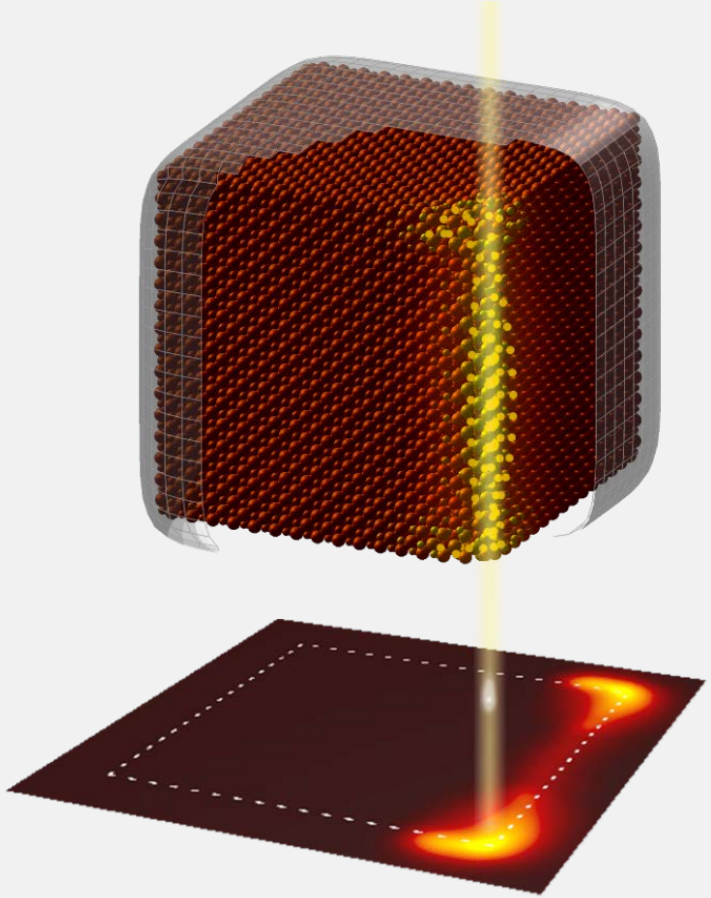
Vibrational bulk and surface losses of swift electrons in ionic nanostructures



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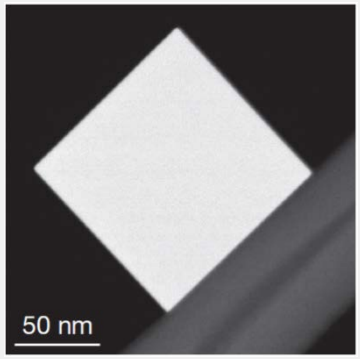
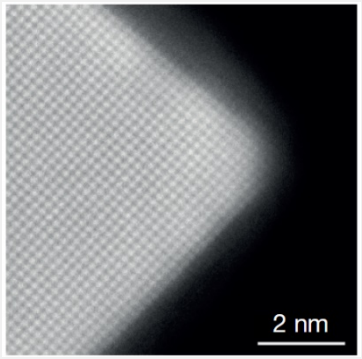
Rutgers University, New Jersey
McMaster University, Hamilton





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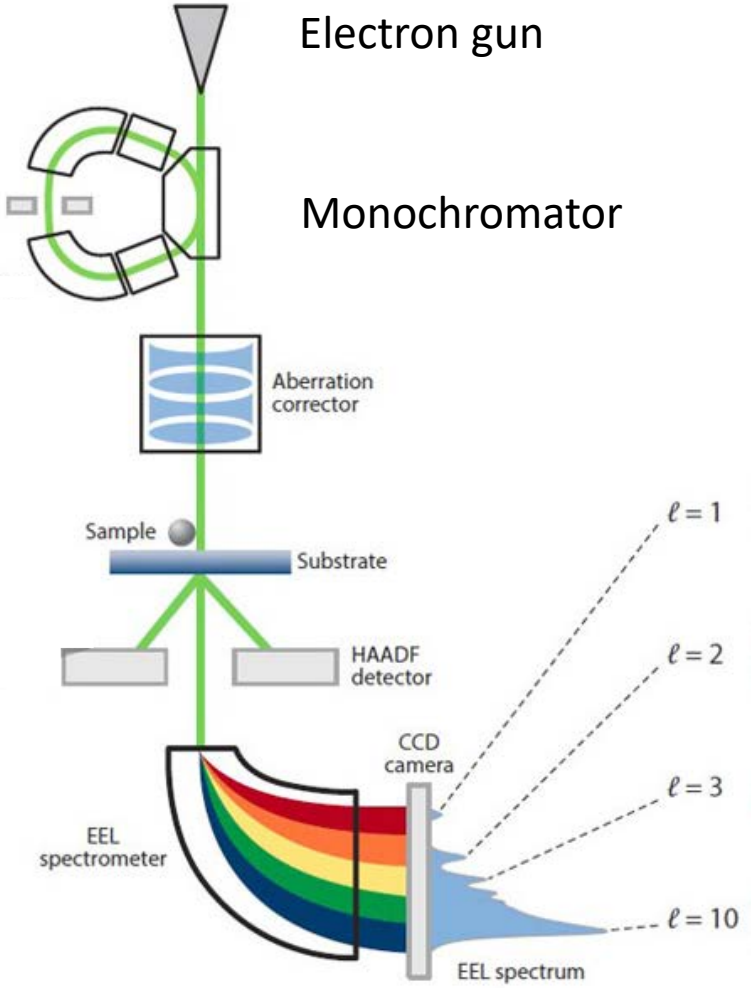
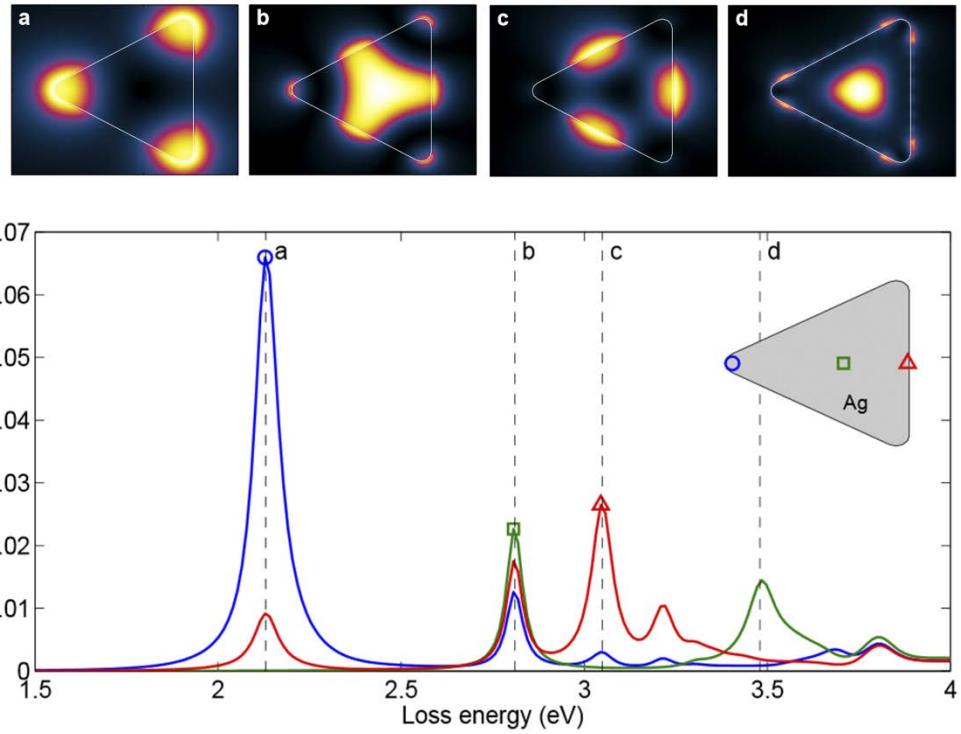
http://physik.uni-graz.at/~atr

