Inelastic scattering -I

Introduction

Cécile Hébert IPHYS-LSME

Layout

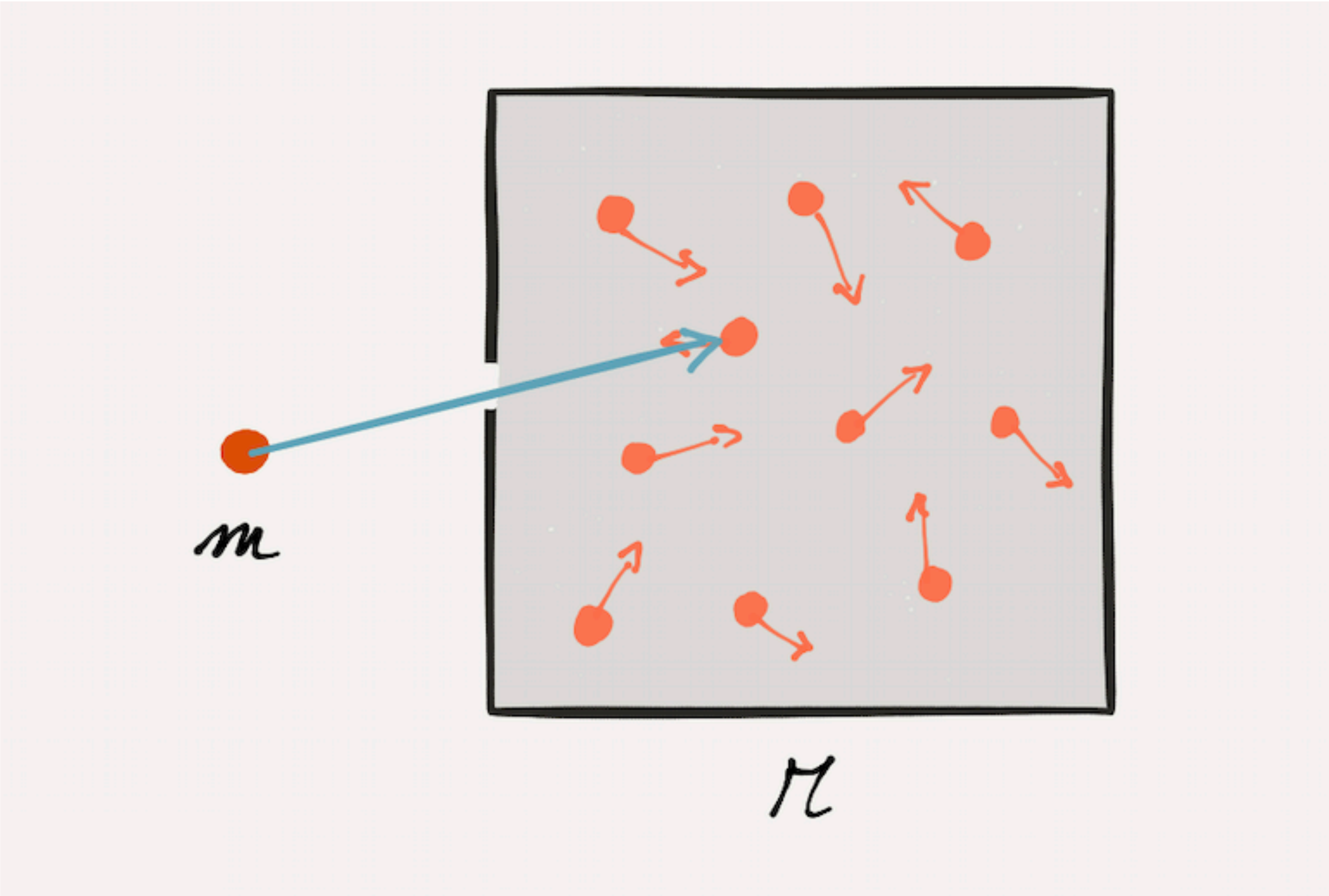
- Inelastic?
- Scattering geometry and notation
- Accessible energy and scattering angle ranges

Inelastic ?

Can you define elastic scattering ? Inelastic scattering ?

Inelastic ?

A thought experiment



Inelastic ?

Definition of elastic / inelastic scattering

Inelastic ?

In the TEM

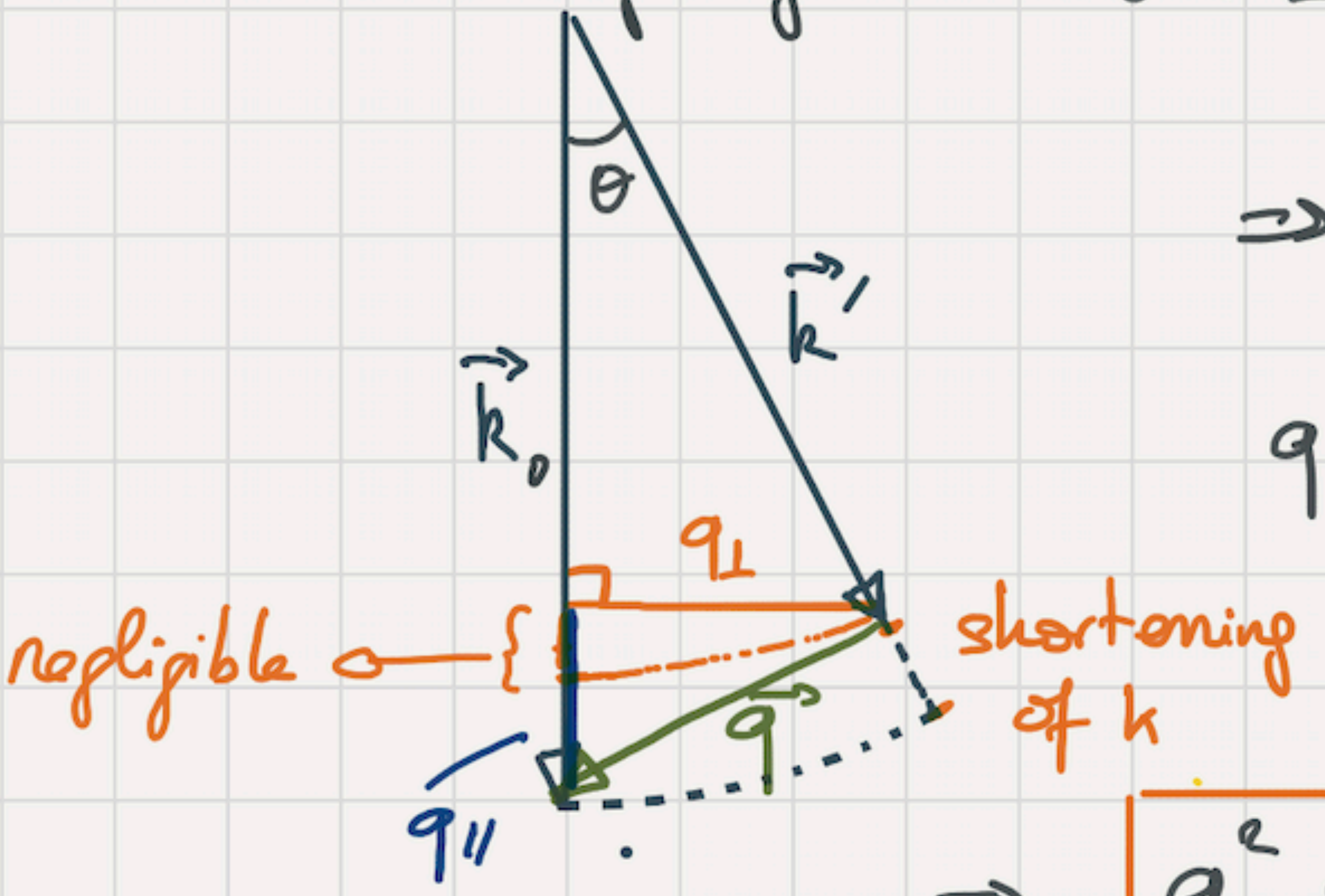
interaction	$\Delta E[eV]$	target apparatus	crystal	atom	electron
Bragg	10^{-23}	in	**el**	-	-
phonon	0.01	in	in	**el**	-
at. displacement	> 10	in	in	**el**	**el**
interband	1	in	in	in	**el**
exciton	< 10	in	in	in	**el**
plasmon	> 10	in	in	-	**el**
ionization	~ 100	in	in	in	**el**
Compton	~ 1000	in	in	in	**el**

“ Inelastic scattering is when the fast electron looses (measurable) energy”

Scattering geometry and notations

Scattering geometry and notations

θ : scattering angle



$\theta \ll 1$ & $k' \simeq k_0$

$$\Rightarrow q^2 \simeq q_{\parallel}^2 + q_{\perp}^2$$

$$q_{\perp}^2 = k_0^2 \theta^2$$

$$q_{\parallel}^2 = k_0^2 \theta_E^2 \ (\text{def})$$

$$\Rightarrow \boxed{q^2 \simeq k_0^2 (\theta^2 + \theta_E^2)}$$

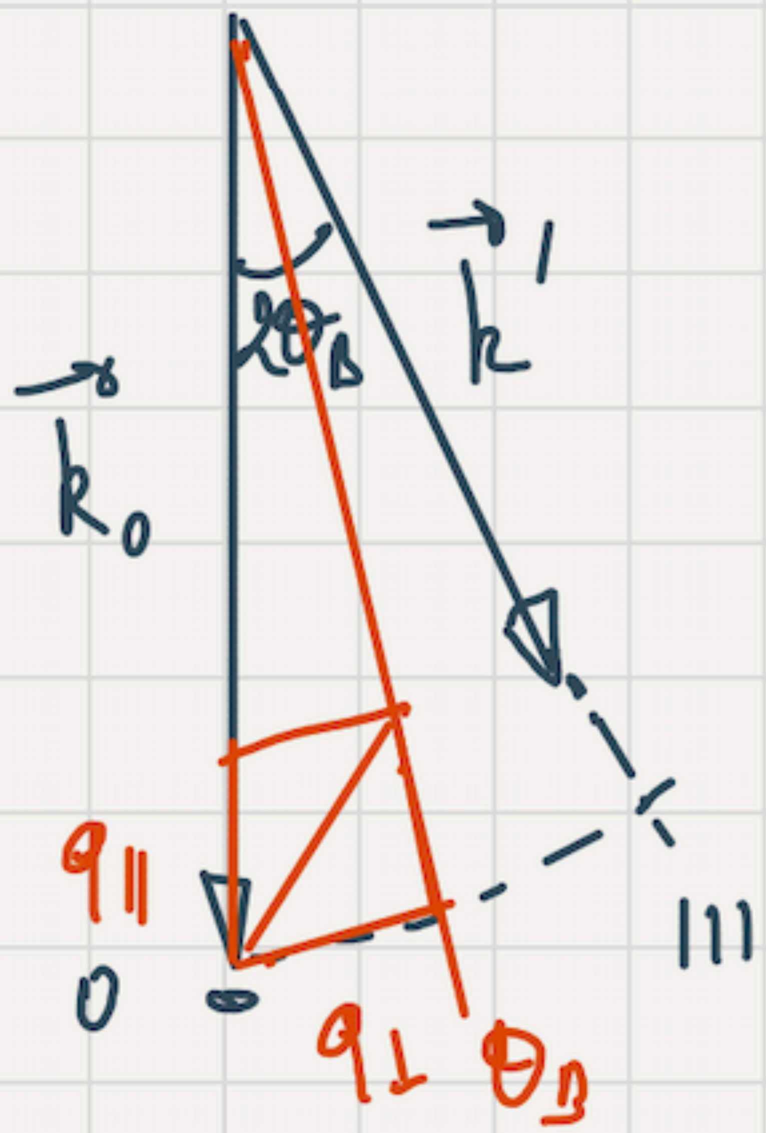
θ & $\theta_E \ll 1$

$$\theta_E = \frac{\Delta E \, m_0 \gamma}{\hbar^2 k_0^2} \simeq \frac{\Delta E}{2 E_0}$$

