Polarization induced renormalization of local interactions in strongly correlated electron systems

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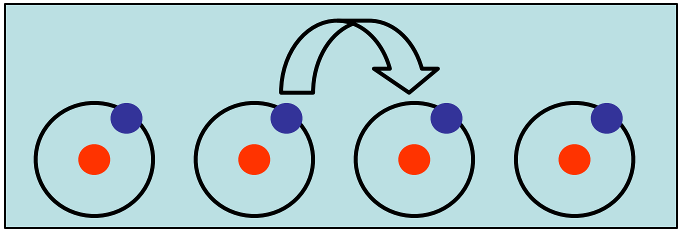
Microscopic screening of short-range Coulomb interactions (e.g. Hubbard U)

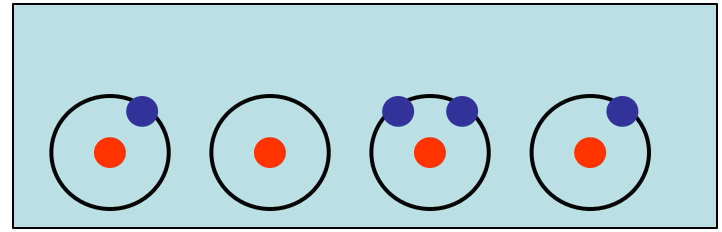
Resonant Inelastic X-ray Scattering (RIXS) on Li_2CuO_2
→ phonon screening of CT energy

Hubbard Hamiltonian

$$H = -t \sum_{\langle ij \rangle,\sigma} (c^\dagger_{i\sigma} c_{j\sigma} + h.c.) + U \sum_i n_{i\uparrow} n_{i\downarrow}$$



Hopping amplitude: ***t***



Coulomb interaction: ***U***

U = 0 — ***Bands: Metallic behaviour***

U >> t — ***Mott-Hubbard Insulator*** → Antiferromagnetism

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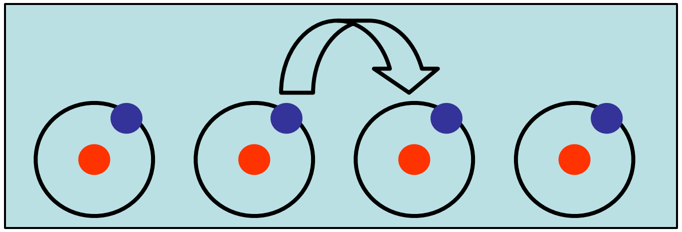

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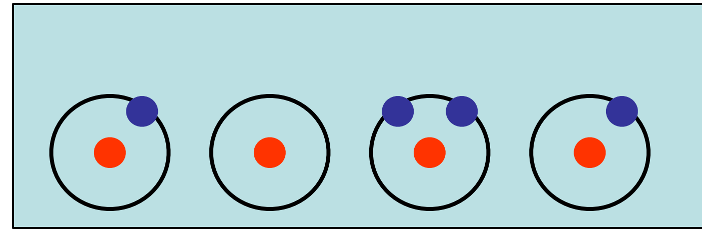

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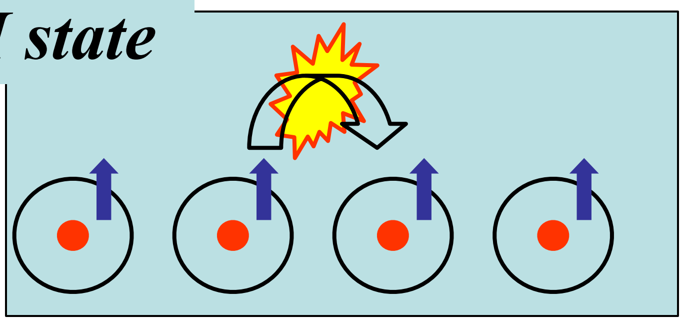
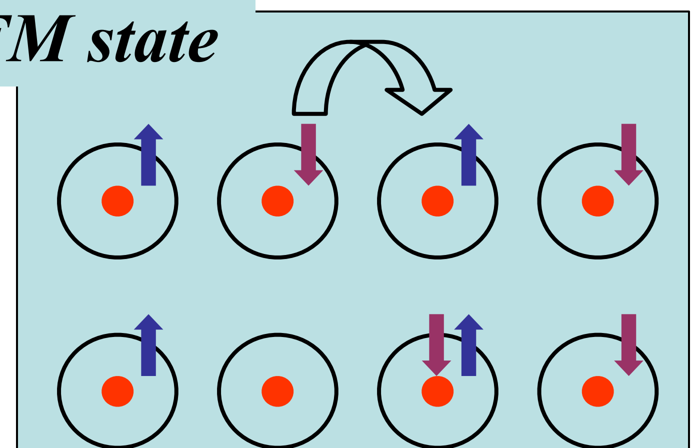
FM state



E = 0

AFM state



E = -t²/U

Atomic U versus solid state U

On-site Coulomb Interaction U				
Ion	*config*	$U_{at}(eV)$	*Solid*	U_{sol} (eV)
Cu	d^9	16.3	CuS	7.0
Ni	d^8	18.0	NiO	7.5
			NiS	5.5
Co	d^7	16.2	CoS_2	4.2
Fe	d^6	14.7	FeO	7.0
Mn	d^5	20.2	MnO	7.0
C_{60}		3.4	C_{60}	1.6

Effective Coulomb interaction for C_{60}		
Geometry	U^{eff}	V^{eff}
Bulk theory	1.75	0.43
Bulk exp. [20]	1.7 ± 0.2	0.4 ± 0.2
(111) Surface	2.04	0.62
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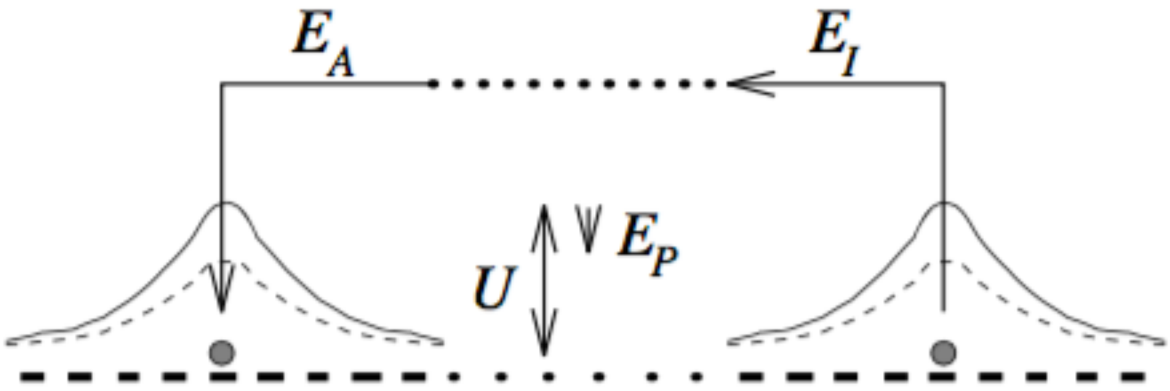
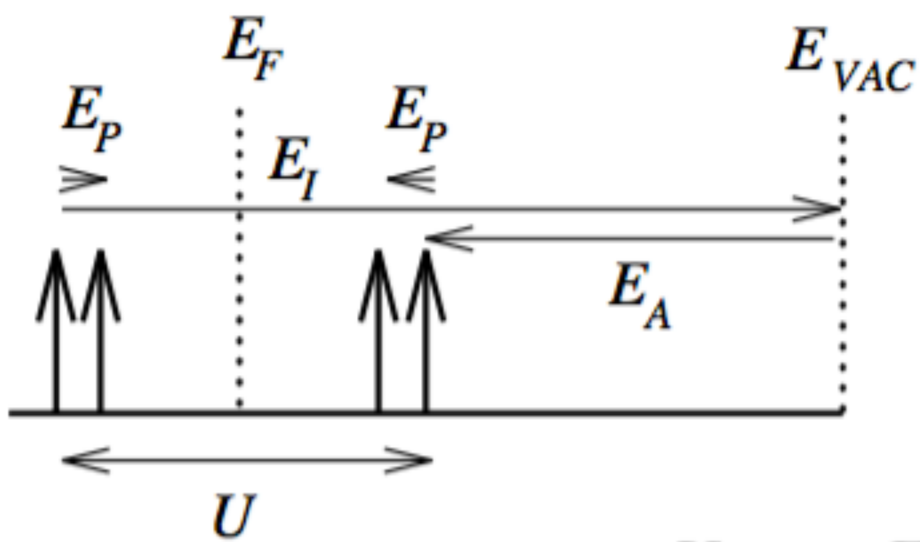
Screening of Hubbard U





