

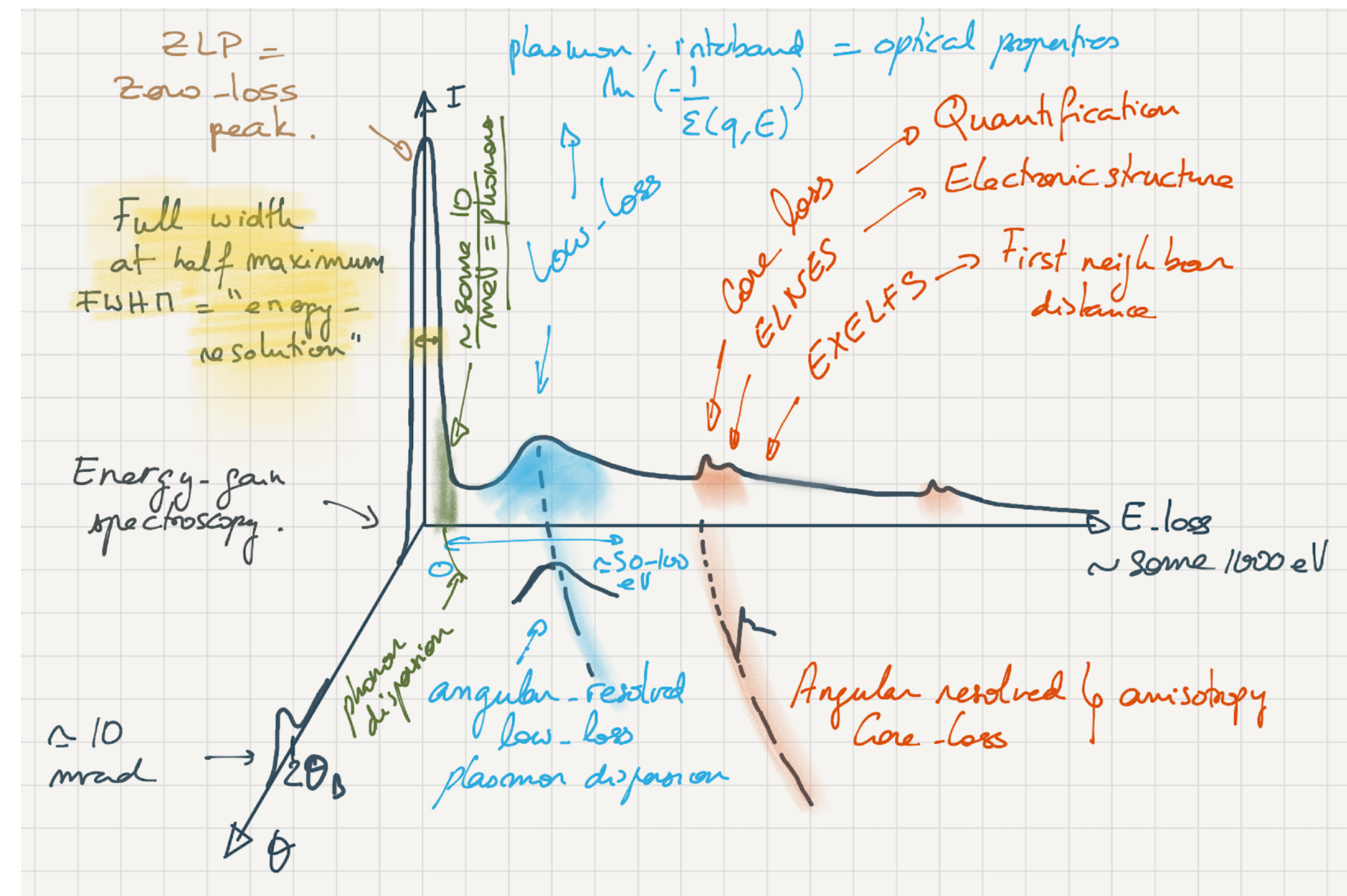
Inelastic scattering - II

Core loss spectroscopy

Cécile Hébert IPHYS-LSME

Layout

- Introduction
- Double differential scattering cross section
- Quantification
- ELNES
- Angular resolved EELS
- Dichroism in the TEM



Dipole approximation

$$\frac{\partial^2 \sigma}{\partial E \partial \Omega} \propto \frac{k'}{k_0} \frac{1}{q^4} \sum_F |\langle I | e^{2\pi i \vec{q} \cdot \vec{R}} | F \rangle|^2 \delta(E_F - E_I - \Delta E)$$

$$\frac{\partial^2 \sigma}{\partial E \partial \Omega} \propto \frac{1}{q^4} \sum_F |\langle I | \vec{q} \cdot \vec{R} | F \rangle|^2 \delta(E_F - E_I - \Delta E)$$

Dipole selection rule. Usually OK.

Dipole approximation: link with XANES

$$\frac{\partial^2 \sigma}{\partial E \partial \Omega} \propto \frac{1}{q^4} \sum_F |\langle I | \vec{q} \cdot \vec{R} | F \rangle|^2 \delta(E_F - E_I - \Delta E)$$

Equivalent to the dipole transition in XANES, with $\vec{q} < - > \vec{e}$ polarisation vector

Angular dependence of X-ray absorption spectra

C Brouder , Journal of Physics: Condensed Matter, Volume 2, Number 3, 1990

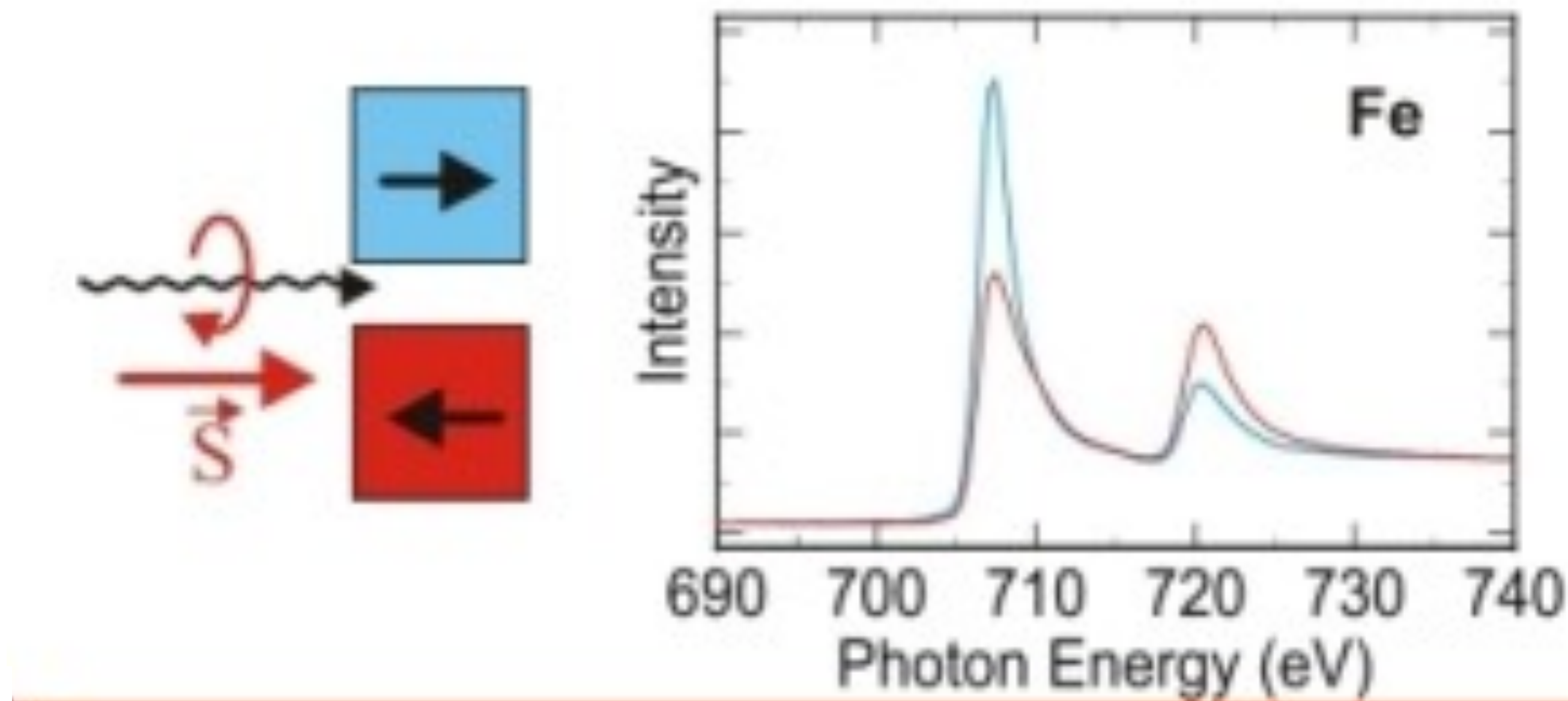
A1.1. Dipole transitions

The dipole absorption cross section can be written

$$\sigma^D(\hat{\epsilon}) = 4\pi^2 \alpha \hbar \omega \sum_{if} |\langle f | \hat{\epsilon} \cdot \mathbf{r} | i \rangle|^2 \delta(E_f - E_i - \hbar \omega) = \sum_{ij} \epsilon_i \epsilon_j \sigma^{ij}. \quad (\text{A1})$$

Magnetic circular dichroism

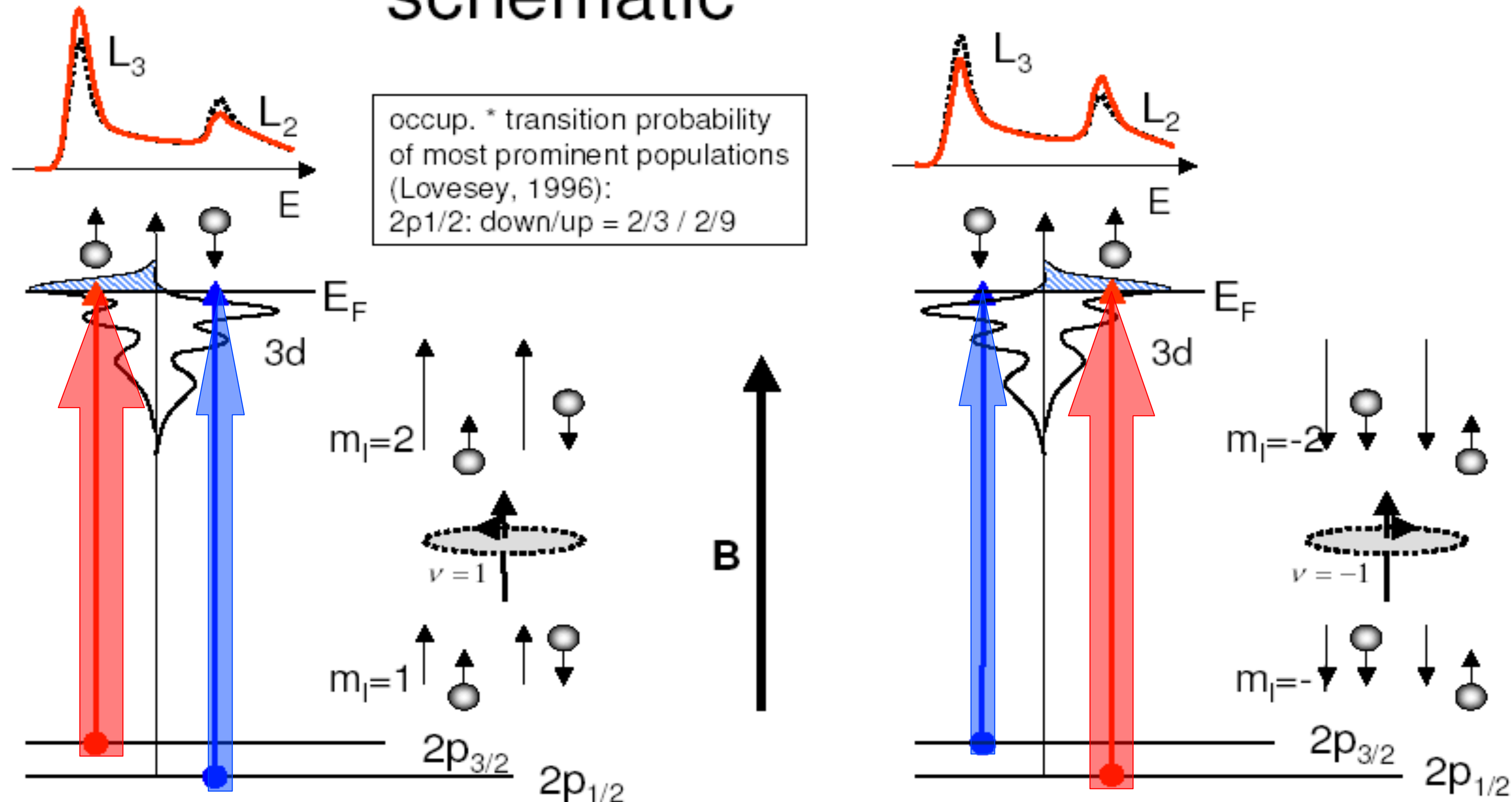
- Prediction: Erskine and Stern, PRB 12, (1975) 737
- Experimental verification (Fe K edge): Schütz et al., PRL 58, (1987) 737
- Breakthrough (Fe L edge): Chen et al. PRL 75, (1995) 152-155.



