Inelastic scattering - III b

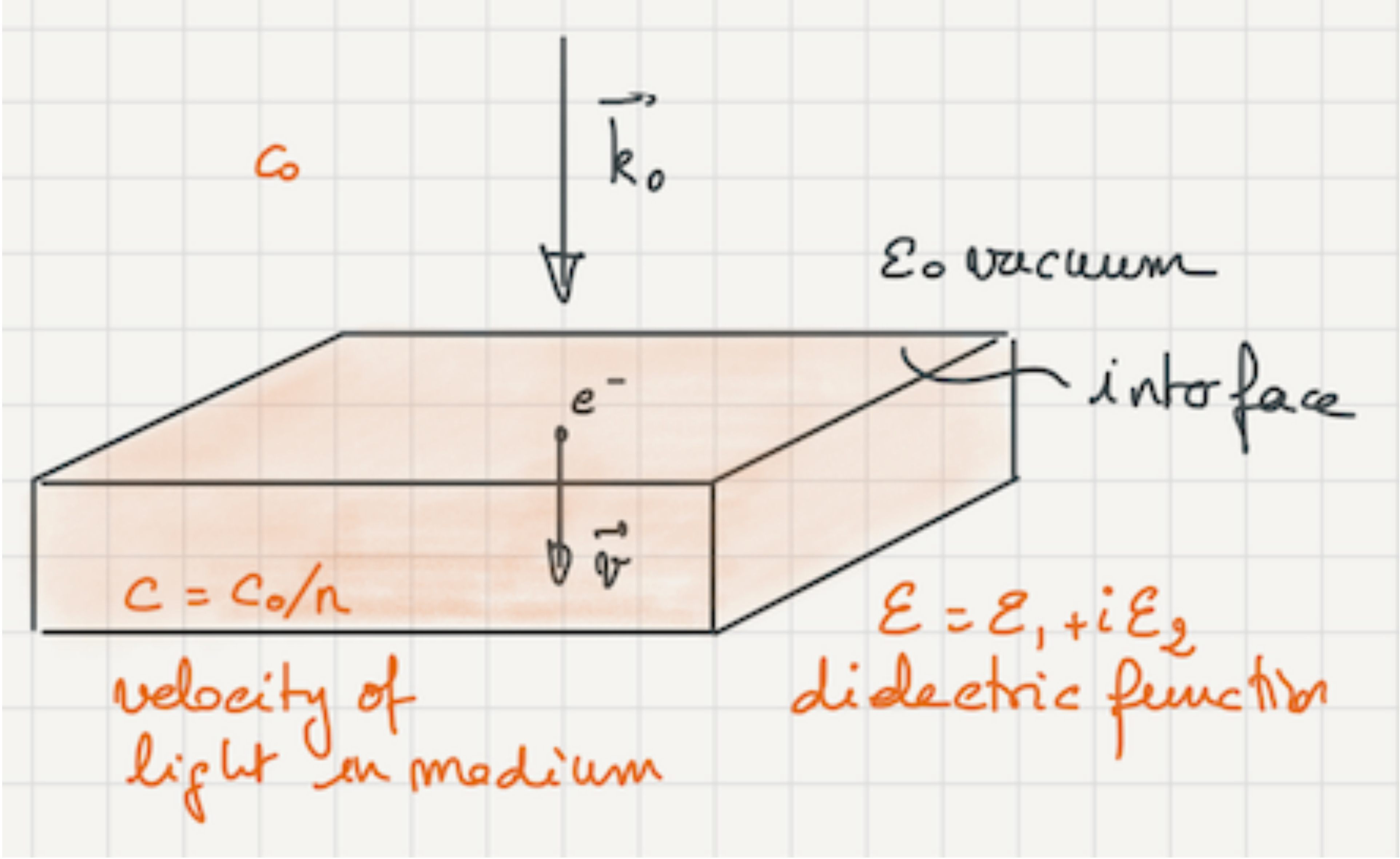
Low loss spectroscopy

Cécile Hébert IPHYS-LSME

Layout

- Introduction
- Formalism
- Experiment, plasmon dispersion
- Plural scattering
- Models for the dielectric function
- Surface effects
- Relativistic effects

Summary



- Single electron description not applicable
- Medium described by the dielectric function
- Linear reaction of medium on the fast travelling charged particle
- $\overrightarrow{D}(\vec{q},\omega) = \epsilon_0\epsilon(\vec{q},\omega)\overrightarrow{E}(\vec{q},\omega)$
- No boundaries, no relativity

$$\frac{\partial^2\sigma}{\partial\Omega\partial\Delta E} = \frac{e^2}{n_v\pi h^2\epsilon_0 v^2}\frac{1}{\theta^2+\theta_E^2}\Im\left(\frac{-1}{\epsilon(\vec{q},\Delta E)}\right) \qquad SSD = \left(\frac{e^2}{\pi h^2\epsilon_0 v^2}\right)D\frac{1}{\theta^2+\theta_E^2}\Im\left(\frac{-1}{\epsilon(\vec{q},\Delta E)}\right)$$

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Plural scattering

I_t Incident intensity on the specimen of thickness t

$$S(E) = I_t n_a t \frac{d\sigma}{d\Delta E}$$

Poisson statistics: the integrated component for n-fold scattering is $I_n = \frac{I_t}{n!}\left(\frac{t}{\lambda}\right)^n e^{-t/\lambda}$

ZLP shape (without specimen) $I_t R(E)$

Zero time scattering $S_{(0)} = I_0 R(E)$

Scatt probability distribution $P(E)$

One time scattering $S_{(1)} = I_1 R(E) * P(E)$

n-times $S_{(n)} = I_n R(E) * P(E) \ldots * P(E)$

Plural scattering

$$I_n = \frac{I_t}{n!}\left(\frac{t}{\lambda}\right)^n e^{-t/\lambda} = I_0 \frac{1}{n!}(t/\lambda)^n$$

n-times $S_{(n)} = I_n R(E) * P(E) \ldots * P(E)$

$$I_{exp}(E) = S_{(0)} + \ldots S_{(n)} + \ldots$$

$$\frac{t}{\lambda}P(E) = FT^{-1}\left[\ln \frac{\tilde{I}_{exp}}{I_0 \tilde{R}}\right]$$

Fourier-log deconvolution

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Evaluating the loss function

$$SSD = \left(\frac{e^2}{\pi h^2 \epsilon_0 v^2}\right) D \frac{1}{\theta^2 + \theta_E^2} \Im\left(\frac{-1}{\epsilon(\vec{q}, \Delta E)}\right)$$

Drude Model: quasi-free electron (mass m) in an oscillating field $\overrightarrow{E} = \overrightarrow{E}_0 e^{-i\omega t}$

$$m\ddot{\vec{x}} + m\Gamma\dot{\vec{x}} = -e\overrightarrow{E}$$

solution: $$\vec{x} = \frac{e\overrightarrow{E}}{m}\frac{1}{\omega^2 + i\Gamma\omega}$$

A displacement $\vec{x}$ gives a polarisation $\overrightarrow{P} = -en\vec{x} = \epsilon_0 \chi \overrightarrow{E}$

$$\omega_p = \sqrt{\frac{ne^2}{\epsilon_0 m}}$$

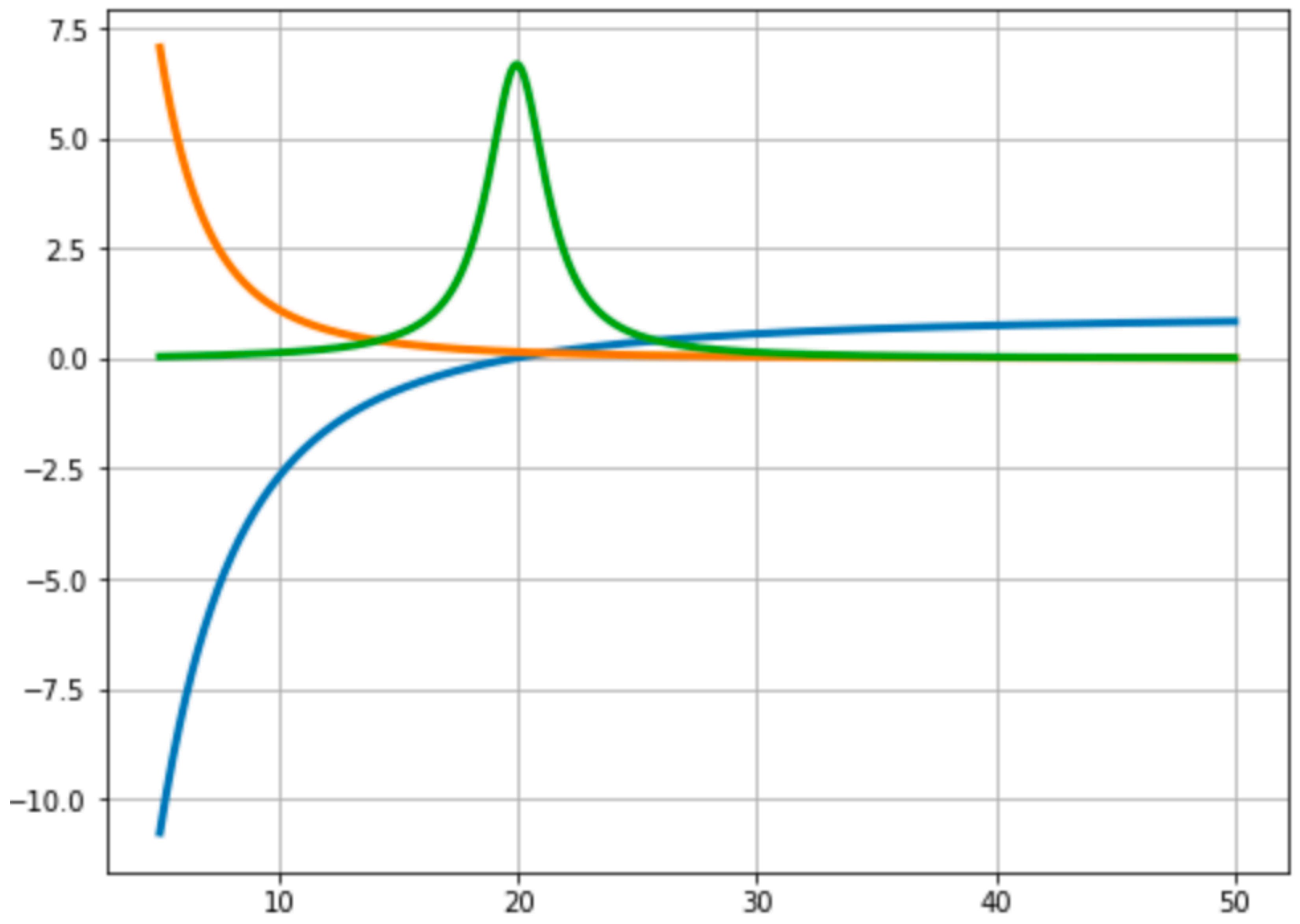
$$\chi = -\frac{\omega_p}{\omega}\left[\frac{\omega - i\Gamma}{\omega^2 + \Gamma^2}\right]$$

$$\epsilon = 1 + \chi = 1 - \frac{\omega_p^2}{\omega^2 + \Gamma^2} + i\omega_p^2 \frac{\Gamma/\omega}{\omega^2 + \Gamma^2}$$

