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Beyond vibrational spectroscopy: hunting the signature of elusive quasiparticles with monochromated STEM-EELS

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Engineering the structural or chemical architecture of functional materials at the nano or even atomic level enables emergent properties that rely on the interplay between fundamental properties of matter such as charge, local atomic-scale chemistry, and recently spin. Methodological developments in high-resolution energy-loss spectroscopy have made it possible to study these phenomena at near-atomic scale in the scanning transmission electron microscope. For instance, recent results exploiting the energy loss near-edge structure, which arises from core-level excitations, pave the way towards mapping electronic orbitals, such as π* and σ* states in epitaxial graphene, in real space. [1] In the low energy loss regime, a dark field EELS detection geometry is used to reveal atomic-scale variations in acoustic and optical phonon excitations in materials, while single-atom impurities can even be shown to have a characteristic vibrational response [2]. In a system consisting of Bi_2Se_3 films grown by chemical vapor deposition on epitaxial graphene, observations using these techniques highlight the interplay between the various phonon modes and the Dirac plasmons in the topological insulator Bi_2Se_3, in addition to a further direct interrogation of the chemical bonds.

Finally, the prospects for observing the excitation of spin waves, or magnons, arising from the collective excitation of the electrons' spin in a lattice and which qualitatively occupy the same energy range as phonons, are explored through preliminary experiments and the development of a theoretical framework based on the diffuse scattering of electrons due to magnons. [3,4]

[1] M. Bugnet *et al.*, *Phys. Rev. Lett.* **128** (2022), 116401
[2] F. S. Hage *et al.*, *Science* **367** (2020), aba1136
[3] K. Lyon *et al., Phys. Rev. B* **104** (2021), 214418
[4] J.A. Castellanos-Reyes *et al.*, *Phys. Rev. B* **108** (2023), 134435