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Stanford

Magnetic circular dichroism

Circular dichroism in $L_{2,3}$ edges of d-metals schematic



Magnetic circular dichroism

Polarization vector in XANES $\triangleq$ Momentum transfer q in ELNES

$$\vec{\epsilon}_1 + i\vec{\epsilon}_2$$

$$\vec{q} + i\vec{q}'$$

Matrix elements of DFF in dipole approximation:

$$\begin{aligned} \sum_{i,f} |\langle f | (\vec{q} + i\vec{q}') \cdot \hat{R} | i \rangle|^2 = \\ \sum_{i,f} |\langle f | \vec{q} \cdot \hat{R} | i \rangle|^2 + \sum_{i,f} |\langle f | \vec{q}' \cdot \hat{R} | i \rangle|^2 + i \left\{ \sum_{i,f} \langle i | \vec{q}' \cdot \hat{R} | f \rangle \langle f | \vec{q} \cdot \hat{R} | i \rangle - \sum_{i,f} \langle i | \vec{q} \cdot \hat{R} | f \rangle \langle f | \vec{q}' \cdot \hat{R} | i \rangle \right\} \\ \propto \Im \left\{ \text{MDFF}(\vec{q}, \vec{q}', E) \right\} \propto \vec{q} \wedge \vec{q}' \end{aligned}$$

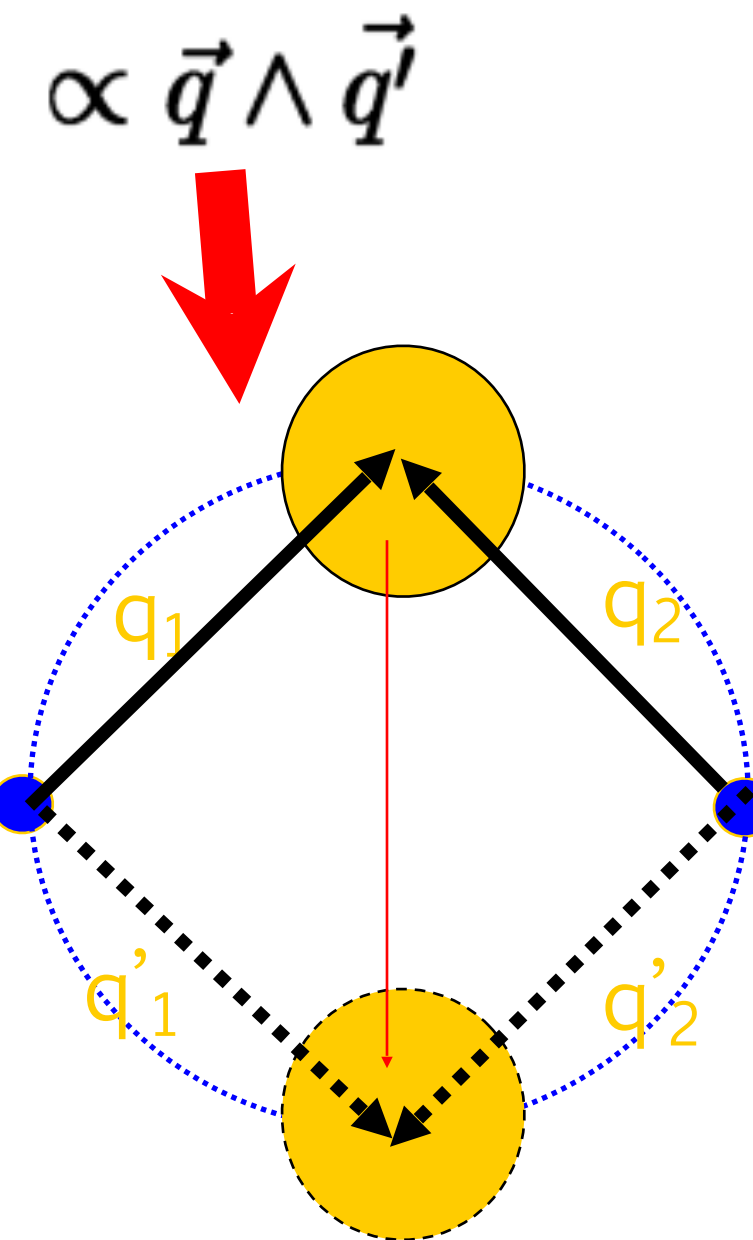
Non zero only for magnetic transitions

Magnetic circular dichroism

$$\sum_{i,f} |\langle f | \vec{q} \cdot \hat{R} | i \rangle|^2 + \sum_{i,f} |\langle f | \vec{q}' \cdot \hat{R} | i \rangle|^2 + i \left\{ \sum_{i,f} \langle i | \vec{q}' \cdot \hat{R} | f \rangle \langle f | \vec{q} \cdot \hat{R} | i \rangle - \sum_{i,f} \langle i | \vec{q} \cdot \hat{R} | f \rangle \langle f | \vec{q}' \cdot \hat{R} | i \rangle \right\}$$

Change phase shift from
+ $\pi/2$ to $-\pi/2$

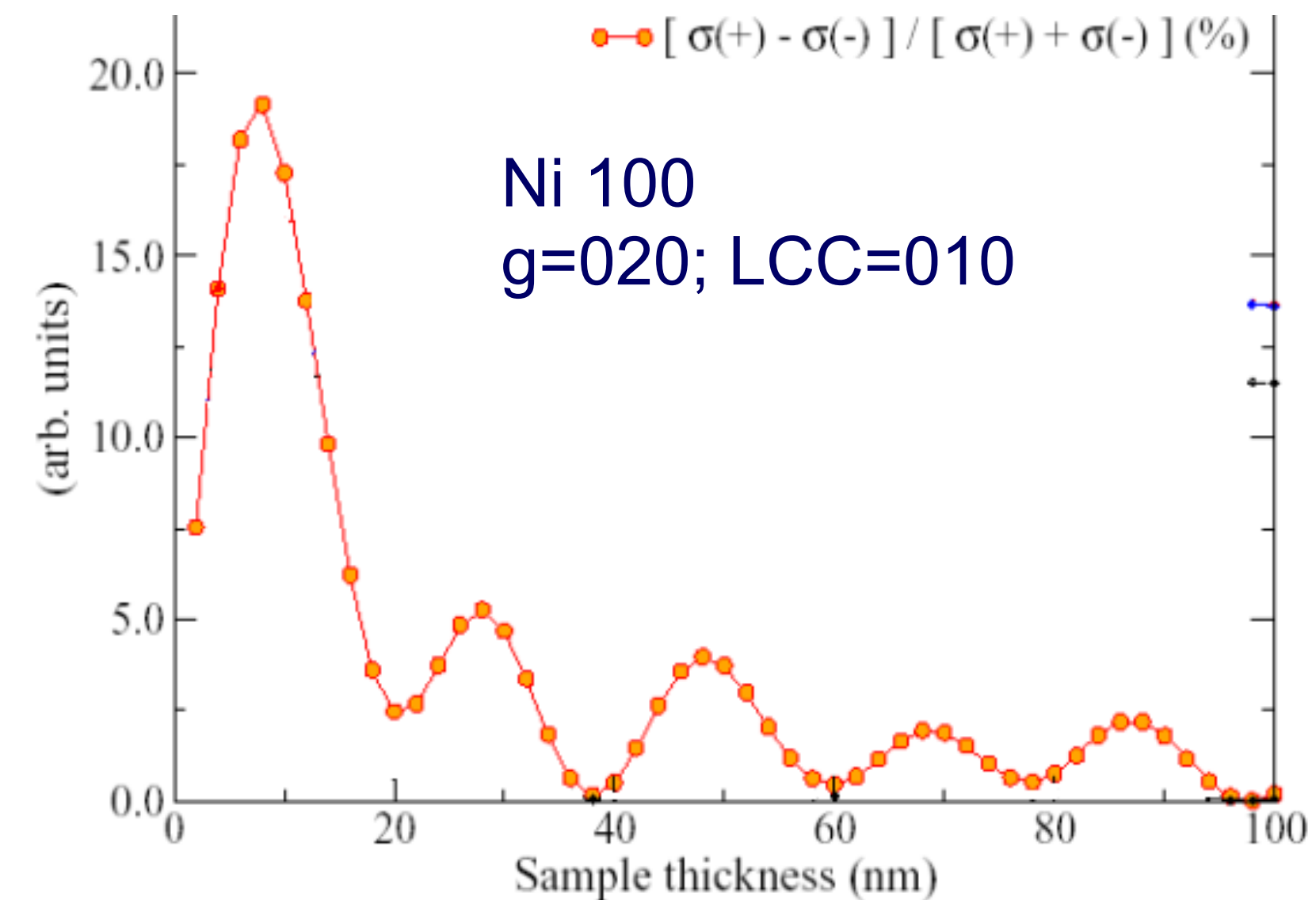
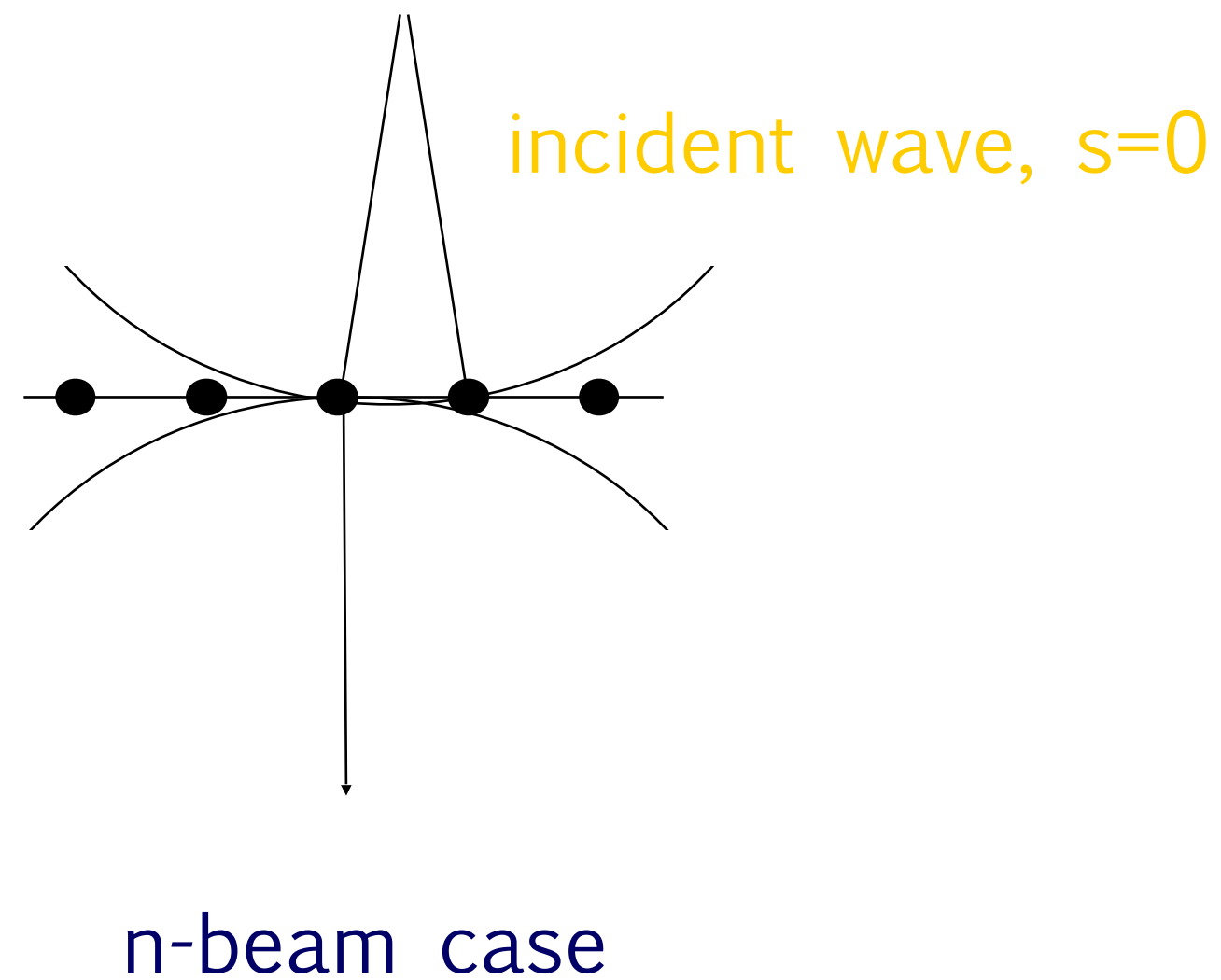
$$\vec{q} + i\vec{q}' \longrightarrow \vec{q} - i\vec{q}'$$