$$U_{eff} = E_I - E_A - 2Ep.$$

Microscopic origin polarization energy

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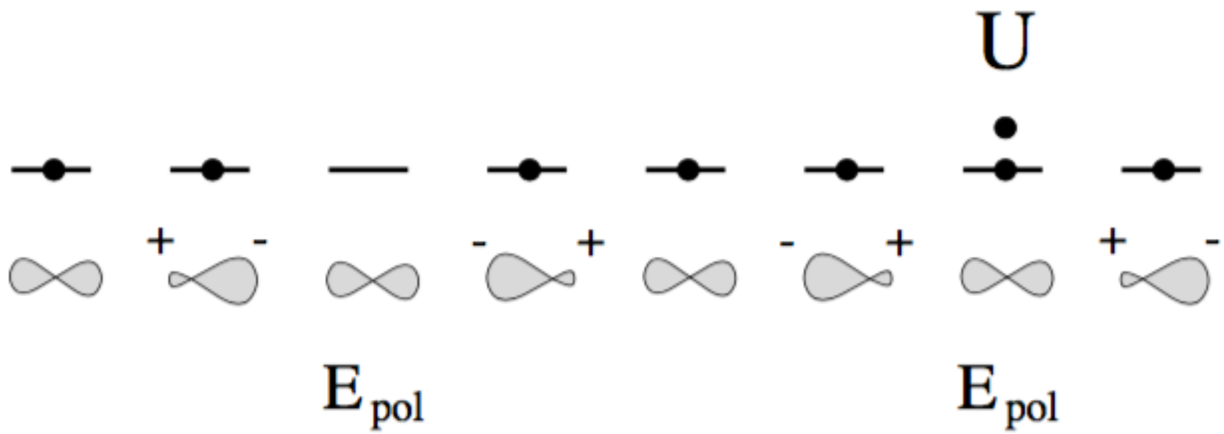

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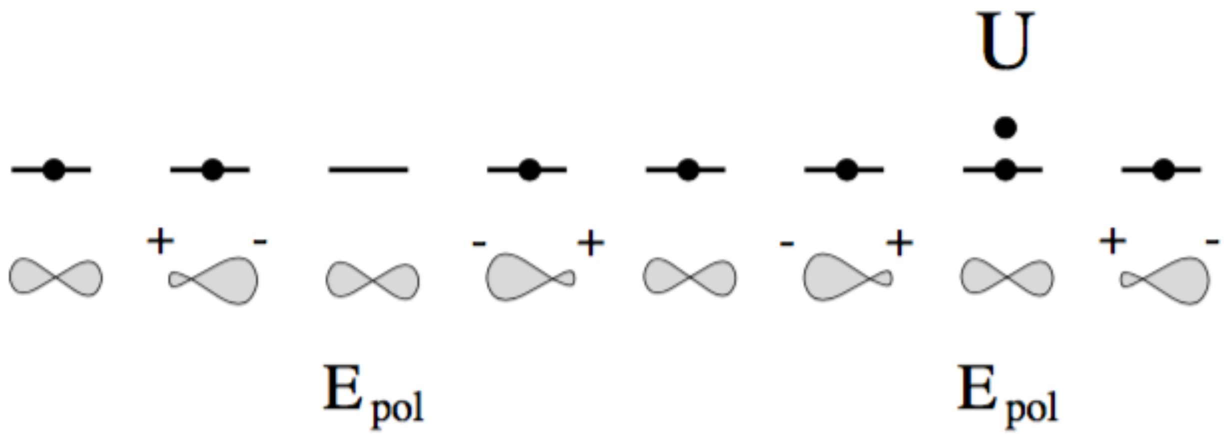

$$\mathbf{p}(\mathbf{r}_i) = \alpha_i^{\mathrm{el}} \mathbf{F}(\mathbf{r}_i) \qquad \Delta E_i = -\tfrac{1}{2}\alpha_i^{\mathrm{el}} \, |\mathbf{F}(\mathbf{r}_i)|^2$$

Clausius-Mossotti

$$\frac{\epsilon - 1}{\epsilon + 2} = 4\pi\alpha/3$$

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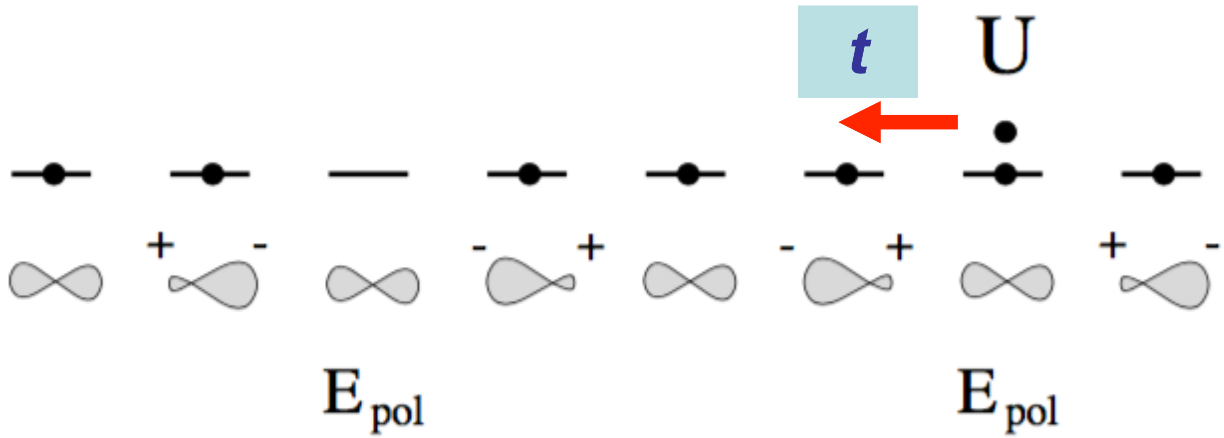

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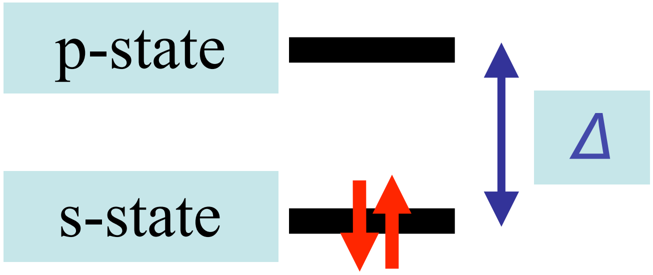
Microscopic polarization

$$
\begin{aligned}
H_{\text{pol}}^{1\text{D}} &= \frac{e^2}{a^2}|X_{sp}|\sum_i(\hat{n}_{i+1} - \hat{n}_{i\ \ 1})(p_i^\dagger s_i + s_i^\dagger p_i) \\
&+ \Delta_{\text{sp}} p_i^\dagger p_i .
\end{aligned}
$$



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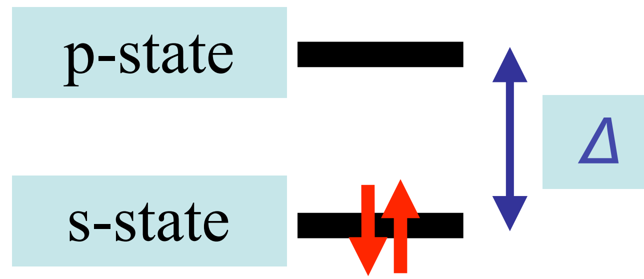