Nanolight 2018
15. March 2018, Benasque

Plasmonic EELS A versatile measurement tool

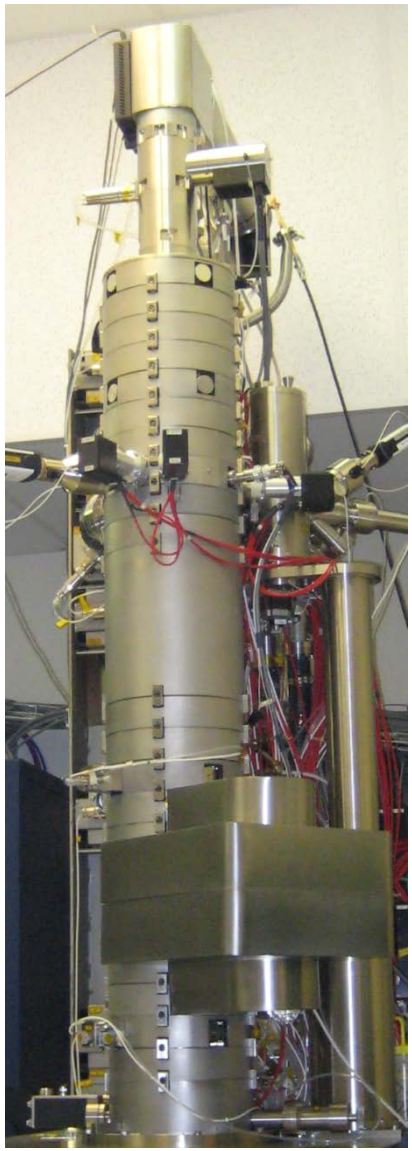


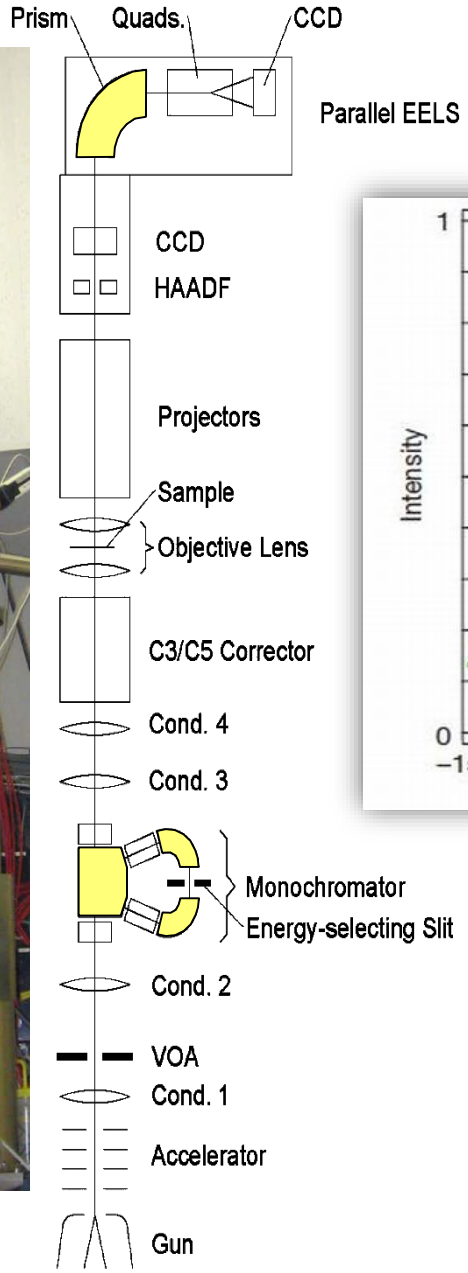


- based on electron microscopy
- nm spatial resolution
- sub-eV energy resolution
- breaks optical selection rules
  allows detection of optically dark states

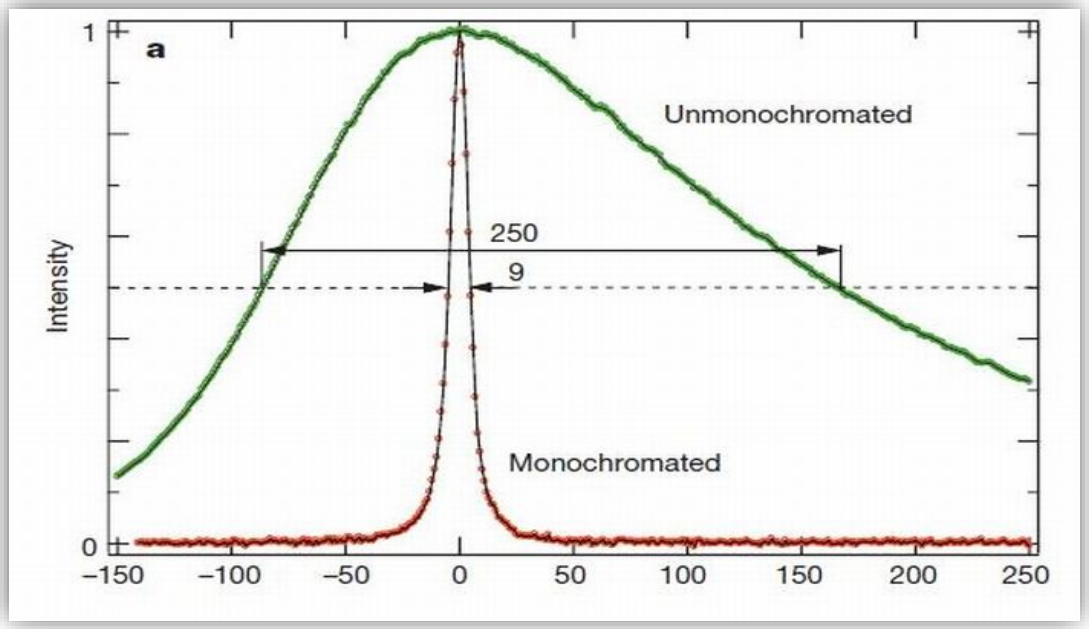
Annual Review of Physical Chemistry 67, 331 (2016)

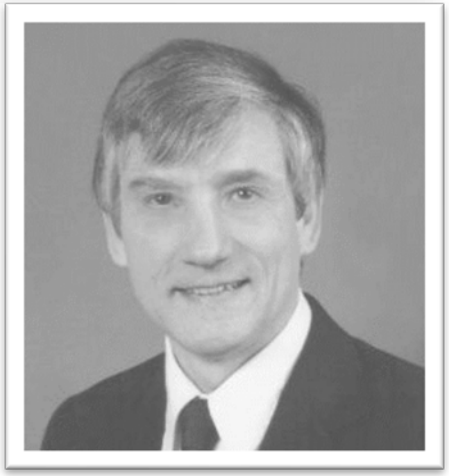
Bosman et al., Nanotechnology **18**, 165505 (2007); Nelayah et al., Nature Phys. **3**, 348 (2007).





O.Krivanek *et al.* Nature 514, 209 (2014).





Collaboration with Maureen Lagos and Phil Batson, Rutgers Univ.

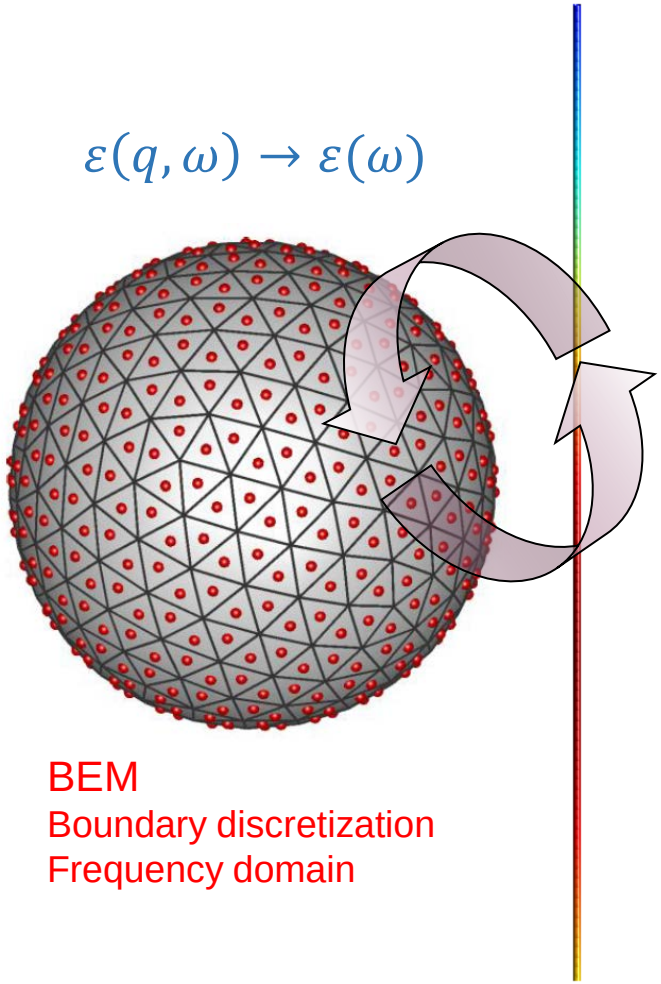
Boundary element method

Discretize boundary rather than volume

works only for bodies with homogeneous material properties (!!!)

Garcia de Abajo and Howie, Phys. Rev. B 65, 115418 (2002).