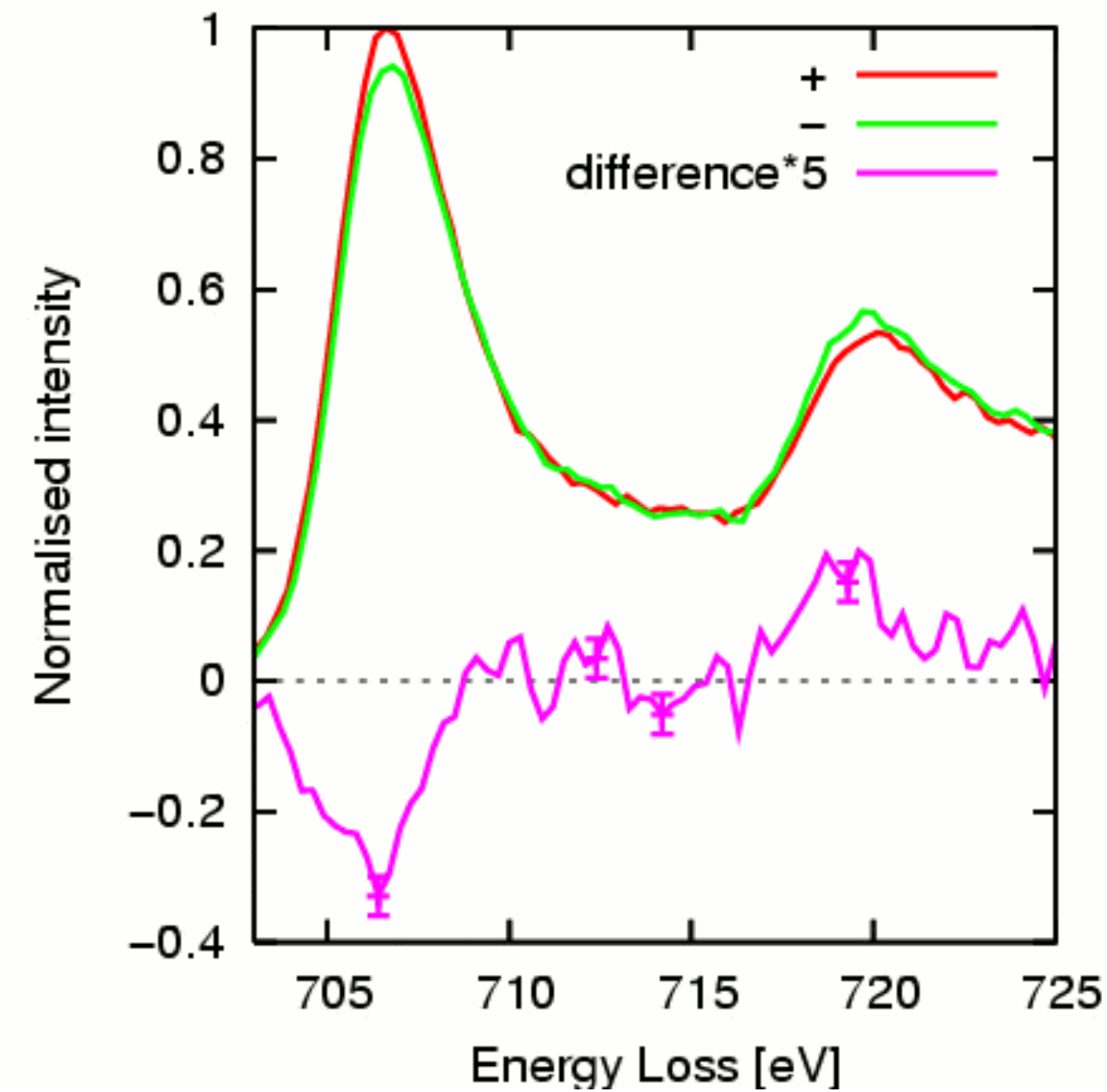
Magnetic circular dichroism

Laue circle centre and thickness determine phase shift

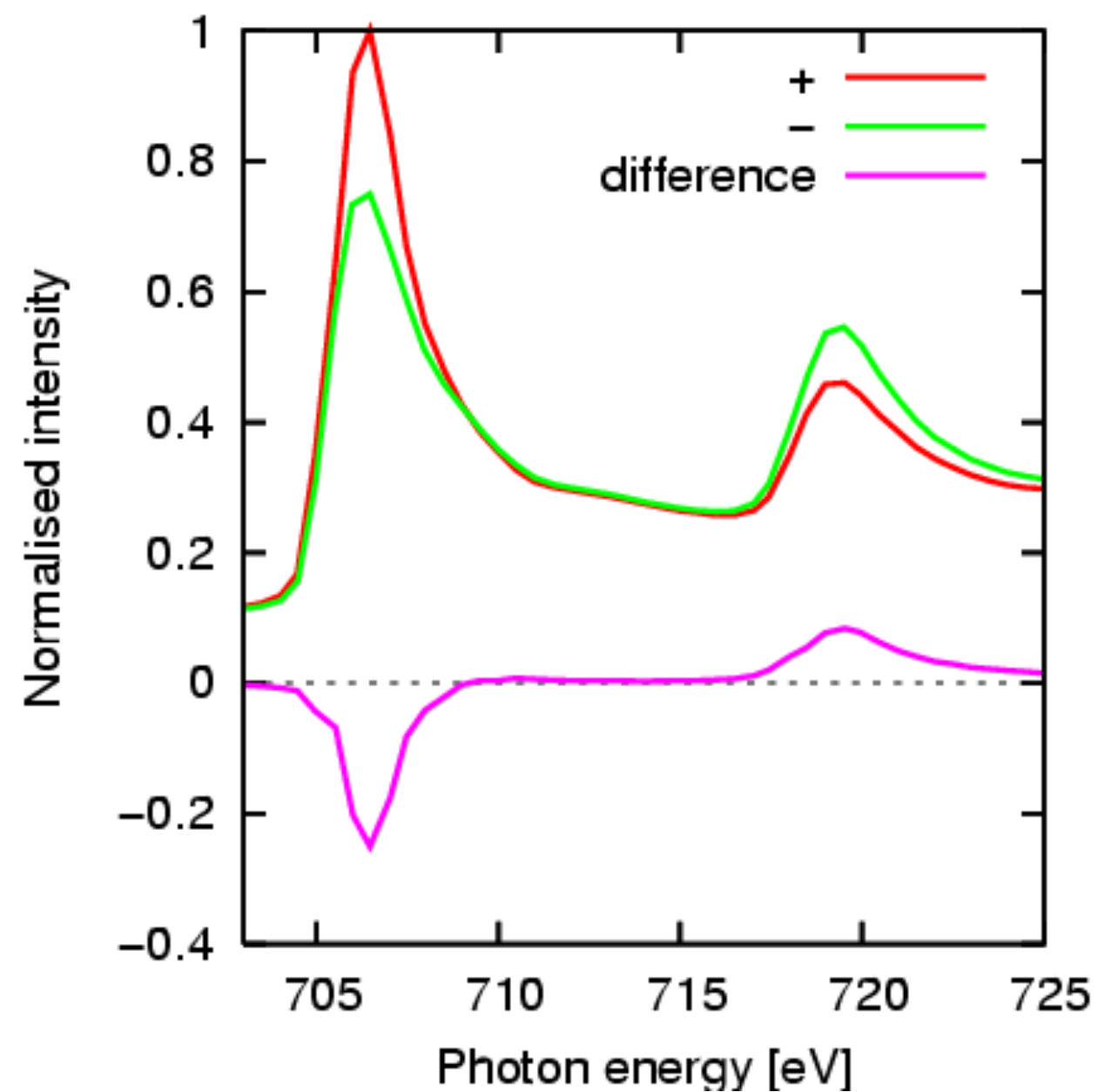
$$\frac{\partial^2 \sigma}{\partial E \partial \Omega} = \frac{4\gamma^2 k_f}{a_0^2 k_i} \left\{ \sum_{j=1}^{2n} \frac{\text{DFF}(\vec{q}_j, E)}{q_j^4} + \sum_{i \neq j} C_{ij} \frac{\text{MDFF}(\vec{q}_i, \vec{q}_j, E)}{q_i^2 q_j^2} \right\}$$



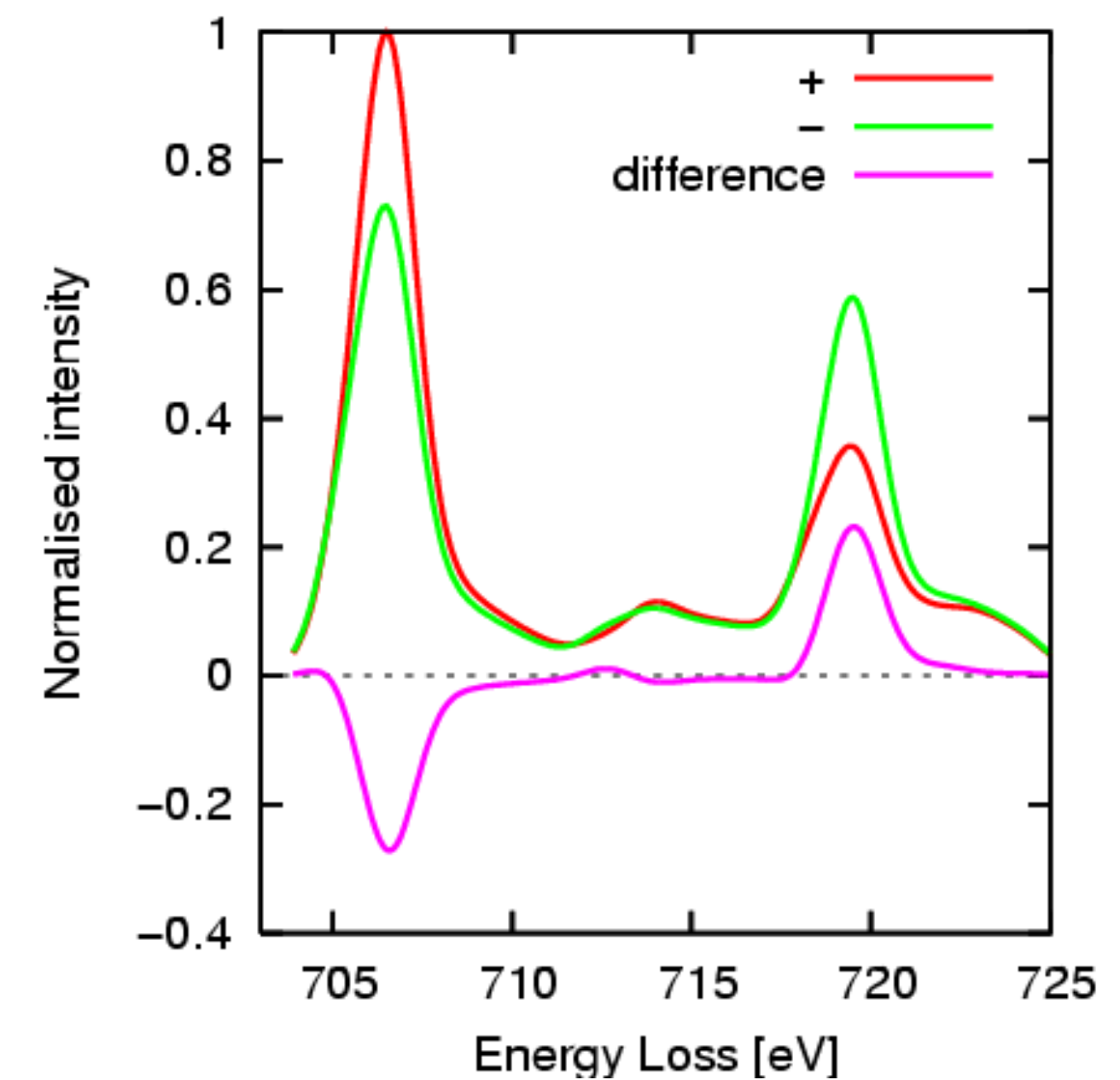
Magnetic circular dichroism



EMCD



XMCD



WIEN2k calc.