ΔE: Energy loss

Scattering geometry and notations

Orders of magnitude and geometrical consequences



at 200 kV $2\theta_{B_{111}}$ in Cu at ~ 10 mrad

at 200 kV losses between 0 & 2000 eV

θ_E at 2000 eV $= \frac{2000}{2 \times 200\,000} = \frac{1}{200} = 5$ mrad

scattering at θ_B $\quad q_\perp \sim q_\parallel$

for 2000 eV loss

Scattering geometry and notations

Collection and illumination (semi) angle

Accessible energy and scattering angles ranges

Instrumental limitations

- The range of accessible energies and scattering angles will govern the type of phenomenon that can be addressed.
- For the high scattering angles, there are hard cutoffs by the optics
- For high energy losses, issues are signal intensity and alignment
- For small energy losses, the width of the zero-loss peak is an issue
- For small scattering angles, the convergence of the beam is a challenge

Accessible energy and scattering angles range

Extremely high energy losses (>2500 eV)

EELS at very high energy losses

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Abstract

Electron energy-loss spectroscopy (EELS) has been investigated in the range from 2 to >10 keV using an optimized optical coupling of the microscope to the spectrometer to improve the high loss performance in EELS. It is found that excellent quality data can now be acquired up until about 5 keV, suitable for both energy loss near edge structure (ELNES) studies of oxidation and local chemistry, and potentially useful for extended energy loss fine structure (EXELFS) studies of local atomic ordering. Examples studied included oxidation in Zr, Mo and Sn, and the ELNES and EXELFS of the Ti-K edge. It is also shown that good quality electron energy-loss spectroscopy can even be performed for losses above 9.2 keV, the energy loss at which the collection angle becomes 'infinite', and this is demonstrated using the tungsten L_3 edge at about 10.2 keV.

Accessible energy and scattering angles range

Extremely high energy losses (>2500 eV)

Unfortunately, acquiring high quality EELS data at higher energy losses is not simple. As Craven and Buggy [8] showed, it is possible to improve the behaviour of the postspecimen lens system in a microscope to better transfer higher energy-loss electrons into an EELS spectrometer. Some of the studies quoted above used minor tweaks of existing lens setups to improve performance in a phenomenological manner. But recent work by some of the present authors [9] has shown a method for producing vastly improved performance in transferring higher loss electrons into the spectrometer, extending the useful range for quantitative EELS out to at least 5 keV. This was done by altering the optical path of the electrons through the postspecimen lenses to make some produce virtual images as the object of the next lens, whilst other produce real images – the movement of these image positions with electron energy change is opposite and allows balancing of the effects of energy loss in the movement of the final crossover that forms the object for the spectrometer.

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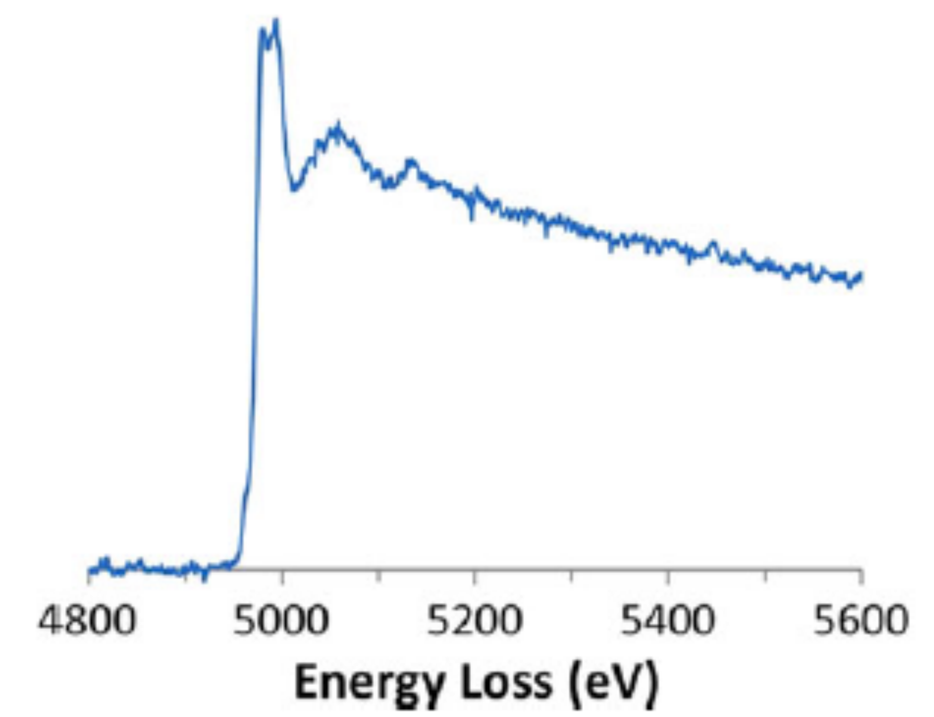


Fig. 3. A background subtracted, deconvolved Ti-K edge from amorphous TiO_2 ($t/\lambda = 0.70$, 1000 s acquisition).

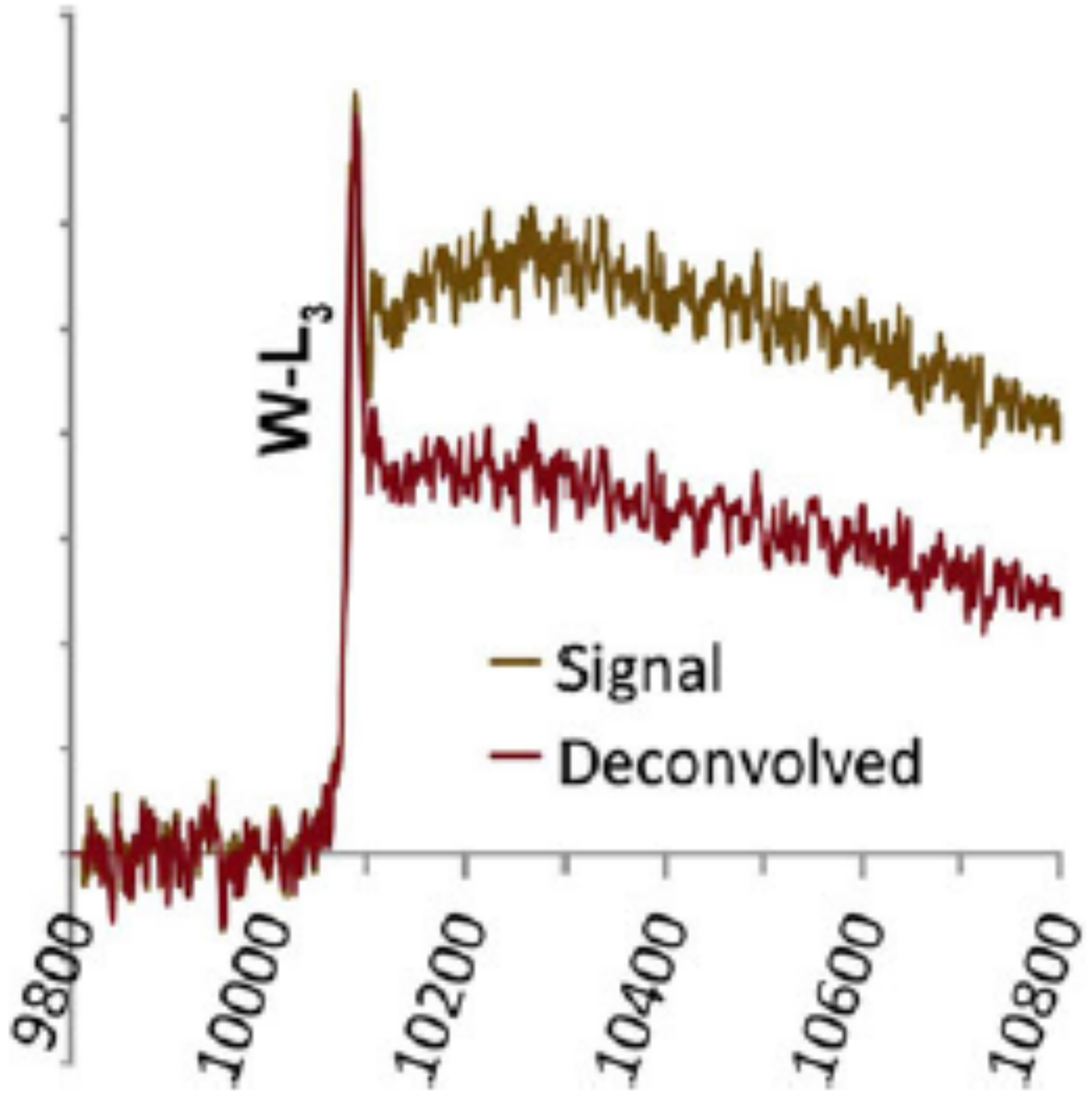


Fig. 4. The tungsten L_3 edge, both background-subtracted raw data and Fourier-ratio deconvolved using the low loss ($t/\lambda = 0.51$, 100 s acquisition).