$\varepsilon(q,\omega) \rightarrow \varepsilon(\omega)$



BEM

Boundary discretization

Frequency domain

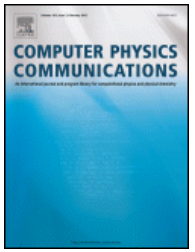
Electrons excite surface plasmons and lose energy

$$P(\hbar\omega) \propto \int_{-\infty}^{\infty} \mathrm{Re}[\boldsymbol{J}_{\mathrm{el}}^{*}(\boldsymbol{r},\omega) \cdot \boldsymbol{E}_{\mathrm{ind}}(\boldsymbol{r},\omega)]\, dz$$

MNPBEM – A Matlab toolbox for plasmonics

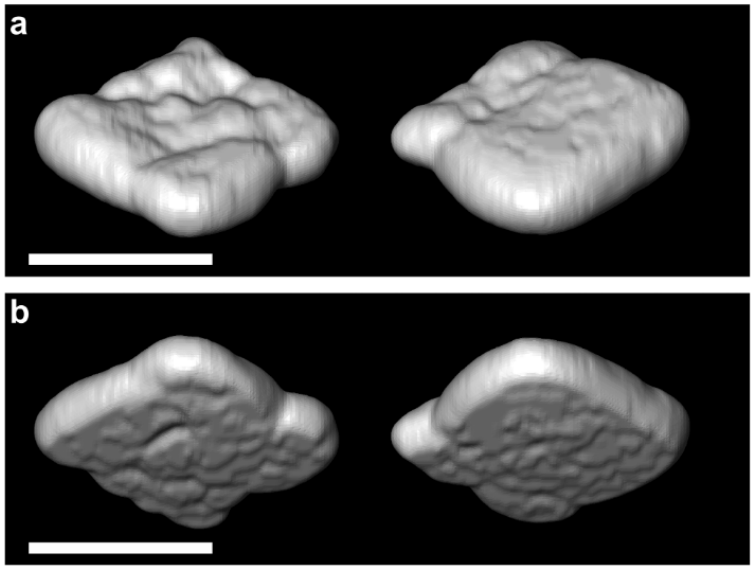
Hohenester, Comp. Phys. Commun., 185, 1177 (2014).

Hohenester and Trügler, Comp. Phys. Commun., 183, 370 (2012).



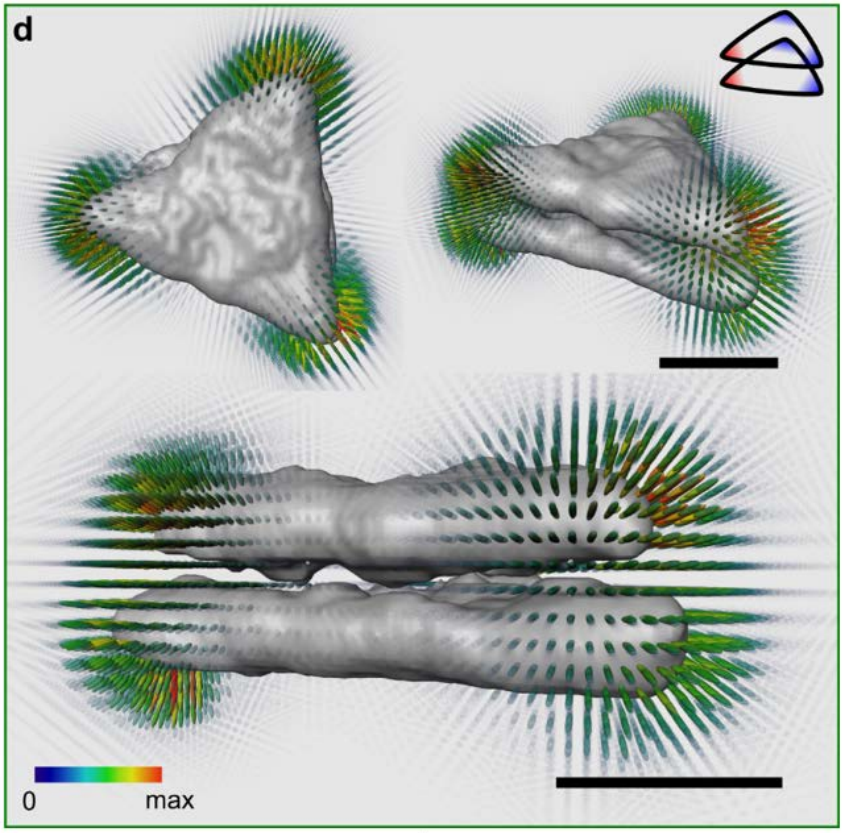
EELS tomography

3D reconstruction

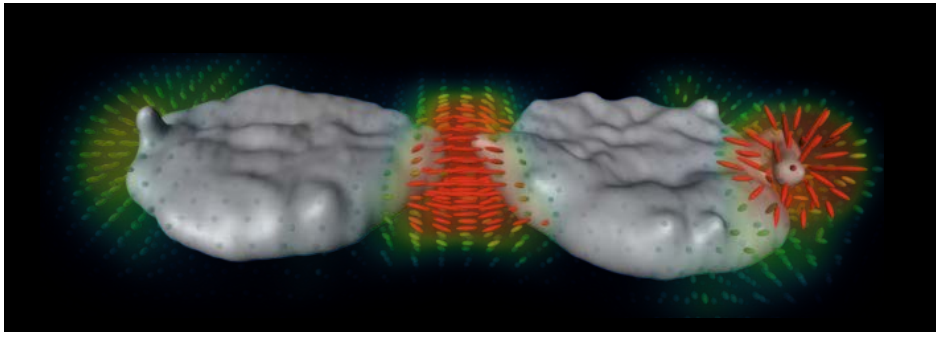


HAADF tilt series

Reconstructed 3D geometry



Tomographic reconstruction of plasmonic fields



Hörl et al., Nature Commun. 8, 37 (2017); Haberfehlner et al., Nano Letters (2017).

Ionic crystals

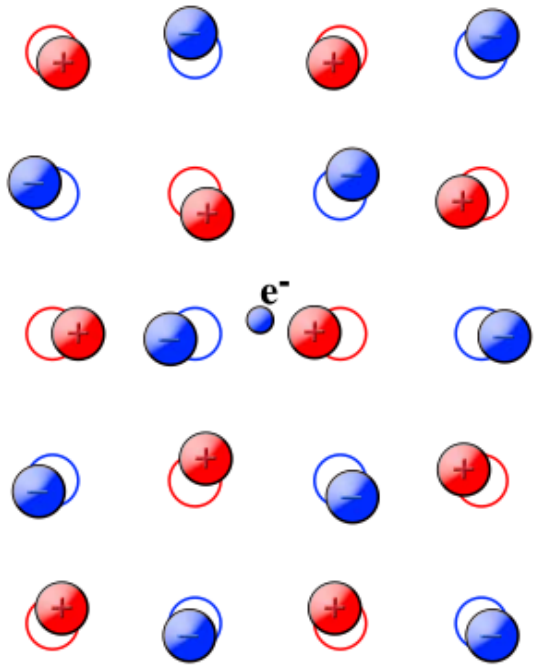
Surface phonon polaritons (10 – 100 meV) rather than surface plasmons (1 eV)

$$\varepsilon_{\text{ion}}(\omega) = 1 + \frac{\omega_{LO}^2 - \omega_{TO}^2}{\omega_{TO}^2 - \omega(\omega + i\gamma)}, \qquad \varepsilon_{\text{metal}}(\omega) = \varepsilon_\infty - \frac{\omega_{\text{pl}}^2}{\omega(\omega + i\gamma)}$$