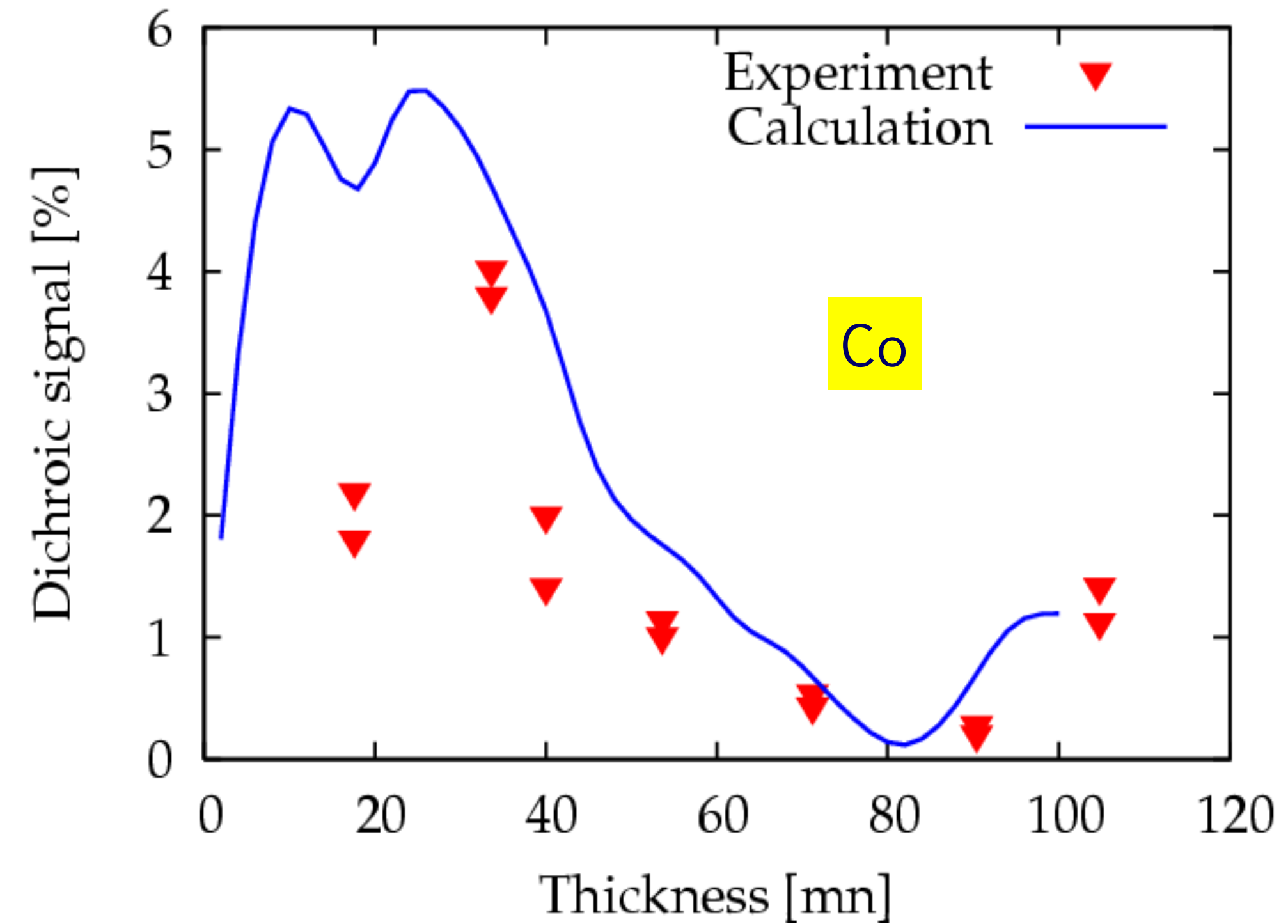
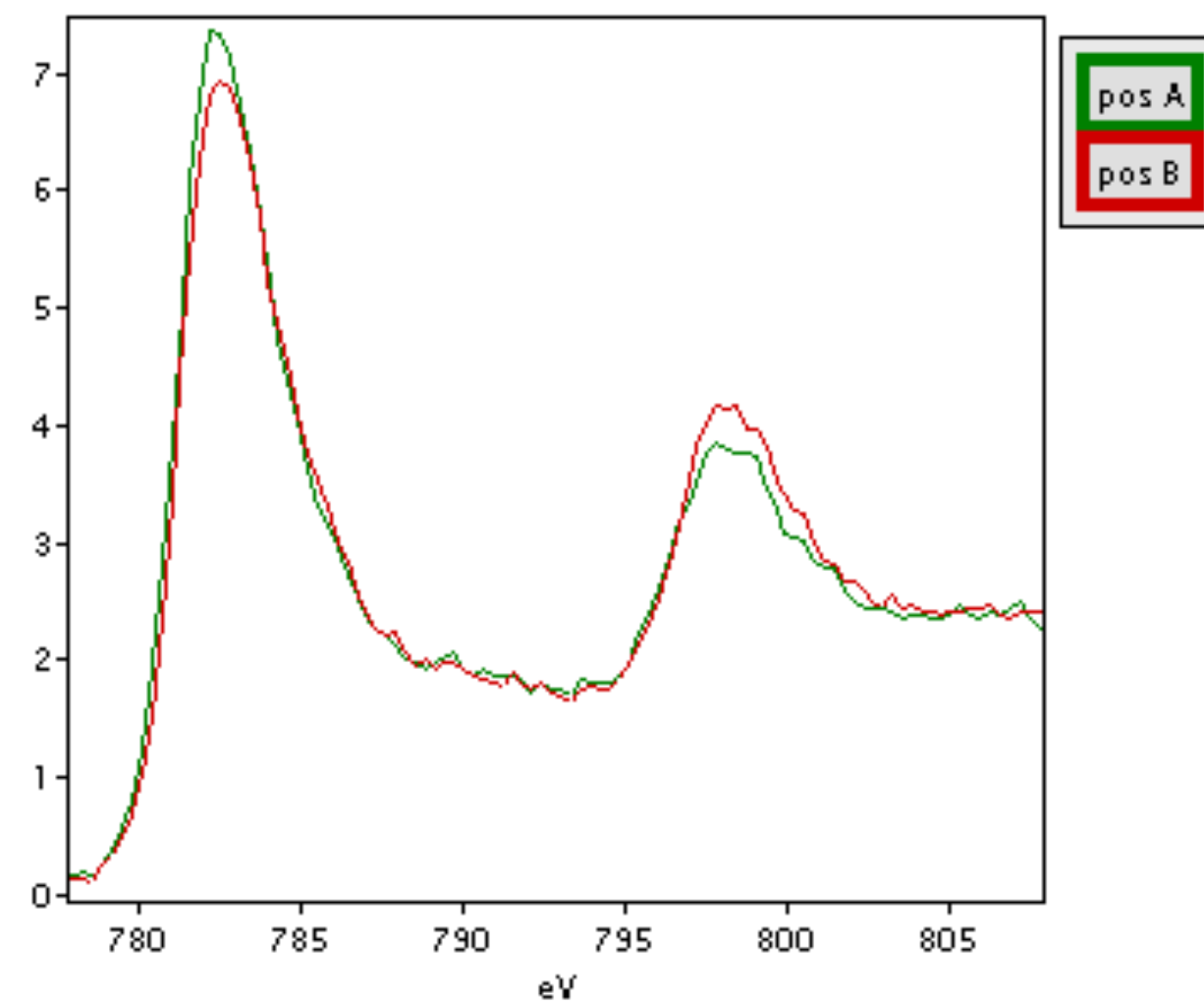
- EMCD (exposure 60 s, 200 nm): S. Rubino, TU Vienna
- Specimen and XMCD: E. Carlino et al., TASC, Elettra
- Calculation: P. Novak et al., Inst. of Physics, Prague

P. Schattschneider et al.,
Nature 441,486-488

Magnetic circular dichroism

Thickness dependence

Cobalt single crystal



2006: Detector shift method
Co [001] $g = 1,0,0$ LCC=1/2,0,0
200 nm lateral resolution
Optimized illumination
60 s acquisition time

Magnetic circular dichroism



Microscopy, 2018, i60–i71

doi: 10.1093/jmicro/dfx129

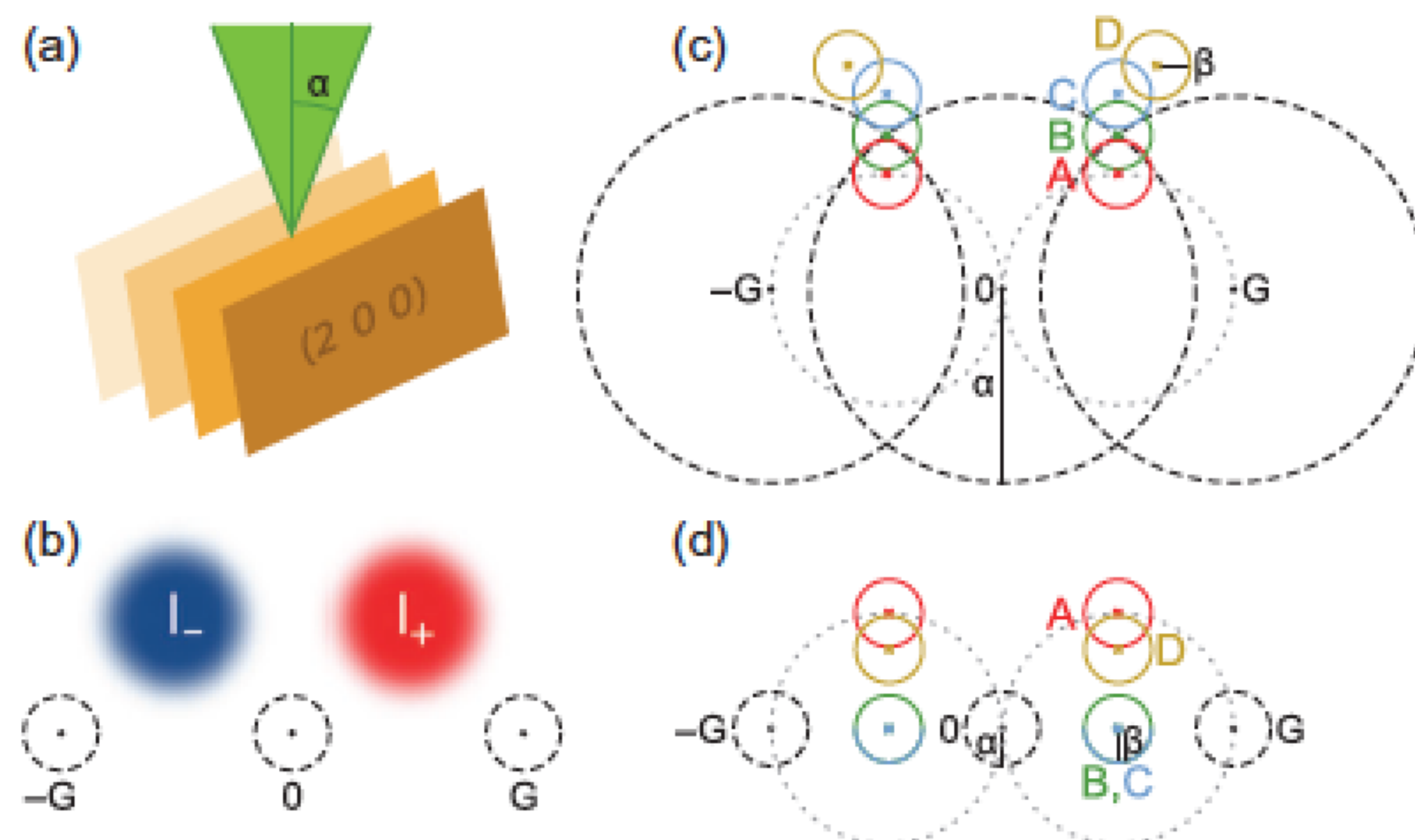
Advance Access Publication Date: 23 January 2018



Article

Convergent-beam EMCD: benefits, pitfalls and applications

S. Löffler^{1,2,*} and W. Hetaba^{1,3}



BCC Fe 10 nm
300 kV
10° from [100]
020 spot (systematic row)

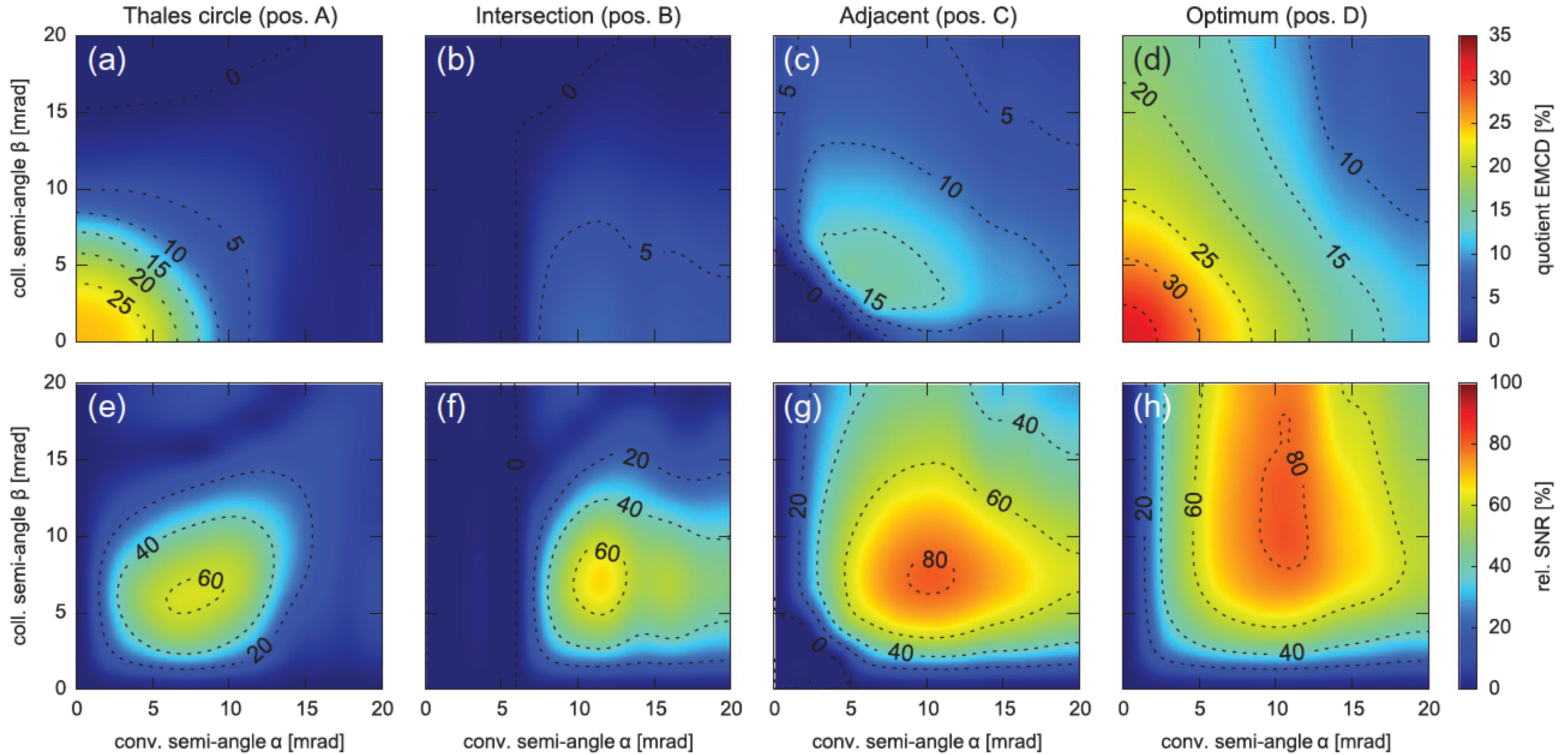


Fig. 4. EMCD effect S (a–d) and SNR $S/\delta S$ (e–h) for the four sets of detector positions A–D as a function of convergence and collection semi-angles. The SNR is given for a jump ratio of $r = 2$ in fractions of the maximum SNR.