Accessible energy and scattering angles range

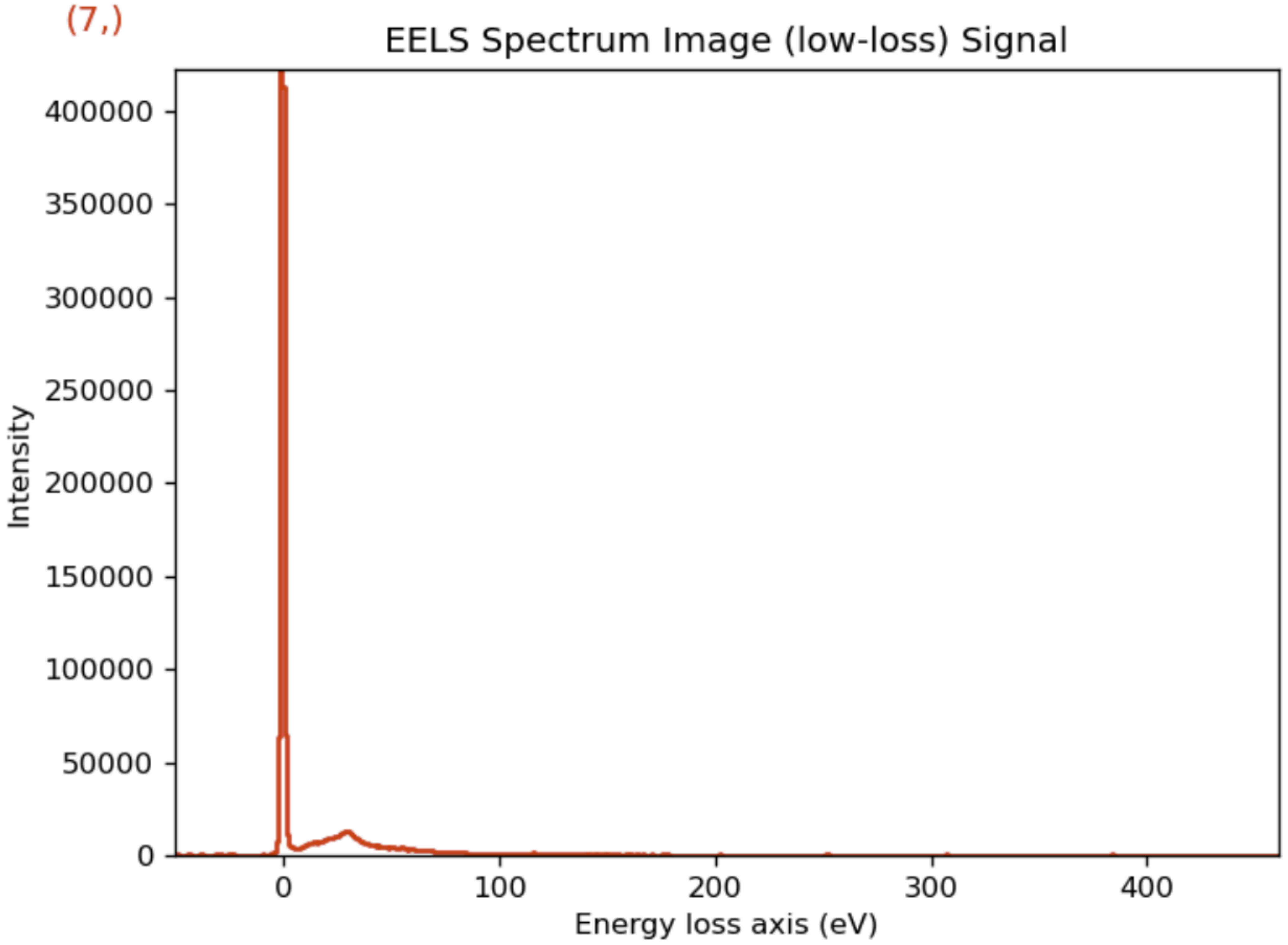
energy resolution and accessibility of low eggy losses

- The experiment geometry is a transmission one

- With a super-thin specimen, most of electrons will *not* experience an energy loss

- With a thick specimen, many electrons will experience *multiple consecutive energy losses that will sum up*

- At the end, a thin specimen is better, but then many electrons will cross without energy losses.



Accessible energy and scattering angles range
around the zero loss peak

Ultramicroscopy
Volume 203, August 2019, Pages 60-67

Progress in ultrahigh energy resolution EELS

O.L. Krivanek[a,b,*], N. Dellby[a], J.A. Hachtel[c], J.-C. Idrobo[c], M.T. Hotz[a], B. Plotkin-Swing[a], N.J. Bacon[a], A.L. Bleloch[a], G.J. Corbin[a], M.V. Hoffman[a], C.E. Meyer[a], T.C. Lovejoy[a]

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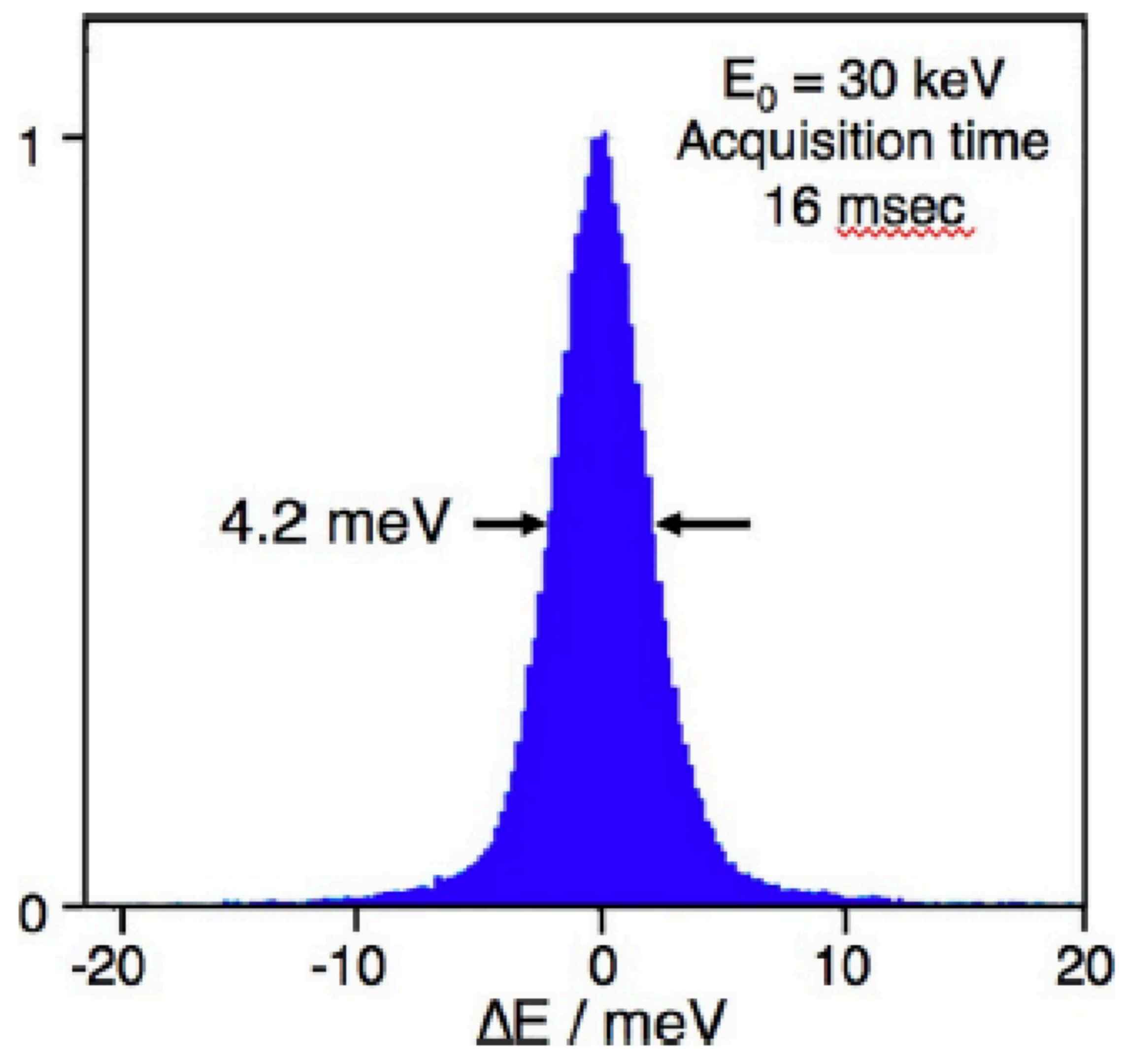


Fig. 2. Zero loss peak (ZLP) recorded in the vacuum outside a sample with the Nion Iris. 30 keV primary energy, 20 mrad illumination and collection half-angles, 0.2 meV/channel, 16 msec acquisition time. The intensity of the ZLP tail at 20 meV energy loss is ~0.001 of the ZLP maximum.

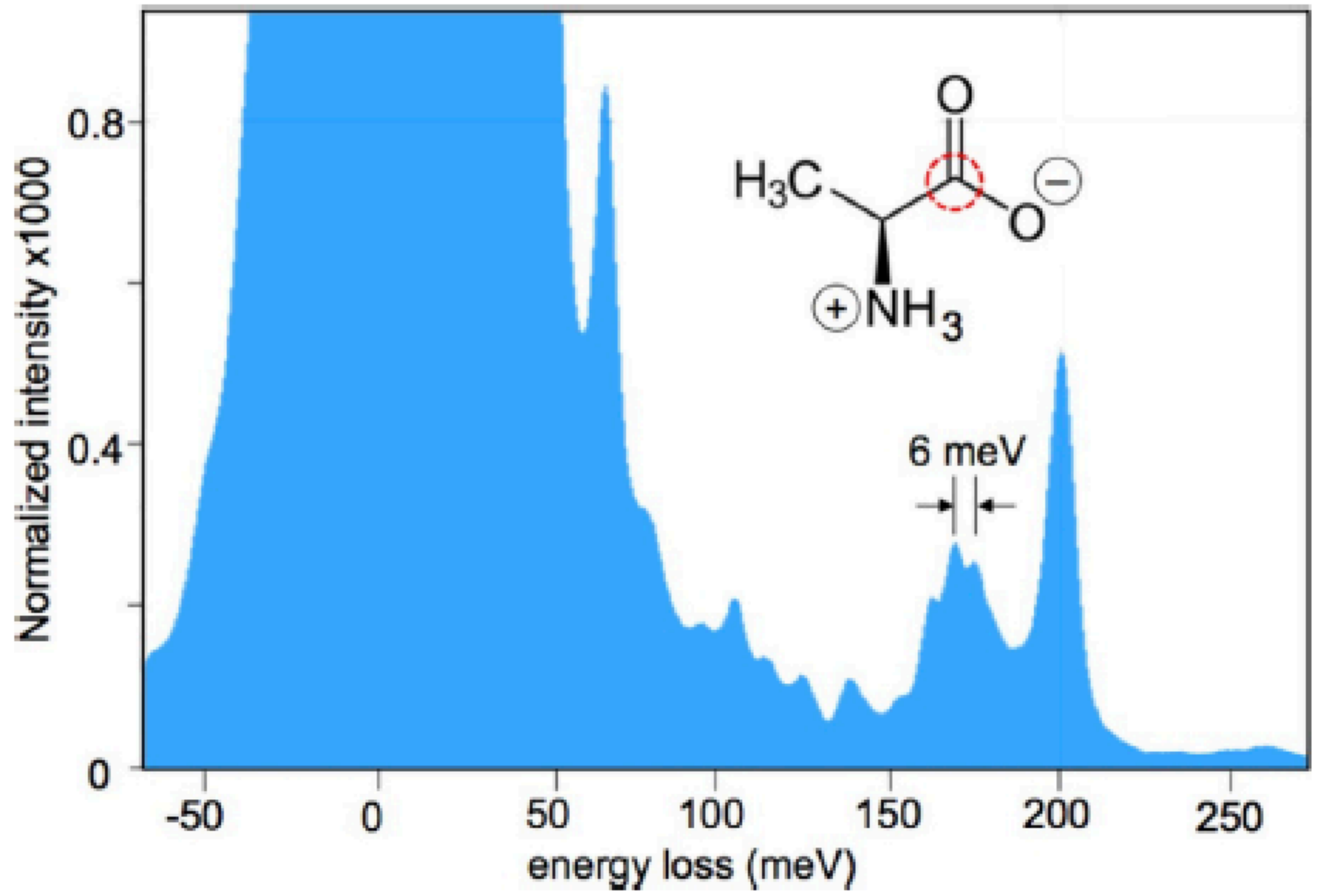


Fig. 5. Linear plot of spectrum of L-alanine zwitterion showing the 40–250 meV region in greater detail. Aloof acquisition with d ~ 20 nm at 30 keV. 100 separate spectra were acquired with acquisition time of 1.5 s each, aligned in energy, and added up, for a total acquisition time of 150 s. The intensity was scaled so that ZLP = 1. See Ref. [57] for similar spectra acquired from isotope-substituted L-alanine.