Evaluating the loss function

Drude Model:

$$\epsilon = 1 + \chi = 1 - \frac{\omega_p^2}{\omega^2 + \Gamma^2} + i\omega_p^2 \frac{\Gamma/\omega}{\omega^2 + \Gamma^2}$$



$$SSD = \left(\frac{e^2}{\pi h^2 \epsilon_0 v^2}\right) D \frac{1}{\theta^2 + \theta_E^2} \Im\left(\frac{-1}{\epsilon(\vec{q}, \Delta E)}\right)$$

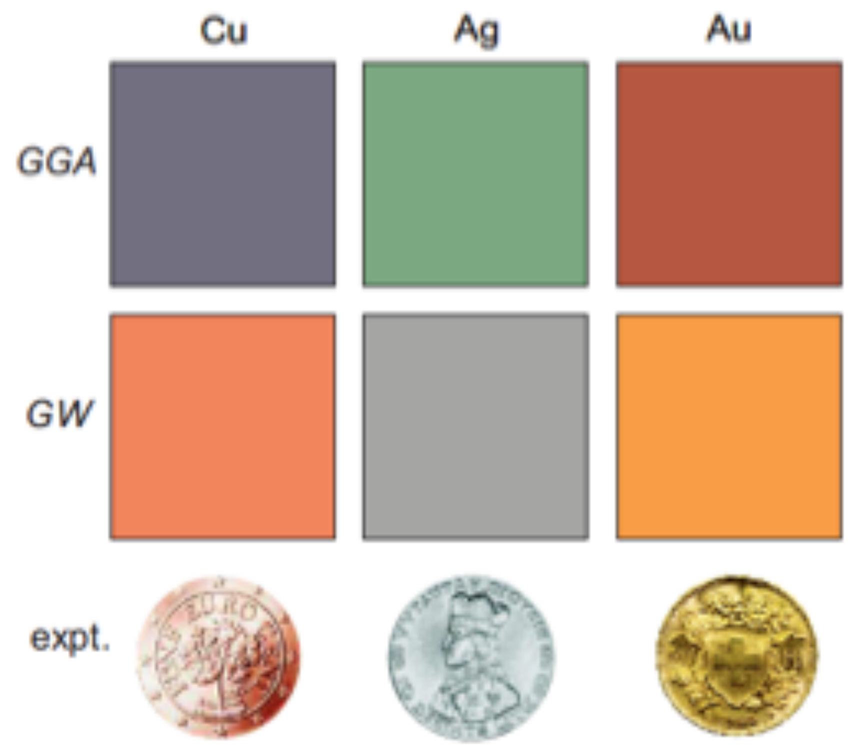
Evaluating the loss function

Ab initio calculations

PHYSICAL REVIEW B 88, 195124 (2013)

Alkauskas & al.

Dynamic structure factors of Cu, Ag, and Au: Comparative study from first principles



Evaluating the loss function

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PHYSICAL REVIEW B **88**, 195124 (2013)

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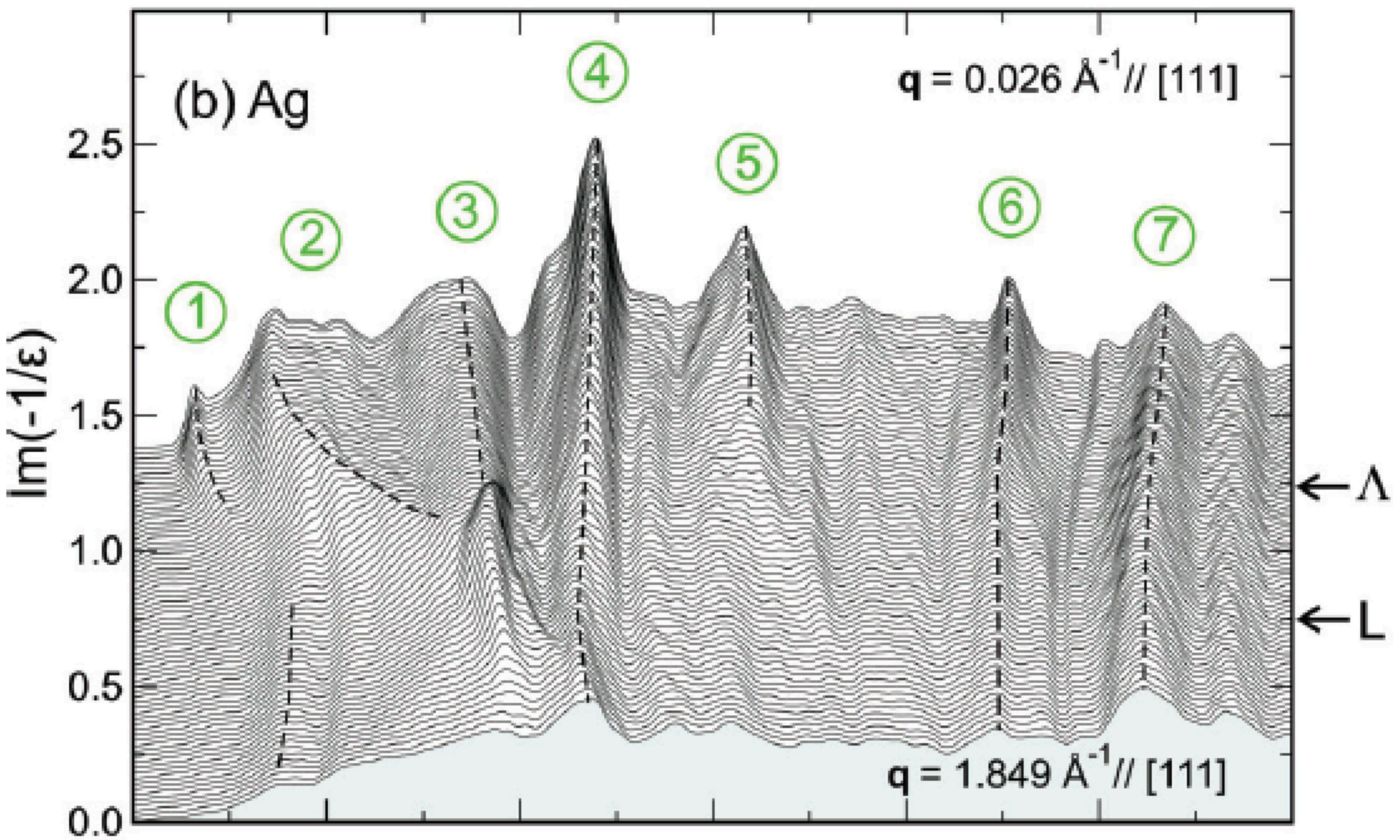