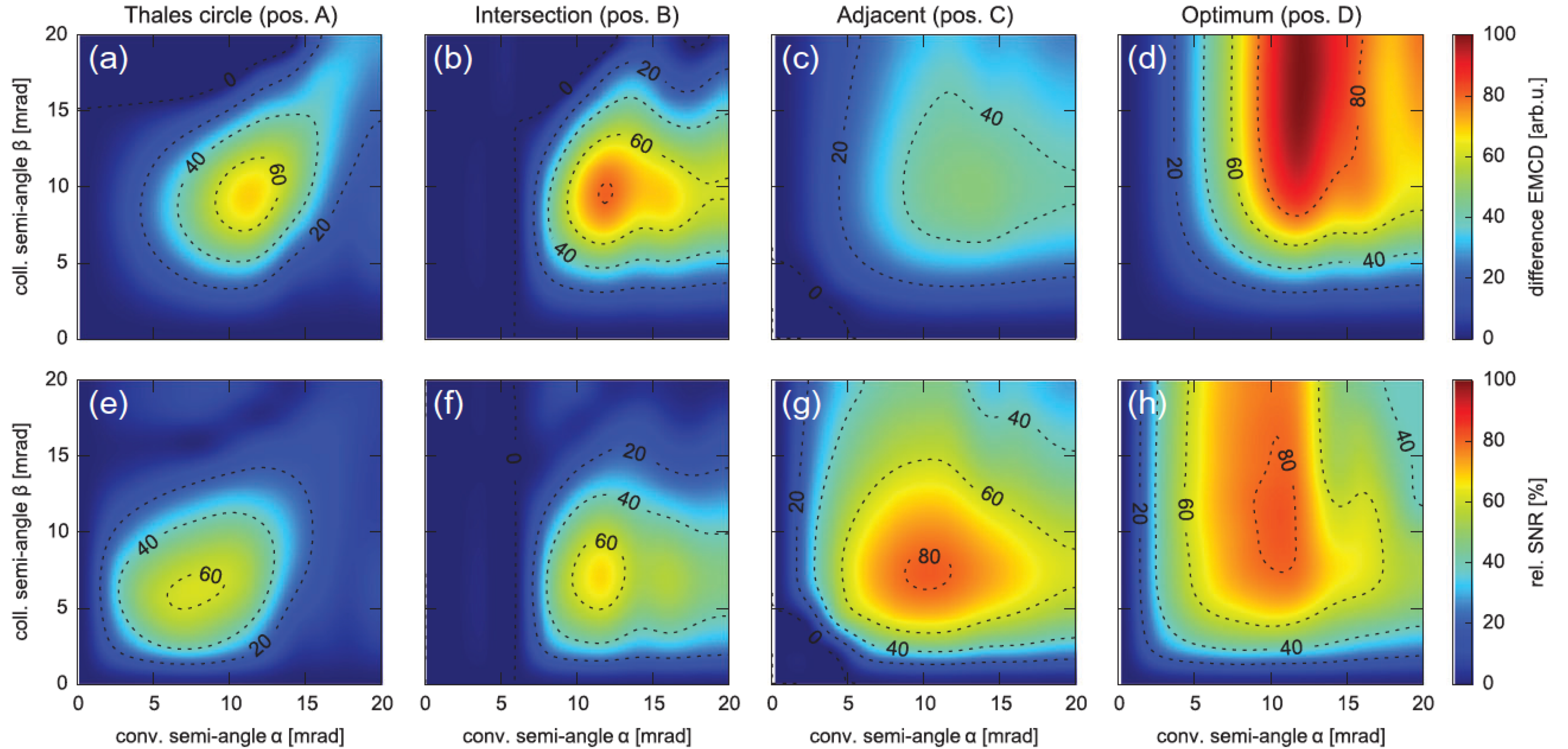


Fig. 3. Difference signal ΔI (a–d) and SNR $\Delta I/\delta\Delta I$ (e–h) for the four sets of detector positions A–D as a function of convergence and collection semi-angles. The SNR is given for a jump ratio of $r = 2$ in fractions of the maximum SNR.

ARTICLE

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OPEN

Magnetic measurements with atomic-plane resolution

Ján Ruzs¹, Shunsuke Muto², Jakob Spiegelberg¹, Roman Adam³, Kazuyoshi Tatsumi², Daniel E. Bürgler³, Peter M. Oppeneer¹ & Claus M. Schneider³

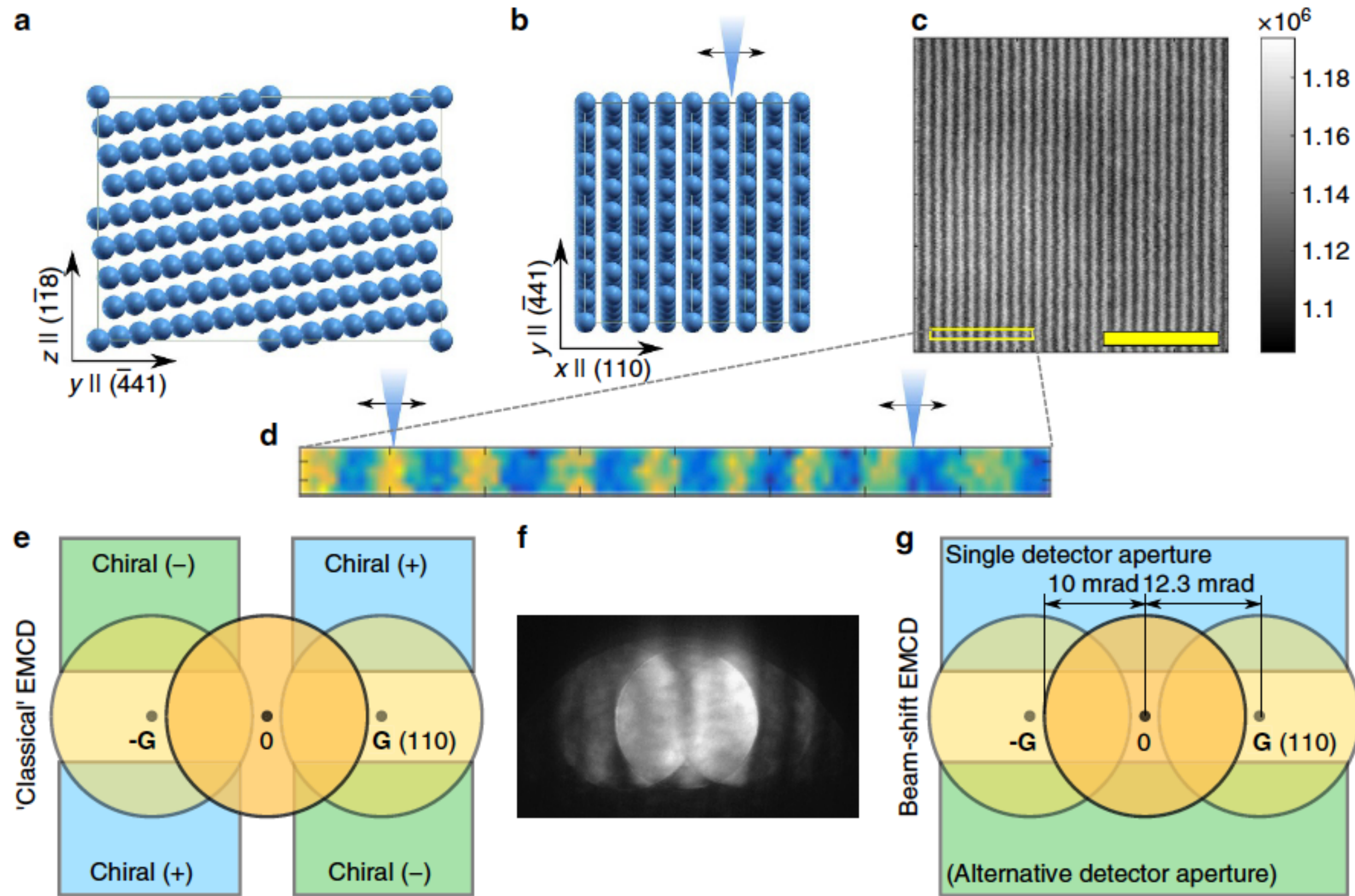


Figure 1 | Scheme of the experimental set-up. (a) Side-view of the structure model of bcc iron tilted into a 3-beam orientation with the beam along the $(1\bar{1}8)$ direction. (b) Top view of the structure model. Scanning direction is schematically shown by blue cone with arrows. (c) ADF survey image (acceleration voltage 200 kV, convergence semi-angle 10 mrad) showing atomic planes. The yellow scale bar denotes 2 Å. (d) ADF image acquired along with the acquisition of a spectrum image. Its size corresponds to the yellow rectangle marked in the ADF survey image. (e) Schematic placement of detectors for a classical EMCD approach in the 3-beam orientation. (f) Experimental diffraction pattern acquired in the same settings as the ADF survey image. (g) Detector placement in the here-proposed APR-EMCD approach. Note that in the classical approach we need two measurements at each beam position to acquire both chiral (+) and chiral (-) spectra, while in the APR-EMCD a single detector aperture is sufficient.

