Inelastic X-ray Scattering at Synchrotron and FEL sources

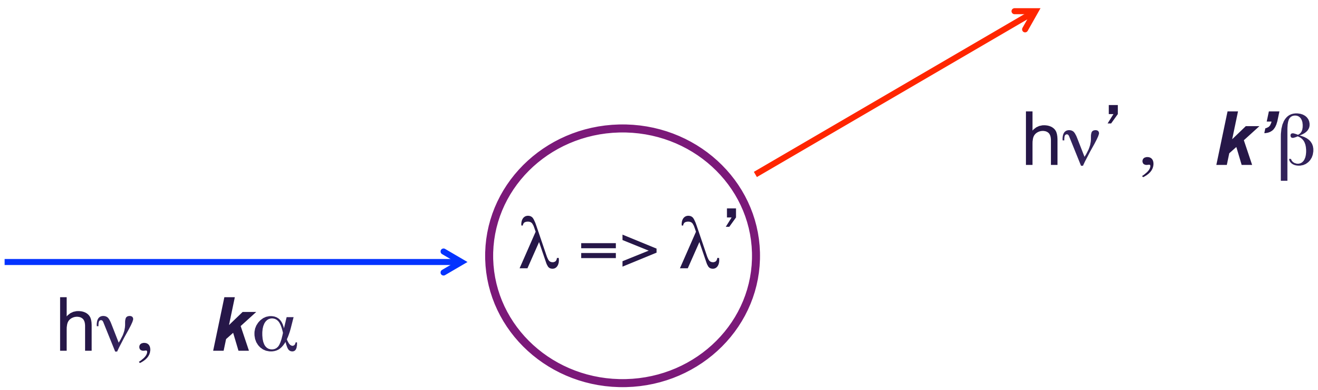
Massimo Altarelli

European XFEL, Schenefeld, Germany

Summary

- Introduction – Scattering Cross Section
- High Resolution Inelastic Scattering at Synchrotrons
- The promise of X-ray Free-Electron Lasers

X-ray scattering processes



- X-rays interact primarily with electrons

Hamiltonian for electronic matter

- With some simplifying assumptions:

Spin-orbit interaction

$$H_{el} = \sum_{i=1}^{N} \left[\frac{\mathbf{p}_i{}^2}{2m} + V(\mathbf{r}_i) + (e\hbar/2m^2c^2)\mathbf{s}_i \cdot (\nabla V(\mathbf{r}_i) \times \mathbf{p}_i) \right]$$

One-electron self-consistent potential

In presence of a radiation field $\mathbf{A}\,(\mathbf{r})$:

$$\mathbf{p}_i \longrightarrow \mathbf{p}_i - (\mathrm{e/c})\,\mathbf{A}(\mathbf{r}_i)$$

Interaction of electrons with the radiation field

- In the weakly relativistic limit ($E^{el} - mc^2 << mc^2$, $h\nu << mc^2$) :

$$H = H_{el} + H_{rad} + H_{int}\,,\ \text{with:}$$

- $$H_{el} = \sum_{i=1}^{N} \left[\frac{\mathbf{p}_i^{\,2}}{2m} + V(\mathbf{r}_i) + (e\hbar/2m^2c^2)\mathbf{s}_i \cdot (\nabla V(\mathbf{r}_i) \times \mathbf{p}_i) \right],$$

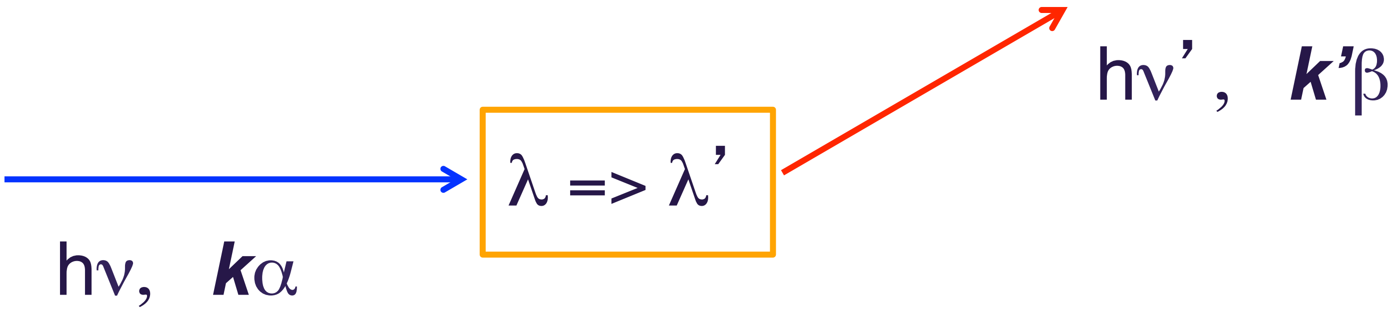
- $$H_{rad} = \sum_{\mathbf{k},\alpha} \hbar\omega_{\mathbf{k}} \left(a^{\dagger}(\mathbf{k},\alpha)a(\mathbf{k},\alpha) + 1/2 \right)$$

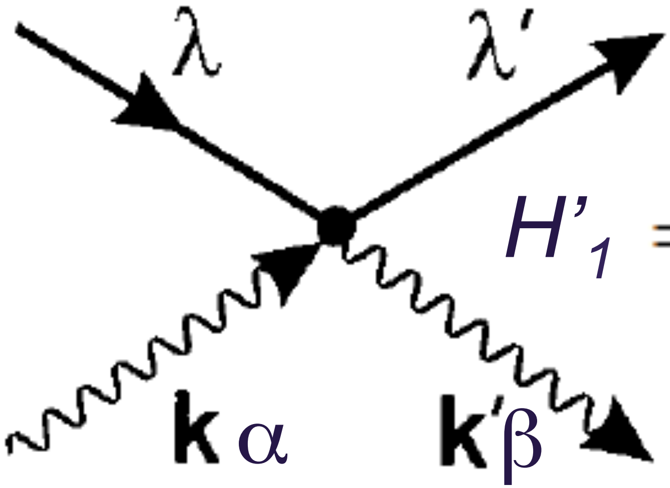
- $$\begin{aligned} H_{int} = \sum_{i=1}^{N} & [(e^2/2mc^2)\mathbf{A}^2(\mathbf{r}_i) - (e/mc)\mathbf{A}(\mathbf{r}_i)\cdot\mathbf{p}_i - \\ & (e\hbar/mc)\mathbf{s}_i \cdot (\nabla \times \mathbf{A}(\mathbf{r}_i)) + \\ & (e\hbar/2m^2c^3)\mathbf{s}_i \cdot [(\partial\mathbf{A}(\mathbf{r}_i)/\partial t) \times (\mathbf{p}_i - (e/c)\mathbf{A}(\mathbf{r}_i))]] \\ \equiv\ & H_1' + H_2' + H_3' + H_4'. \end{aligned}$$

Inelastic scattering processes

	Elastic, non-res	Inelastic, Non-res.	Elastic Resonant	Inelastic, Resonant	
H'_1	Thomson/ Bragg scattering	Compton, Raman, S(**q**,ω)			
H'_2	Orbital magnetic scattering		Resonant Elastic (REXS: charge, spin, orbital order…)	Absorption, XMCD, Emission, Res. Inelastic (RIXS)	
H'_3	Spin Magnetic				
H'_4	Spin Magnetic				