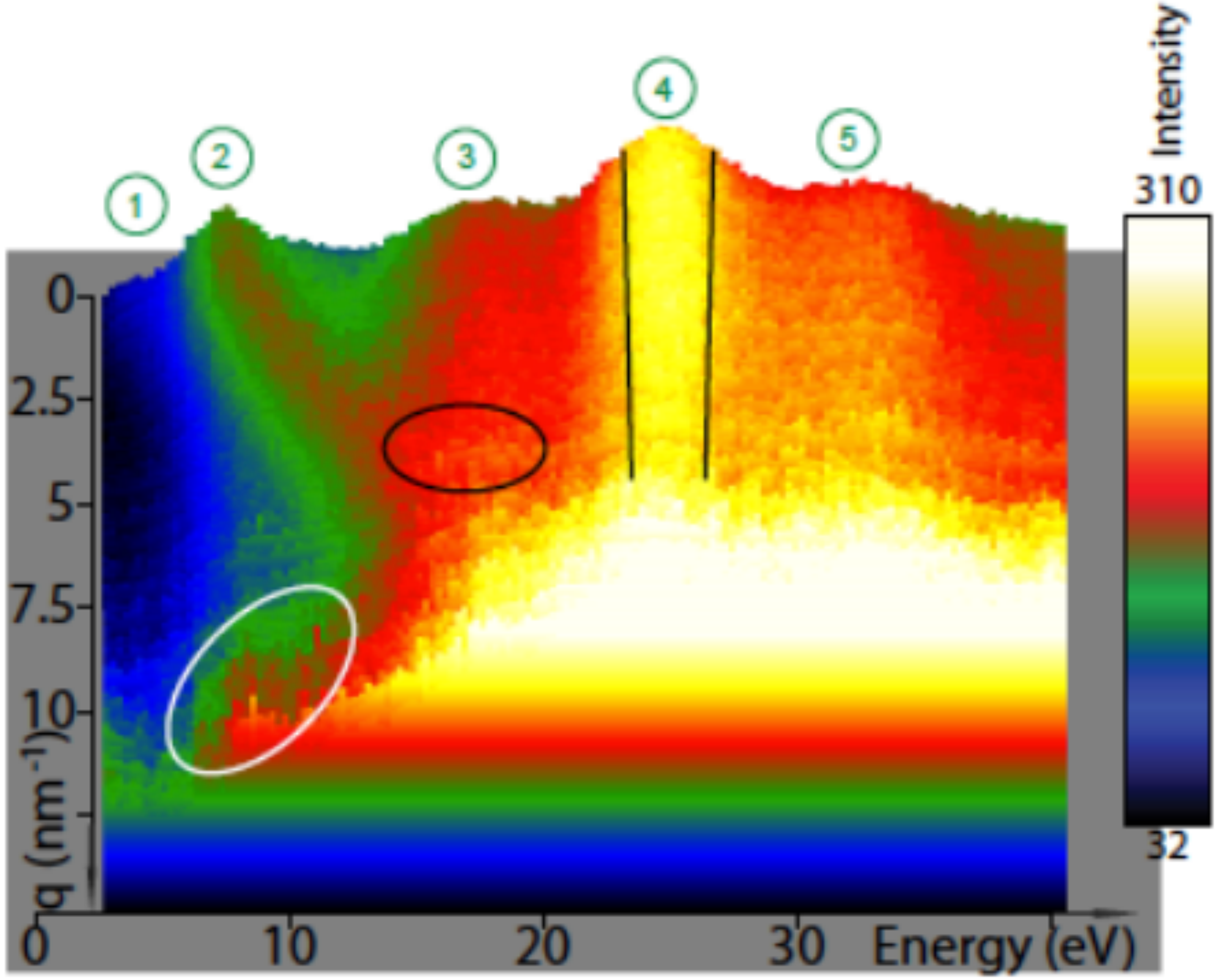

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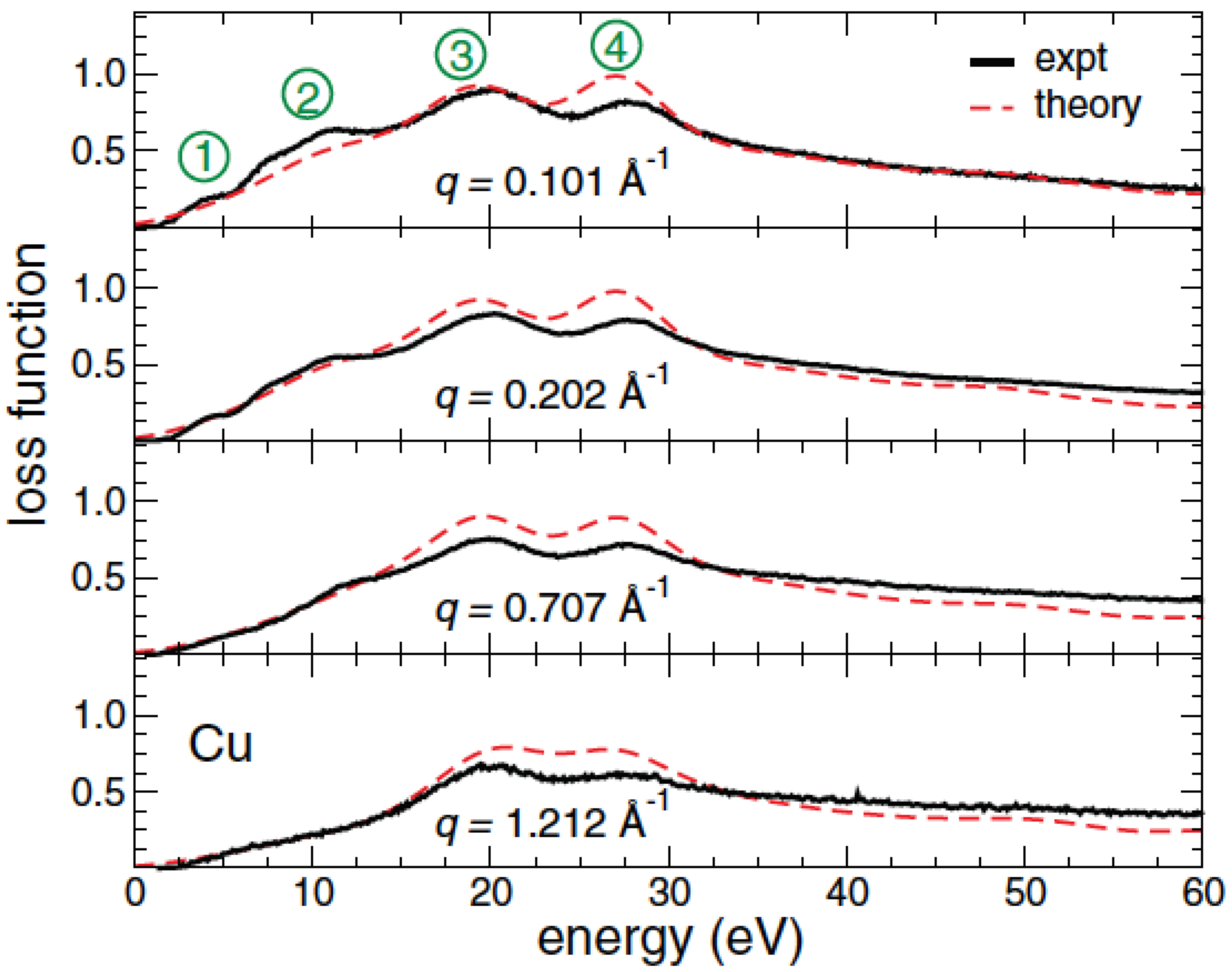

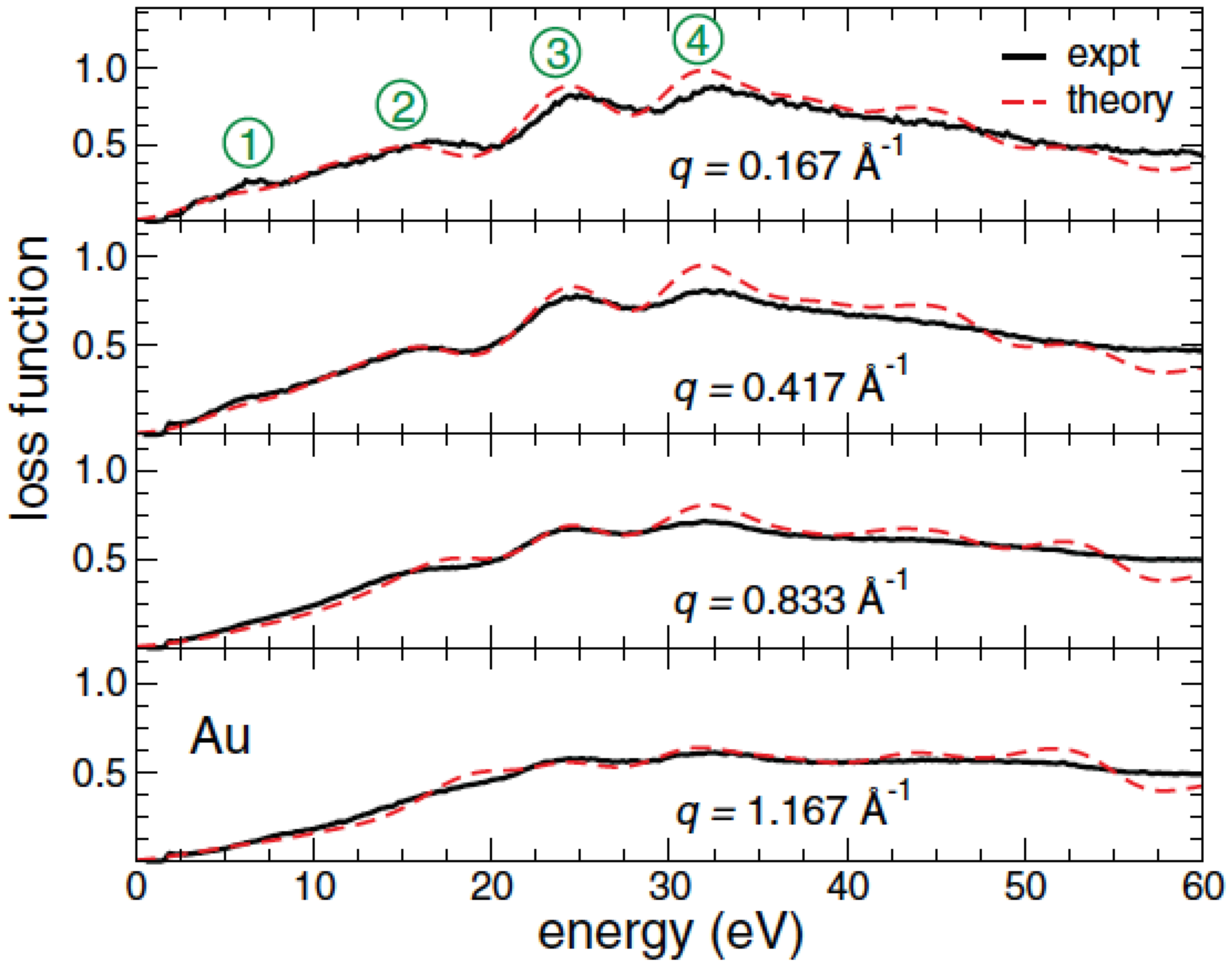

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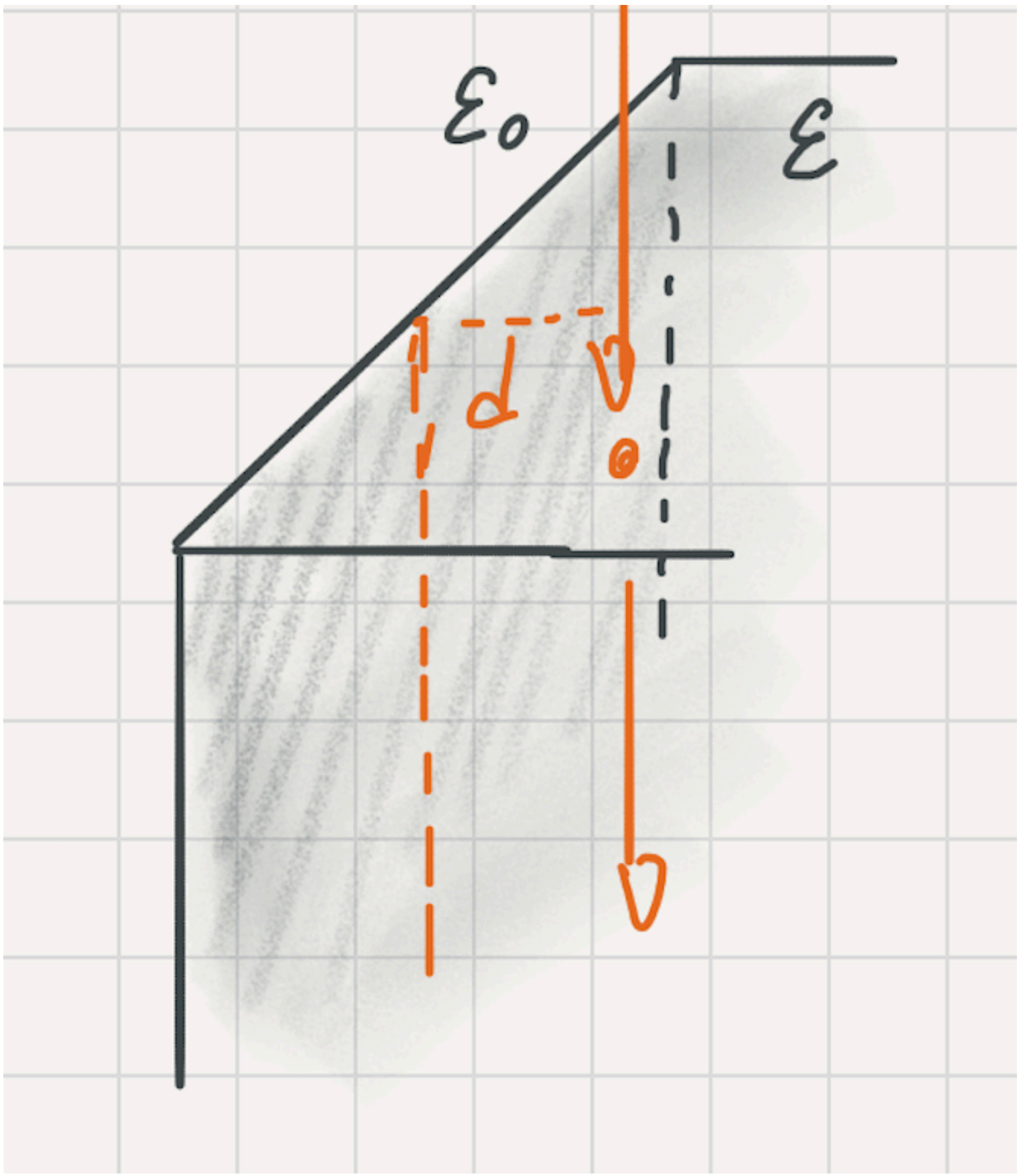
Surface effects