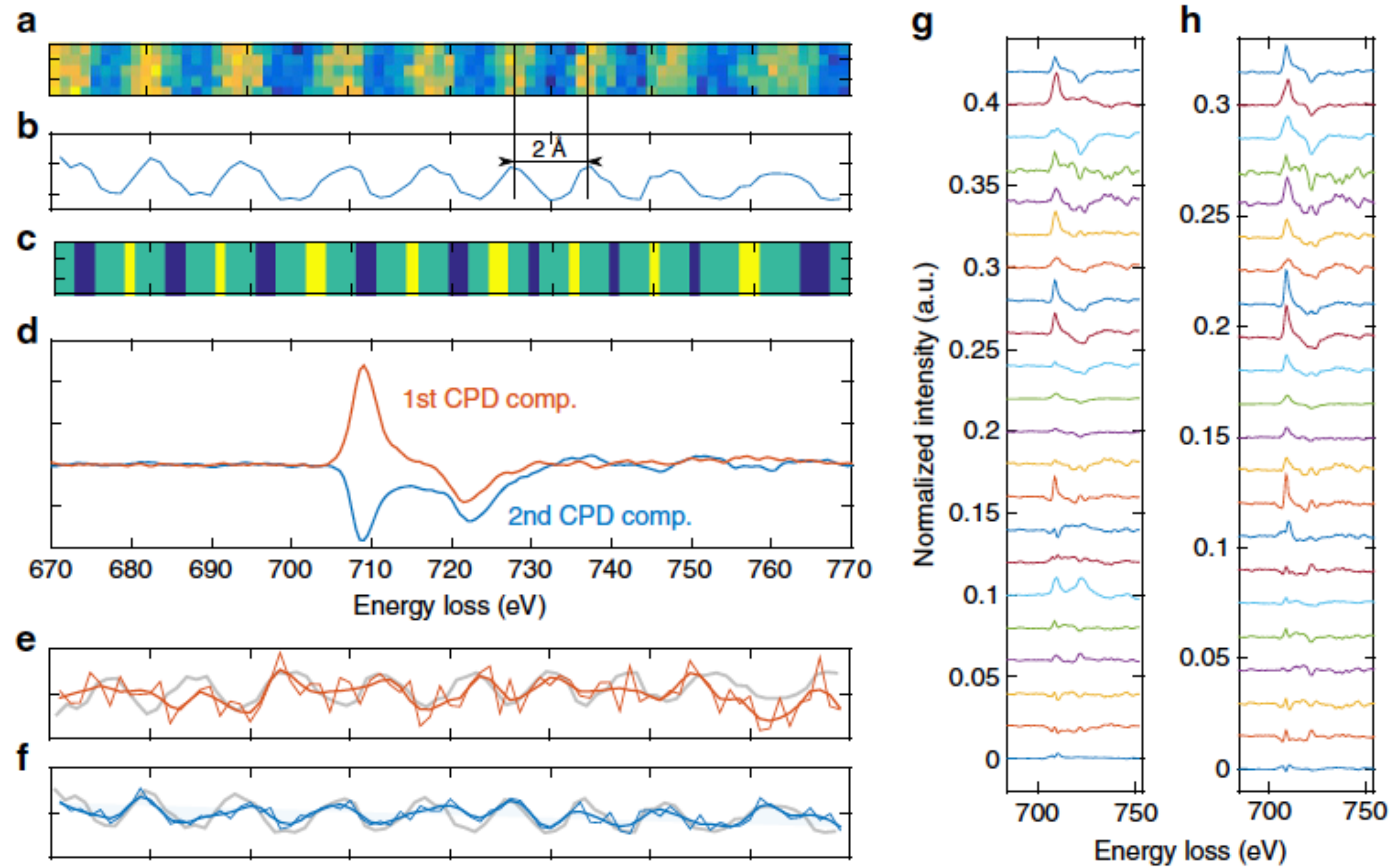


Figure 3 | Experimental detection of APR-EMCD. (a) Representative ADF image of the scanned area and (b) its line profile. A smoothed profile was used to identify individual minima and maxima, based on which a mask has been defined (c) with blue and yellow regions marking summation areas centred at $d/4$ to the right or left of the atomic columns. Their width is determined by local steepness of the ADF line profile. (d) Example of the two CPD components with line profiles of their maps. (e,f) Note the strong correlation between the second CPD component (blue) and the overlaid ADF line profile shown in grey colour. The EMCD-like component also shows such a tendency, but the correlation is weaker. (g) Spectral differences obtained before the subtraction of the second CPD component and (h) after the subtraction. Note how the subtraction of the second CPD component leads to a higher number of spectra showing clear EMCD signature in (h). Spectral differences were calculated from scaled spectra, so that their average peak value at L_3 edge equals to one. In g and h, the individual spectral differences were vertically offset by 0.02 and 0.015, respectively.

Inelastic scattering - III

Low loss spectroscopy

Cécile Hébert IPHYS-LSME

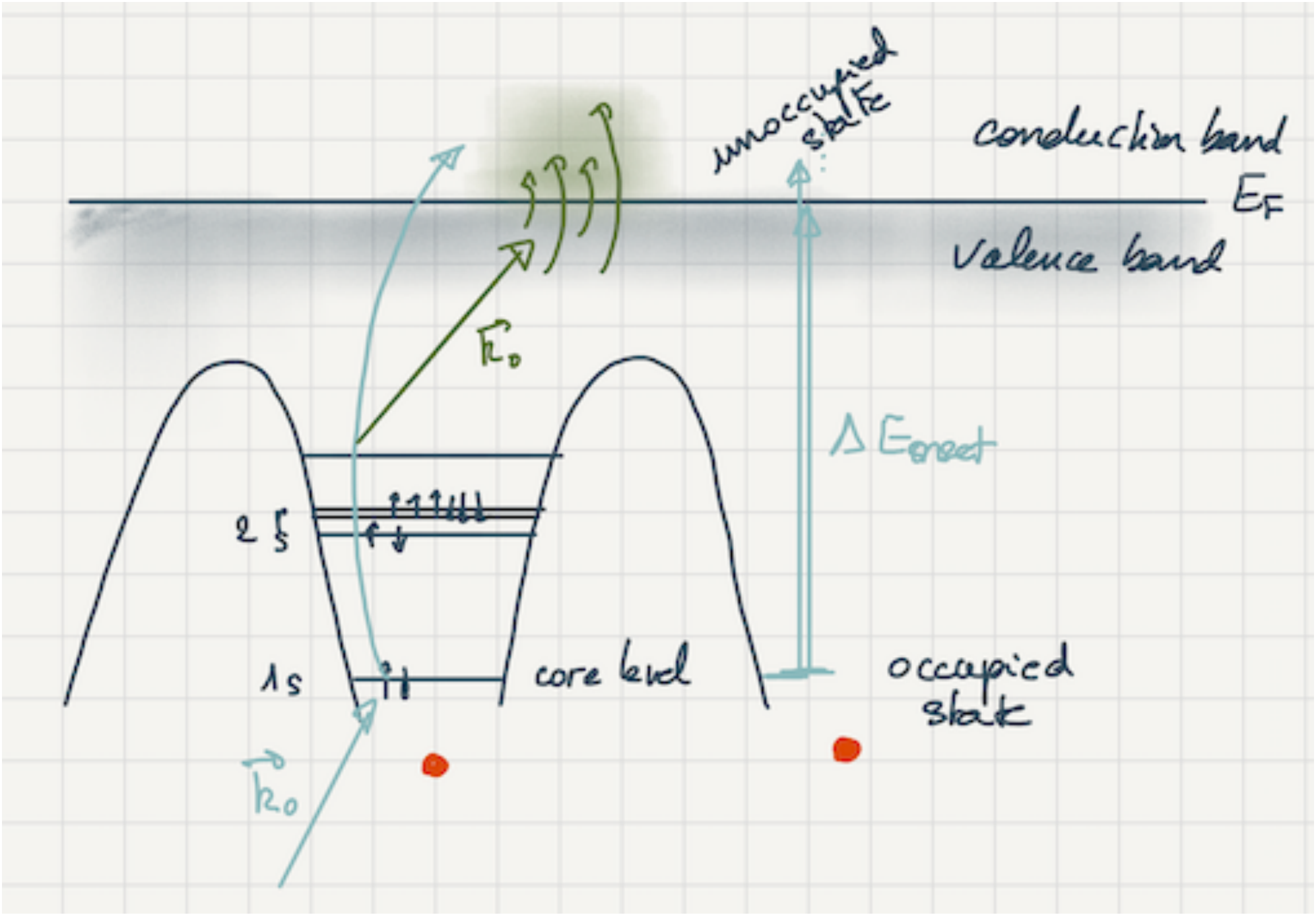
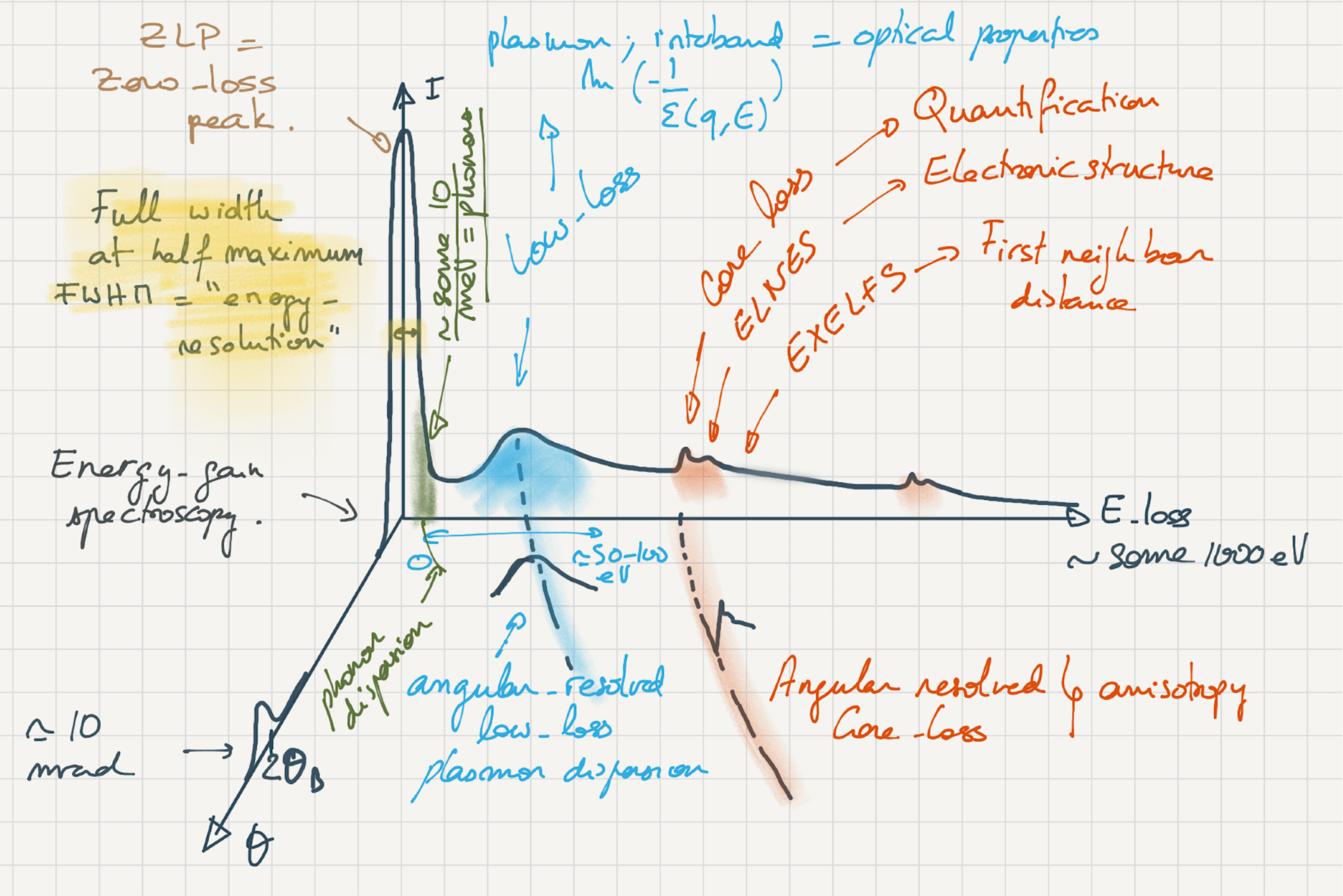
Layout

- Introduction
- Formalism
- Experiment, plasmon dispersion
- Plural scattering
- beyond linear response
- Surface effects
- Relativistic effects

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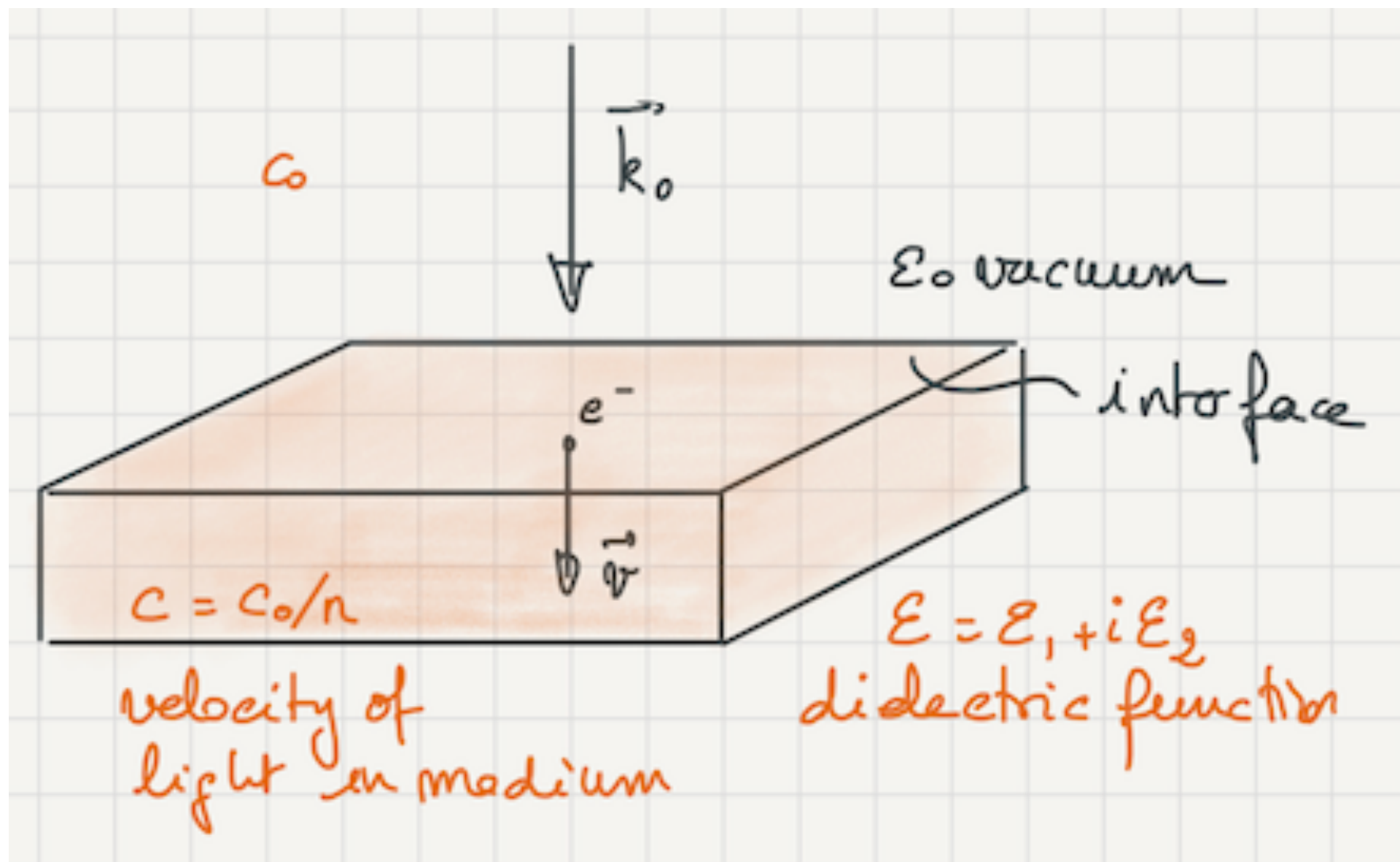
Introduction