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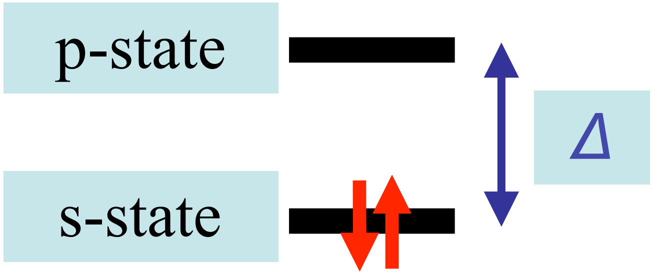
\begin{aligned}
E &= E_0 - \sum_i \frac{|\langle\phi_i|H^{1\mathrm{D}}_{\mathrm{pol}}|\phi_0\rangle|^2}{\Delta_{\mathrm{sp}}} \\
&= E_0 - \sum_i \frac{(eX_{sp})^2}{\Delta_{\mathrm{sp}}}\mathbf{F}_i^2 .
\end{aligned}
$$

↓ α

atomic polarizability

Microscopic polarization

$$H_{\rm pol}^{\rm 1D} = \frac{e^2}{a^2}|X_{sp}|\sum_i(\hat{n}_{i+1} - \hat{n}_{i\;\,1})(p_i^\dagger s_i + s_i^\dagger p_i) + \Delta_{\rm sp}p_i^\dagger p_i.$$



static doublon+hole

t/Δ →0

$$E = E_0 - \sum_i \frac{|\langle\phi_i|H_{\rm pol}^{\rm 1D}|\phi_0\rangle|^2}{\Delta_{\rm sp}} = E_0 - \sum_i \frac{(eX_{sp})^2}{\Delta_{\rm sp}}\mathbf{F}_i^2.$$

↓ α

atomic polarizability

Polarizability screening of Hubbard U

polarization field:

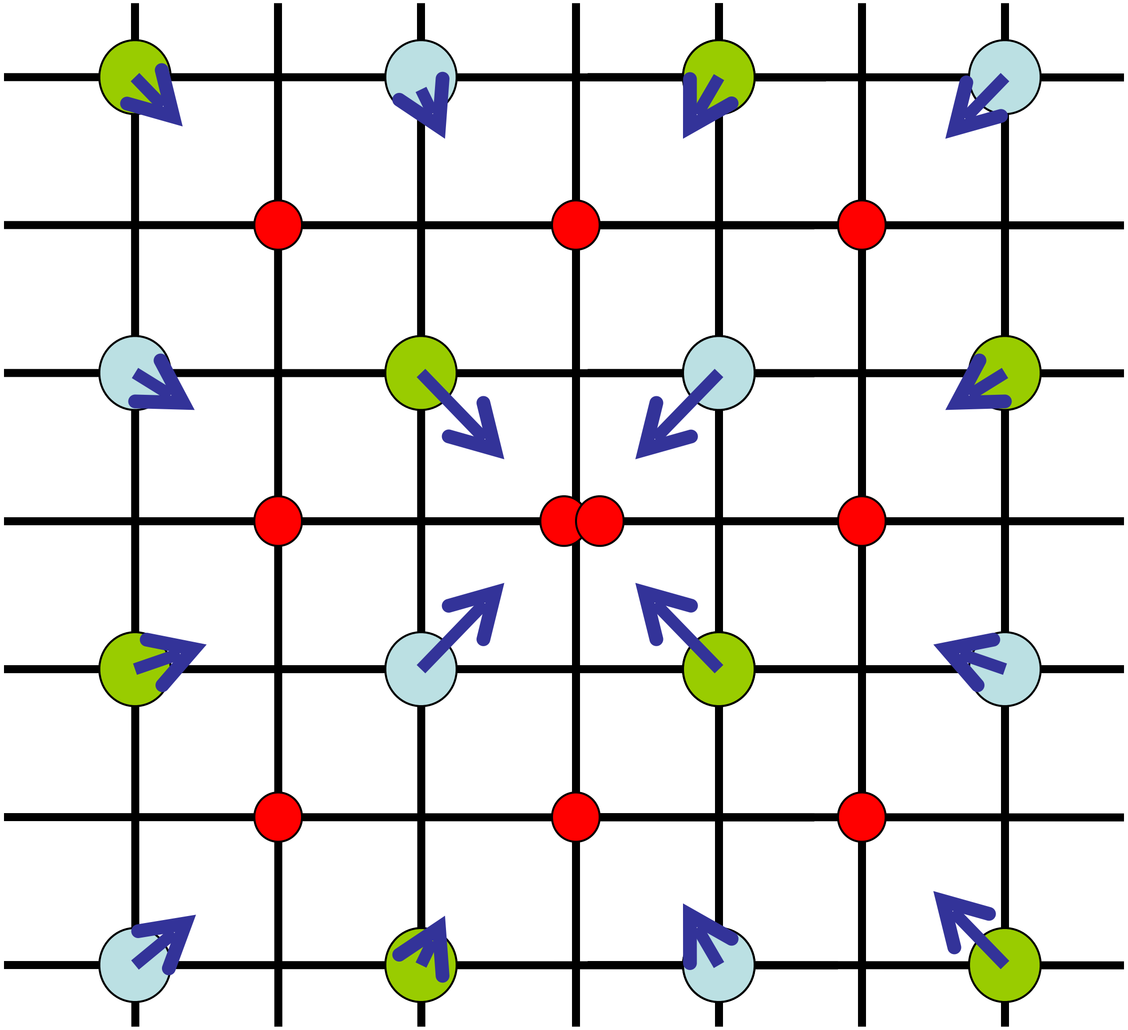
$$\vec{P}_i = \alpha \vec{E}_i$$

atomic polarizability: α

pol. energy:

$$E_{pol} = \frac{1}{2} \sum_i \vec{E}_i \cdot \vec{P}_i$$

$$\alpha_{oxygen} \approx 2A^3$$



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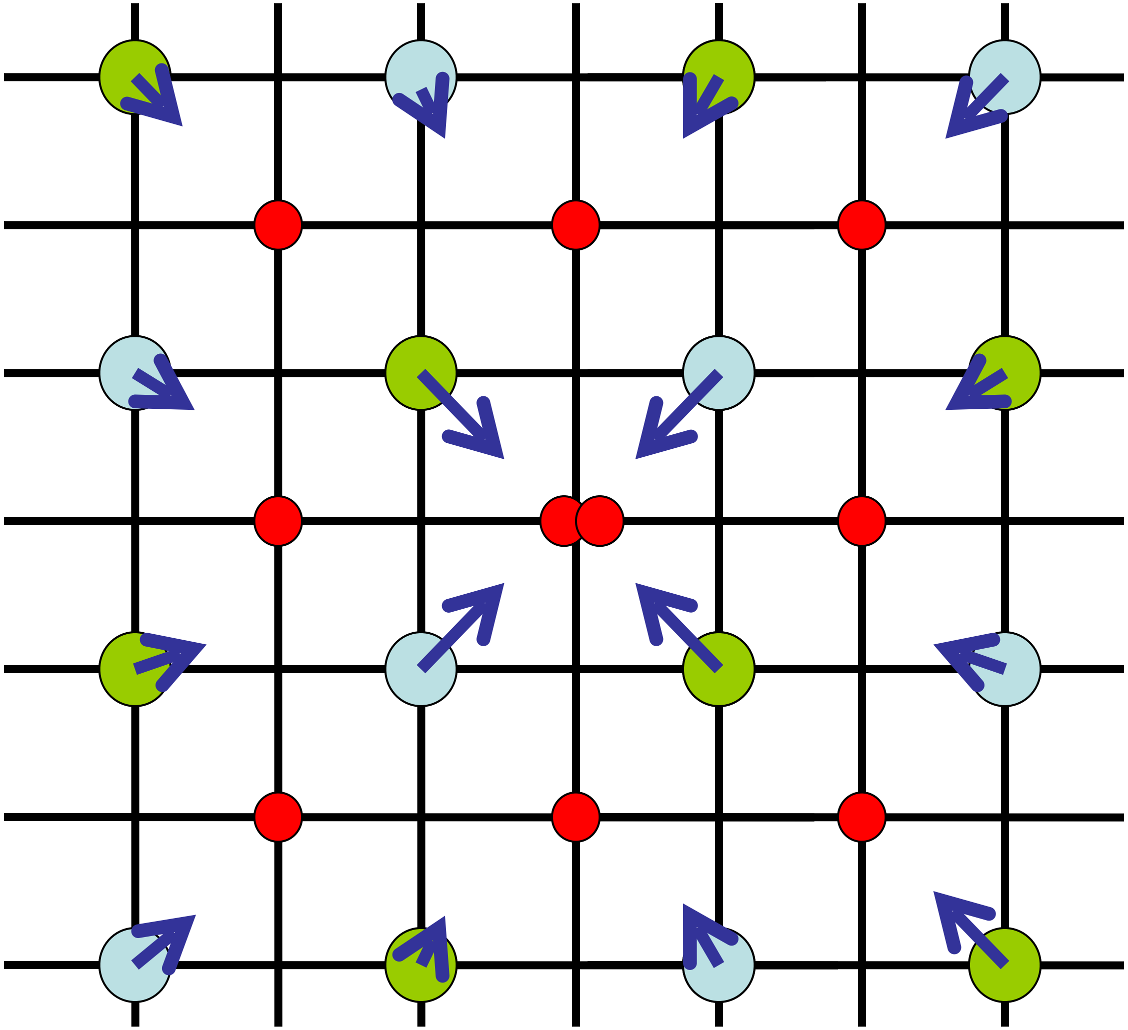
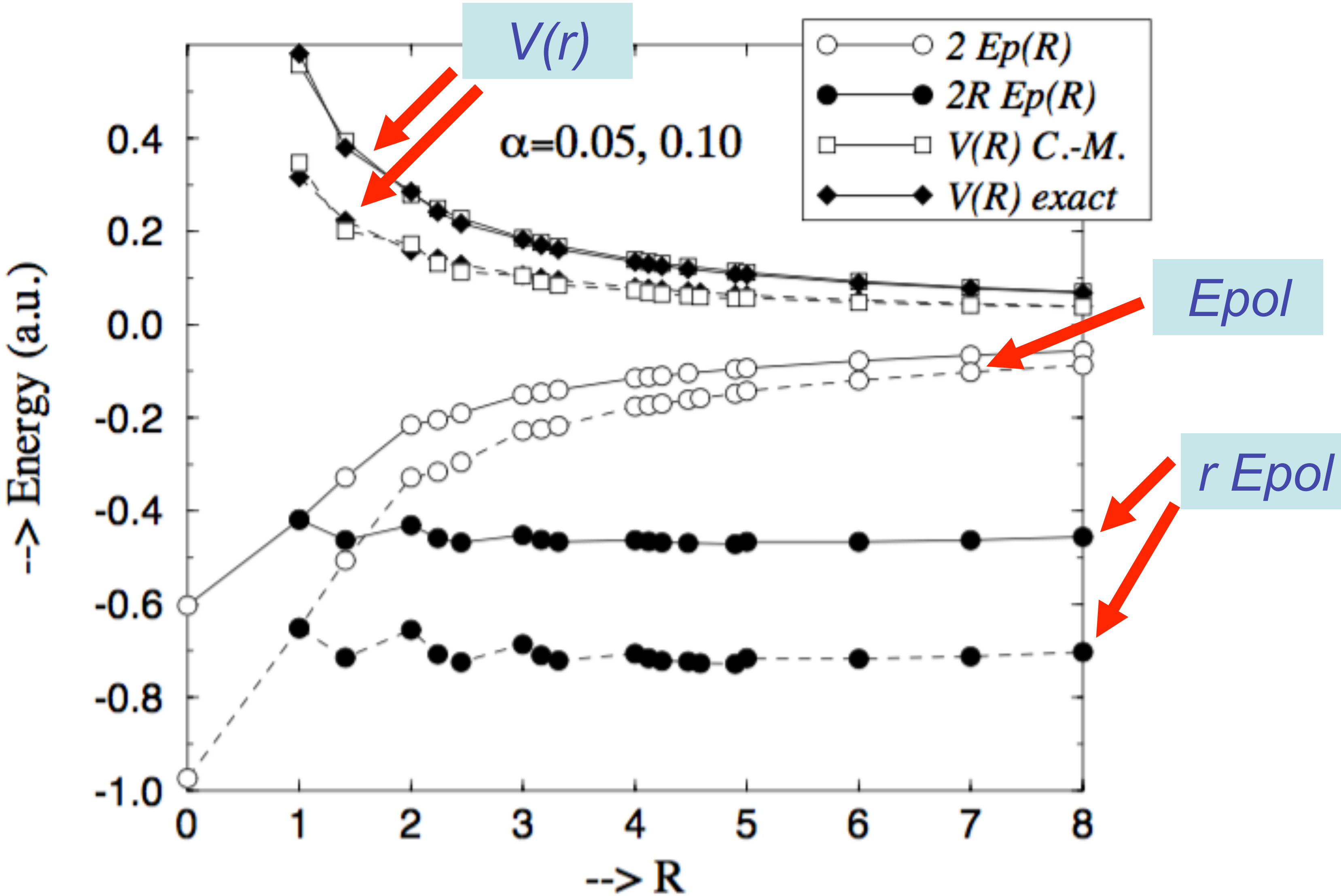


$$U = U_{atomic} - 2E_{pol}$$

but does this hold at large distances?