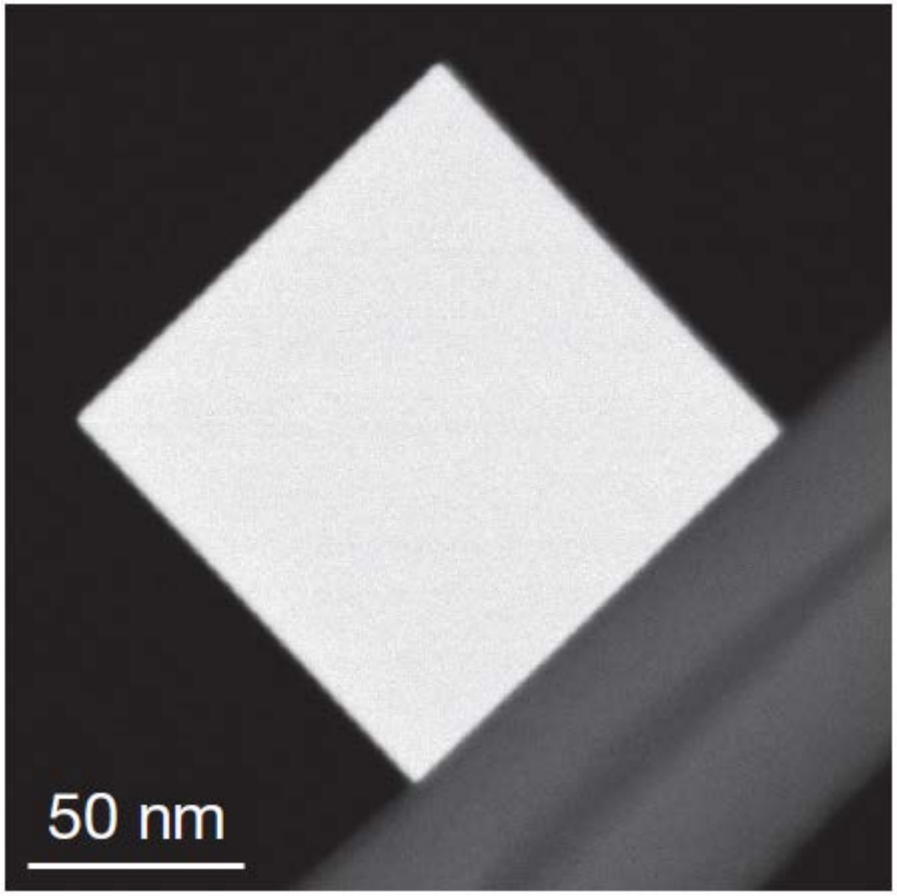
This talk :

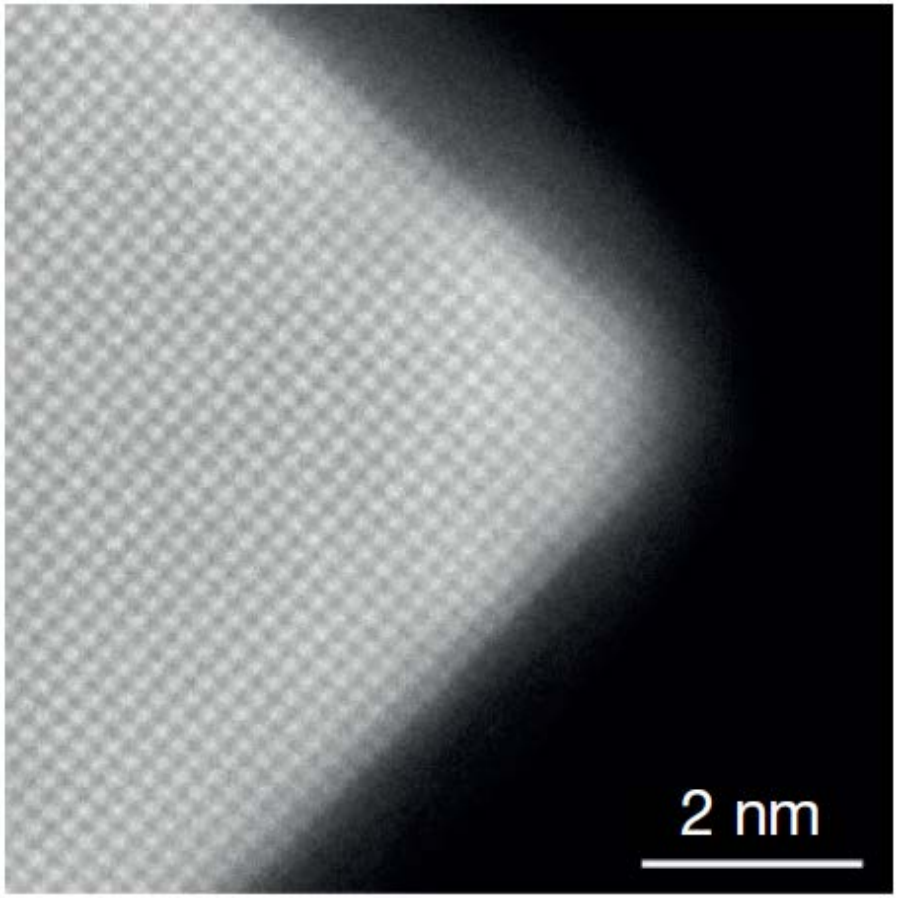
- ❖ EELS with high <10 meV energy resolution
- ❖ Molecular dynamics description

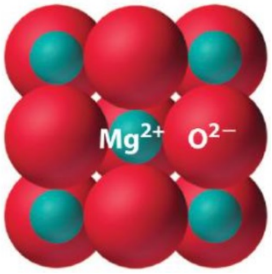
M. Lagos, A. Trügler, et al., Nature 543, 533 (2017)
M. Lagos, A. Trügler, et al., Microscopy 1-11 (2018)
U. Hohenester, A. Trügler, et al., submitted (2018)



