to van der Waals heterostructures of a few layers or other thin crystalline materials, provided a difference in the displacement probability of an atomic species can be uniquely determined.

[1] Susi, Hofer et al., *Nat. Commun.* **7**, 13040 (2016).