Polarizability versus dielectric screening in 3D



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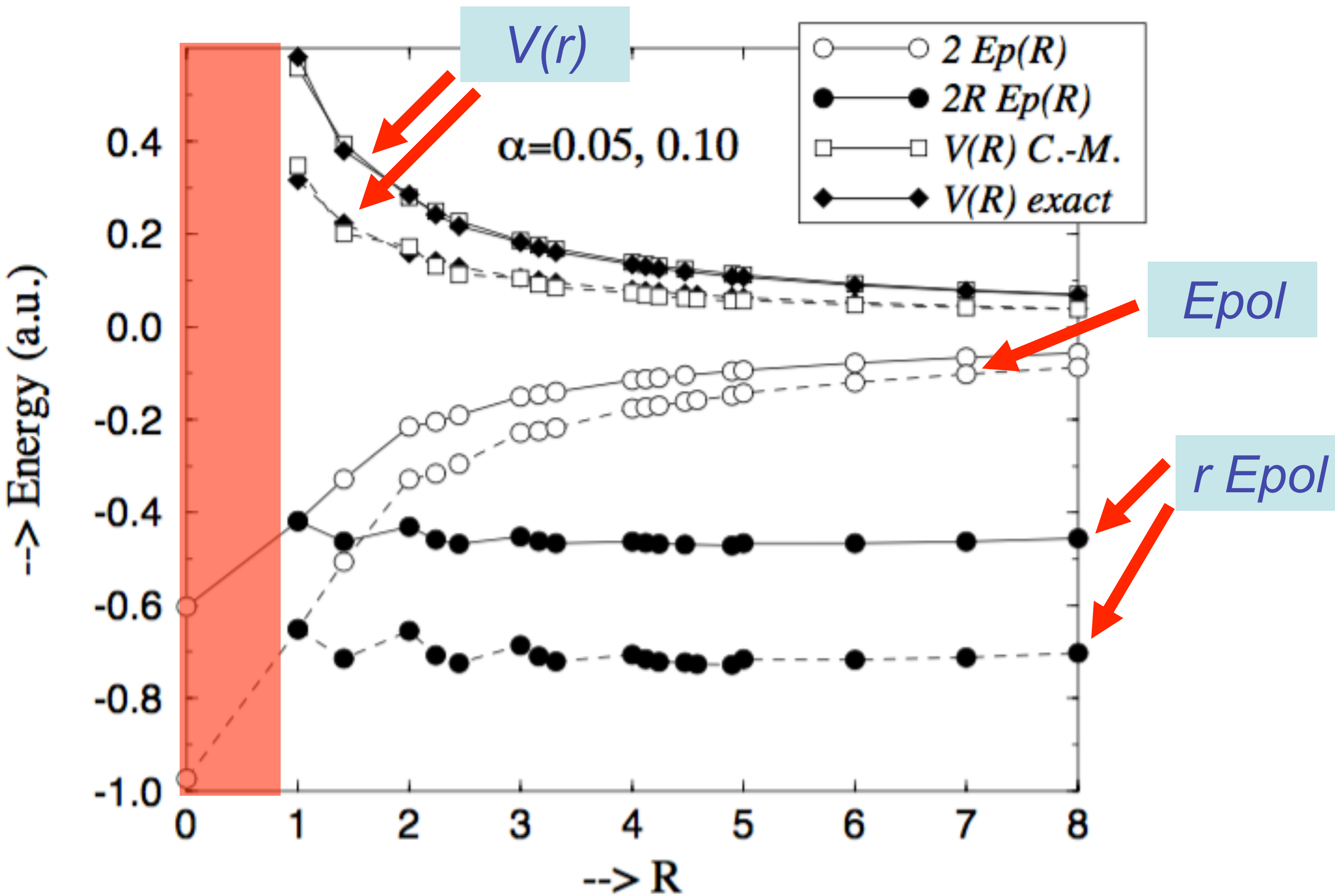
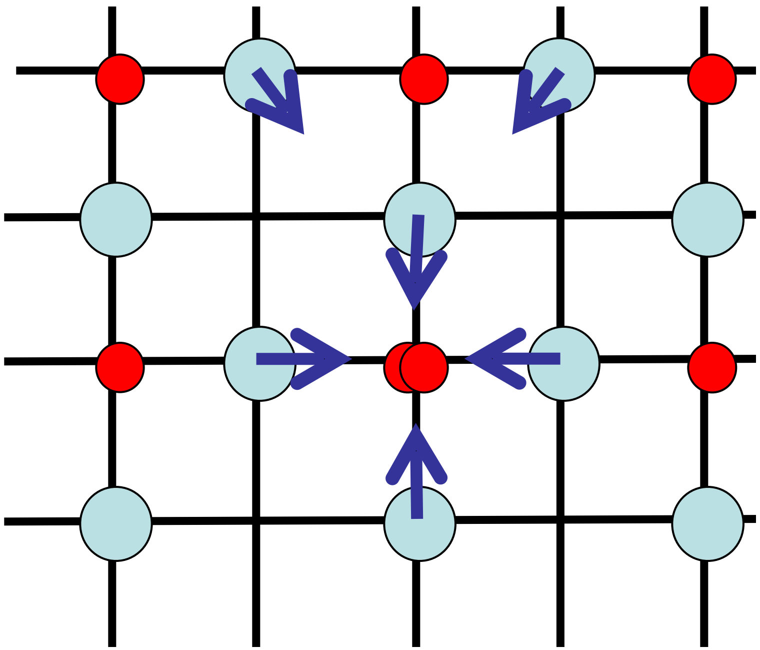
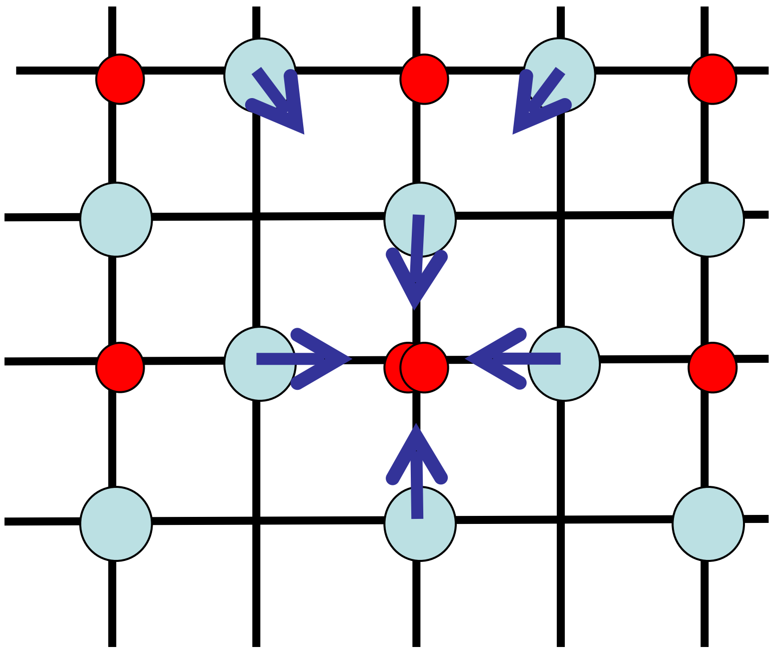


Screening of neighbor V on a 2D CuO_2 lattice





$$\alpha_{oxygen} \approx 2A^3$$

Cu-O-Cu bond angle 180°

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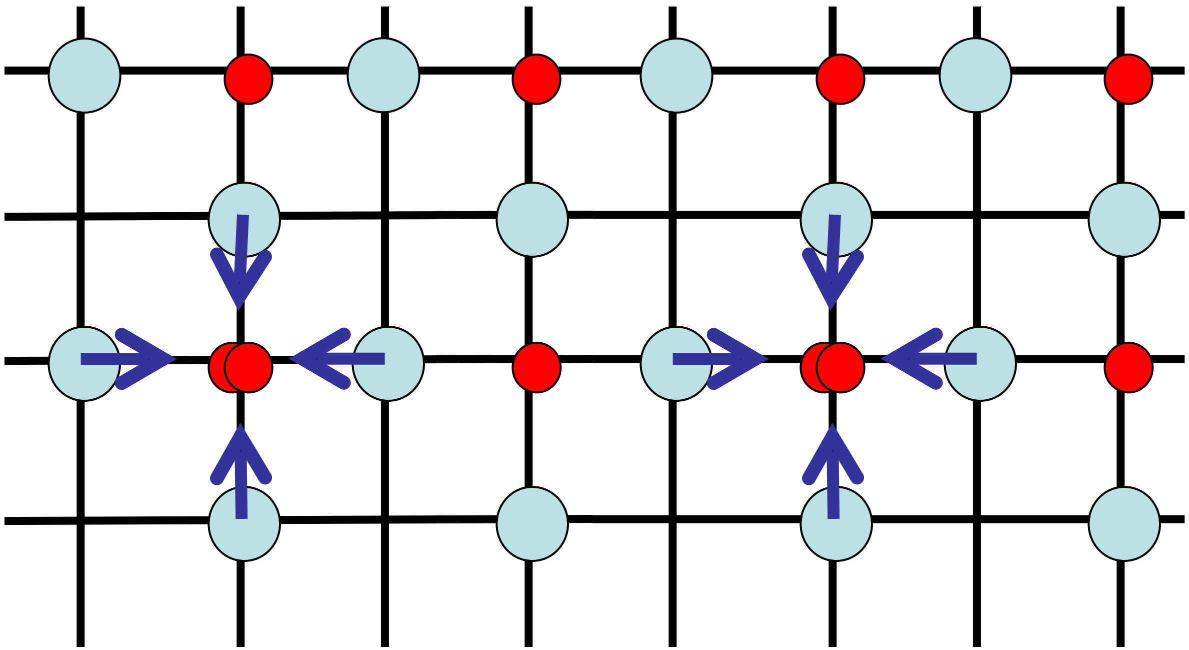