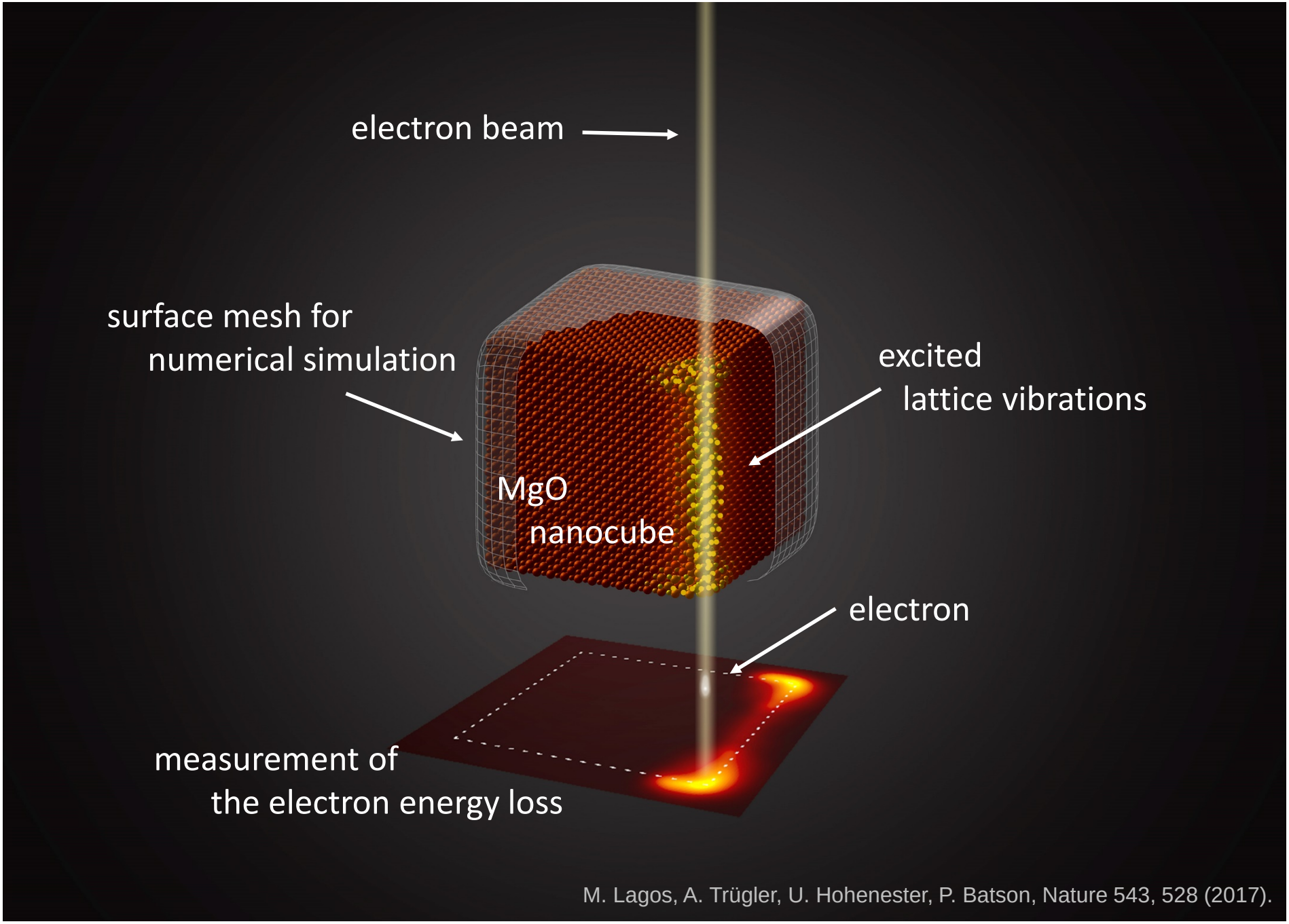
MgO cube



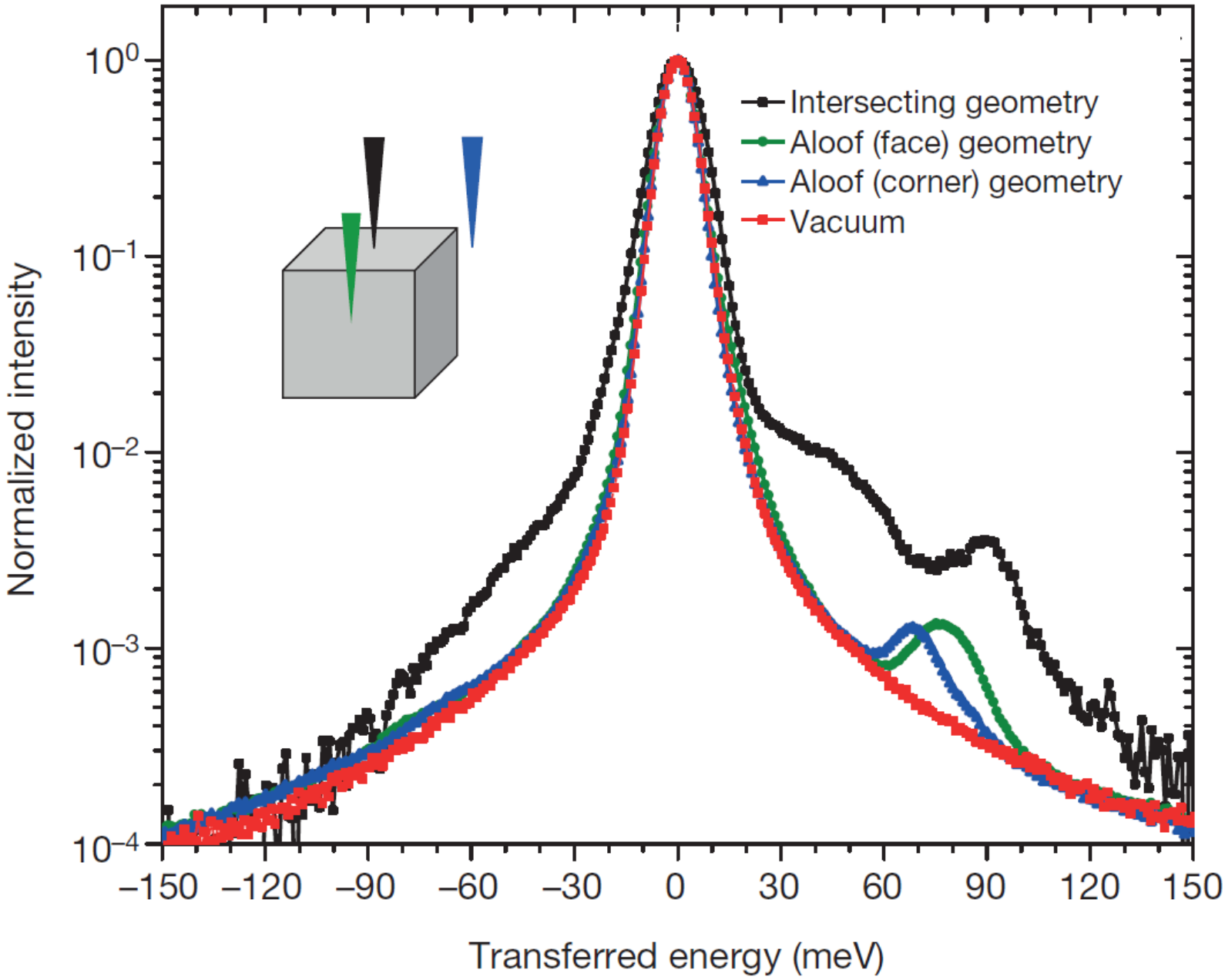




M. Lagos, A. Trügler, U. Hohenester, P. Batson, Nature 543, 528 (2017).



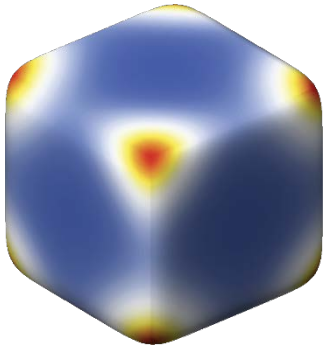
M. Lagos, A. Trügler, U. Hohenester, P. Batson, Nature 543, 528 (2017).



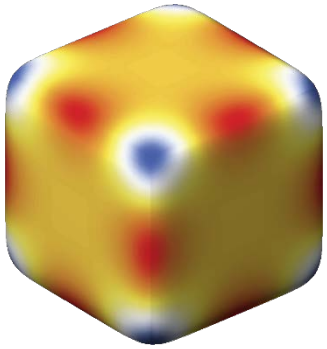
M. Lagos, A. Trügler, U. Hohenester, P. Batson, Nature 543, 528 (2017).