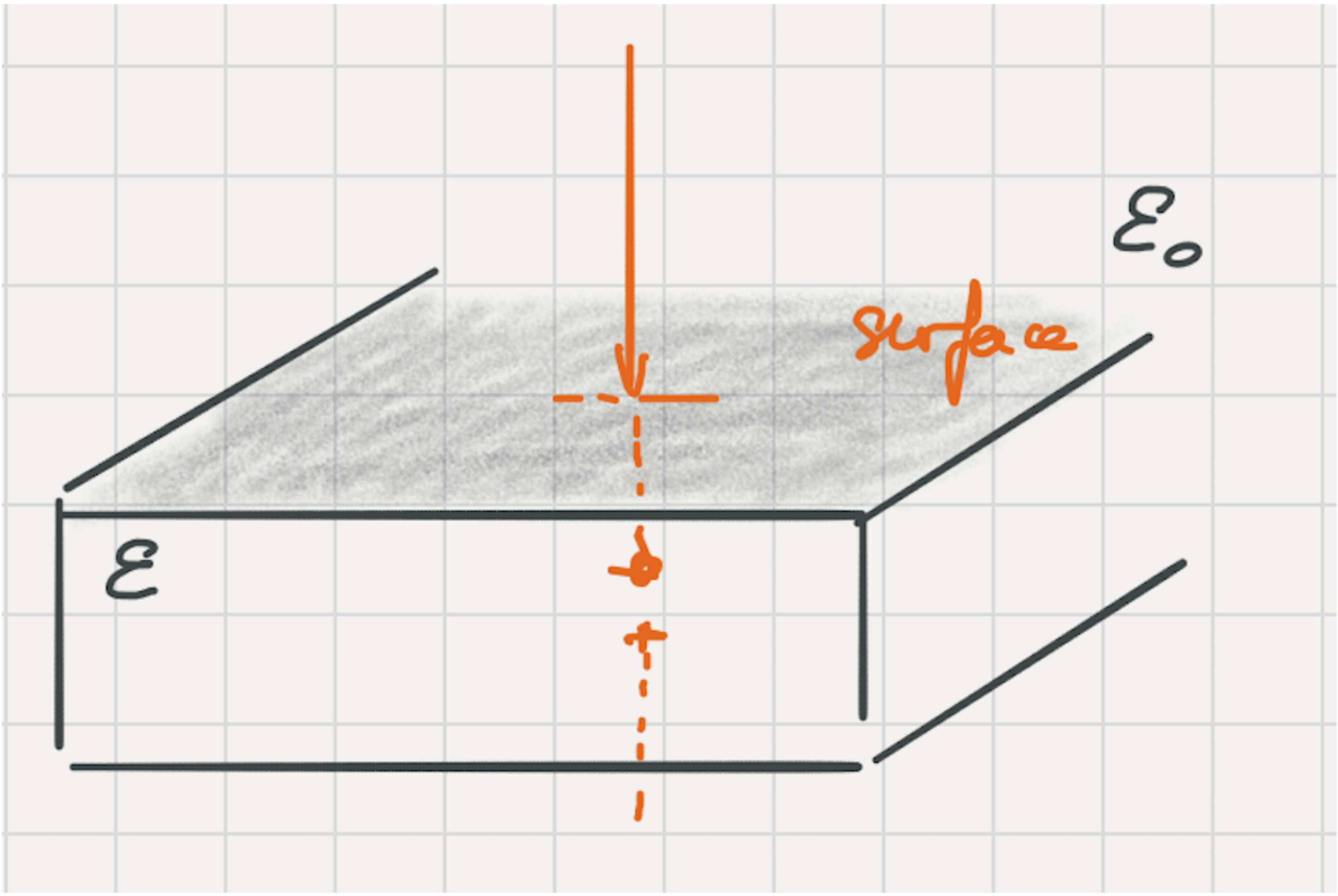
Maxwell + continuity of $\overrightarrow{D}$ across the interface

surface resonance at $\omega_s = \omega_p/\sqrt{2}$

diminution of bulk contribution

Transmission geometry

derivation by Ritchie 1957



Surface effects

Physical review, vol 106, number 5

Plasma Losses by Fast Electrons in Thin Films*

R. H. Ritchie
Health Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee
(Received February 7, 1957)

$$P(k_\perp,\omega)=\frac{e^2}{\pi^2\hbar v^2}\left\{\mathrm{Im}\left(\frac{1}{\epsilon}\right)\frac{a}{(k_\perp{}^2+\omega^2/v^2)}+\frac{2k_\perp}{(k_\perp{}^2+\omega^2/v^2)^2}\,\mathrm{Im}\left[\frac{1-\epsilon}{\epsilon}\cdot\frac{2(\epsilon-1)\cos(\omega a/v)+(\epsilon-1)\exp(-k_\perp a)+(1-\epsilon^2)\exp(k_\perp a)}{(\epsilon-1)^2\exp(-k_\perp a)-(\epsilon+1)^2\exp(k_\perp a)}\right]\right\},$$

where $\epsilon=[1-\omega_p{}^2/(\omega^2-ig\omega)]$.

One notes that the effect of the boundary is to cause a decrease in loss at the plasma frequency and an additional loss at $\omega=\omega_p/\sqrt{2}$. Call the probabilities for these

Surface effects

Separation of bulk and surface-losses in low-loss EELS measurements in STEM

K.A. Mkhoyan[a,*], T. Babinec[a,b], S.E. Maccagnano[a], E.J. Kirkland[a], J. Silcox[a]

Ultramicroscopy 107 (2007) 345–355

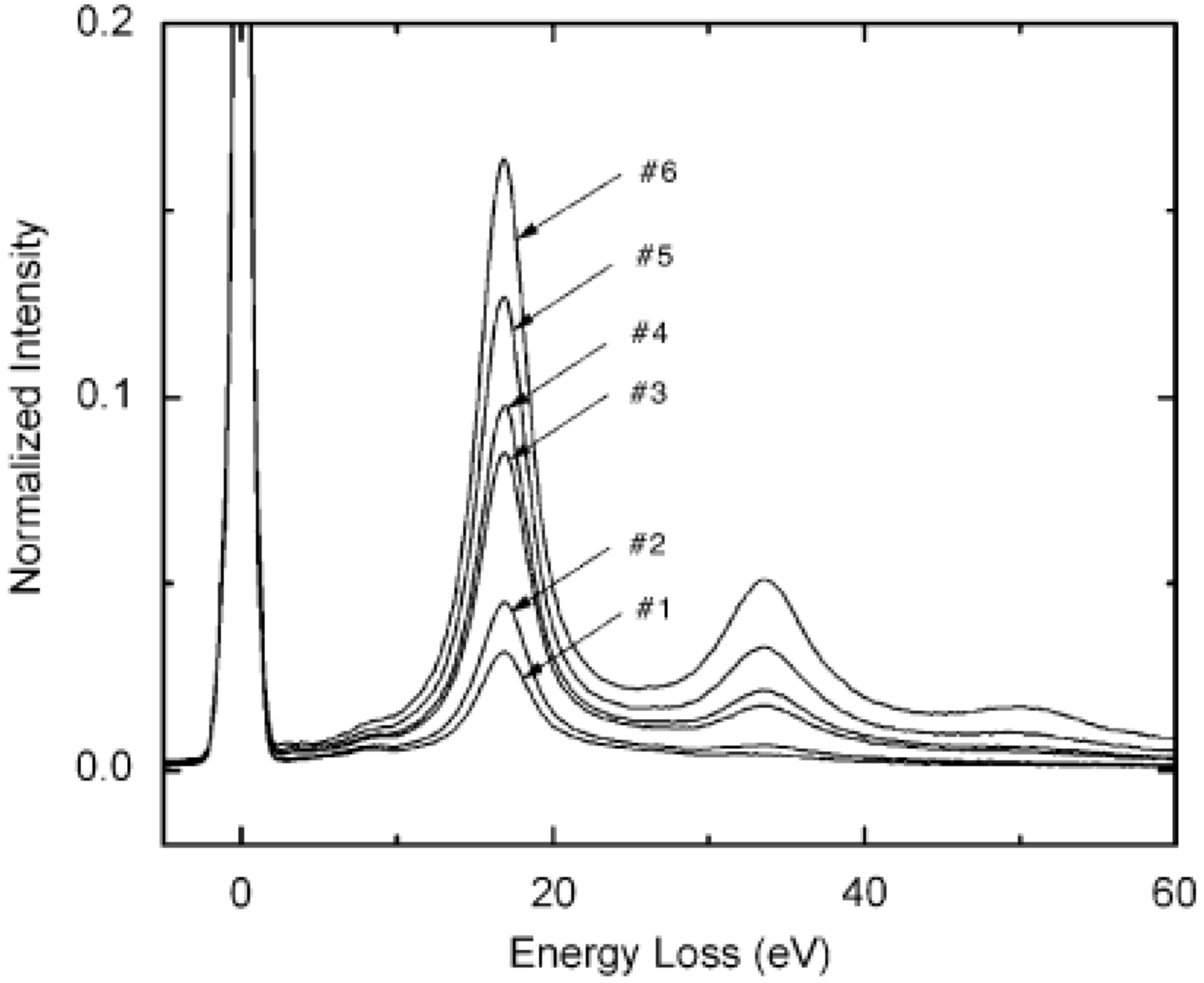


Fig. 2. Six low-loss EELS data recorded from a Si sample at different thicknesses. Spectra are scaled to have the same zero-loss peak intensity (normalized to one). The labels are from the thinnest, #1, to the thickest, #6.

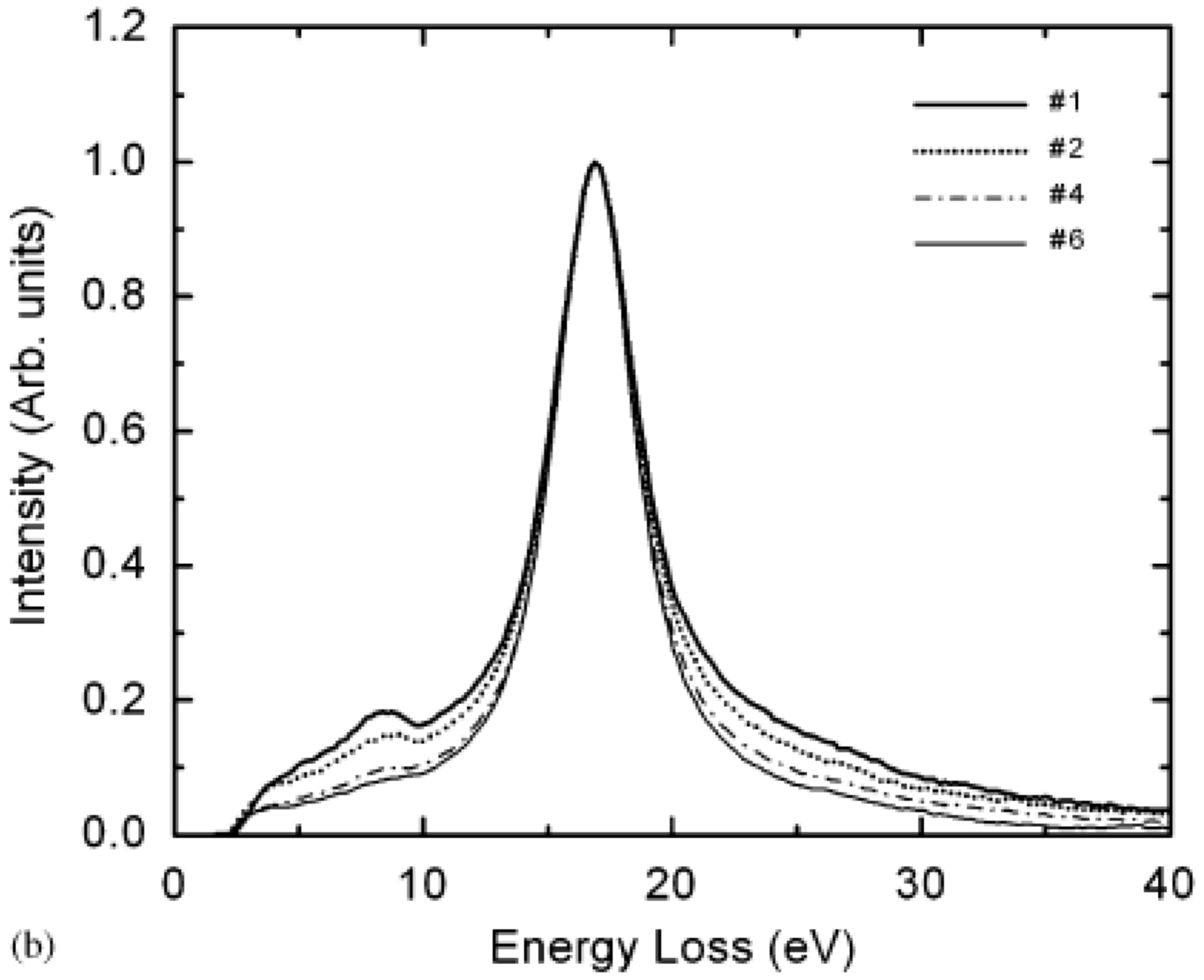


Fig. 4. (a) Low-loss spectra from the silicon sample before and after Fourier-log deconvolution: (b) the single scattering distributions obtained for four different thicknesses. The original EELS data are presented in Fig. 2. The spectra are scaled to the peak of the bulk plasmon-loss.