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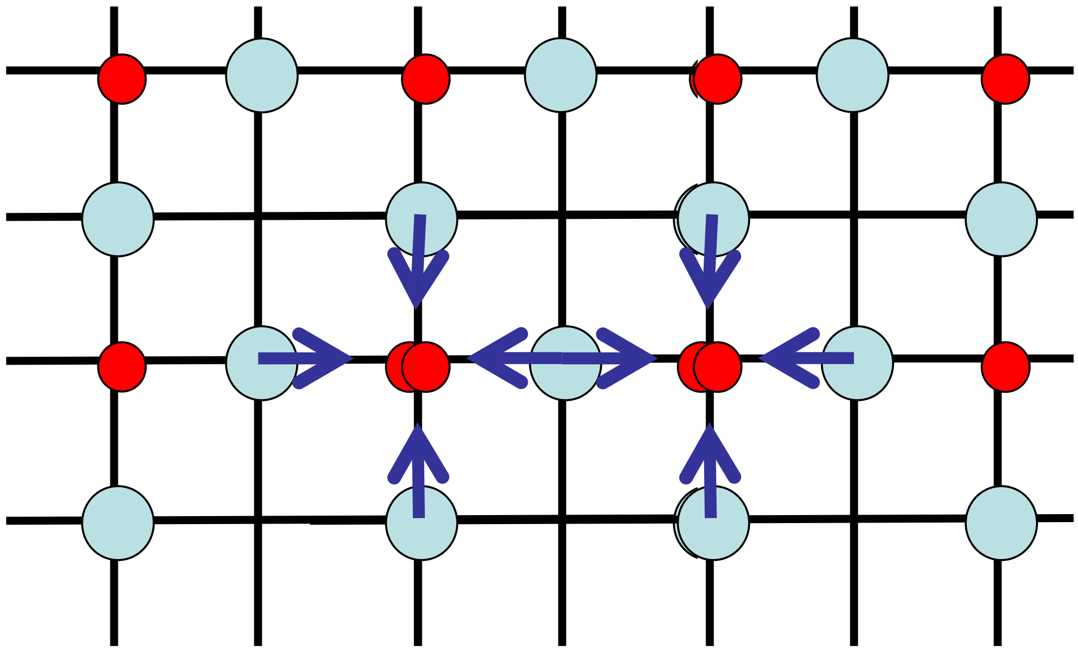

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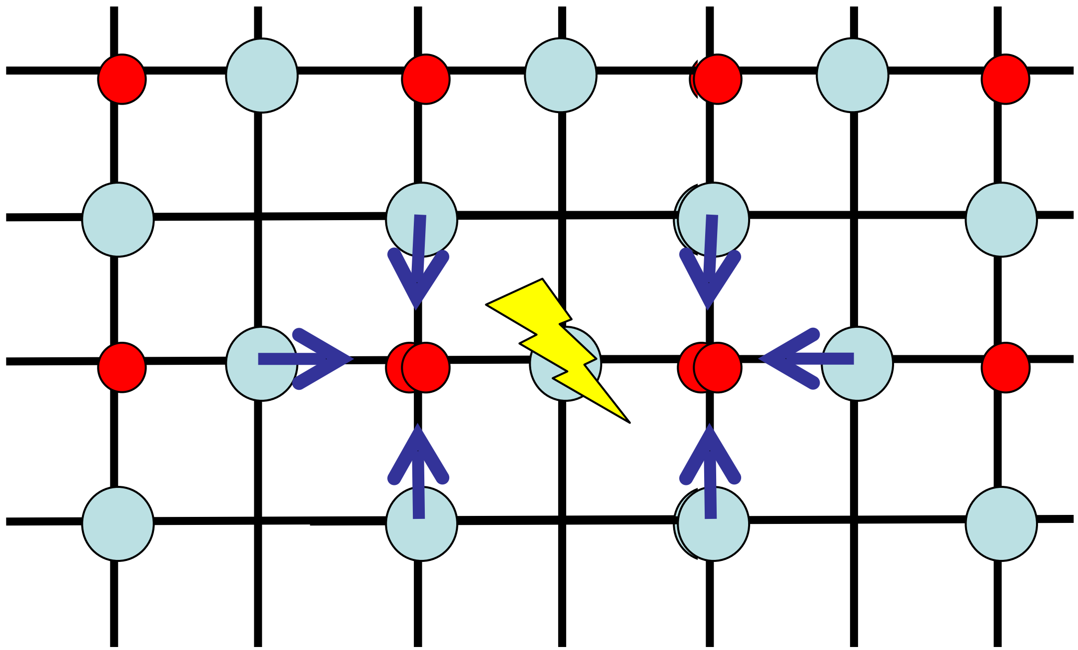


polarization partially antiparallel
nearest neighbor V is antiscreened

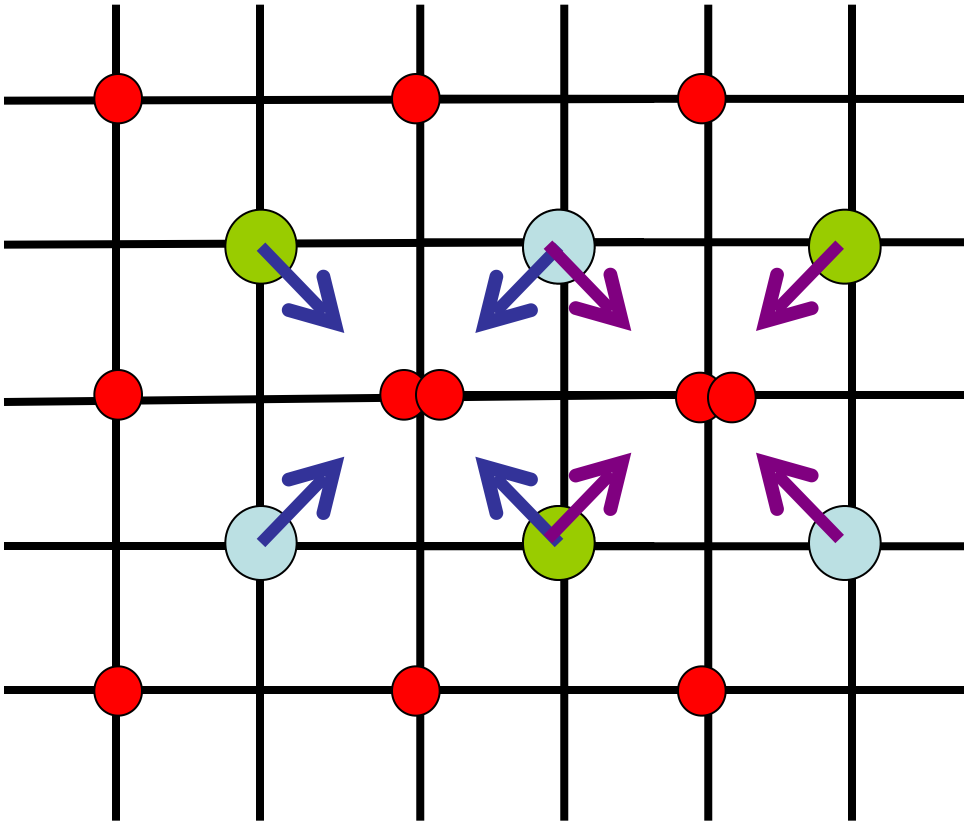
$$\alpha_{oxygen} \approx 2A^3$$

Cu-O-Cu bond angle 180°

Screening of neighbor V on 2D cubic lattice

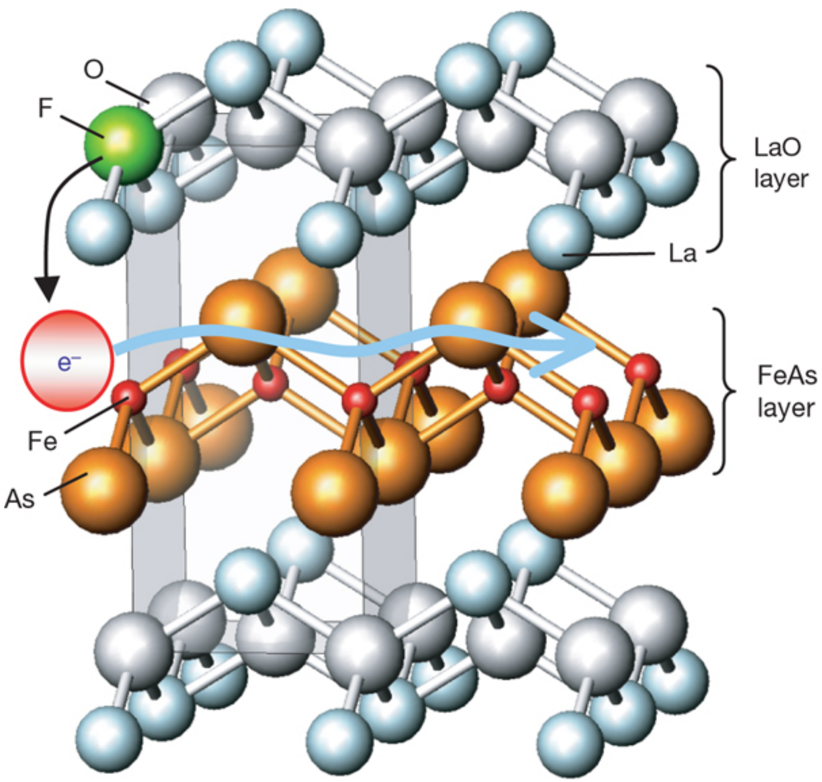
two polarization fields partially perpendicular