Basics of (non-res.) Inelastic X-ray Scattering





$$H'_1 = \frac{e^2}{2mc^2} \sum_i \delta(\boldsymbol{r} - \boldsymbol{r}_i) \boldsymbol{A}(\boldsymbol{r}) \cdot \boldsymbol{A}(\boldsymbol{r})$$

Golden rule, etc.,…

$$\langle k'\beta\lambda'|\, H'_1 \,|k\alpha\lambda\rangle = \frac{1}{L^3}\left(\frac{e^2}{mc^2}\right)\frac{\hbar c}{(\omega_k\omega_{k'})^{1/2}}(\varepsilon_\alpha^* \cdot \varepsilon_\beta)\left\langle \lambda' \left| \sum_i \mathrm{e}^{-\mathrm{i}(\boldsymbol{k}-\boldsymbol{k}')\cdot\boldsymbol{r}_i} \right| \lambda \right\rangle$$

i = all electrons

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{k\alpha\to k'\beta} = (k'/k)\left(\frac{e^2}{mc^2}\right)^2 |\varepsilon_\alpha^* \cdot \varepsilon_\beta|^2 S(\boldsymbol{q},\omega)$$

$$S(\boldsymbol{q},\omega) = \sum_{\lambda\lambda'}\sum_{ij} p_\lambda \langle\lambda|\,\mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i}\,|\lambda'\rangle\langle\lambda'|\,\mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_j}\,|\lambda\rangle\,\delta(E_\lambda - E_{\lambda'} + \hbar\omega)$$

S(q, ω), the dynamic structure factor

$$S(q,\omega) = \sum_{\lambda\lambda'} \sum_{ij} p_\lambda \langle\lambda|\,\mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i}\,|\lambda'\rangle\langle\lambda'|\,\mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_j}\,|\lambda\rangle\delta(E_\lambda - E_{\lambda'} + \hbar\omega)$$

….**and with some simple mathematics:**

$$\sum_i \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i} = \int d\,\boldsymbol{r}\ \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}} \sum_i \delta\,(\boldsymbol{r}\text{-}\boldsymbol{r}_i\,) = \rho\,(\boldsymbol{q}\,)$$

$$\sum_{\lambda'} \langle\lambda|A|\lambda'\rangle\langle\lambda'|B|\lambda\rangle\delta(E_\lambda - E_{\lambda'} + \hbar\omega)$$

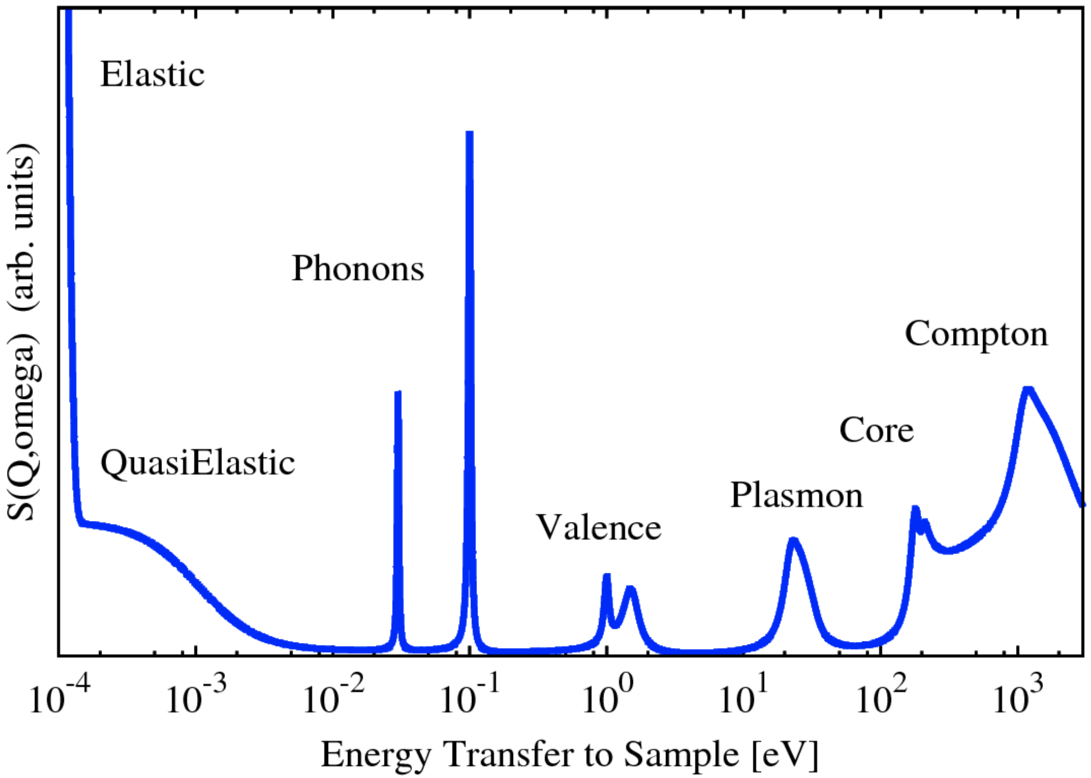
$$= \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} \mathrm{d}t\,\mathrm{e}^{-\mathrm{i}\omega t} \sum_{\lambda'} \langle\lambda|A|\lambda'\rangle\langle\lambda'|B|\lambda\rangle\,\mathrm{e}^{\mathrm{i}(E_{\lambda'}-E_\lambda)t/\hbar}$$

S(q, ω) and the density density correlations

$$S(q,\omega) = \sum_{\lambda\lambda}\sum_{ij} p_\lambda \langle\lambda|\,e^{-iq\cdot r_i}\,|\lambda'\rangle\langle\lambda'|\,e^{iq\cdot r_j}\,|\lambda\rangle\delta(E_\lambda - E_{\lambda'} + \hbar\omega)$$

$$= \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} dt\, e^{-i\omega t}\langle\rho(q,0)\rho^+(q,t)\rangle$$