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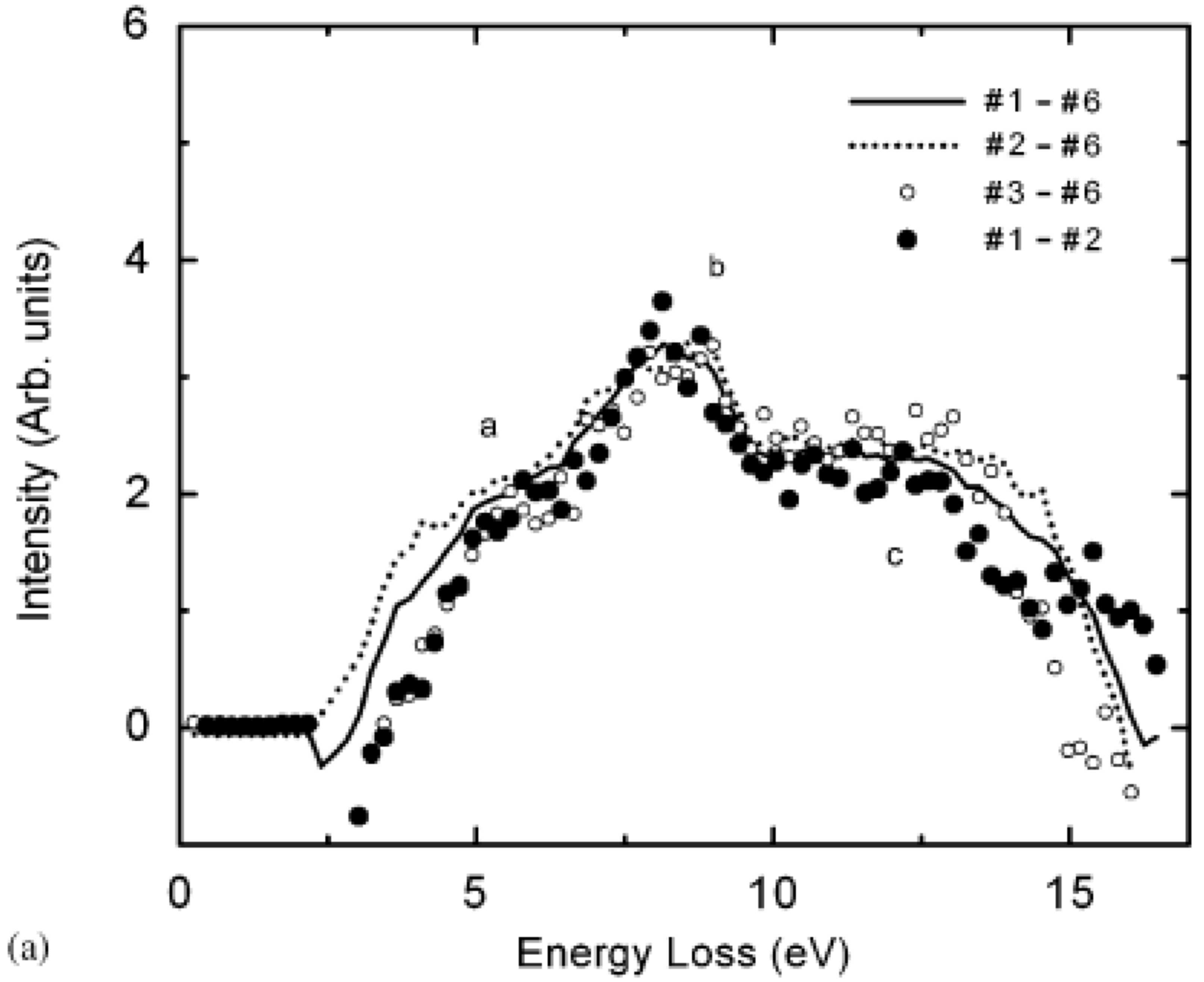
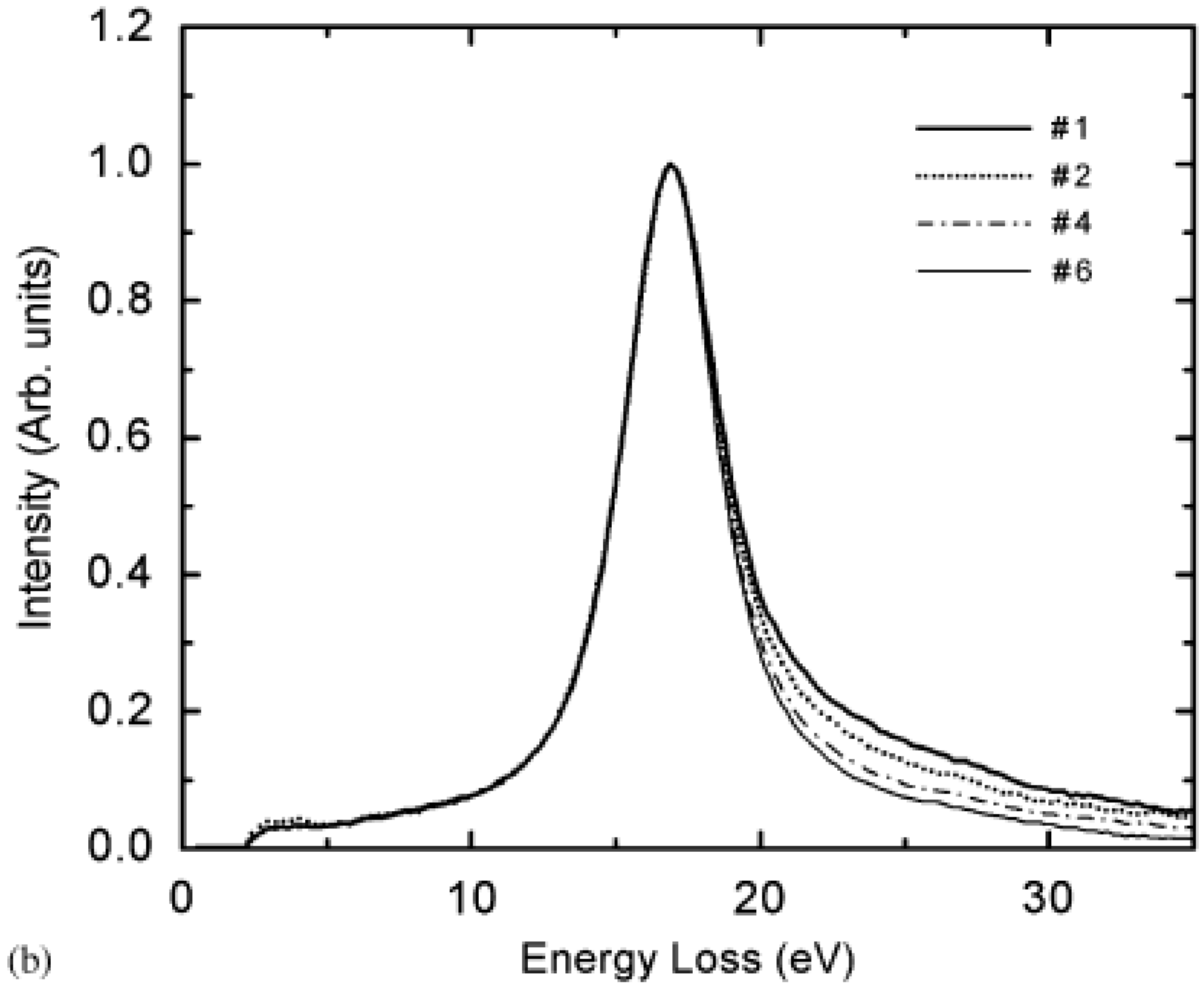
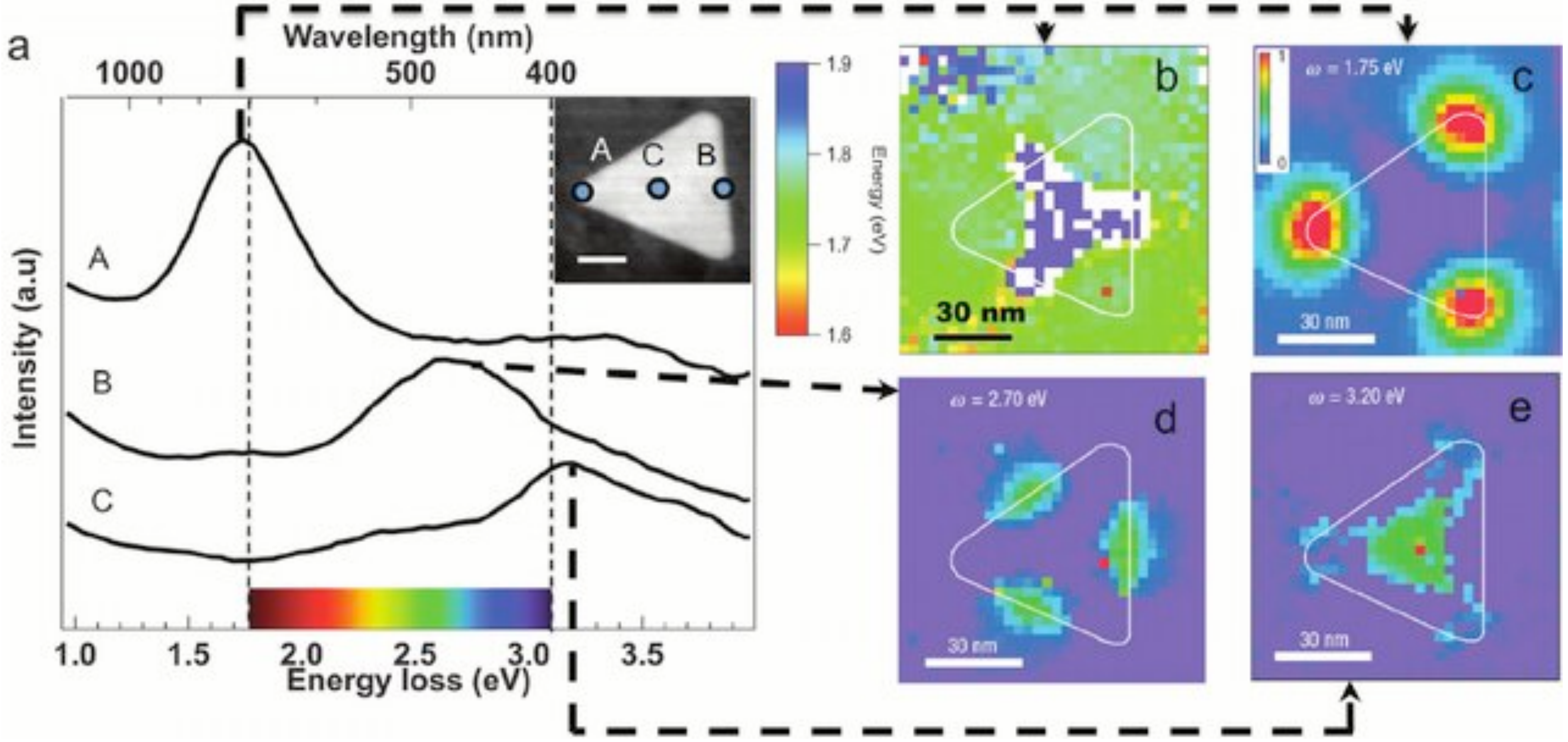