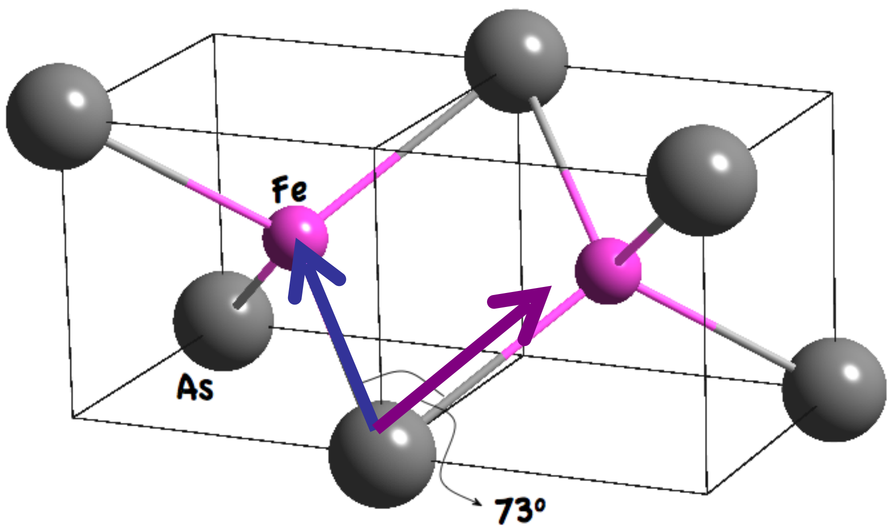
nearest neighbor V is (partially) unscreened



bond angle 90^o

Screening of neighbor V on $BaFe_2As_2$ lattice



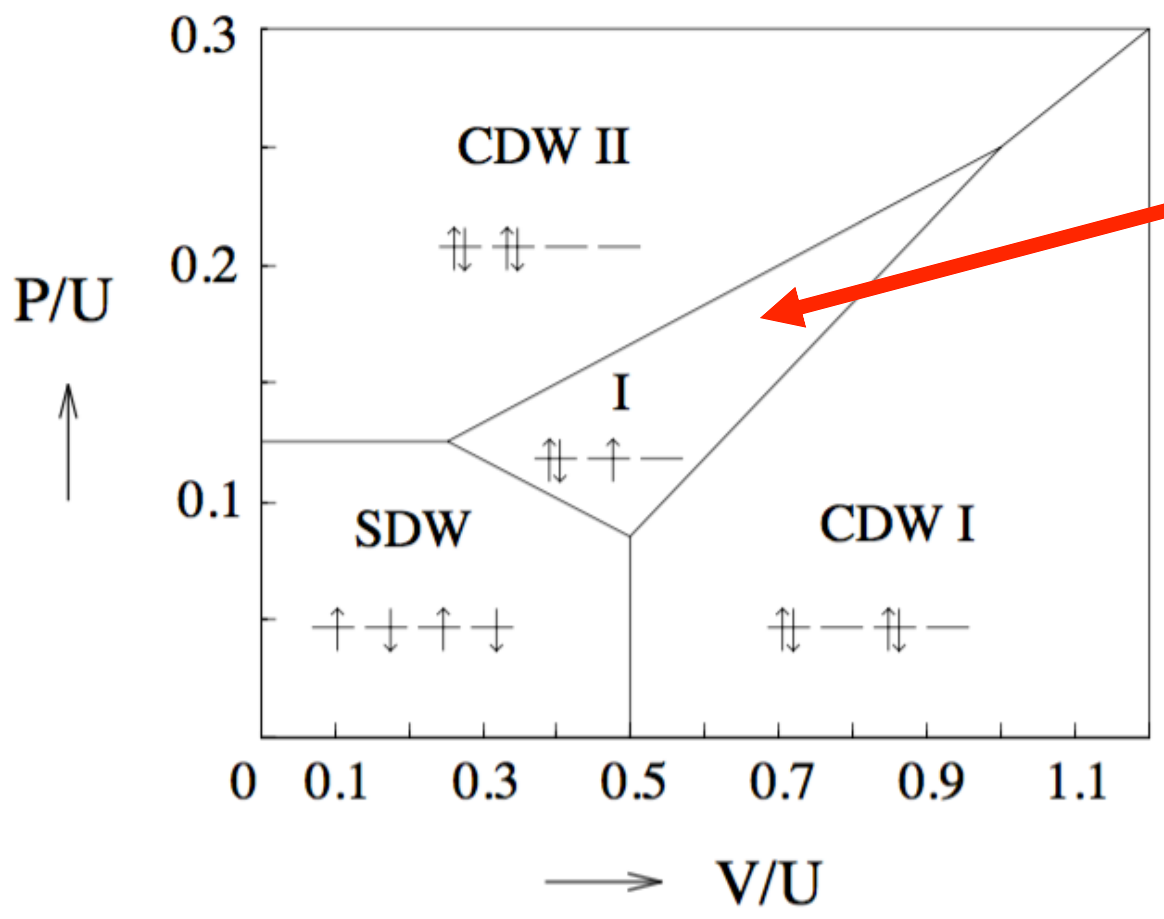


$$\alpha_{arsenic} \approx 10\,A^3$$

angle of 73°:
nearest neighbor V is overscreened

Sawatzky, Elfimov, JvdB, Zaanen
EPL 86, 17006 (2009)