L. van Hove,
Phys Rev **95**, 249 (1954)



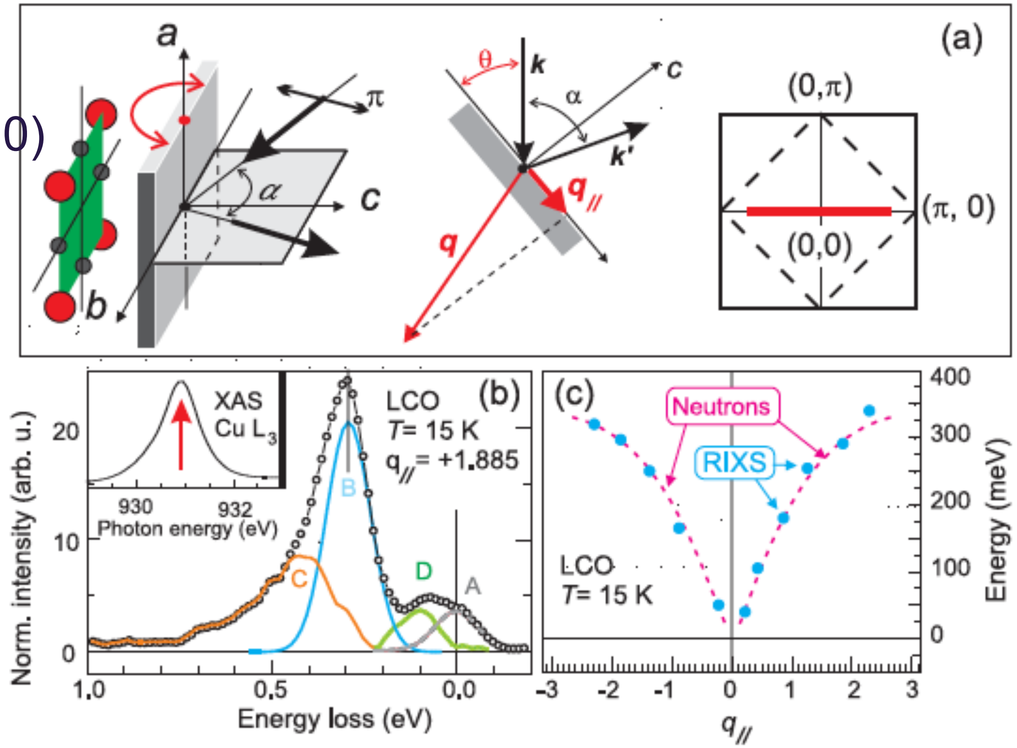
The beauty of non-resonant inelastic scattering

- One knows exactly what is being measured! **A well-defined correlation function**

- No such luck in RIXS! (…although RIXS is **wonderful**!)

Magnetic excitations in La_2CuO_4

Braicovich et al., PRL **104**, 077002 (2010)



Why phonons with X-rays? Better use neutrons?

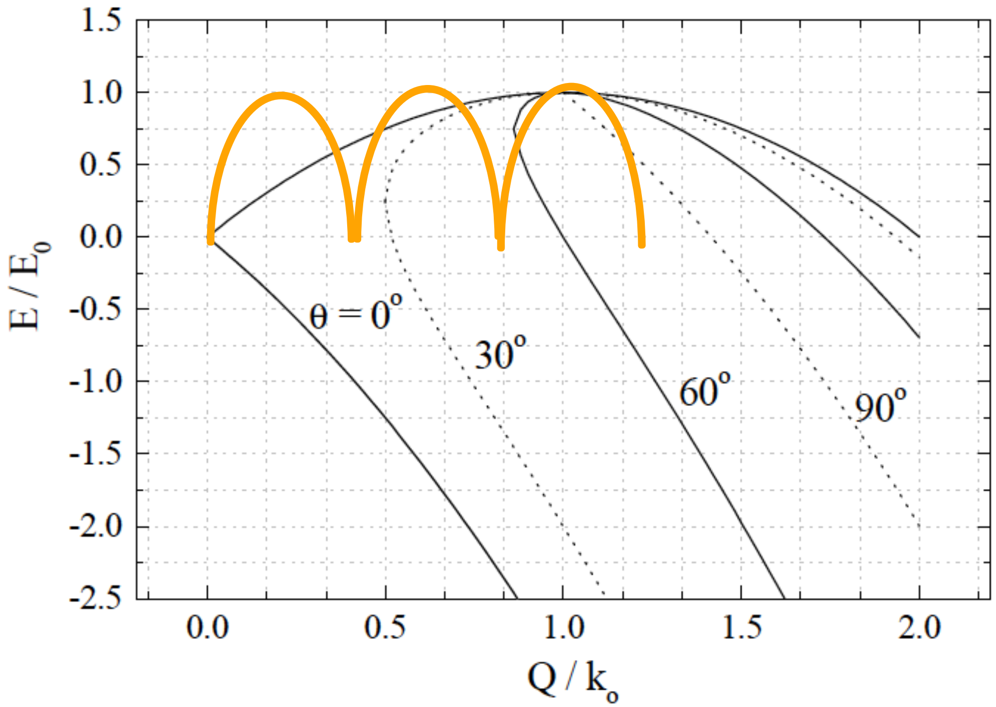


Figure 2 – *The kinamtcs region accessible to neutron scattering experiments (region inside the curves) is reported for different scattering angles (θ=0, 30, 60 and 90) in the E/E_i vs Q/k_i plane.*

- Neutrons: Kinematics forbids excitations with velocity > ~4 000 m/s

Solids: get away with higher Brillouin zones.

Disordered systems:

no such way out!

Neutrons λ =0.1 nm, E ~ 81.2 meV

X-rays λ =0.1 nm, E ~ 12.4 keV

- X-rays:

meV resolution

means $\Delta E/E \sim 10^{-7}$!!

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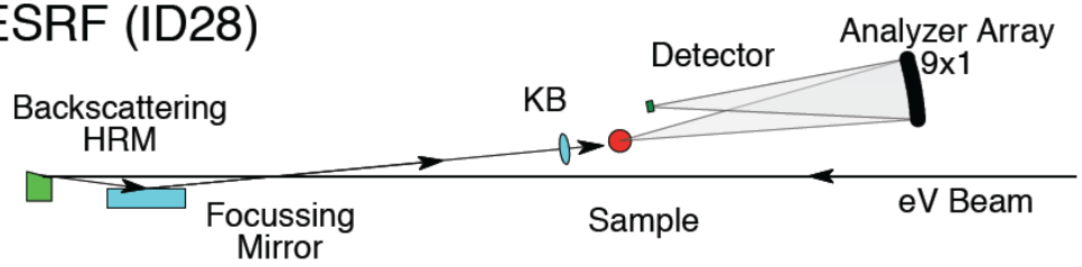
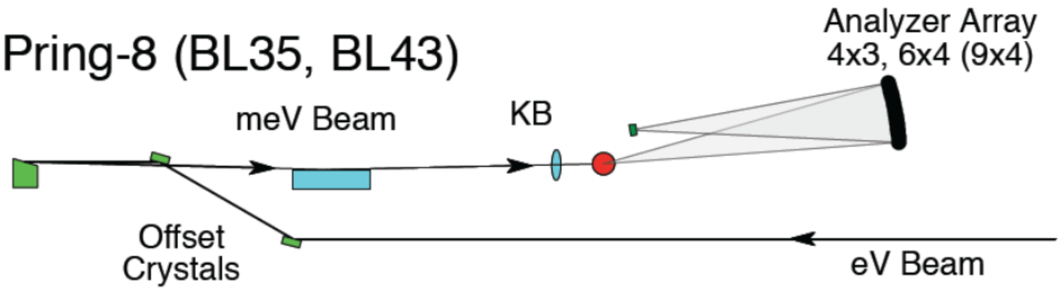
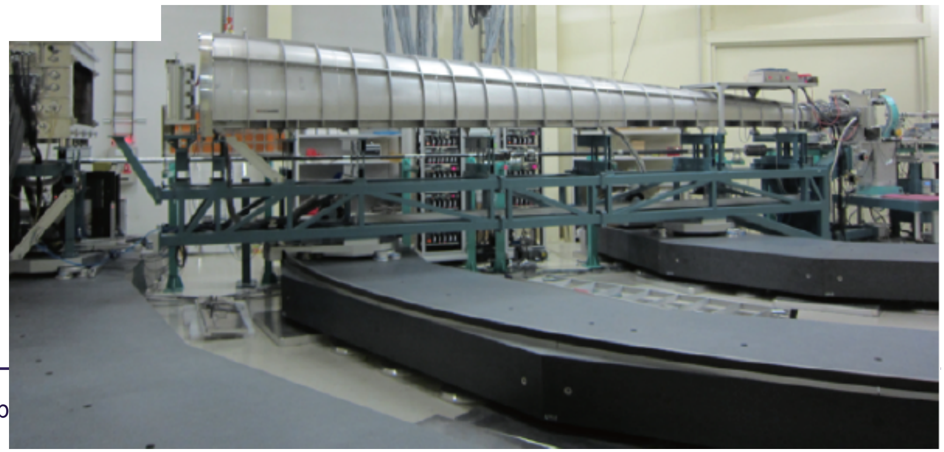
Phonon spectroscopy with ~ meV resolution

ESRF (ID28)



SPring-8 (BL35, BL43)





- High order near back-reflections of Si 17.79 keV (999), 21.75 keV (11 11 11)), 25.7 keV (13 13 13)

And of course….