M. Lagos, A. Trügler, U. Hohenester, P. Batson, Nature 543, 528 (2017).

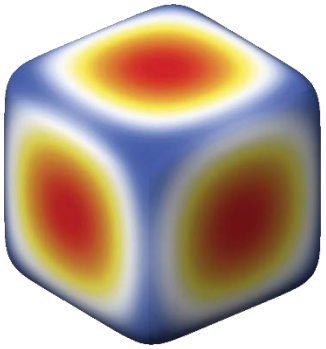
Phonon eigenmodes



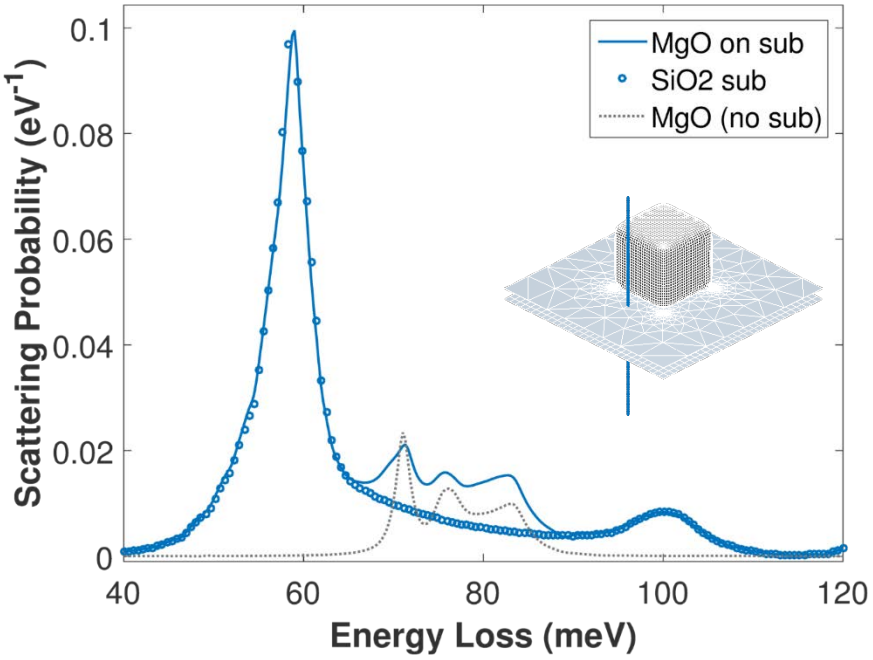
corner mode

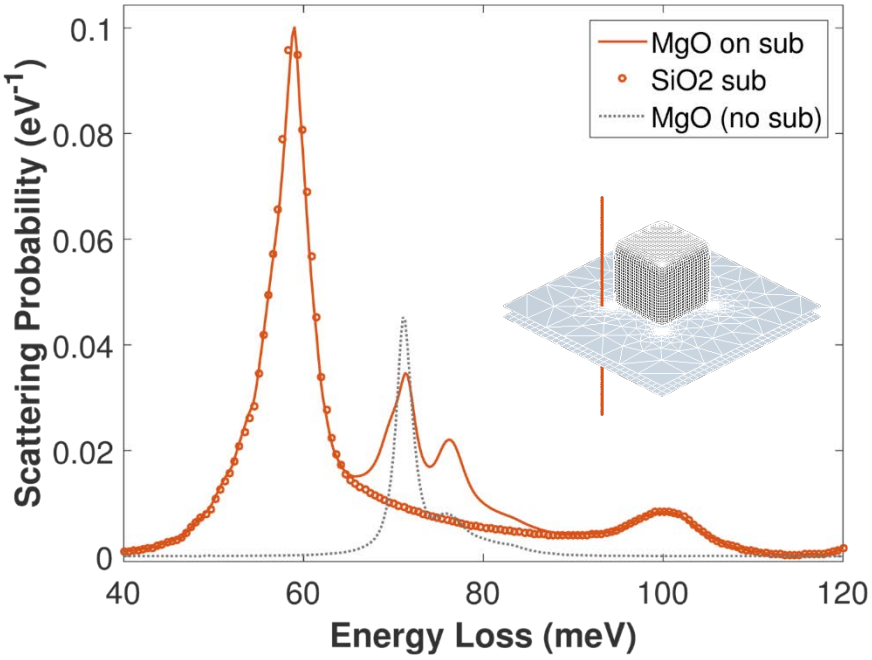


edge mode



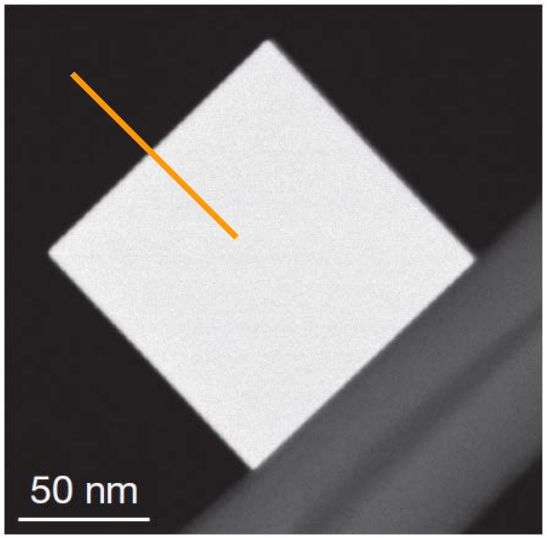
face mode

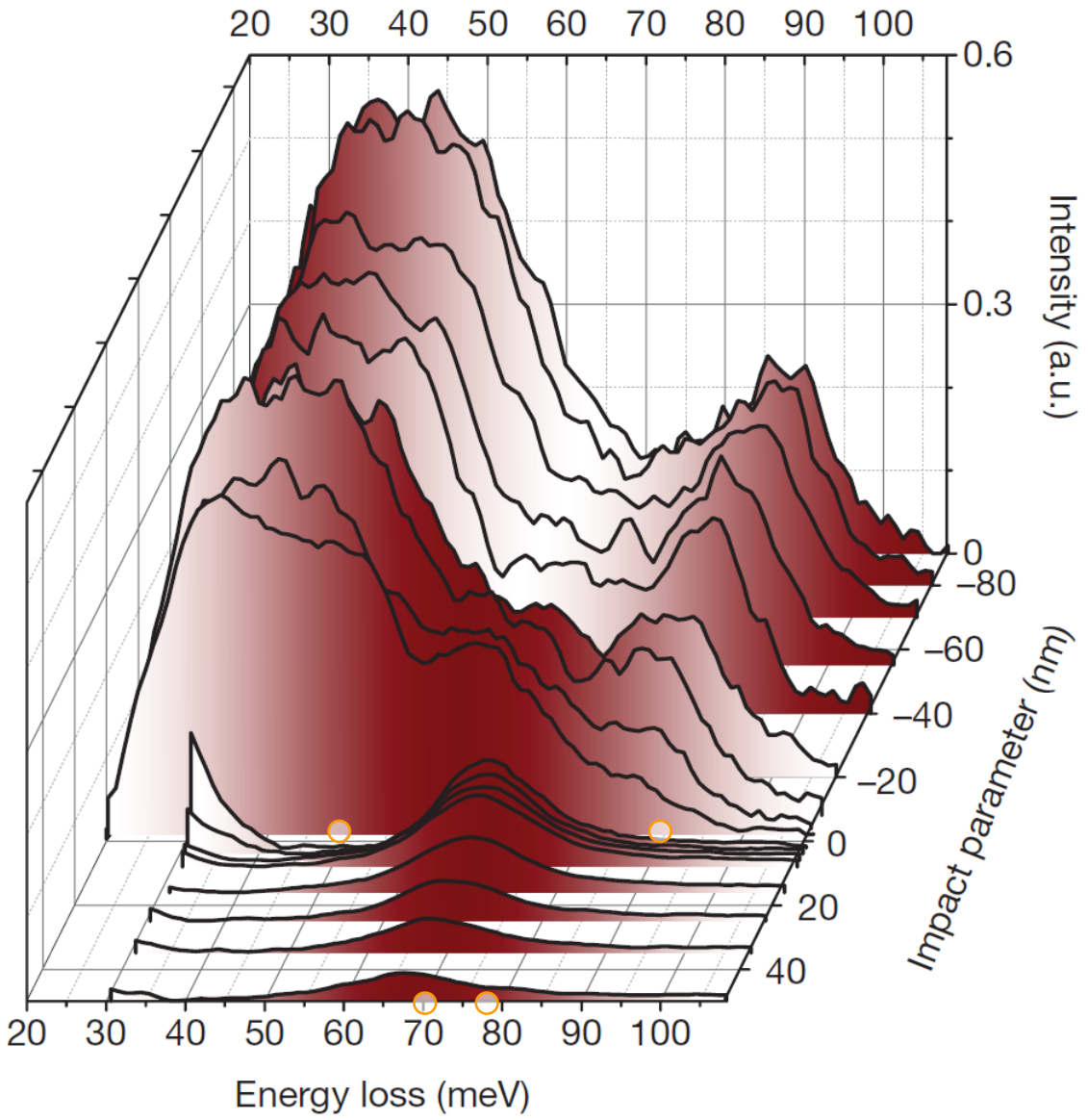




M. Lagos, A. Trügler, et al., Microscopy 1-11 (2018)

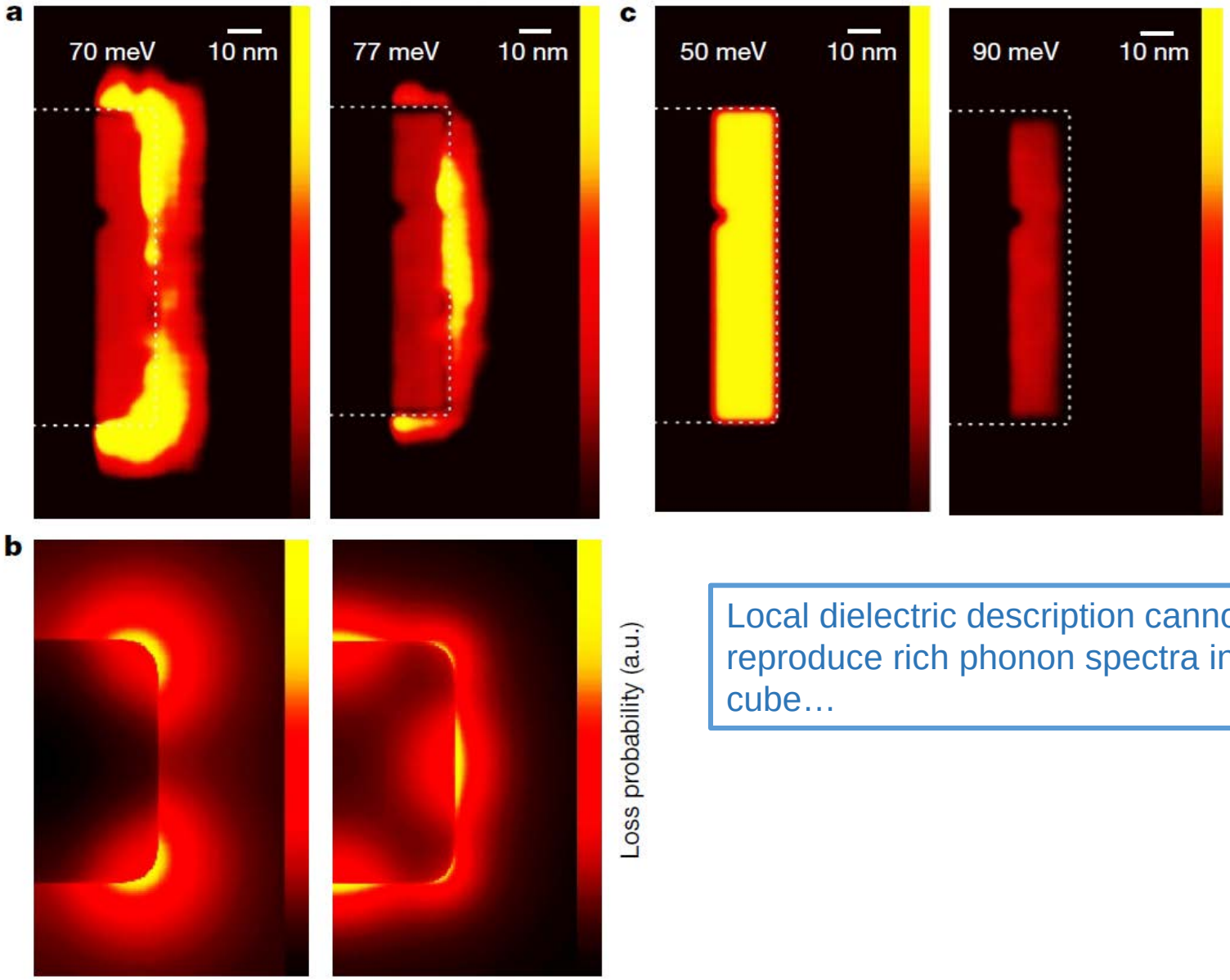
MgO phonon polaritons





M. Lagos, A. Trügler, U. Hohenester, P. Batson, Nature 543, 528 (2017).

Results Intensity maps (experiment & theory)