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- Introduction
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Formalism



- Single electron description not applicable
- Medium described by the dielectric function
- Linear reaction of medium on the fast travelling charged particle
- Effect of boundaries (surfaces)
- Retardation effect (speed of electron versus speed of light)

Formalism

Infinite medium; no retardation, linear response

$$\vec{\nabla} \cdot \vec{D}(\vec{r}, t) = \rho_f(\vec{r}, t) \quad \text{divergence of electric displacement linked to density of free charge}$$

$$\rho_f(\vec{r}, t) = -e\delta(\vec{r} - \vec{v}t)$$

$$FT[\vec{\nabla} \vec{f}] = 2\pi i \vec{q} FT[\vec{f}]$$

$$2\pi i \vec{q} \cdot \vec{D}(\vec{q}, \omega) = \tilde{\rho} = -e\delta(2\pi \vec{q} \cdot \vec{v} - \omega)$$

$$\vec{D}(\vec{q}, \omega) = \epsilon_0 \epsilon(\vec{q}, \omega) \vec{E}(\vec{q}, \omega) \quad \text{Response of medium to perturbation}$$

Formalism

Infinite medium; no retardation

$$\vec{E}(\vec{r}, t) = - \vec{\nabla} \Phi(\vec{r}, t) \quad \text{Electric field and potential}$$

$$\vec{E}(\vec{q}, \omega) = - 2\pi i \vec{q} \Phi(\vec{q}, \omega)$$

$$\Phi(\vec{q}, \omega) = - \frac{e \delta(2\pi \vec{q} \cdot \vec{v} - \omega)}{4\pi^2 q^2 \epsilon_0 \epsilon(\vec{q}, \omega)}$$

Electron travels along z direction, a distance $dz=v \cdot dt$ during dt

$$\text{Stopping power } SP = \frac{-dW}{dz} \text{ with } dW \text{ work of electric force over distance } dz$$

Formalism

Infinite medium; no retardation

$$SP = -e \frac{\partial \Phi}{\partial z} \Big|_{x=y=0, z=vt} \quad \text{Back to real space...}$$

$$\Phi(\vec{r}, t) = \iint -\frac{e\delta(2\pi\vec{q} \cdot \vec{v} - \omega)}{4\pi^2 q^2 \epsilon_0 \epsilon(\vec{q}, \omega)} e^{2\pi i \vec{q} \cdot \vec{r} - i\omega t} d^3 q d\omega \quad \vec{q} \cdot \vec{v} = v q_z$$

$$SP = \frac{e^2}{4\pi^2 \epsilon_0} \iint 2\pi i q_z \frac{\delta(2\pi v q_z - \omega)}{(q_{\perp}^2 + q_z^2) \epsilon(\vec{q}, \omega)} e^{2\pi i \vec{q} \cdot \vec{r} - i\omega t} d^3 q d\omega \Big|_{x=y=0, z=vt}$$

integration over q_z with delta function picks value of integrand (normalize by $2\pi v$)

Formalism

Infinite medium; no retardation

$$SP = \frac{e^2 i}{8\pi^3 \epsilon_0 v^2} \int_{q_\perp} \int_{\omega=-\infty}^{+\infty} \frac{\omega}{(q_\perp^2 + (\frac{\omega}{2\pi v})^2) \epsilon(\vec{q}_\perp, \omega)} d^2 q_\perp d\omega$$

$\epsilon(\vec{r}, t)$ real therefore $\epsilon(-\omega) = \epsilon^*(\omega)$

$$SP = \frac{e^2}{4\pi^3 \epsilon_0 v^2} \int_{q_\perp} \int_{\omega=0}^{+\infty} \frac{\omega}{(q_\perp^2 + (\frac{\omega}{2\pi v})^2)} \Im \frac{-1}{\epsilon(\vec{q}, \omega)} d^2 q_\perp d\omega$$

$$\Delta E = \hbar \omega$$

Formalism

Infinite medium; no retardation

$$SP = \frac{e^2}{4\pi^3\epsilon_0 v^2} \int_{q_\perp} \int_{\omega=0}^{+\infty} \frac{\omega}{(q_\perp^2 + (\frac{\omega}{2\pi v})^2)} \Im \frac{-1}{\epsilon(\vec{q}, \omega)} d^2 q_\perp d\omega$$

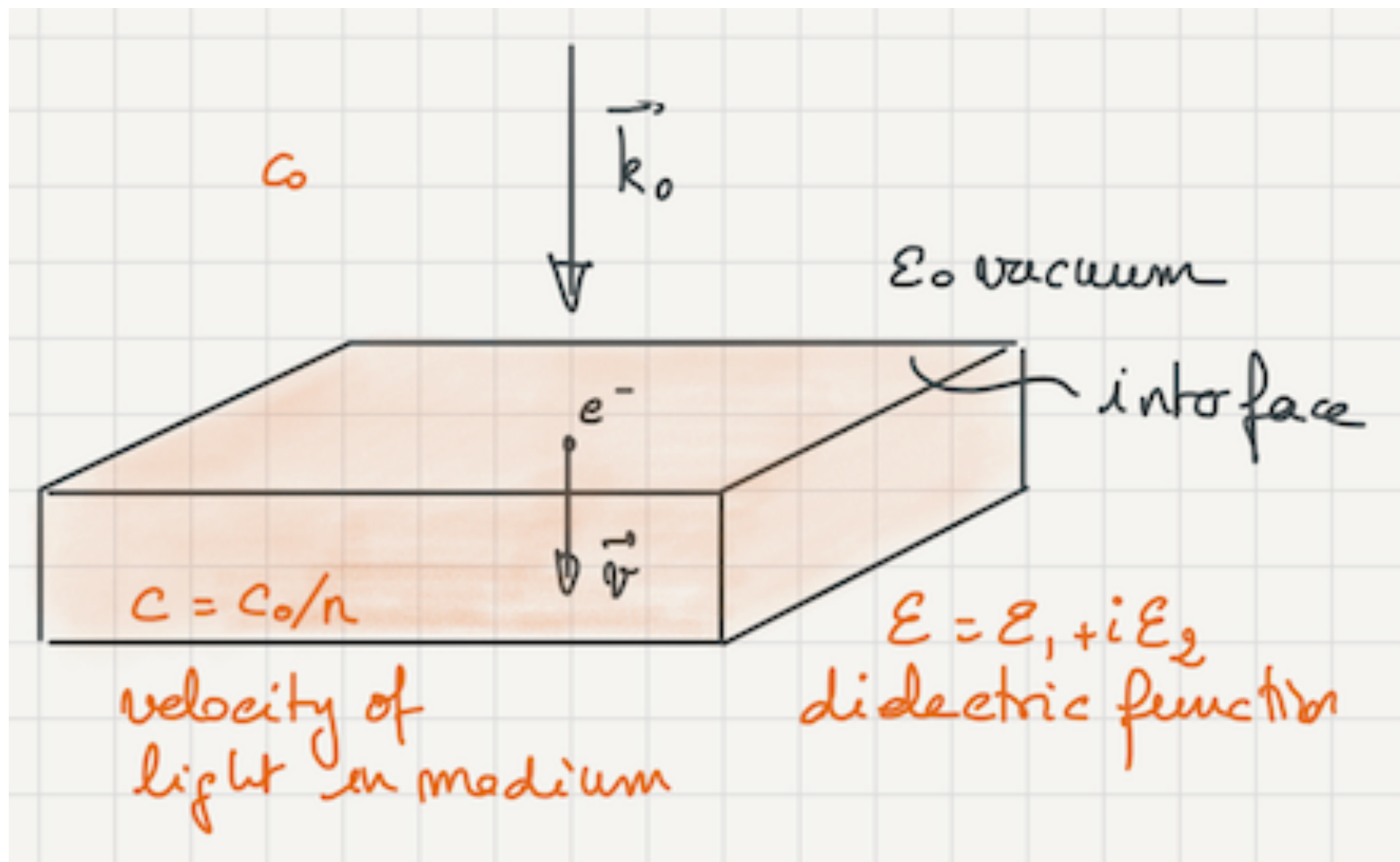
$$SP = \iint \Delta E \frac{\partial P}{\partial q_\perp \partial \omega} d^2 q_\perp d\omega$$

P: E-loss probability per unit path length

$$SP = \iint n_v \Delta E \frac{\partial \sigma}{\partial \Omega \partial \Delta E} d\Omega d\Delta E$$

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

Formalism



- Single electron description not applicable
- Medium described by the dielectric function
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$$\frac{\partial^2 \sigma}{\partial \Omega \partial \Delta E} = \frac{e^2}{n_v \pi \hbar^2 \epsilon_0 v^2} \frac{1}{\theta^2 + \theta_E^2} \Im \left(\frac{-1}{\epsilon(\vec{q}, \Delta E)} \right)$$

Formalism

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

$$\frac{\partial \sigma}{\partial \Delta E} = \frac{e^2}{n_v h^2 \epsilon_0 v^2} \ln(1 + \beta^2 / \theta_E^2) \mathfrak{I} \left(\frac{-1}{\epsilon(\vec{q}, \Delta E)} \right)$$

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

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Experiment

PHYSICAL REVIEW B

VOLUME 27, NUMBER 9

1 MAY 1983

Experimental energy-loss function, $\text{Im}[-1/\epsilon(q, \omega)]$, for aluminum