- APS sectors 3 and 30…

HERIX, sector 30

Important parameters

- Resolution: e.g. Si (9 9 9) > 1.8 meV
  Si (11 11 11) > 0.77 meV
  Si (13 13 13) > 0.32 meV

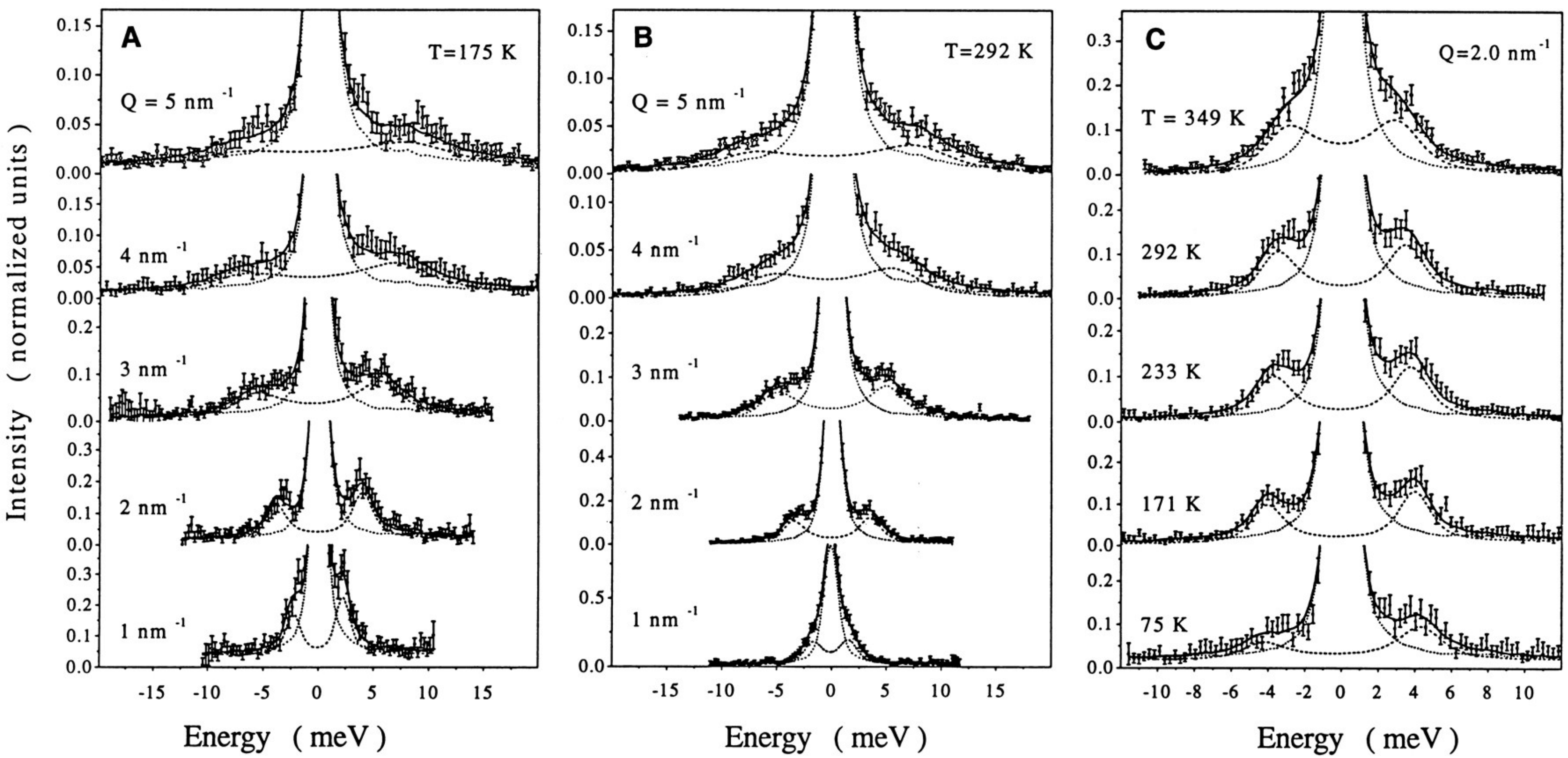
- Energy transfer: max ~ 200 meV (ESRF ID28, HERIX,…)

- Flux on sample: 10^9 to ~10^{11} photons/s (depending on resolution)

