Extending the Capabilities of Energy- and Momentum-Resolved STEM

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Although vibrational electron energy-loss spectroscopy had already been demonstrated in the 1960s [1], the possibility to do so in an actual electron microscope with a sub-nm probe was only established in 2014 [2]. While specialized optical vibrational spectroscopy techniques have attained sub-nm resolution by now [3], the electron microscope is still unique in being the only instrument capable of atomically resolving bulk-like specimens not just surfaces. Recently, we have demonstrated atomically-resolved phonon EELS of extended defects – in excellent agreement with calculations [4]. This study has shown that grain boundaries are not only barriers to phonon transport but can also support localized phonon modes and thus potentially act as waveguides for phonons in future devices. Another promising application is momentum-resolved vibrational EELS first demonstrated by Hage et al. in 2018 [5]. Here, we experimentally map phonon dispersion surfaces (in 2D) from momentum-resolved vibrational EELS [6] combined with quantitative comparison with suitable theory [7]. The technique could be used to visualize anisotropies in phonon transport, e.g. in steady states, or to investigate mode softening.

The utilization of vibrational EELS to probe biologically-relevant materials without damage (in aloof geometry) was shown by Rez et al. in 2016 [8]. We have advanced this technique and will demonstrate the first combination of vibrational spectroscopy and cryo-transfer to investigate beam damage in vitrified samples [9]. We observe that hydrogen atoms are break off first and we also observe the partial reforming of bonds.

Lastly, we will demonstrate how to energy-resolve 4D-STEM at the atomic scale, leading to rich 5-dimensional data sets. The practical implementation using the registration of 4D-STEM [10] and first results for core-loss [11] as well as vibrational energies will be shown and potential applications discussed.

[1] J. Geiger, *Z. Phys.* **181**,413 (1964).
[2] O.L. Krivanek et al., *Nature* **514**, 209 (2014).
[3] Y. Zhang, *Natl. Sci. Rev.* **6**, 1169 (2019).
[4] B. Haas et al., *Nano Lett.* **23**, 5975 (2023).
[5] F.S. Hage et al., *Sci. Adv.* **4**, eaar7495 (2018).
[6] B. Haas et al., *Microsc. Microanal.* **29**, 356 (2023).
[7] P. Rez and A. Singh, *Ultramicroscopy* **220**, 113162 (2021).
[8] P. Rez et al., *Nature Commun.* **7**, 10945, (2016).
[9] B. Haas et al., *MC2023 Proceedings*, IM4.004, 501 (2023).
[10] C.M. O'Leary & B. Haas et al., *Microsc. Microanal.* **28**, 1417 (2022).
[11] B. Haas and C.T. Koch, *Microsc. Microanal.* **28**, 406 (2023).