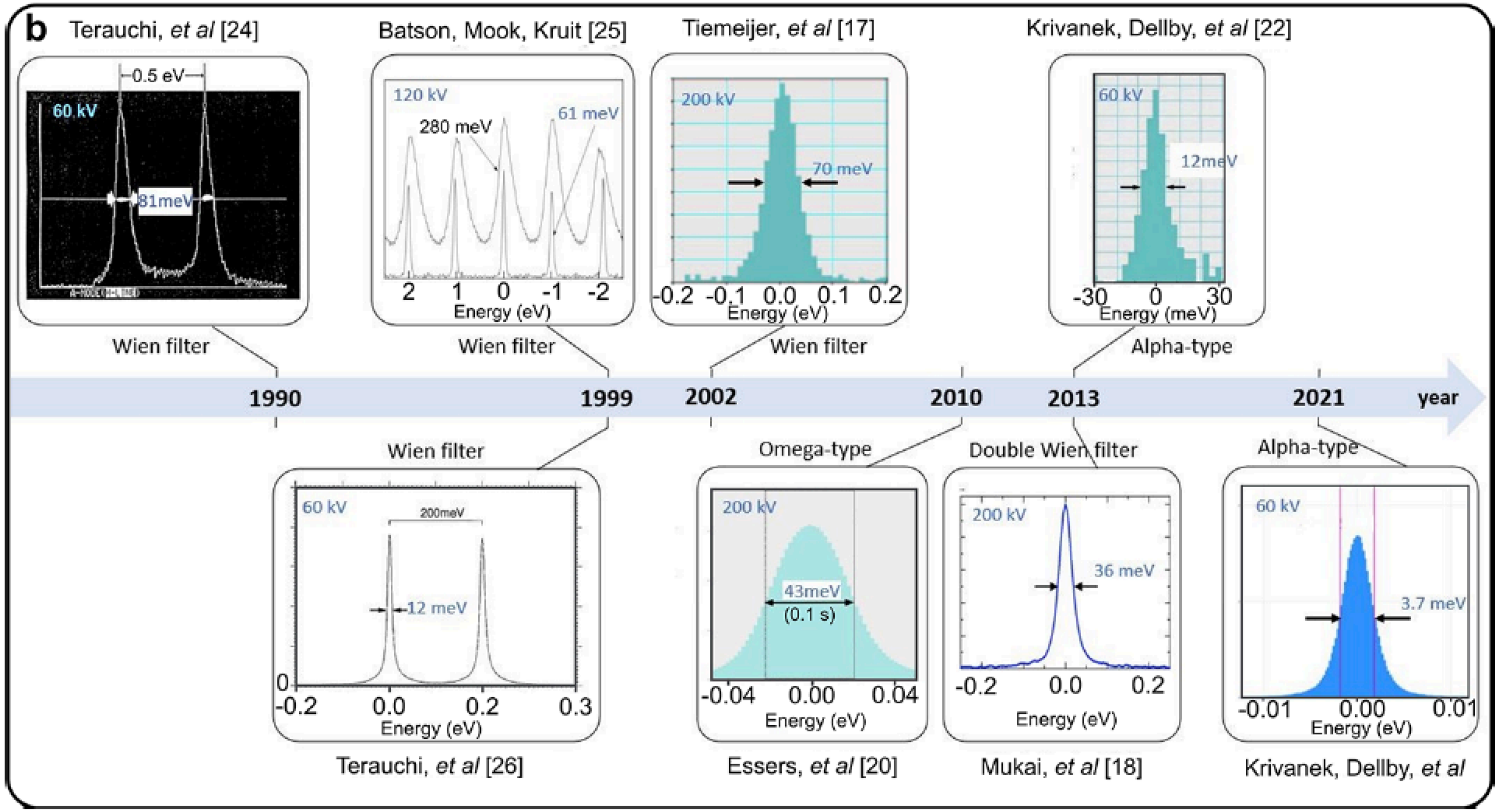
Accessible energy and scattering angles range

around the zero loss peak

Microscopy, 2022, **71(S1)**, i174–i199
DOI: https://doi.org/10.1093/jmicro/dfab050
Supplement Paper

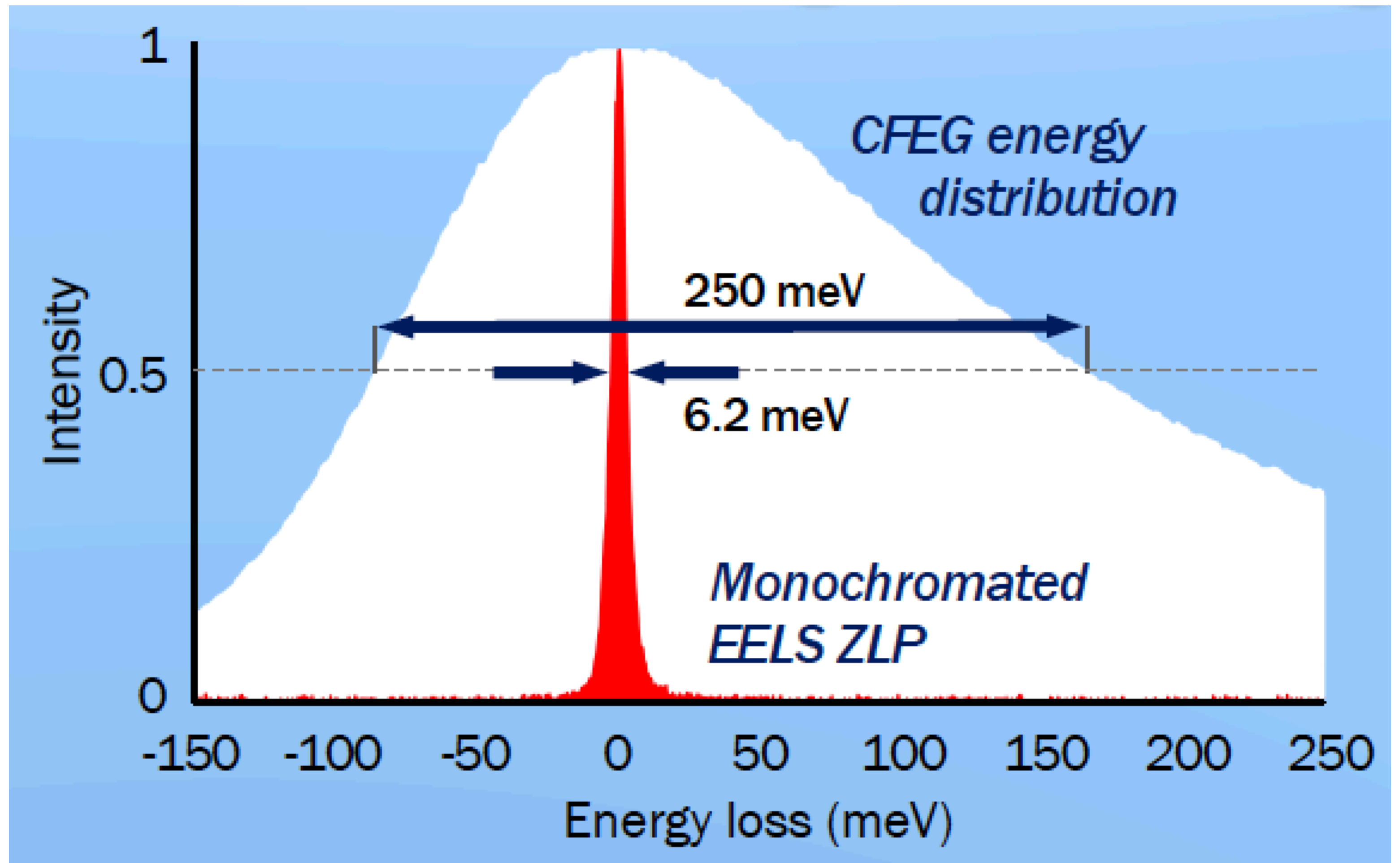
Advances in ultrahigh-energy resolution EELS: phonons, infrared plasmons and strongly coupled modes

Maureen J Lagos[1,2,*], Isobel C Bicket[2], S Shayan Mousavi M[1] and Gianluigi A Botton[1,3,*]



Accessible energy and scattering angles range

around the zero loss peak



Zero loss peak (ZLP) acquired with Nion HERMES™, 1 ms, 60 keV primary voltage, compared to the unmonochromated ZLP. Courtesy UC Irvine.