European XFEL

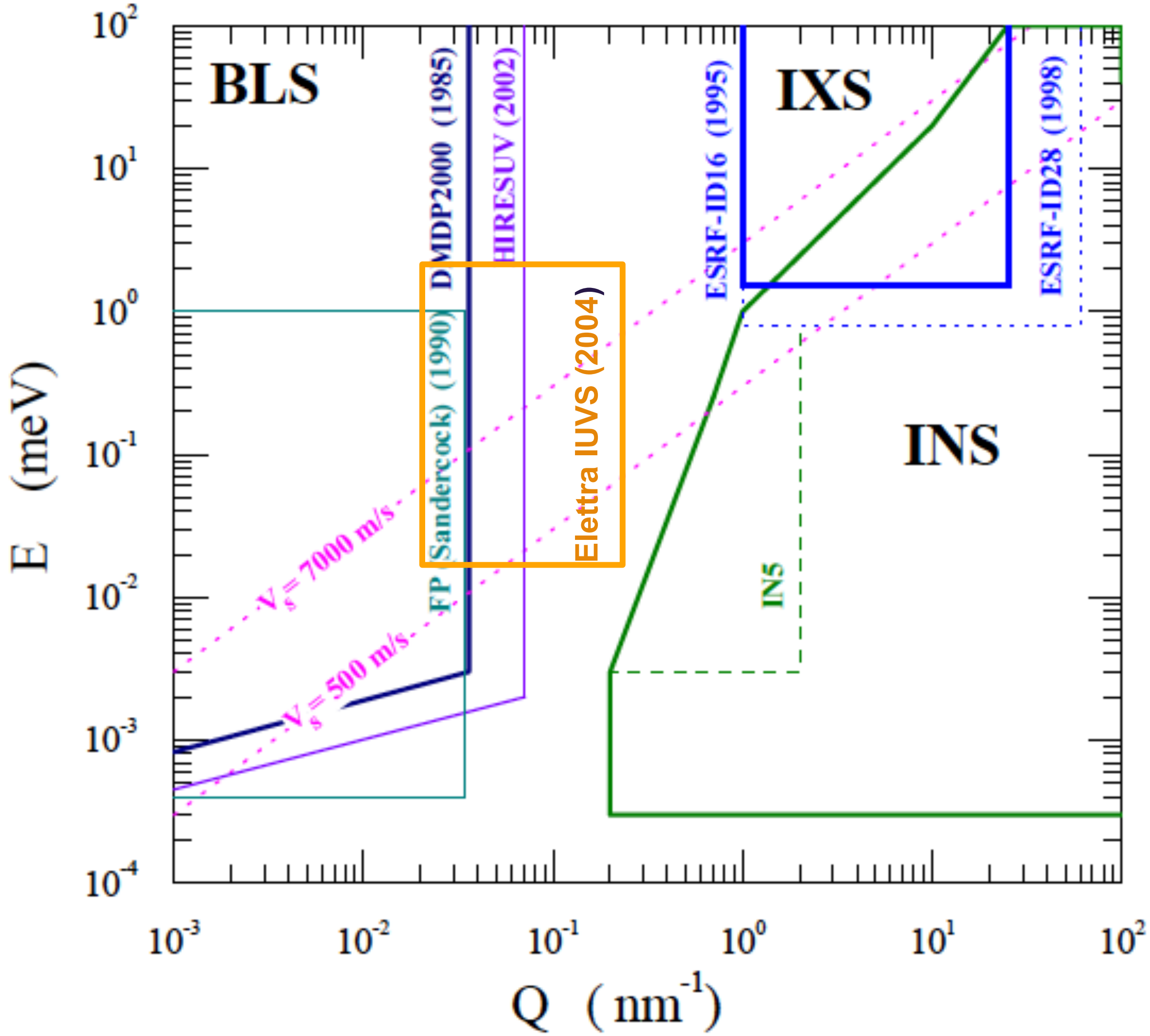
Francesco Sette *et al.*, *Science* 280, 1550 (1998)

IXS spectra of glassy glycerol at 175 K at the indicated Q values.



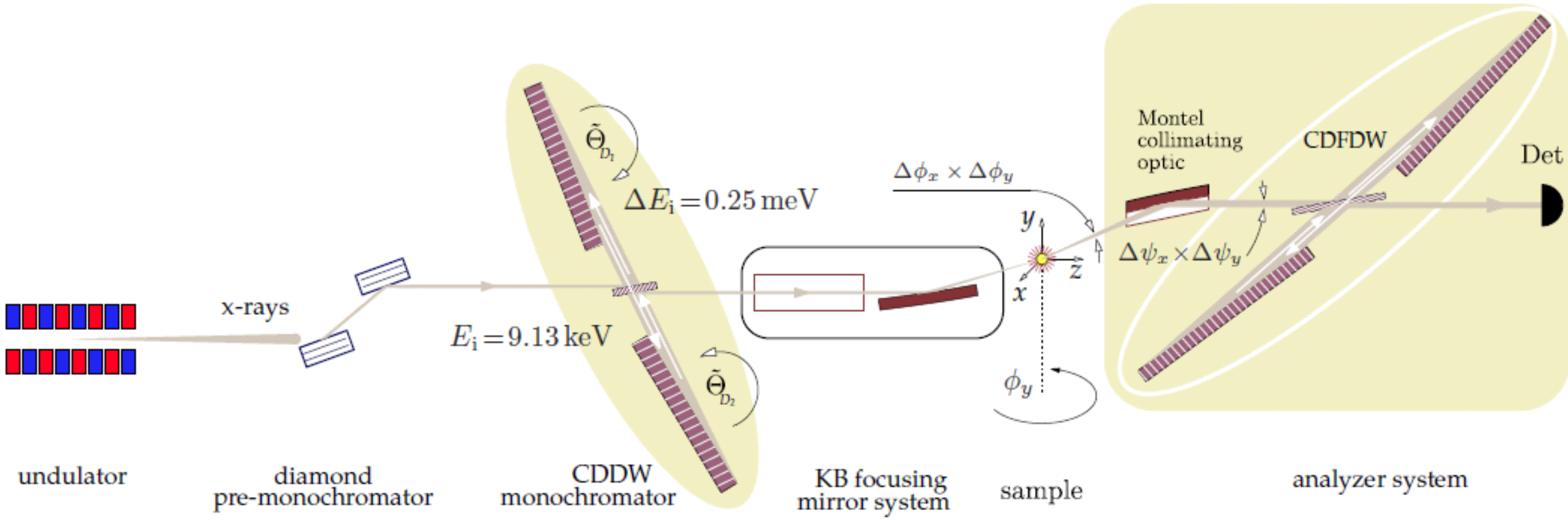
Energy-momentum ranges of inelastic scattering

See e.g. C. Masciovecchio et al., AIP Conf. Proceedings **705**, 1190 (2004)



UHRIX: Ultra-High-Resolution IXS Spectrometer

Joint R&D Effort of APS and DLS



Courtesy of Y. Shvyd'ko, ANL, APS

Electronic excitations are also important…

PRL **99,** 026401 (2007) PHYSICAL REVIEW LETTERS week ending 13 JULY 2007

Nonresonant Inelastic X-Ray Scattering and Energy-Resolved Wannier Function Investigation of *d*-*d* Excitations in NiO and CoO

B. C. Larson,[1] Wei Ku,[2] J. Z. Tischler,[1] Chi-Cheng Lee,[2,3] O. D. Restrepo,[1,4] A. G. Eguiluz,[1,4] P. Zschack,[5] and K. D. Finkelstein[6]

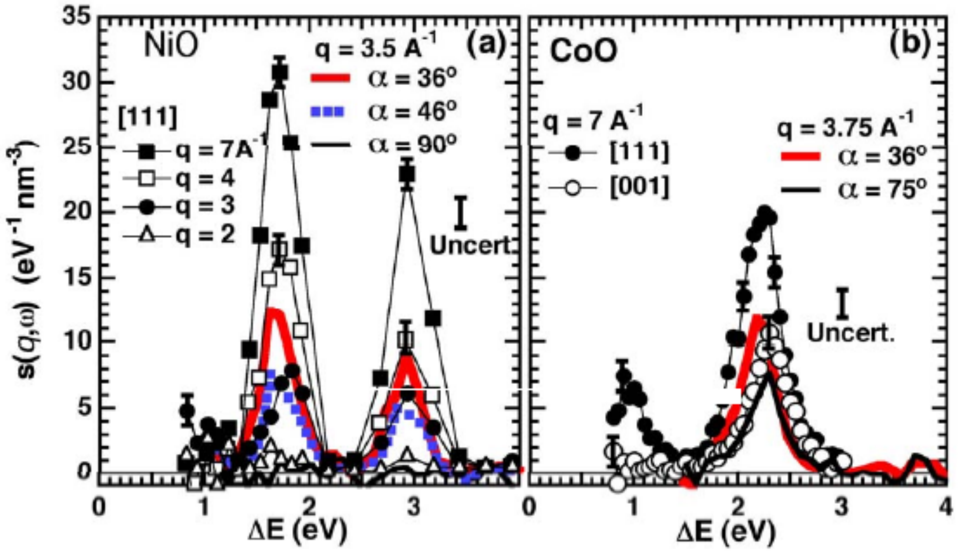


FIG. 2 (color online). High-resolution (0.3 eV) measurements of the q magnitude and orientation dependence of the d-d peak excitations for NiO and CoO; α is the q-orientation angle between the 110 and 001 directions [see Figs. 4(f) and 4(h)].