Local dielectric description cannot reproduce rich phonon spectra inside cube…

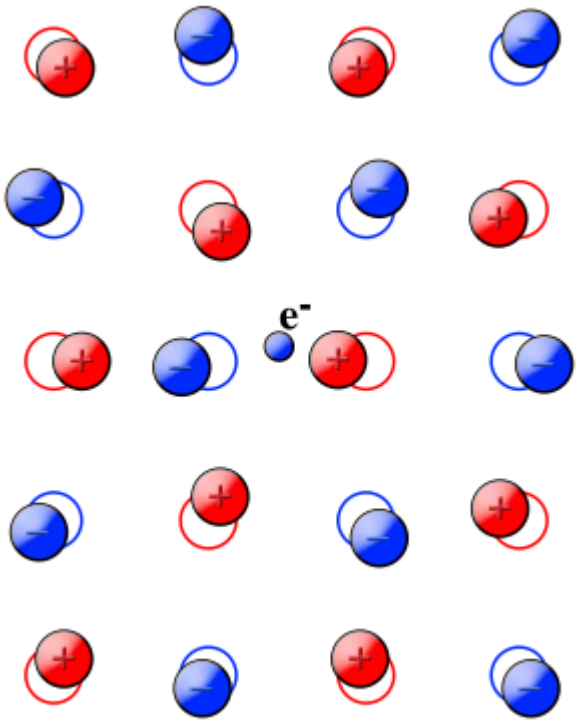
M. Lagos, A. Trügler, U. Hohenester, P. Batson, Nature 543, 528 (2017).

Molecular dynamics simulations

Newton‘s equations of motion for ion dynamics

long-range Coulomb forces (fast multipoles) and short-range interatomic forces

$$M_j \ddot{\boldsymbol{R}}_j + \nabla_{\boldsymbol{R}_j} \sum_{j'} V_{jj'}(\boldsymbol{R}_j - \boldsymbol{R}_{j'}) = eZ_j \, \boldsymbol{E}_{\mathrm{el}}(\boldsymbol{R}_j, t)$$



Chalopin et al., Appl. Phys. Lett. 100, 241904 (2012).

Molecular dynamics simulations

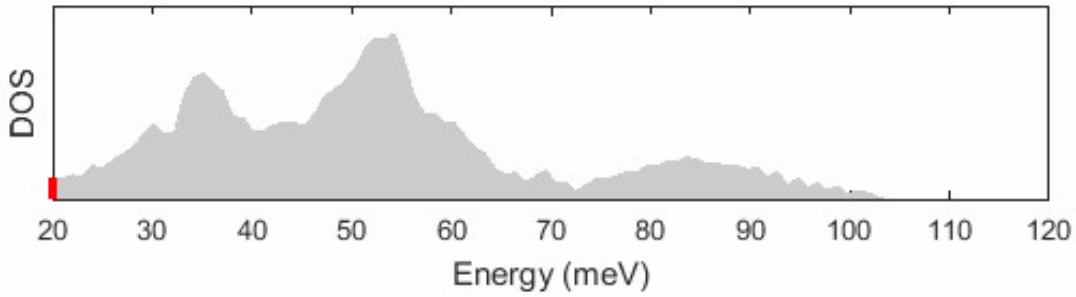
Electron energy loss probability

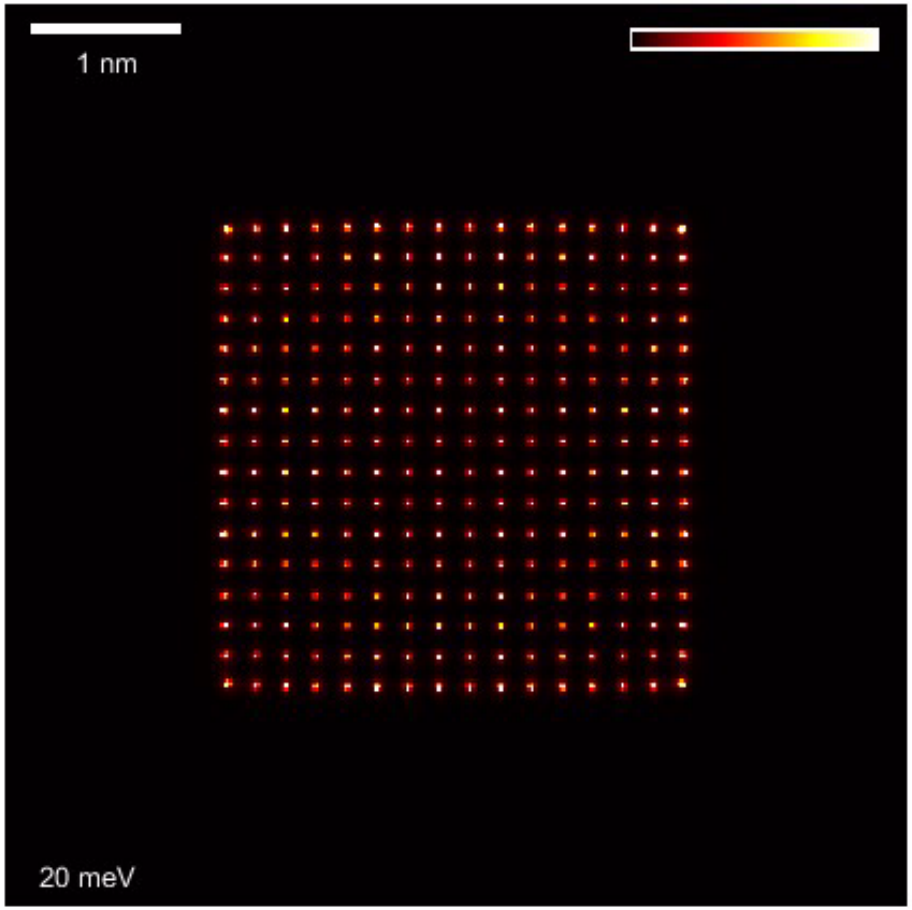
$$P(\hbar\omega) = \frac{1}{\pi\hbar\omega} \int \mathrm{Re}\left[\boldsymbol{E}^*_{\mathrm{el}}(\boldsymbol{r},\omega) \cdot \boldsymbol{J}_{\mathrm{ind}}(\boldsymbol{r},\omega)\right] \, d^3r$$

Ionic polarization current

$$\boldsymbol{J}_{\mathrm{ind}}(\boldsymbol{r},t) = \sum_j \dot{\boldsymbol{R}}_j \, eZ_k \delta(\boldsymbol{r} - \boldsymbol{R}_j)$$

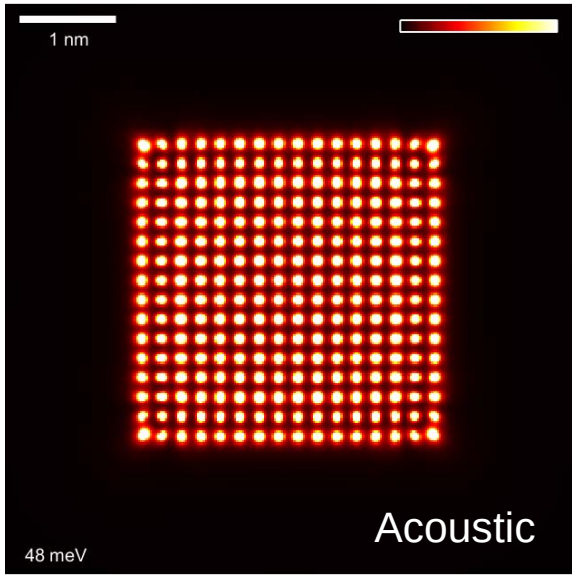
U. Hohenester, A. Trügler et al., submitted (2018).