- Conventional FEG: 800 meV (0.8 eV)
- Cold FEG: 300 meV… but shape!
- Monochromated “conventional” TEM 150-200 meV
- Super high energy resolution < 10 meV
- Still “starting point” much higher

Accessible energy and scattering angles range

around the zero loss peak

Atomic resolution mapping of phonon excitations in STEM-EELS experiments

CrossMark

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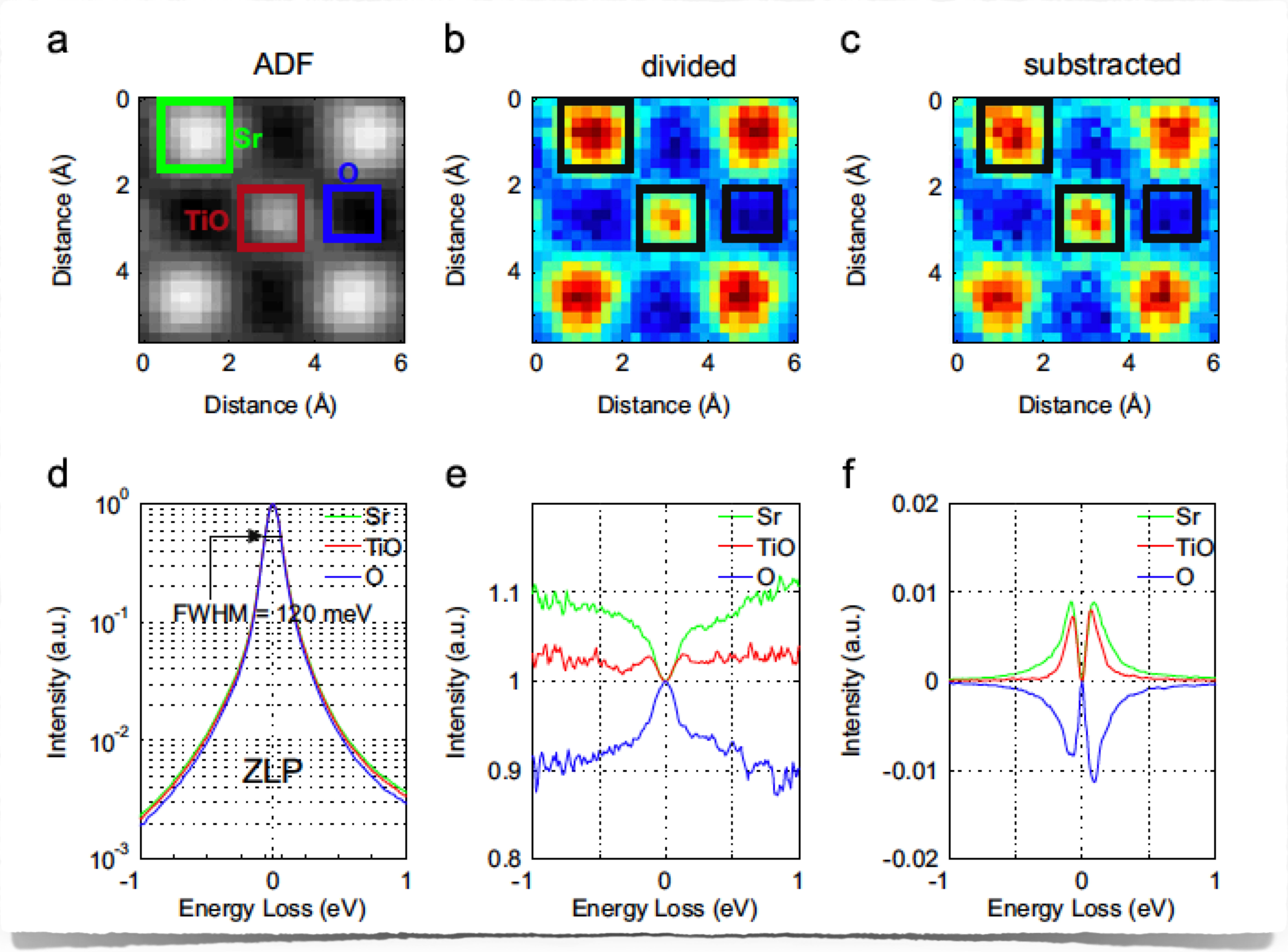
ABSTRACT

Atomically resolved electron energy-loss spectroscopy experiments are commonplace in modern aberration-corrected transmission electron microscopes. Energy resolution has also been increasing steadily with the continuous improvement of electron monochromators. Electronic excitations however are known to be delocalized due to the long range interaction of the charged accelerated electrons with the electrons in a sample. This has made several scientists question the value of combined high spatial and energy resolution for mapping interband transitions and possibly phonon excitation in crystals. In this paper we demonstrate experimentally that atomic resolution information is indeed available at very low energy losses around 100 meV expressed as a modulation of the broadening of the zero loss peak. Careful data analysis allows us to get a glimpse of what are likely phonon excitations with both an energy loss and gain part. These experiments confirm recent theoretical predictions on the strong localization of phonon excitations as opposed to electronic excitations and show that a combination of atomic resolution and recent developments in increased energy resolution will offer great benefit for mapping phonon modes in real space.

Accessible energy and scattering angles range

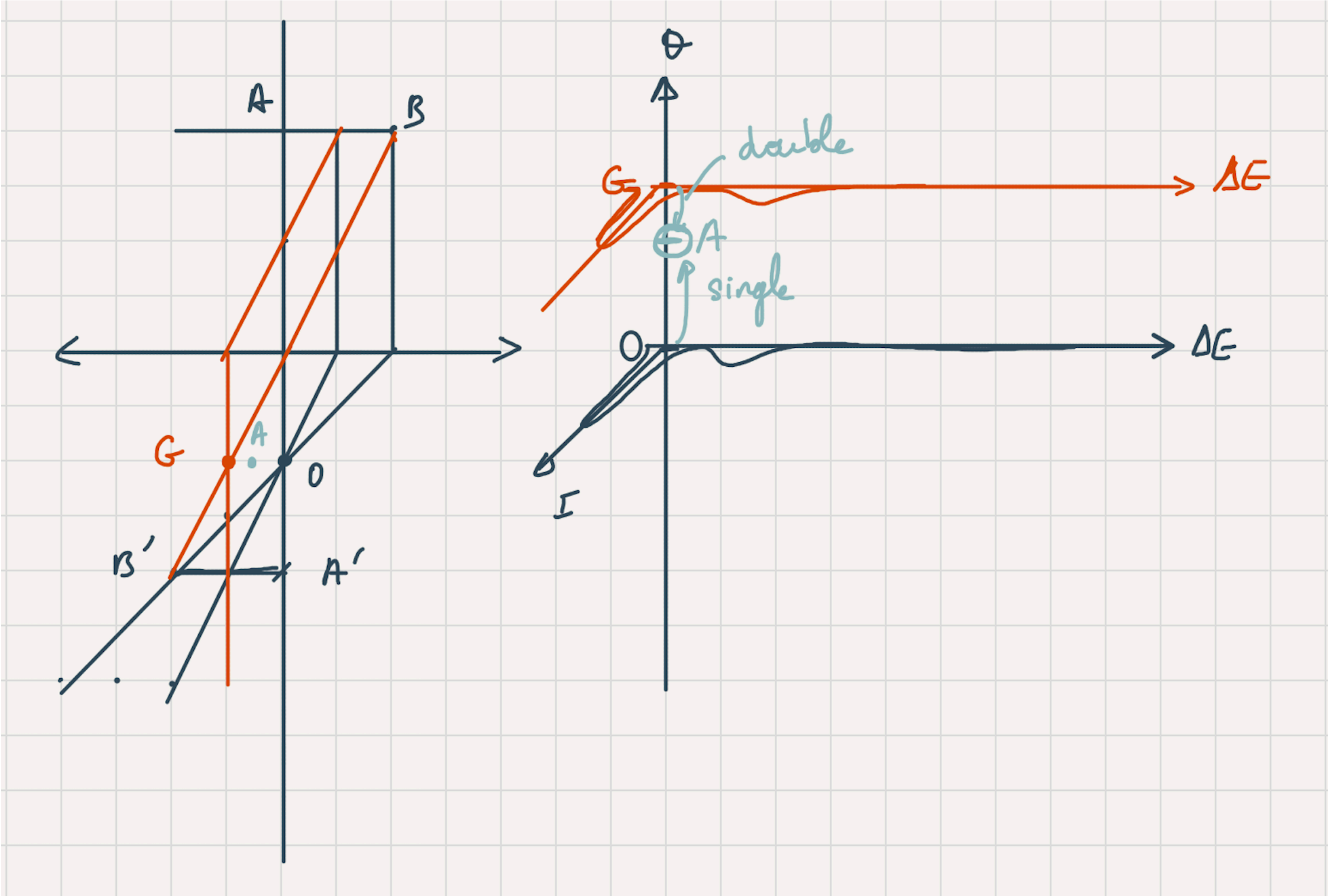
around the zero loss peak



R. Egoavil et al. / Ultramicroscopy 147 (2014) 1–7

Accessible energy and scattering angles range

effect of diffracted spots



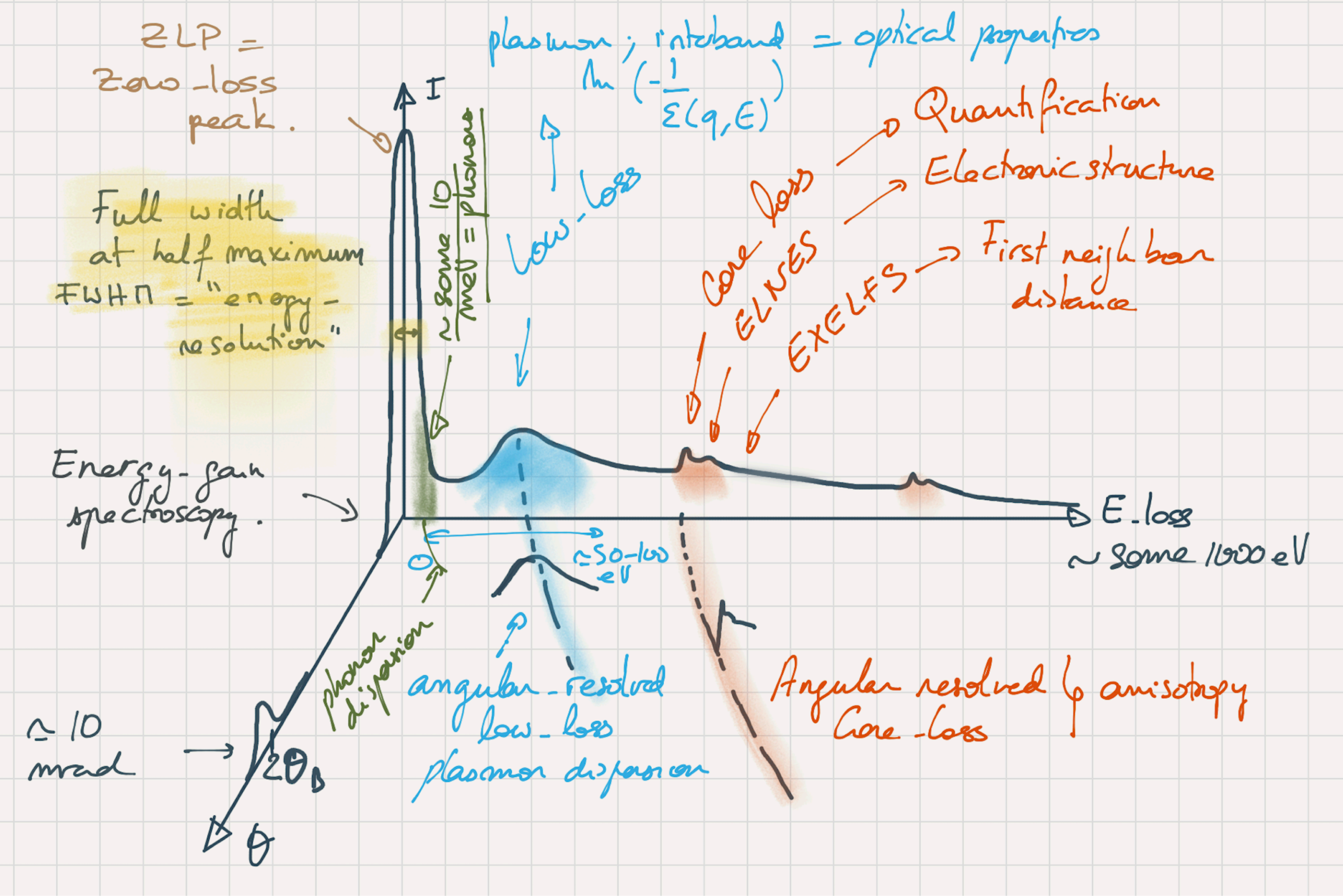
Diffracted spots act as secondary sources. They will hide the vanishing signal from single scattering at higher momentum transfer.