(a)



(b)

Plasmon losses in nanoparticles



… next week

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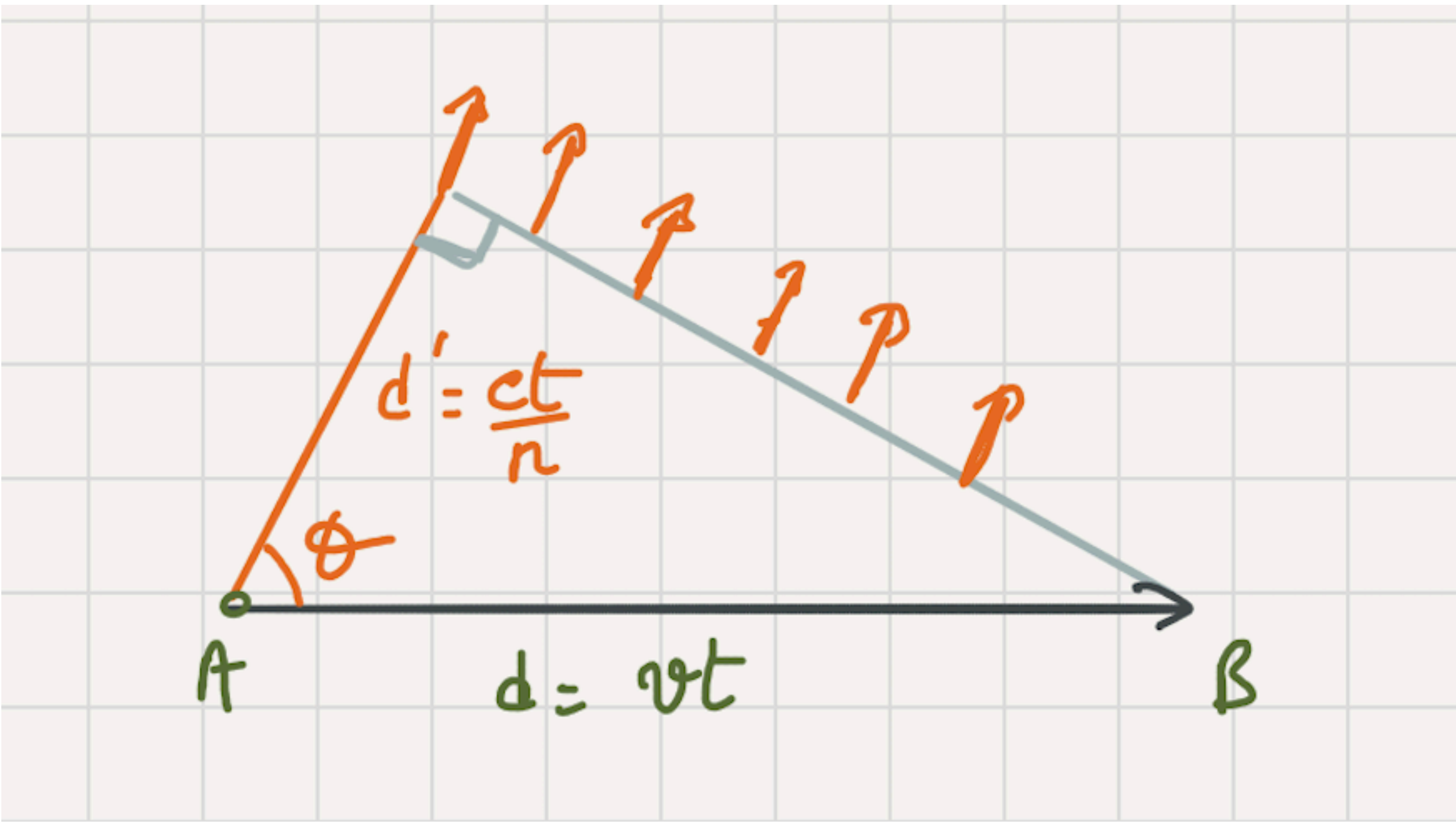
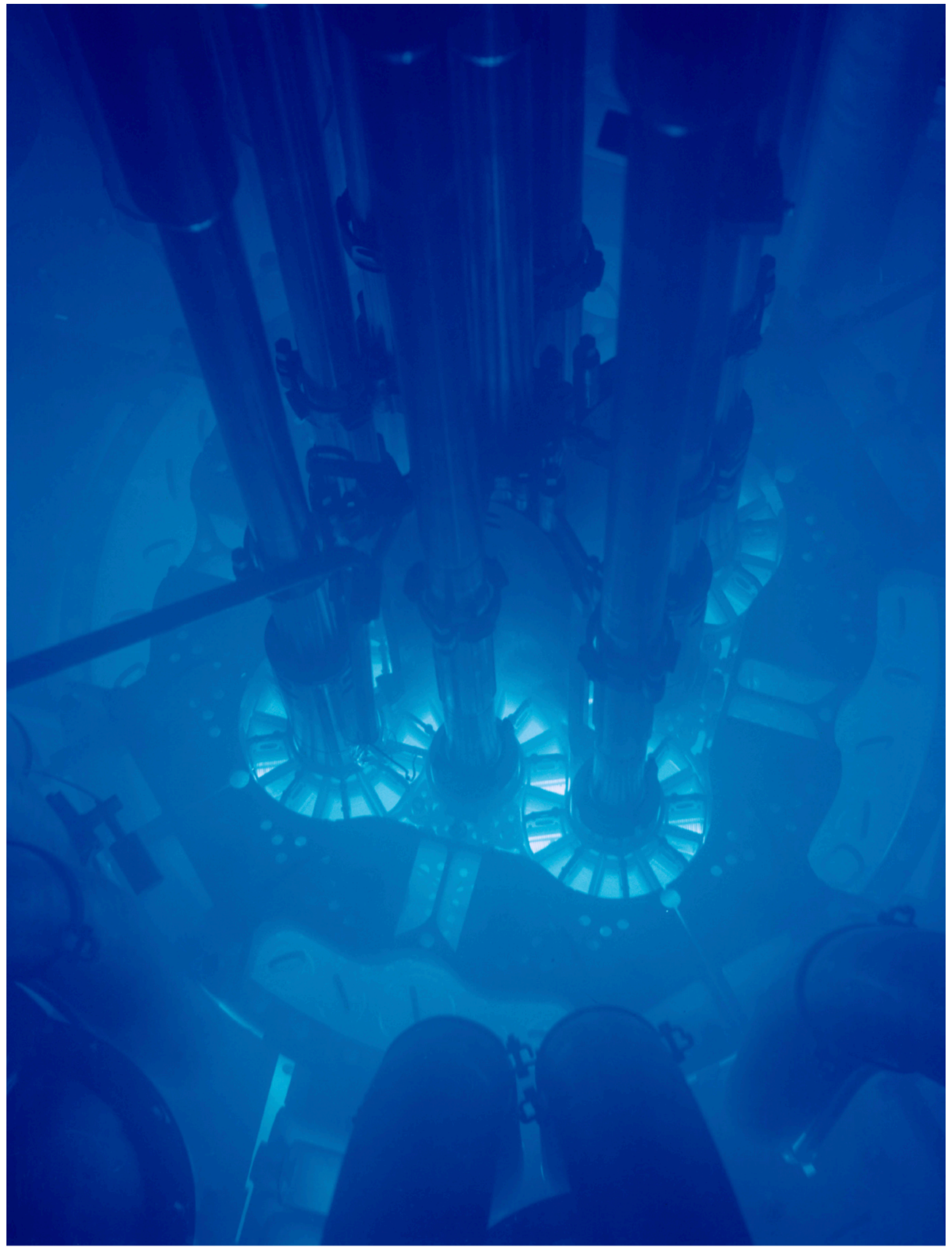
Cerenkov losses

A charged particle traveling faster than the phase velocity of light in a medium emits radiation





Cerenkov losses

Formulation: Kröger, 1968

$$\frac{\partial^2 P_{\mathrm{nr}}}{\partial E \partial \Omega}(E,\theta) = \left(\frac{\mathrm{e}^3}{4\pi^3\hbar^2\varepsilon_0 v^2}\right) \times \mathrm{Im}\left(\frac{t}{\hat{\varepsilon}\varphi^2} - \frac{2\theta^2(\hat{\varepsilon}-\hat{\eta})^2}{\hat{\varepsilon}\hat{\eta}\varphi^4}\frac{\hbar}{m_{\mathrm{e}}v}\left[\frac{\sin^2 d_{\mathrm{e}}}{L^+} + \frac{\cos^2 d_{\mathrm{e}}}{L^-}\right]\right).$$

Zeitschrift für Physik 216, 115—135 (1968)

Berechnung der Energieverluste schneller Elektronen in dünnen Schichten mit Retardierung

E. Kröger

Institut für Angewandte Physik der Universität Hamburg

Eingegangen am 11. Juli 1968

Calculations of the Energy Losses of Fast Electrons in Thin Foils with Retardation

$$\varphi^2 = \lambda^2 + \theta_E^2,$$

$$\lambda^2 = \theta^2 - \hat{\varepsilon}\theta_E^2\beta^2,$$

$$\lambda_0^2 = \theta^2 - \hat{\eta}\theta_E^2\beta^2,$$

$$d_{\mathrm{e}} = (tE)/(2\hbar v),$$

$$L^+ = \lambda_0\hat{\varepsilon} + \lambda\hat{\eta}\tanh\left(\frac{\lambda d_{\mathrm{e}}}{\theta_E}\right)$$

and

$$L^- = \lambda_0\hat{\varepsilon} + \lambda\hat{\eta}\coth\left(\frac{\lambda d_{\mathrm{e}}}{\theta_E}\right).$$