Energy transfer:
~ several eV

S(q, ω) and the Fluctuation-Dissipation Theorem

$$S(q,\omega) = \frac{1}{\pi}\frac{1}{1-\mathrm{e}^{-\beta\omega}}\chi''_{\rho^+(q)\rho(q)}(\omega)$$

***S(q,ω)* is related to the imaginary (= *dissipative*) part of the electron density-density response function…**

- S(q,ω) => χ'' (ω) => Kramers- Kronig => χ' + i χ'' = density-density response => Fourier tr. => density – density response **in time (P. Abbamonte)**

Response of electrons in water

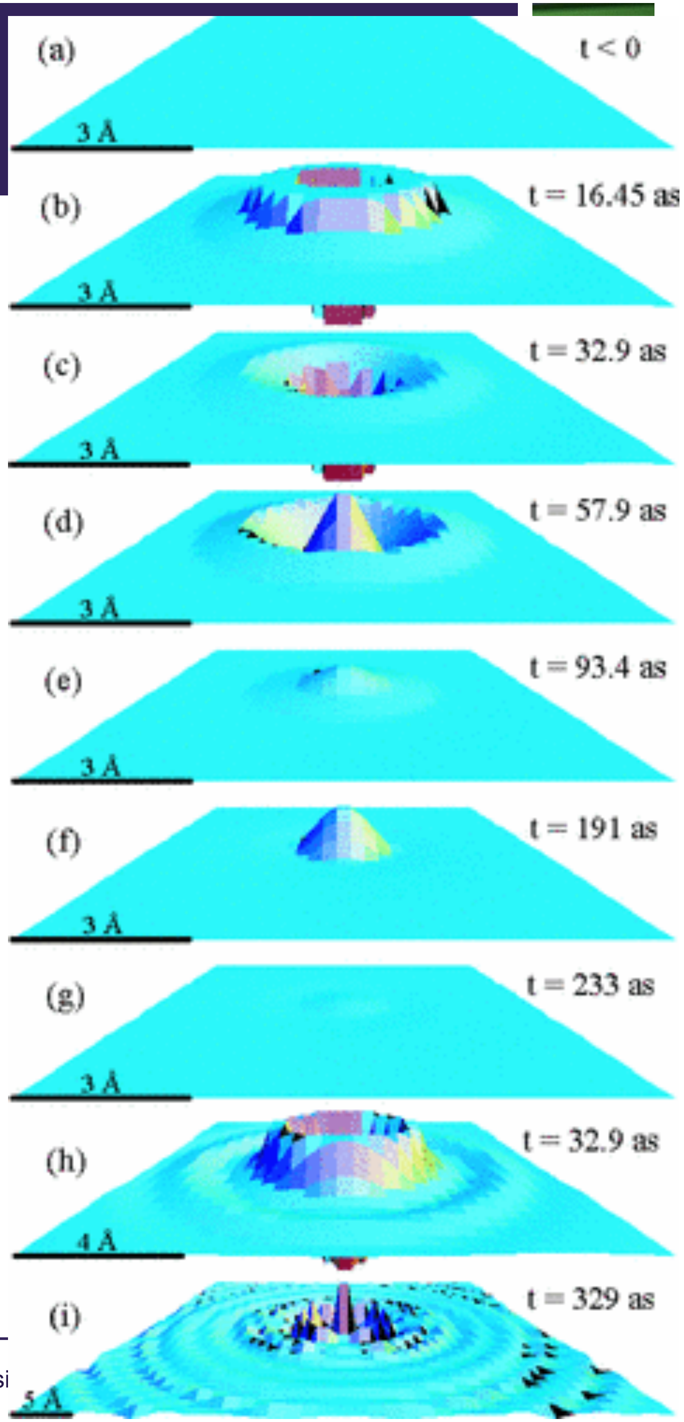
VOLUME 92, NUMBER 23 PHYSICAL REVIEW LETTERS week ending 11 JUNE 2004

Imaging Density Disturbances in Water with a 41.3-Attosecond Time Resolution

P. Abbamonte,[1,*] K. D. Finkelstein,[2] M. D. Collins,[1] and S. M. Gruner[1,2]
[1]Department of Physics, Cornell University, Ithaca, New York 14853-2501, USA
[2]Cornell High Energy Synchrotron Source, Cornell University, Ithaca, New York 14853-2501, USA

Down to ~ 30 as resolution

(with assumptions on extrapolations, Numerical K-K and Fourier transforms…)



Summary

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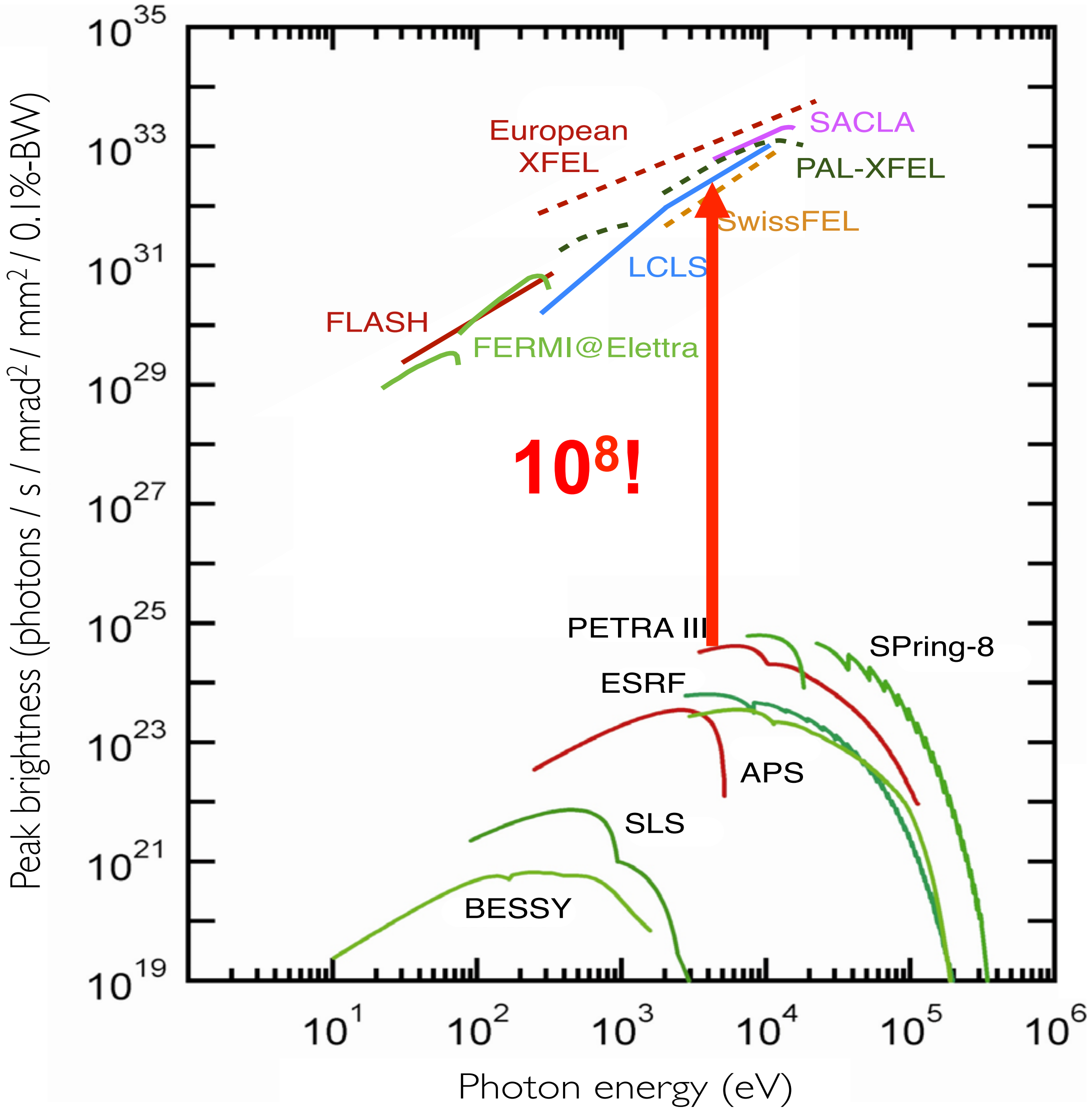
European XFEL