BZ boundary 1/2 way between 0 and G100.

Forbidden reflexions help a lot!

Accessible energy and scattering angles range

obtainable information, link to this lecture



- Core loss spectroscopy
- Angular resolved core loss
- Low loss: plasmon and interband transition
- Angular resolved low losses
- EELS for plasmonics and nano photonics
- (Vibrational spectroscopies)

Inelastic scattering - II

Core loss spectroscopy

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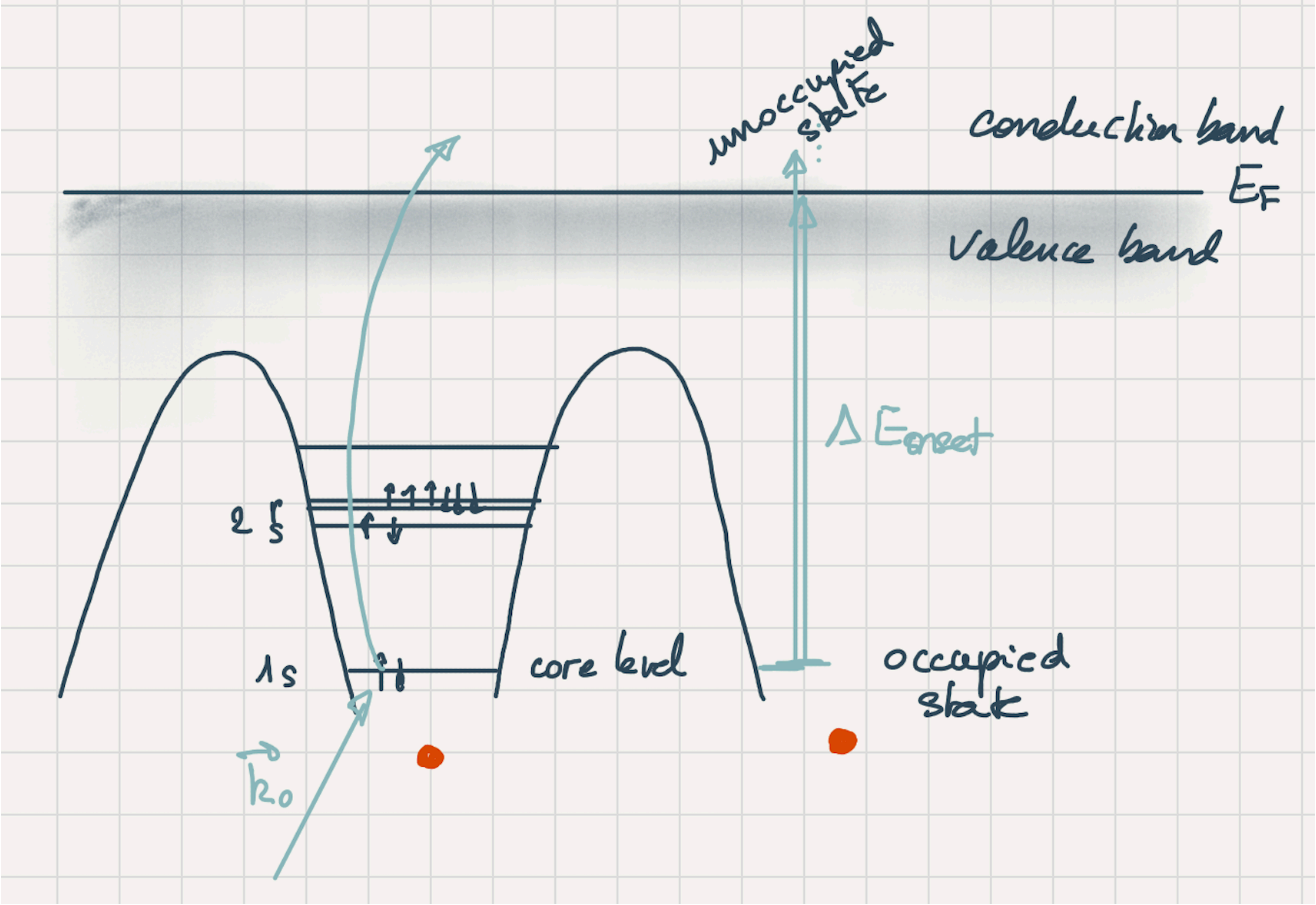
Layout

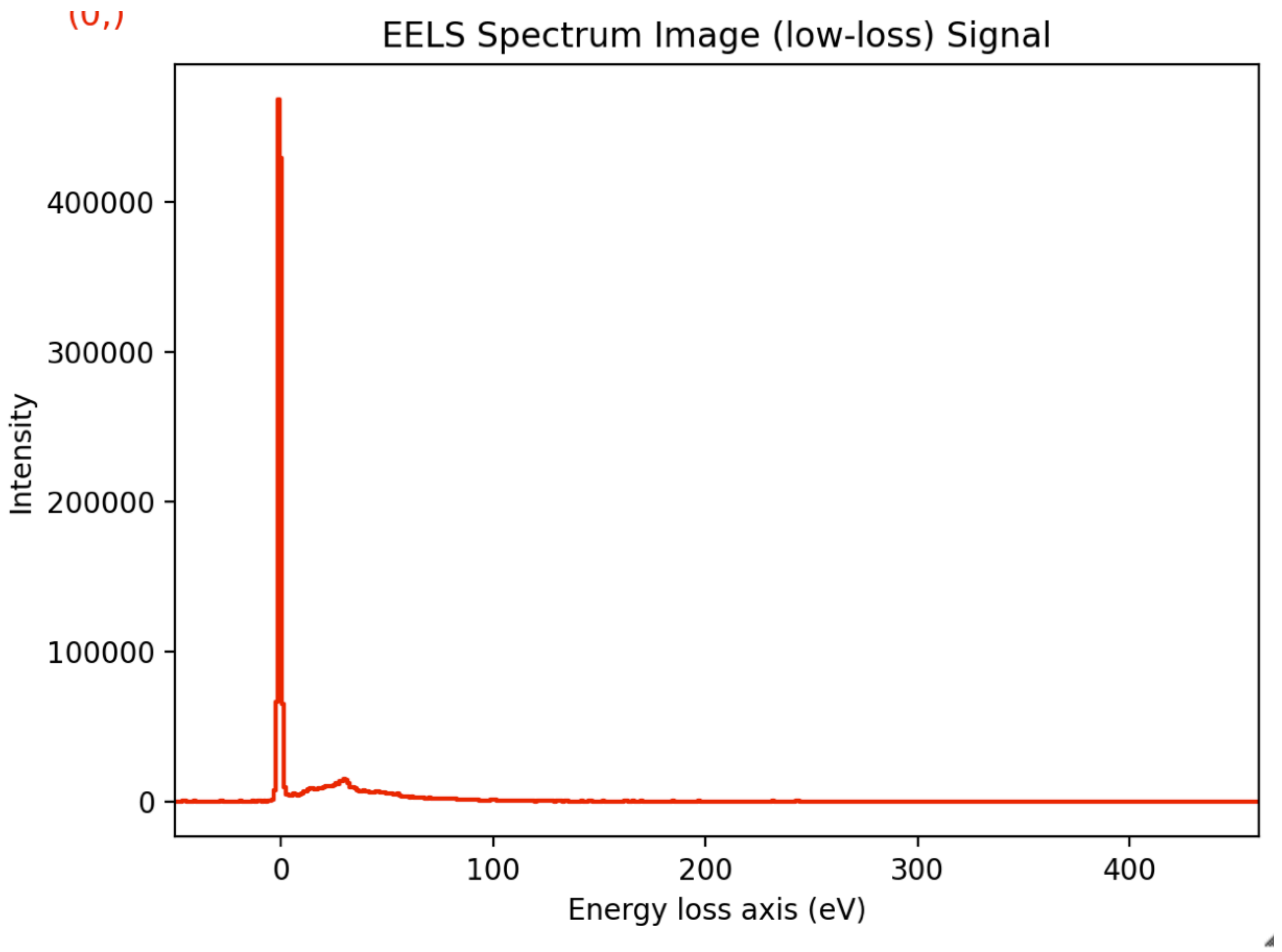
- Introduction
- Double differential scattering cross section
- Quantification
- ELNES