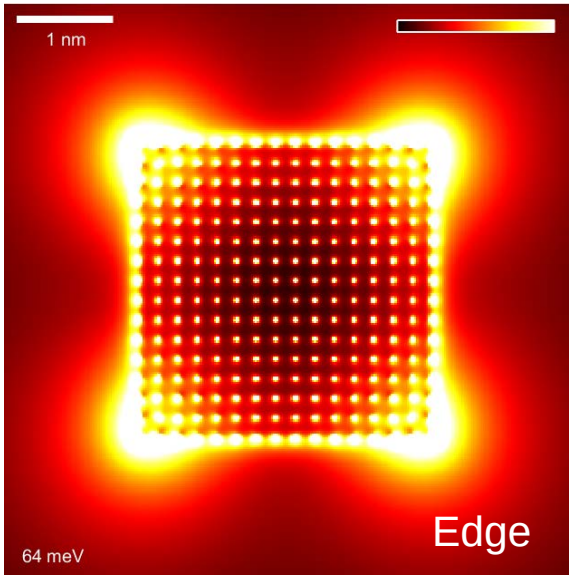


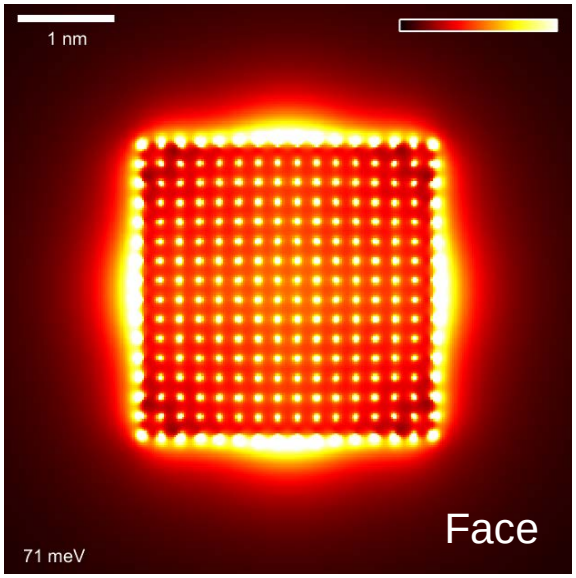


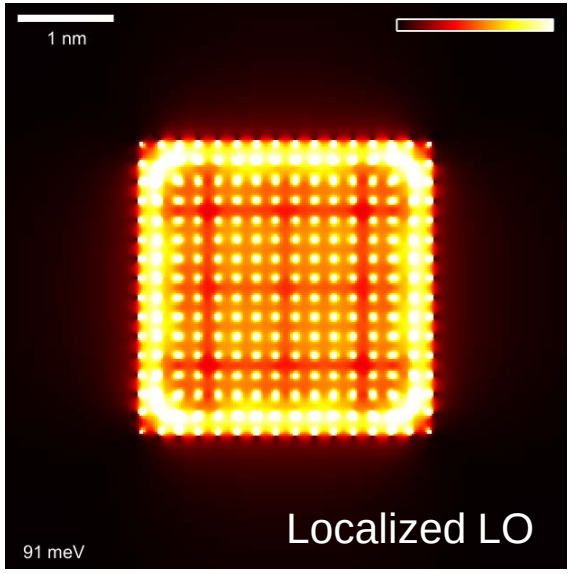
U. Hohenester, A. Trügler et al., submitted (2018).

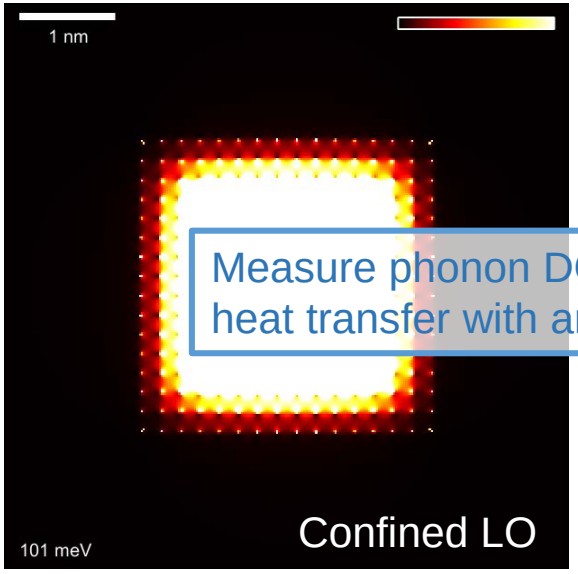
Molecular dynamics







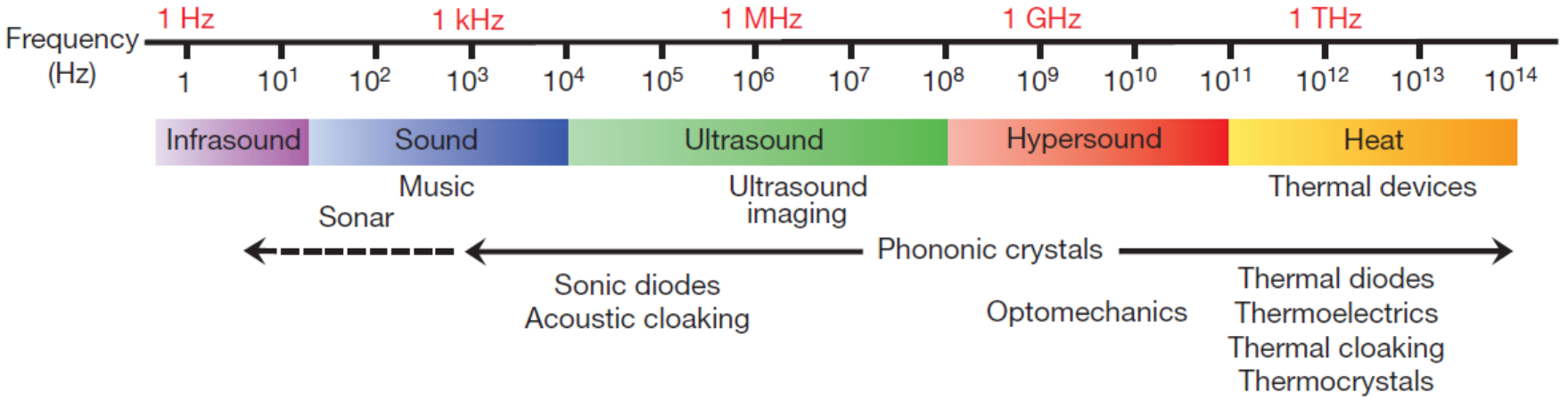




Measure phonon DOS, localized phonons, nearfield heat transfer with an electron microscope !

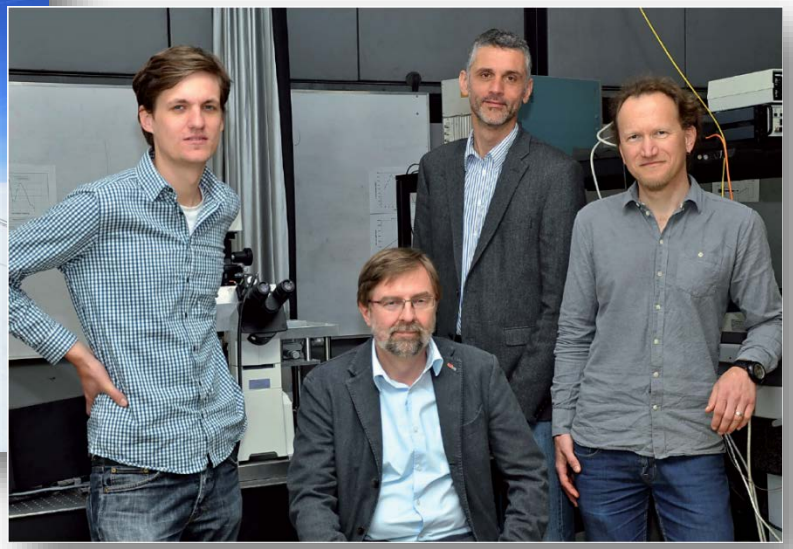
Summary and perspective

- Measurement of localized surface and bulk phonons at single nanoparticles
- Local limit ($q \longrightarrow 0$) works fine for non-penetrating electron beams
- Need to go beyond local description to include rich internal phonon spectra



M.. Maldovan, Sound and heat revolutions in phononics, Nature 503, 209 (2013).

Acknowledgements



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Franz Schmidt, Georg Haberfehlner, Gerald Kothleitner, Ferdinand Hofer, Harald Ditlbacher, Andreas Hohenau, Joachim Krenn

Maureen Lagos, Phil Batson (Rutgers and McMaster University)

FWF Der Wissenschaftsfonds.