Cerenkov losses

The impact of surface and retardation losses on valence electron energy-loss spectroscopy

Rolf Erni[a,*], Nigel D. Browning[b,c]

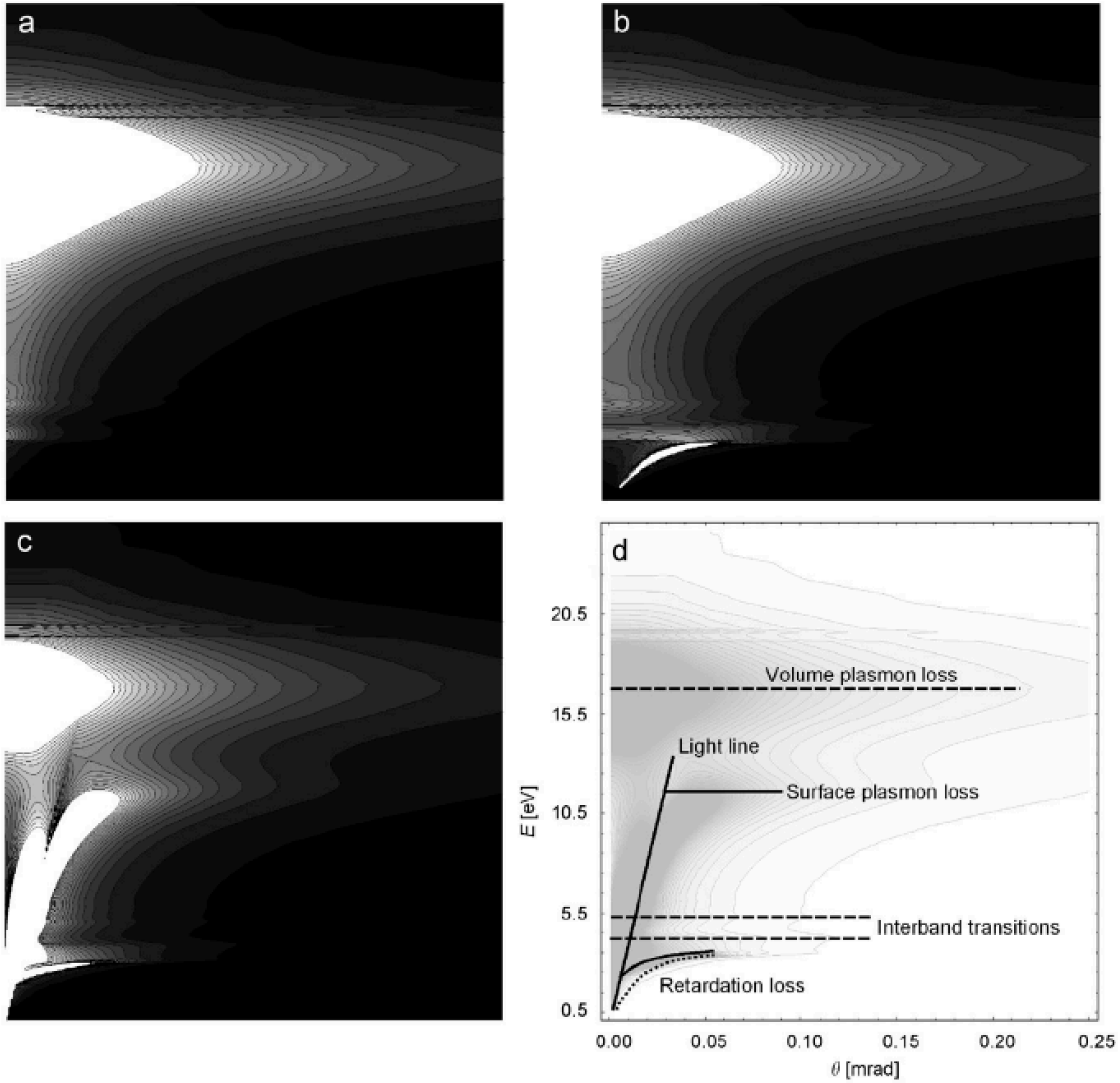
Ultramicroscopy 108 (2008) 84–99

a) volume contribution

b) + Cherenkov

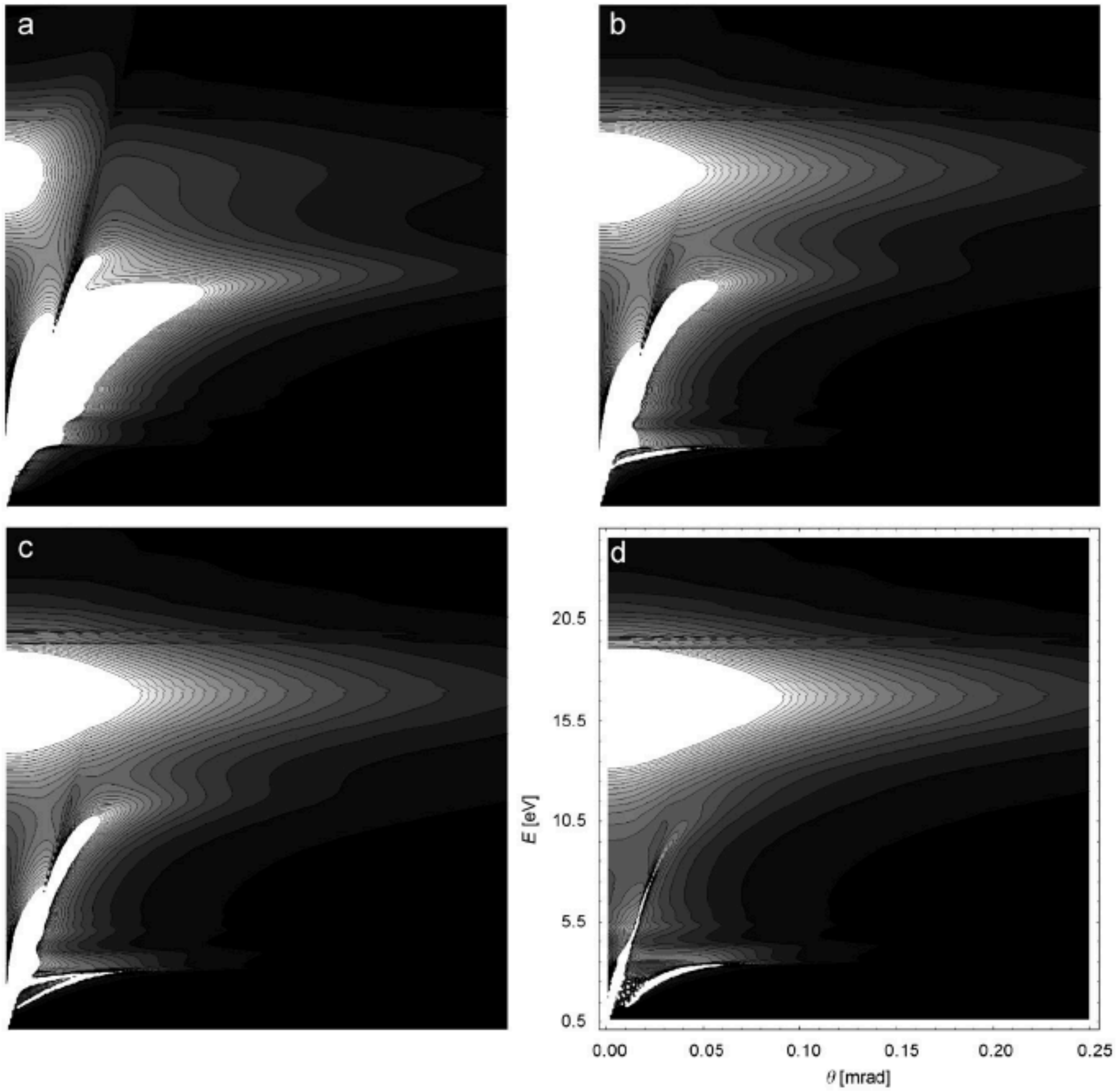
c) + surface term

Si scattering probability calculated using tabulated dielectric function



Cerenkov losses

Si scattering probability calculated using tabulated dielectric function, thickness dependemce: 10, 50, 100 & 1000nm



Cerenkov losses and surface losses

practical issues: kills the dream of obtaining optical properties from EELS in a straightforward manner

Design and application of a relativistic Kramers–Kronig analysis algorithm

Alberto Eljarrat*, Christoph T. Koch

Department of Physics, Humboldt University of Berlin, Newtonstraße 15, Berlin 12489, Germany

Ultramicroscopy 206 (2019) 112825

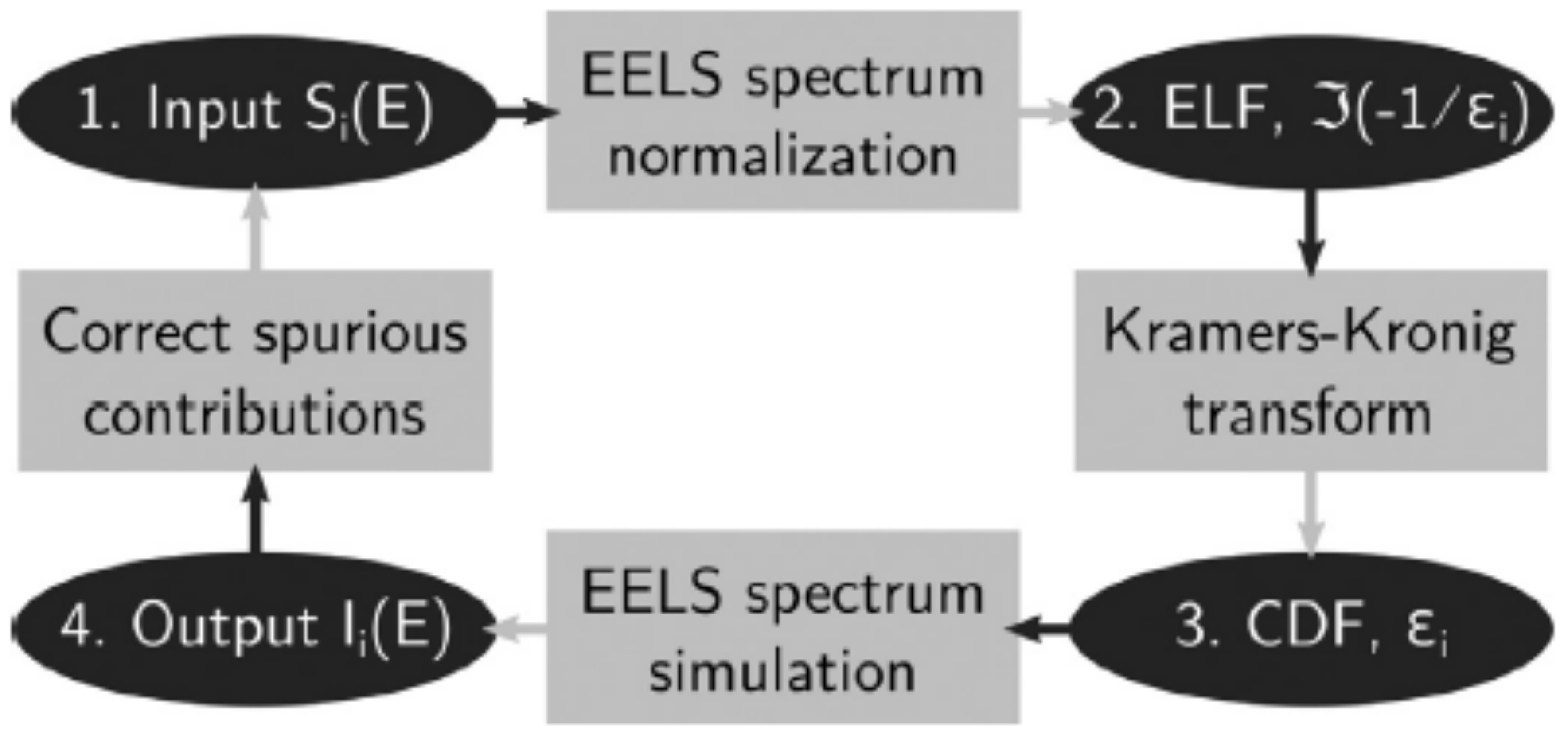


Fig. 3. Diagram showing the individual processing steps of the KKA; starting from the normalization of the spectrum to obtain an estimate of the ELF; the Kramers–Kronig transformation to retrieve the CDF; and simulation of the underlying model to correct the spurious (surface) contributions.