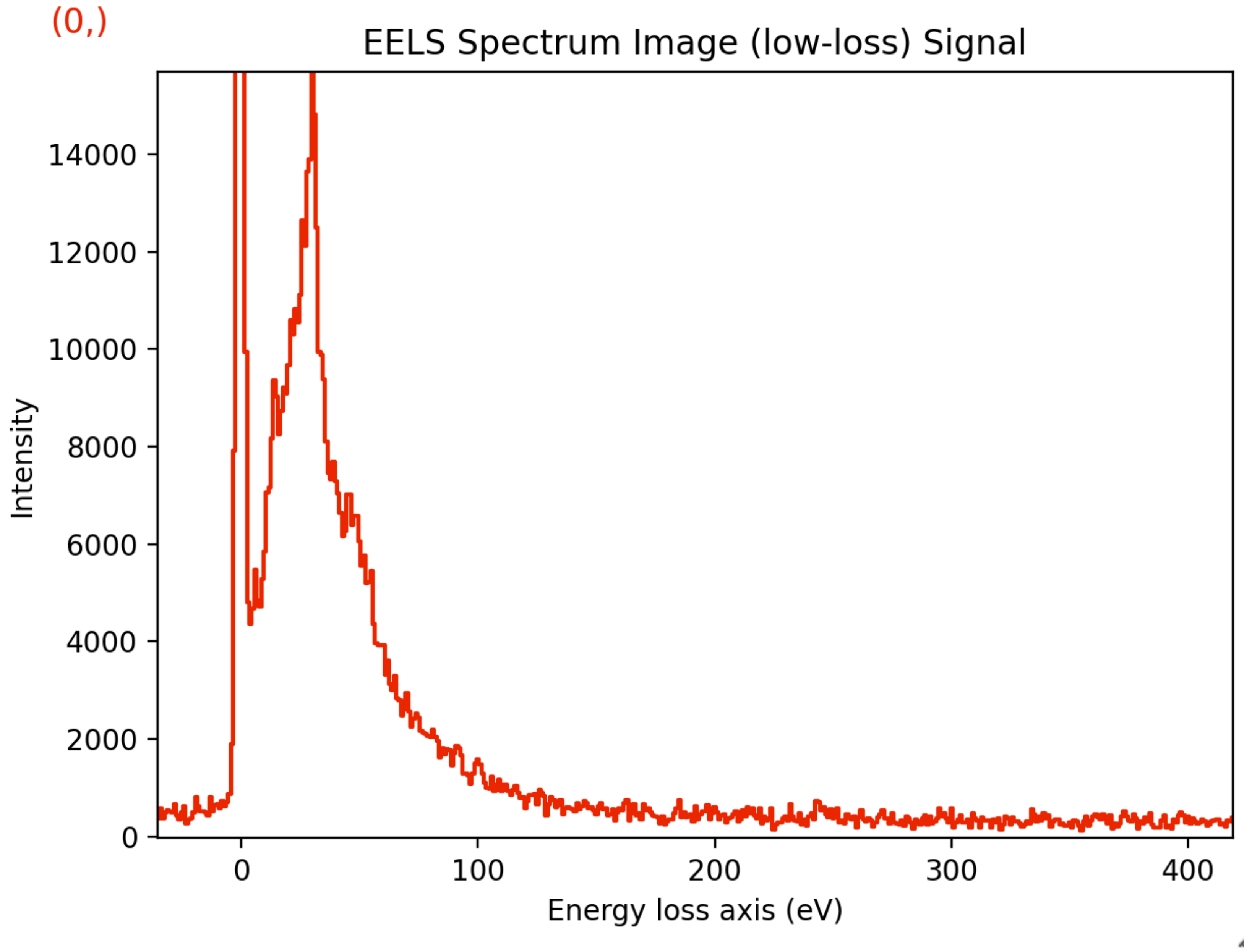
Introduction

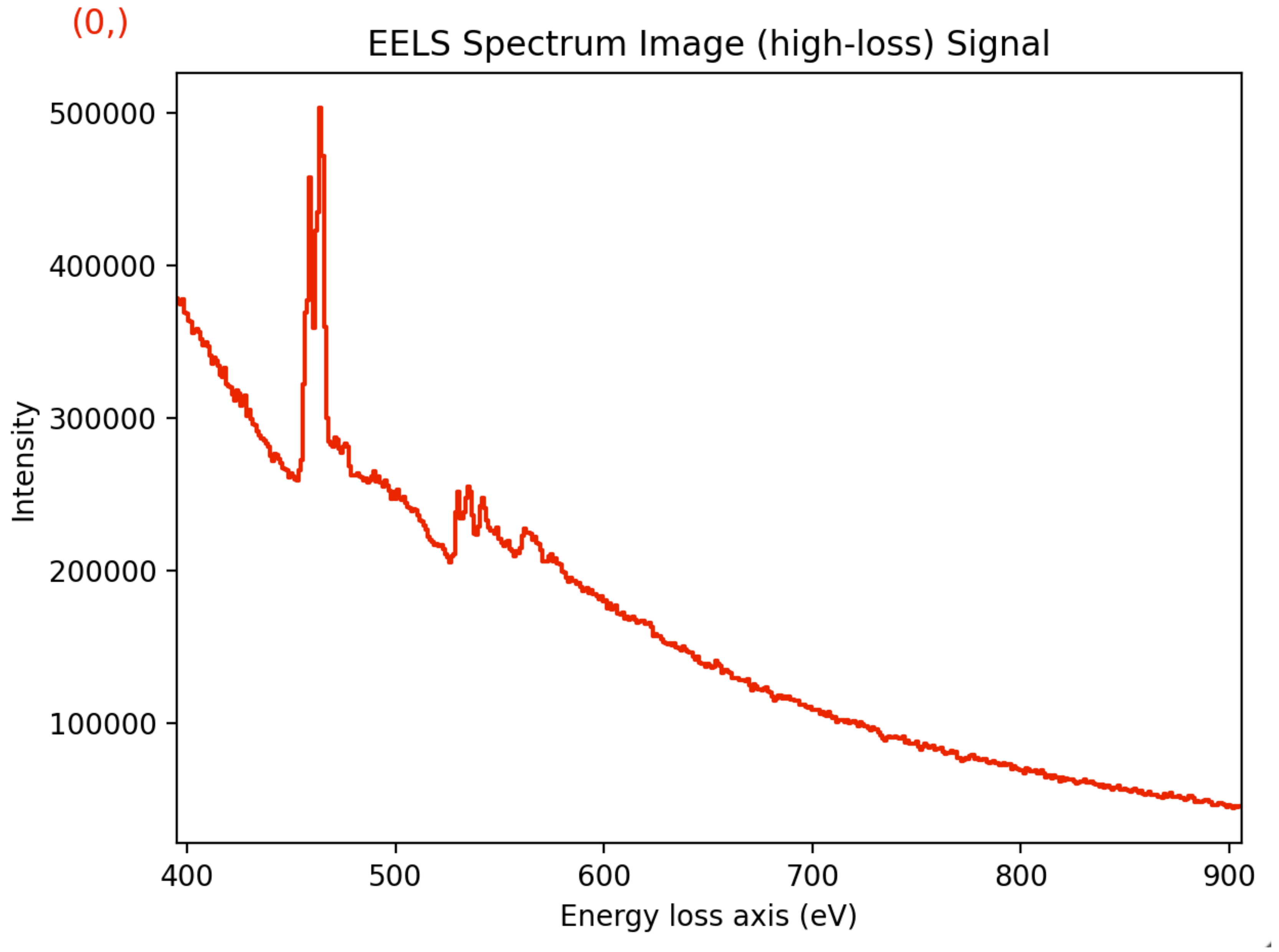


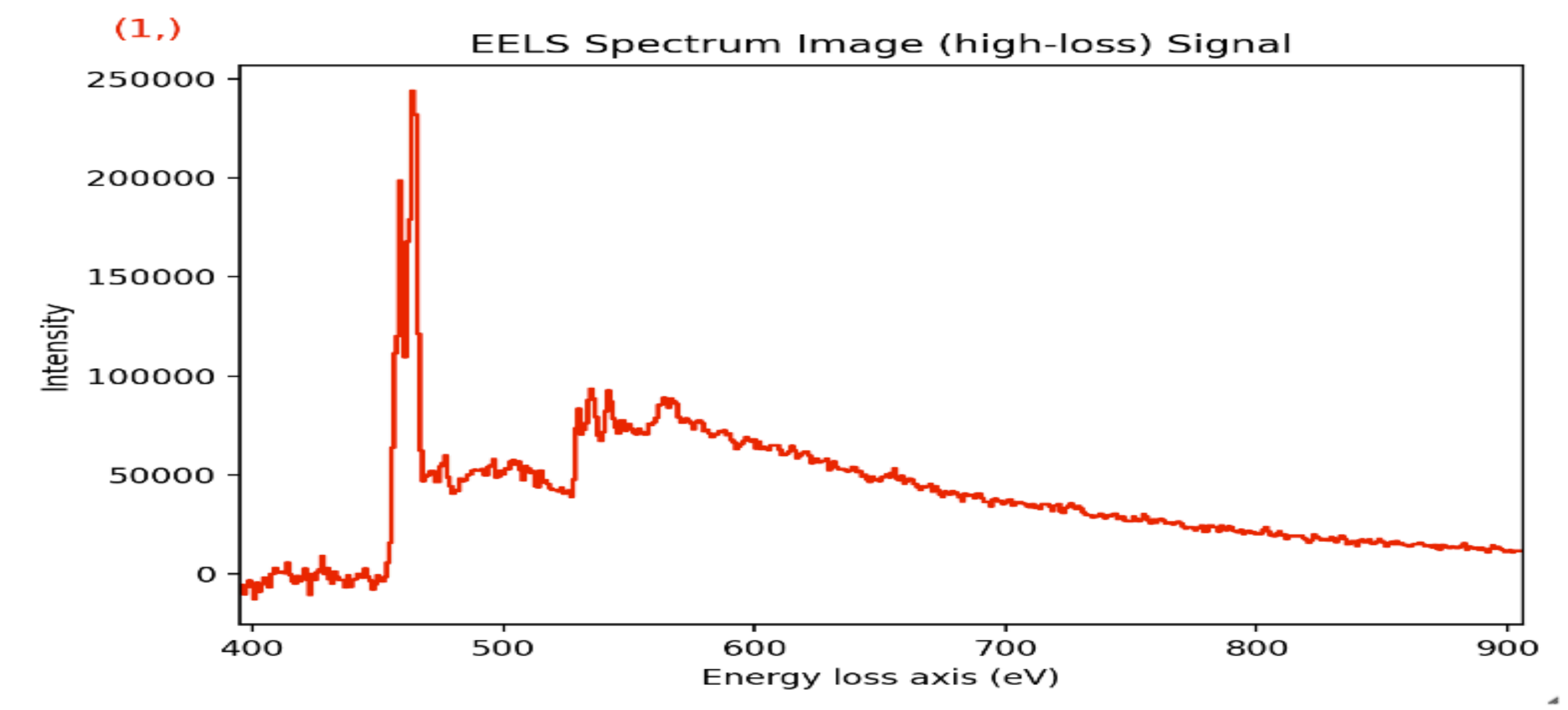
- Core electron: transition from a localised core state to an unoccupied state
- Onset: energy needed to reach Fermi level
- Transition possible above onset