P. E. Batson* and J. Silcox

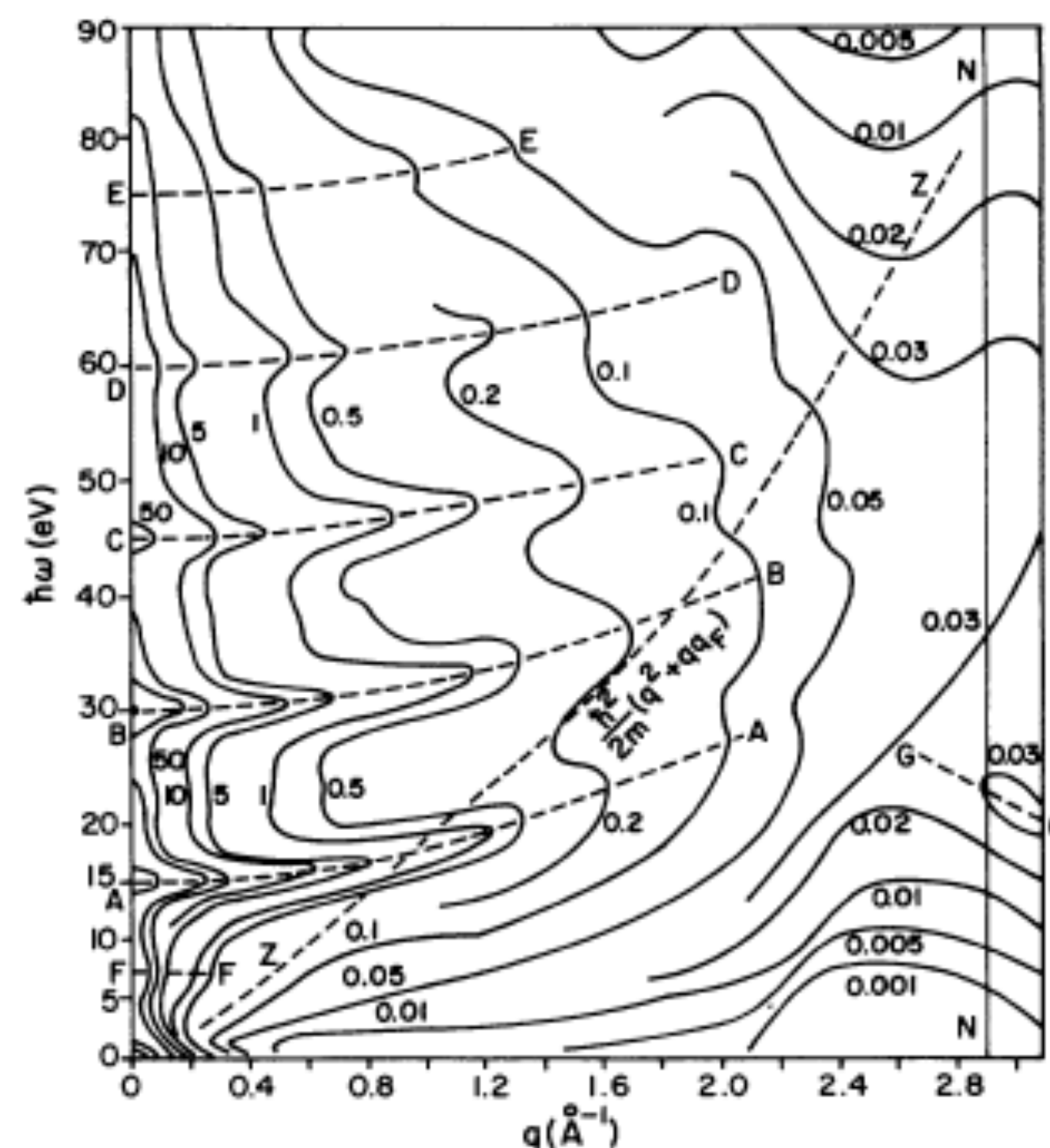
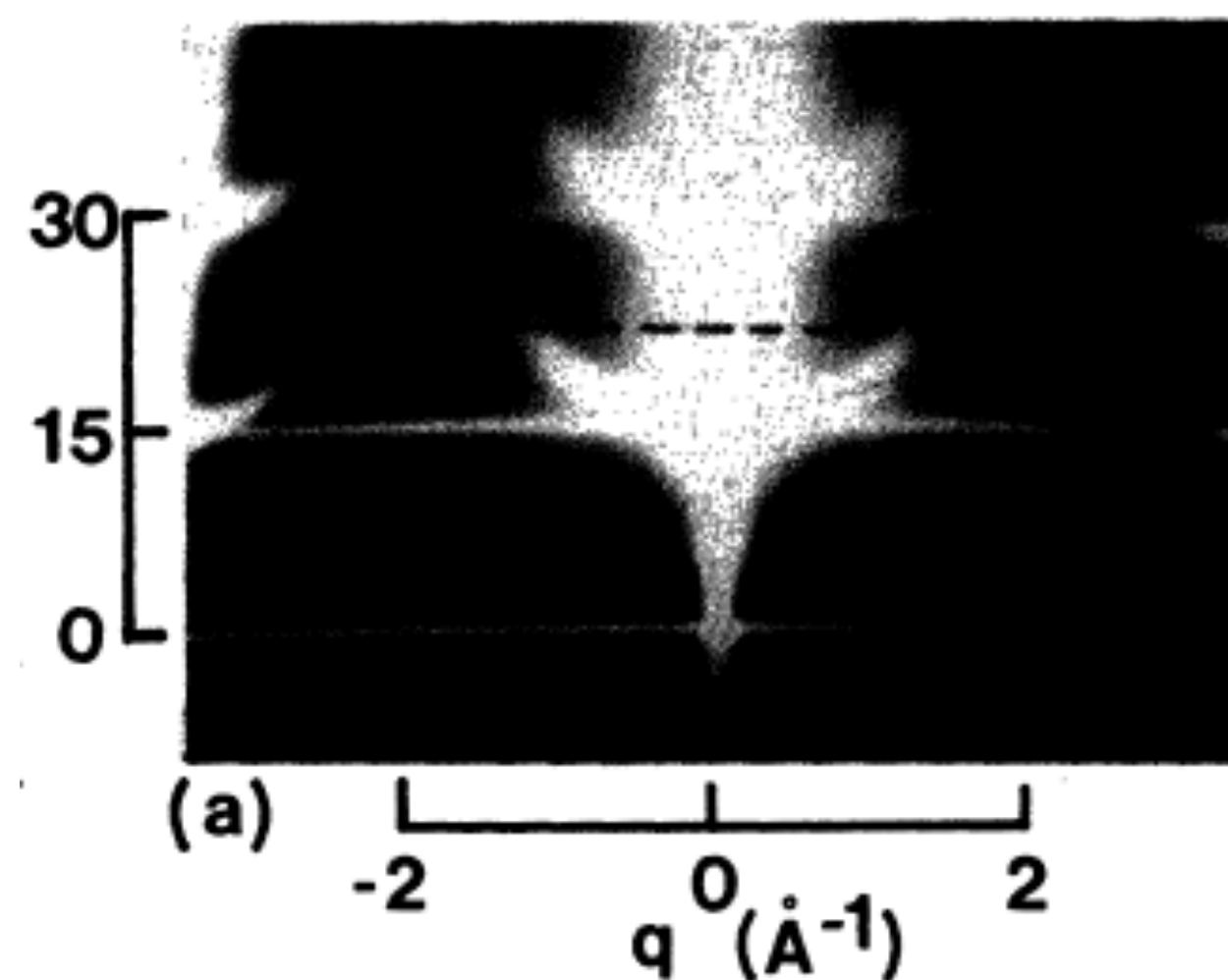


FIG. 11. Intensity contour map after application of the image subtraction. Note the enhancement of the Bragg plus plasmon scattering (GG).

Free electron gas: Jellium model
damped oscillator.

Plasmon parabolic dispersion

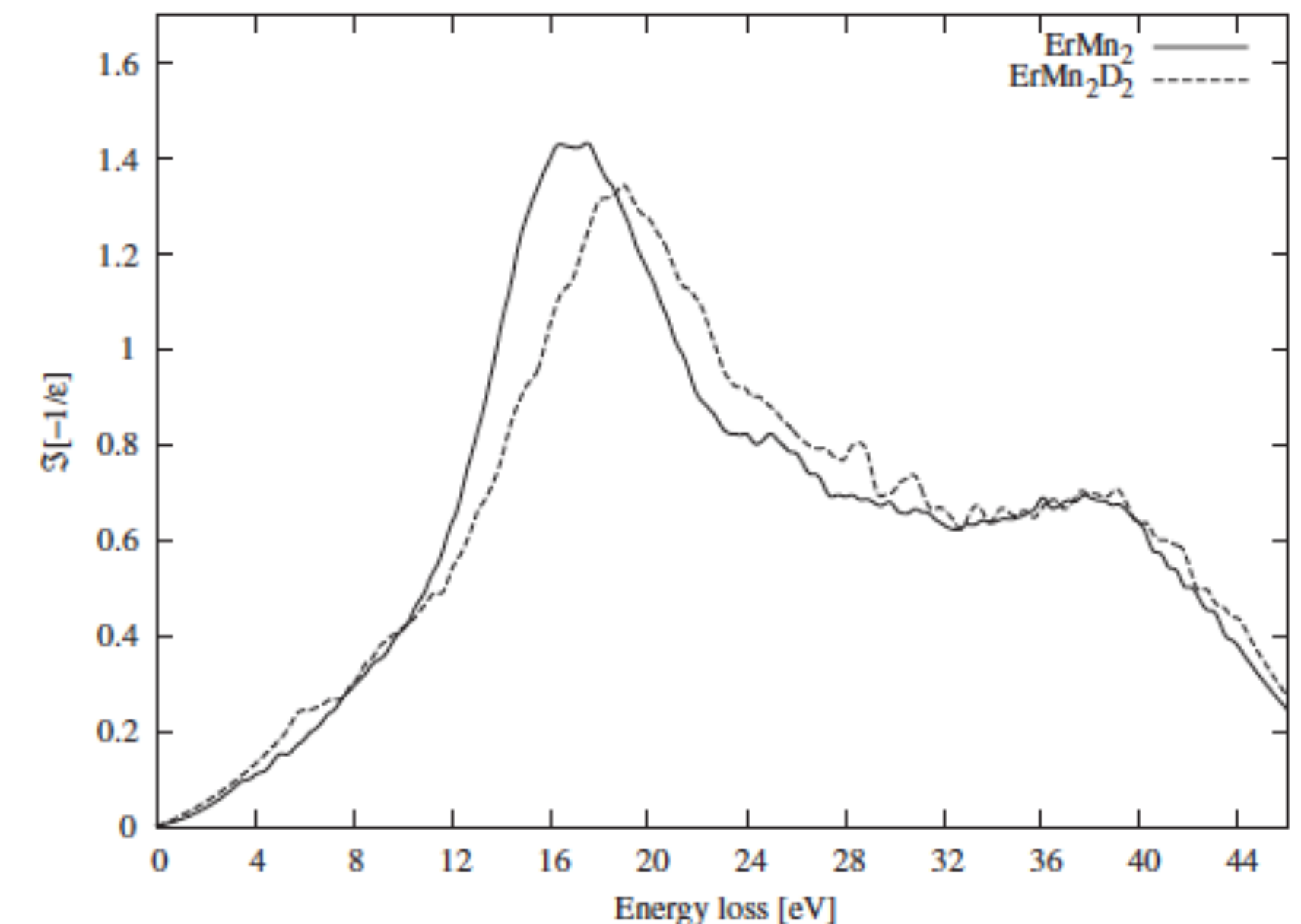
Experiment interpretation

Jellium model: FEG modelled as a damped oscillator, with Γ damping constant.

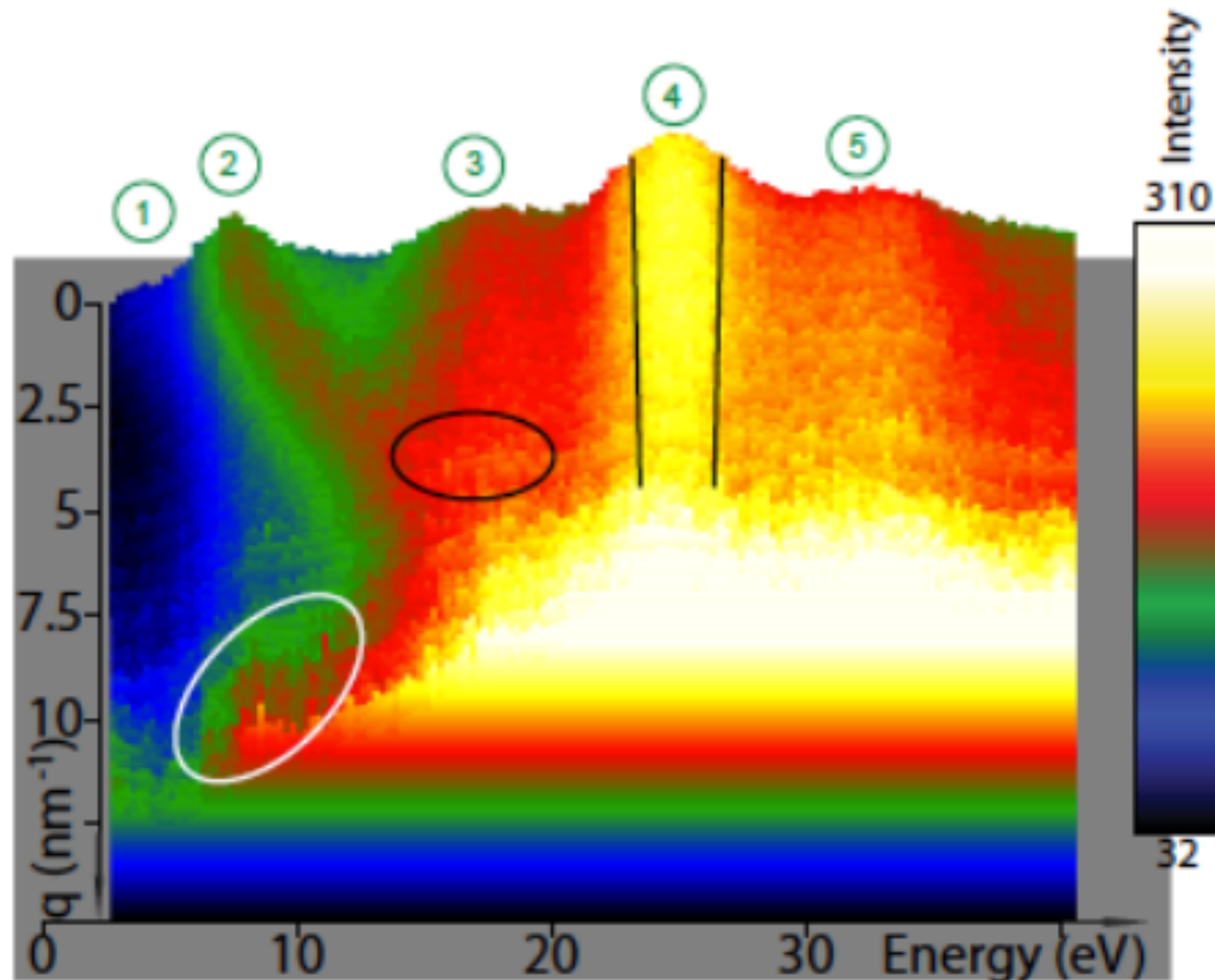
natural frequency of damped oscillator can be calculated $\omega_p = \sqrt{ne^2/(\epsilon_0 m)}$

sharp resonance: peak at ω_p . Broad resonance, peak appears at lower energy

resonance for zero-crossing of ϵ_1 and small ϵ_2



Experiment



Experiment in Ag

structure 2 shows a nice dispersion

other structures have a different behaviour

Simulation: TDDFT