How to retrieve the complex dielectric function ?

Not bullet proof :-/

Combination theory + experiment

including relativistic KKA treatment

communications
materials

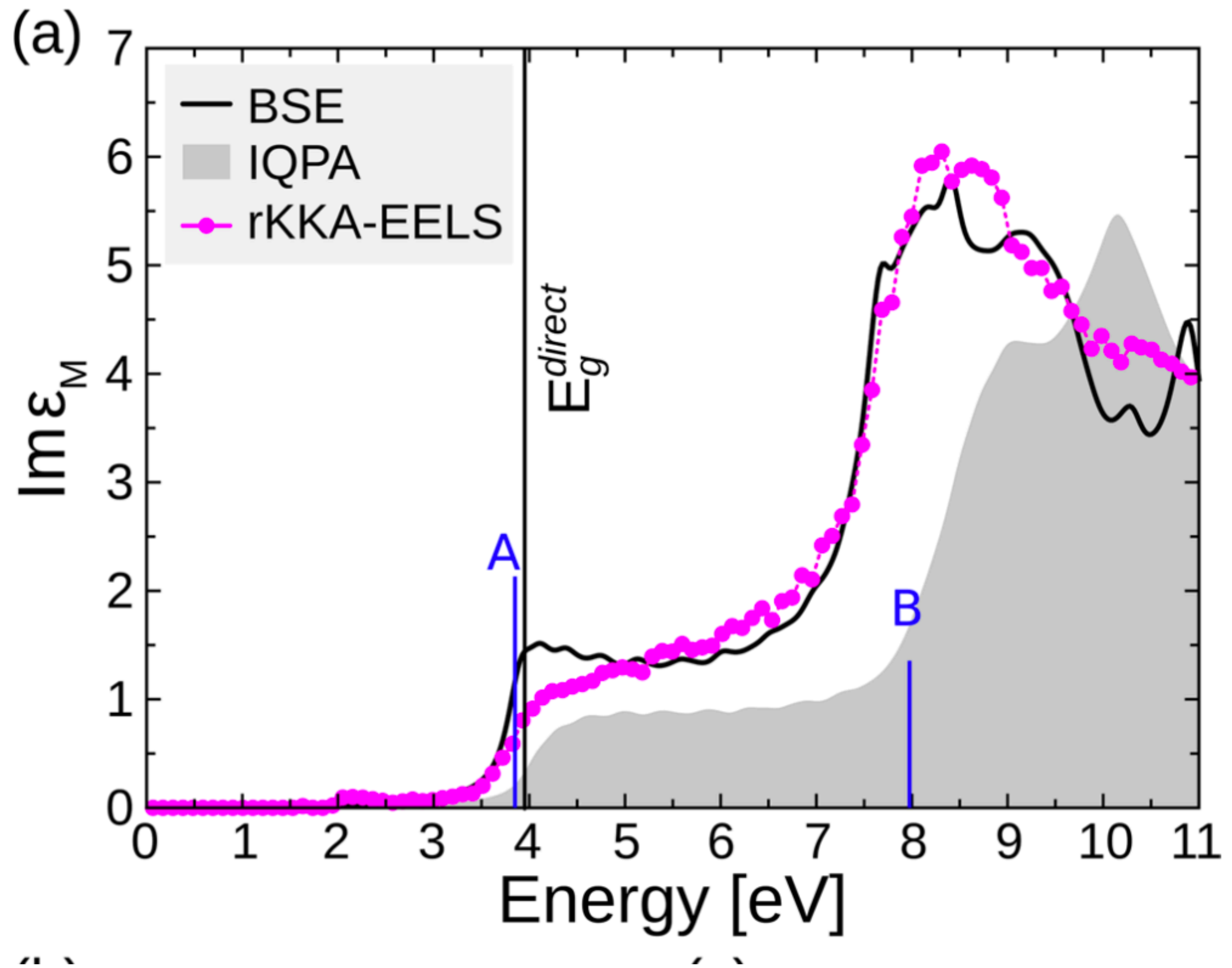
ARTICLE

Check for updates

https://doi.org/10.1038/s43246-022-00234-6 OPEN

A consistent picture of excitations in cubic $BaSnO_3$ revealed by combining theory and experiment

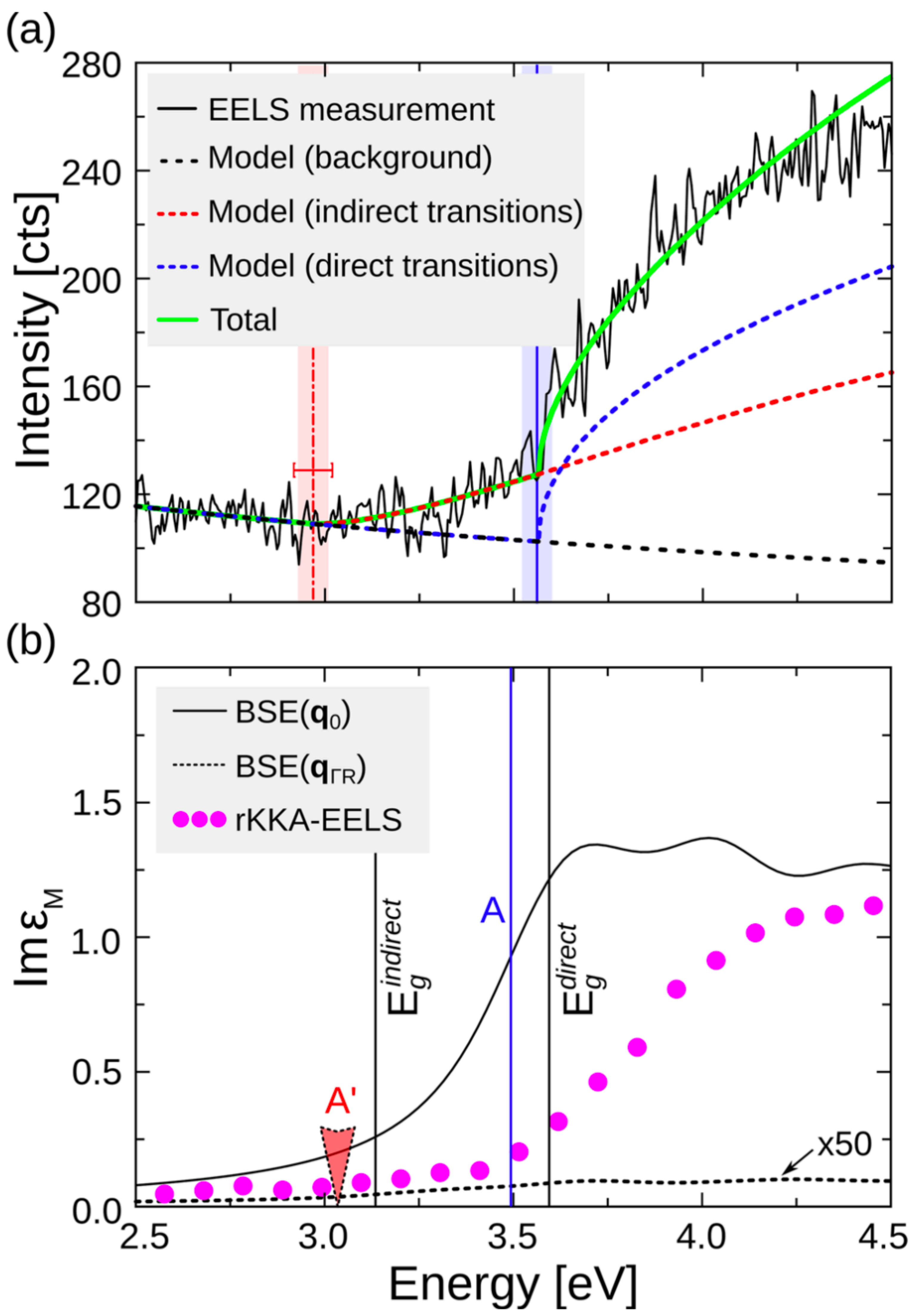
Wahib Aggoune[1,2✉], Alberto Eljarrat[1], Dmitrii Nabok[1], Klaus Irmscher[3], Martina Zupancic[3], Zbigniew Galazka[3], Martin Albrecht[3], Christoph Koch[1] & Claudia Draxl[1,2]



Combination theory + experiment

including relativistic KKA treatment

Fig. 5 Comparison between theory and experiment. a Electron energy-loss spectroscopy (EELS) measurement (black solid line) of a $BaSnO_3$ single crystal in the energy range around the absorption onset. The fits obtained by a model assuming either indirect or direct transitions (see "Methods" section) are shown by the red and blue dashed lines, respectively. The dashed black line refers to the background model while the green solid line shows the sum of the three models. The vertical blue line indicates the onset of direct transitions and the dashed red line that of indirect transitions; the shaded areas represent their error bar. **b** Dielectric function in the energy range of the absorption onset given by Bethe–Salpeter equation (BSE, black solid line) and determined from EELS measurement (magenta circles). The intensity of the BSE spectrum for momentum transfer $\mathbf{q} = \mathbf{q}_{(\Gamma R)} = (1/2,1/2,1/2)$, shown by the dashed line, is scaled by a factor of 50 for better visibility. A Lorentzian broadening of 0.2 eV is applied. The blue line indicates the first direct excitation (optical gap) obtained for $\mathbf{q}_0 = (0,0,0)$ (A), the red arrow refers to the first indirect transition for $\mathbf{q} = \mathbf{q}_{\Gamma R}$ (A′). Both BSE spectra are corrected by a value of -0.367 eV to account for band-gap renormalization due to zero-point vibrations and temperature.



Data availability

Input and output files can be downloaded free of charge from the NOMAD Repository[70] at the following link: https://doi.org/10.17172/NOMAD/2021.05.10-1. The EELS and rKKA-EELS data can be found at https://doi.org/10.17172/NOMAD/2021.11.01-1. The data of the case with optimized lattice parameter can be found at https://doi.org/10.17172/NOMAD/2021.02.25-1.

Code availability

The code `exciting` is available at exciting-code.org.