HARD X-ray FEL sources worldwide

Project	LCLS I, US	SACLA, JP	European XFEL	SwissFEL, CH	PAL-XFEL, KR	LCLS II, US
Max. electron energy (GeV)	14.3	8.5	17.5	5.8	10	4
Wavelength range (nm)	0.1–4.4	0.06–0.3	0.05–4.7	0.1–7	0.06–10	0.25 – 4.7
Photons/pulse	~ 10^{12}	2 x 10^{11}	~ 10^{12}	~ 3.6 x 10^{10}	10^{11}–10^{13}	2 10^{11} – 2 10^{10}
Peak brilliance	2 x 10^{33}	1 x 10^{33}	5 x 10^{33}	7 x 10^{32}	1.3 x 10^{33}	
Pulses/second	120	60	27 000	100	60	10^5 - 10^6
Date of first beam	**2009**	**2011**	**2017**	**2017**	**2016**	**2019**

Potential for high repetition rate XFEL's



- Outstanding performance in peak brilliance of XFEL translates into an outstanding **average brilliance for high rep. rate.**
- At European XFEL self-seeding and tapering are proposed, with ~ 27 000 pulses/s for ultra high spectral flux

Realistic estimates for source and spectrometer

JOURNAL OF SYNCHROTRON RADIATION

ISSN 1600-5775

Ultra-high-resolution inelastic X-ray scattering at high-repetition-rate self-seeded X-ray free-electron lasers

Oleg Chubar,[a] Gianluca Geloni,[b] Vitali Kocharyan,[c] Anders Madsen,[b] Evgeni Saldin,[c] Svitozar Serkez,[c] Yuri Shvyd'ko[d]* and John Sutter[e]

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Accepted 28 December 2015

Edited by M. Yabashi, RIKEN SPring-8 Center,

J. Synchrotron Rad. (2016). **23**, 410–424

European XFEL, SASE2 undulator configuration

Combination of high rep-rate HXRSS and Tapering

Tapering: increases power

HXRSS: decreases bandwidth

Figure of merit for IXS: spectral flux

O. Chubar, G. Geloni, V. Kocharyan, A. Madsen, E. Saldin, Y. Shvyd'ko, J. Sutter

		Units
Undulator period	40	mm
Periods per segment	125	
Total number of segments	35	
K parameter (r.m.s.)	2.658	
Intersection length	1.1	m
Wavelength	0.1358	nm
Energy	17.5	GeV
Charge	250	pC
Horizontal normalized slice emittance (*)	4.0×10^{-7}	m rad
Vertical normalized slice emittance (*)	3.6×10^{-7}	m rad
Peak current	5.0	kA
Energy spread σ_γ (*)	0.96	

Simulations from undulator to sample

Location (method)	Δt (ps)	ΔE (meV)	Flux (photons s^{-1})	Spectral flux (photons s^{-1} meV^{-1})
Undulator exit, $z = 74$ m (*GENESIS*)	0.014	950	2.0×10^{17}	2.1×10^{14}
Sample, $z = 1018$ m (*SRW* wavefront propagation)	225	0.087	6.3×10^{12}	7×10^{13}
Sample, $z = 1018$ m (ray-transfer matrix)	190	0.09		