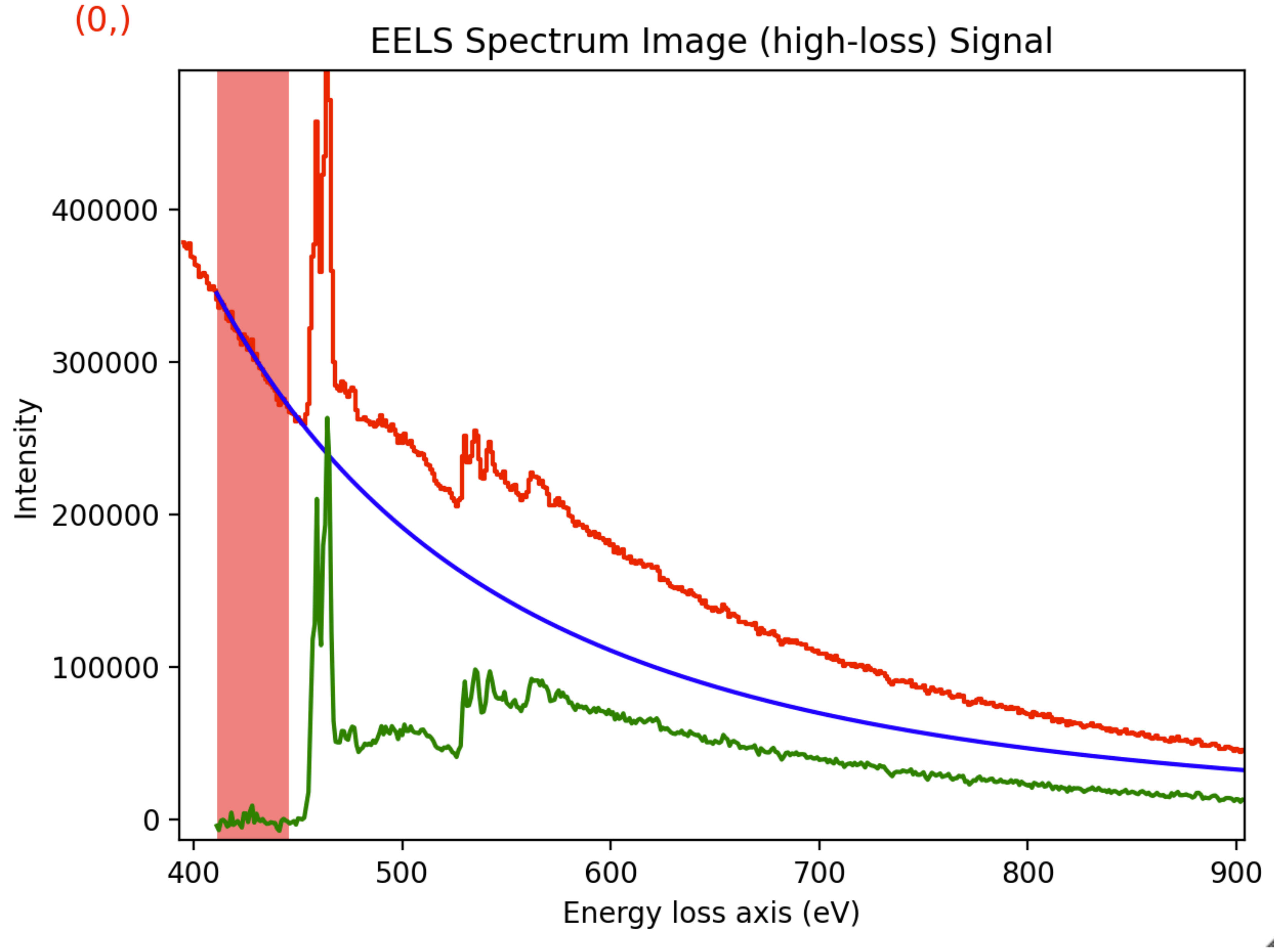
Introduction





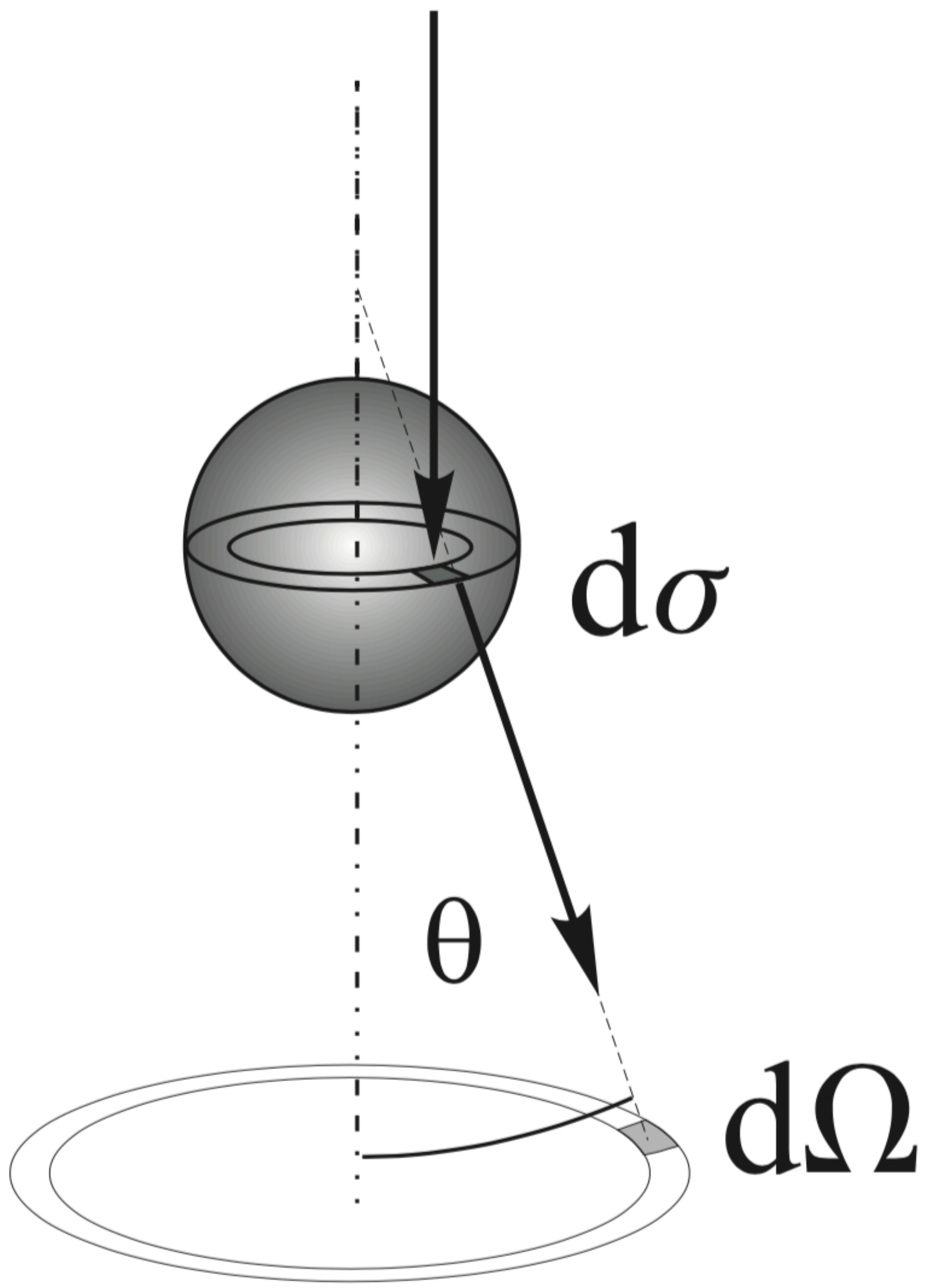


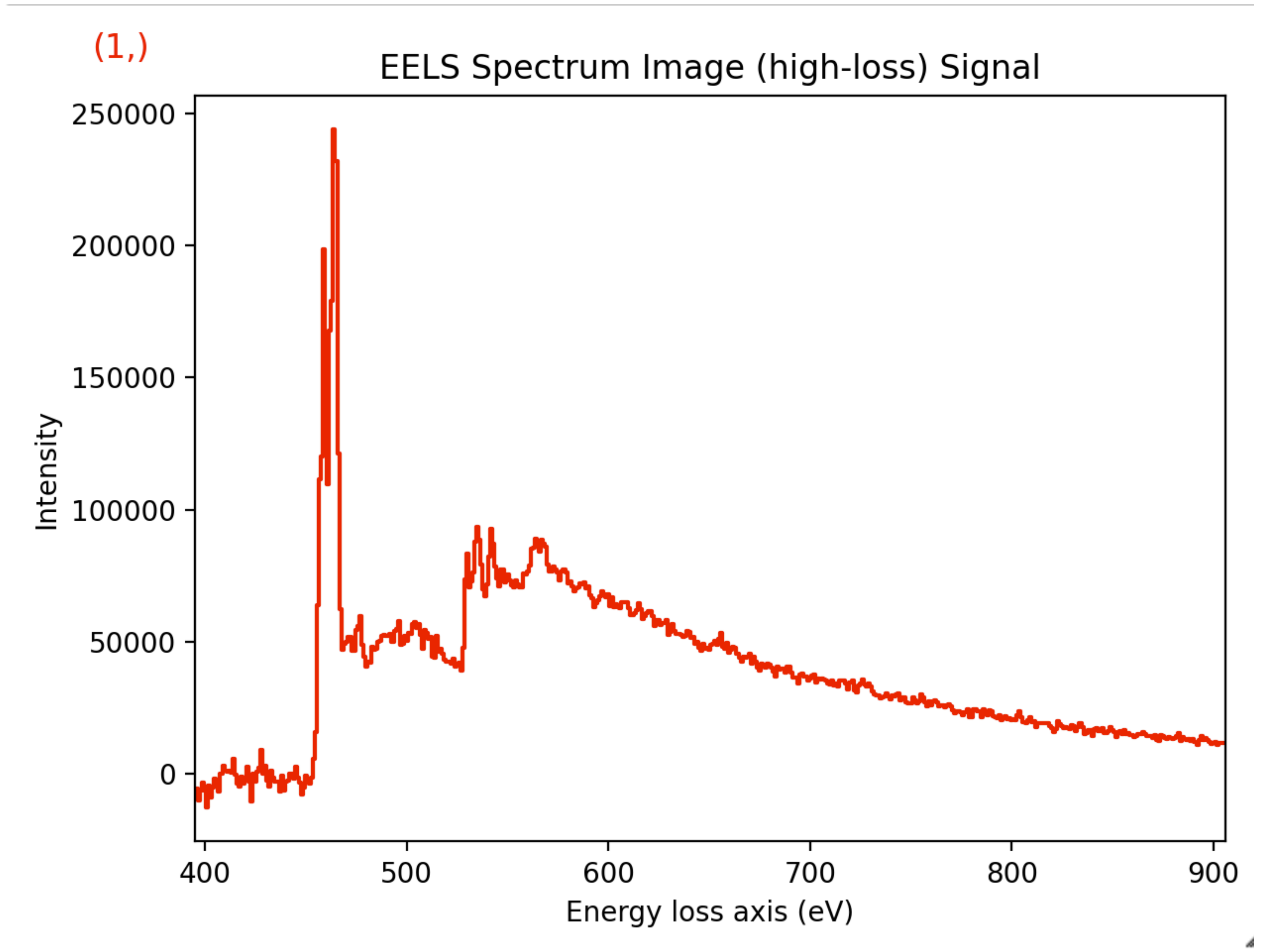




Double differential scattering cross section

Definition





- Apparent area relevant for the scattering
- Function of scattering angle and energy loss
- per unit of solid angle and energy

Double differential scattering cross section

- System: fast (incoming) electron + target electron
- Interaction potential: Coulomb
- First order perturbation theory, first Born approximation

H.A.Bethe: 1930:

Zur Theorie des Durchgangs schneller Korpuskularstrahlen durch Materie

Annalen der Physik, vol. 397, Issue 3, pp.325-400