Filling the gap in energy-momentum space

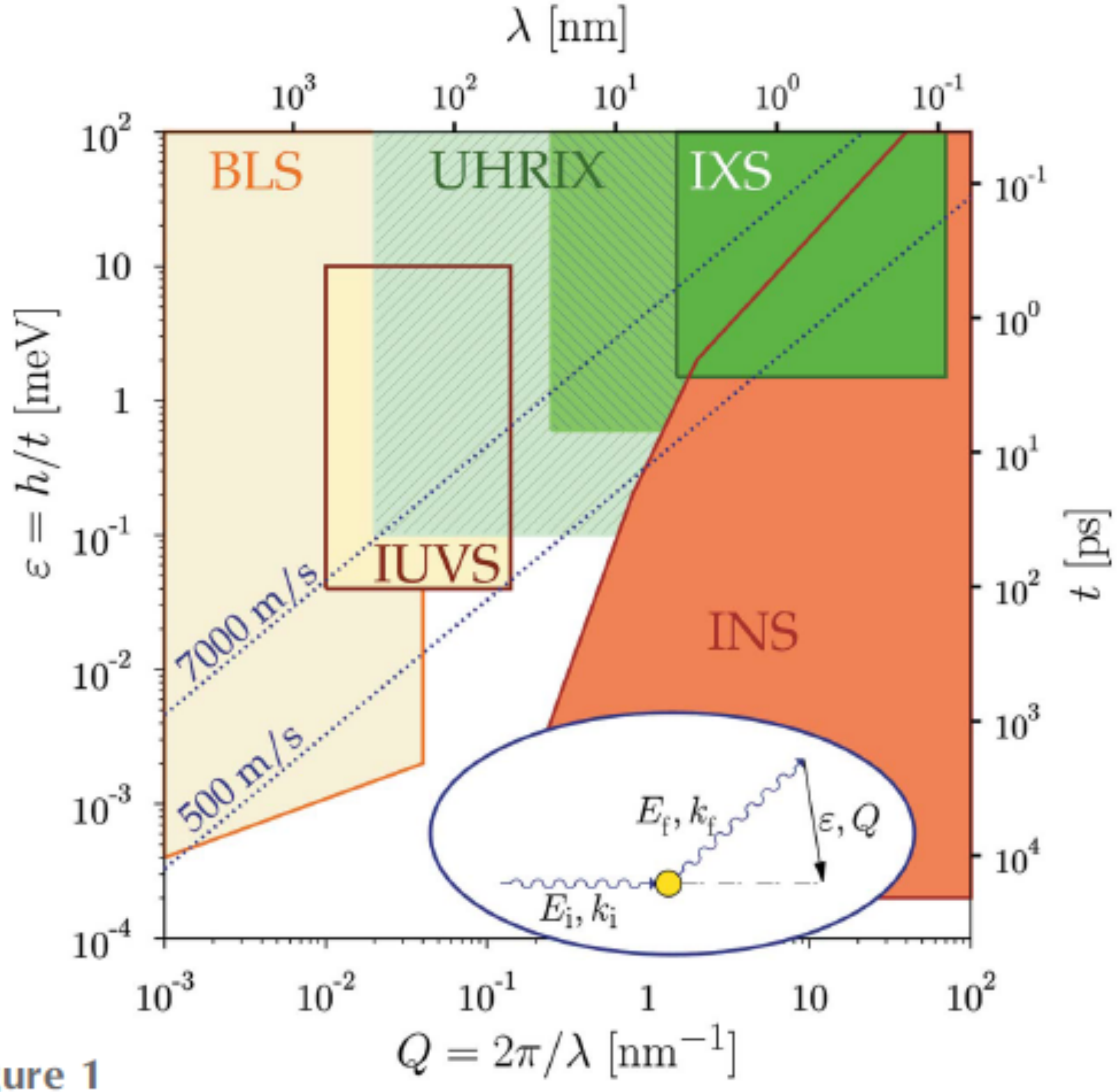


Figure 1

Conclusion:

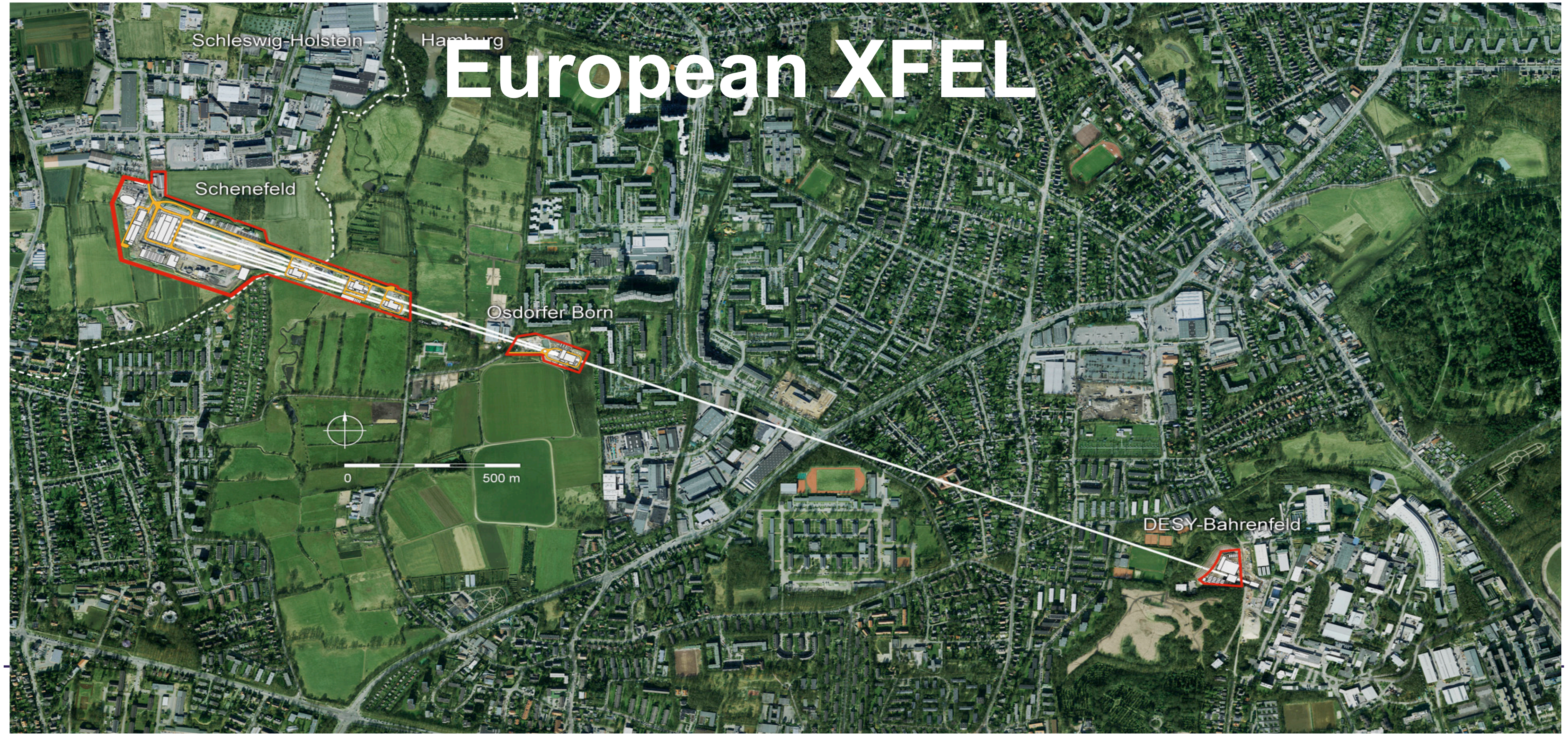
High repetition rate hard x-ray FEL's deliver 2-3 orders of magnitude more flux in a meV bandwidth then synchrotron sources

→ Source: ~ 2 10^{14} ph/s/meV at 9 keV possible at European XFEL, SASE2 self-seeded, tapered undulator

→ ~ 7 10^{12} ph/s in 90 μeV BW at the sample, with UHRIX or X-ray Echo spectrometer

An opportunity not to be missed!

- Ultra-high resolution inelastic scattering can exploit extraordinary average brightness of high rep. rate hard x-rays FEL's for novel research



Thank you for your attention!

Some reviews to learn more…

W. Schulke, "Inelastic Scattering by Electronic Excitations", in *Handbook on Synchrotron Radiation*, Vol. 3, ed. G. Brown and D.E. Moncton, (Elsevier Science Publ., 1991).

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **13** (2001) 7511–7523 PII: S0953-8984(01)25536-2

Theory of inelastic x-ray scattering from condensed matter

Sunil K Sinha

Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439, USA

http://arxiv.org/abs/1504.01098

Introduction to High-Resolution Inelastic X-Ray Scattering

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Research and Utilization Division, SPring-8/JASRI, 1-1-1 Kouto, Sayo, Hyogo 679-5198 Japan

From time response to frequency response…

LETTERS
nature physics
PUBLISHED ONLINE: 27 OCTOBER 2013 | DOI: 10.1038/NPHYS2788

Fourier-transform inelastic X-ray scattering from time- and momentum-dependent phonon–phonon correlations

M. Trigo[1,2]*, M. Fuchs[1,2], J. Chen[1,2], M. P. Jiang[1,2], M. Cammarata[3], S. Fahy[4], D. M. Fritz[3], K. Gaffney[2], S. Ghimire[2], A. Higginbotham[5], S. L. Johnson[6], M. E. Kozina[2], J. Larsson[7], H. Lemke[3], A. M. Lindenberg[1,2,8], G. Ndabashimiye[2], F. Quirin[9], K. Sokolowski-Tinten[9], C. Uher[10], G. Wang[10], J. S. Wark[5], D. Zhu[3] and D. A. Reis[1,2,11]*

…Talk by David Reis => 0.3 meV resolution!