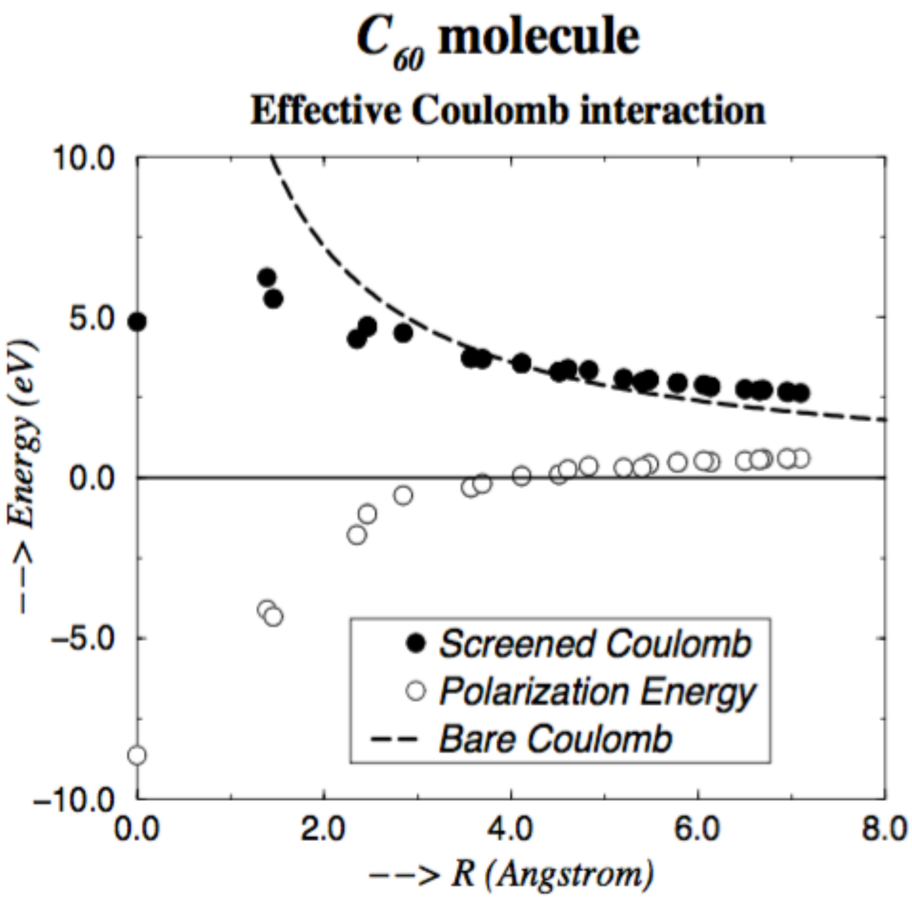
New types of charge order emerge

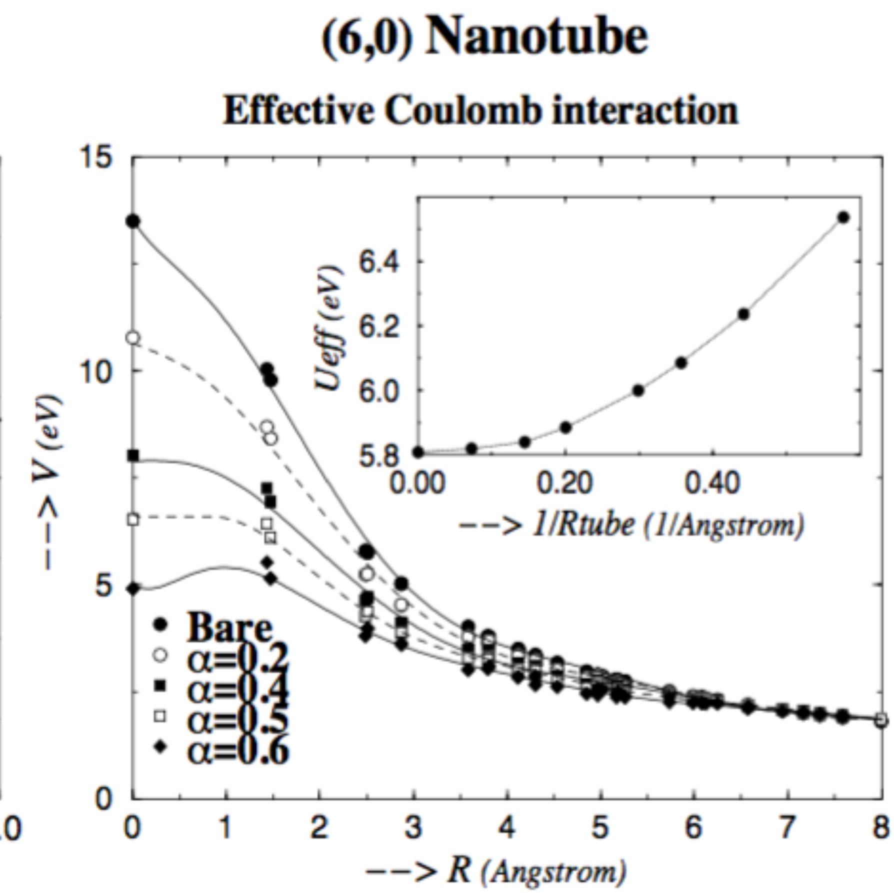


Broken inversion symmetry

JvdB, Meinders, Lorenzana, Eder & Sawatzky, PRL 75, 4658 (1995)

Screened interaction in nano systems



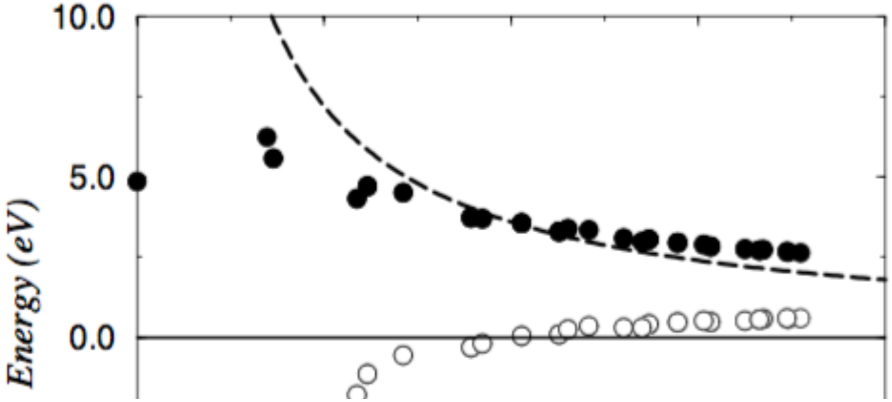


JvdB & Sawatzky , Electronic Properties of Novel Materials - Progress in Molecular Nanostructures, 152 (1998)

Screened interaction in nano systems

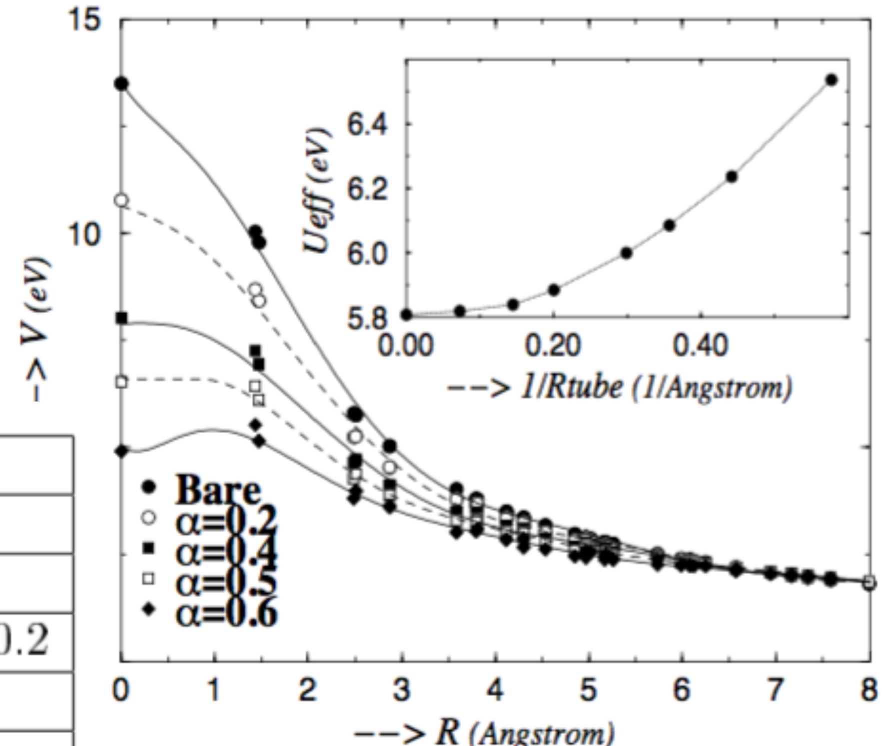
C_{60} molecule

Effective Coulomb interaction



(6,0) Nanotube

Effective Coulomb interaction



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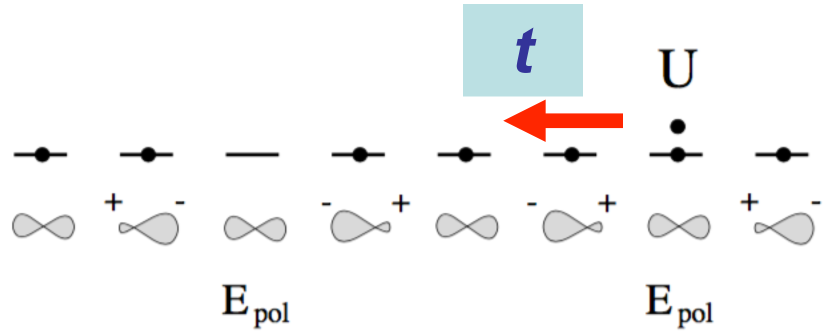
Perturbative dependence of Epol on t/Δ



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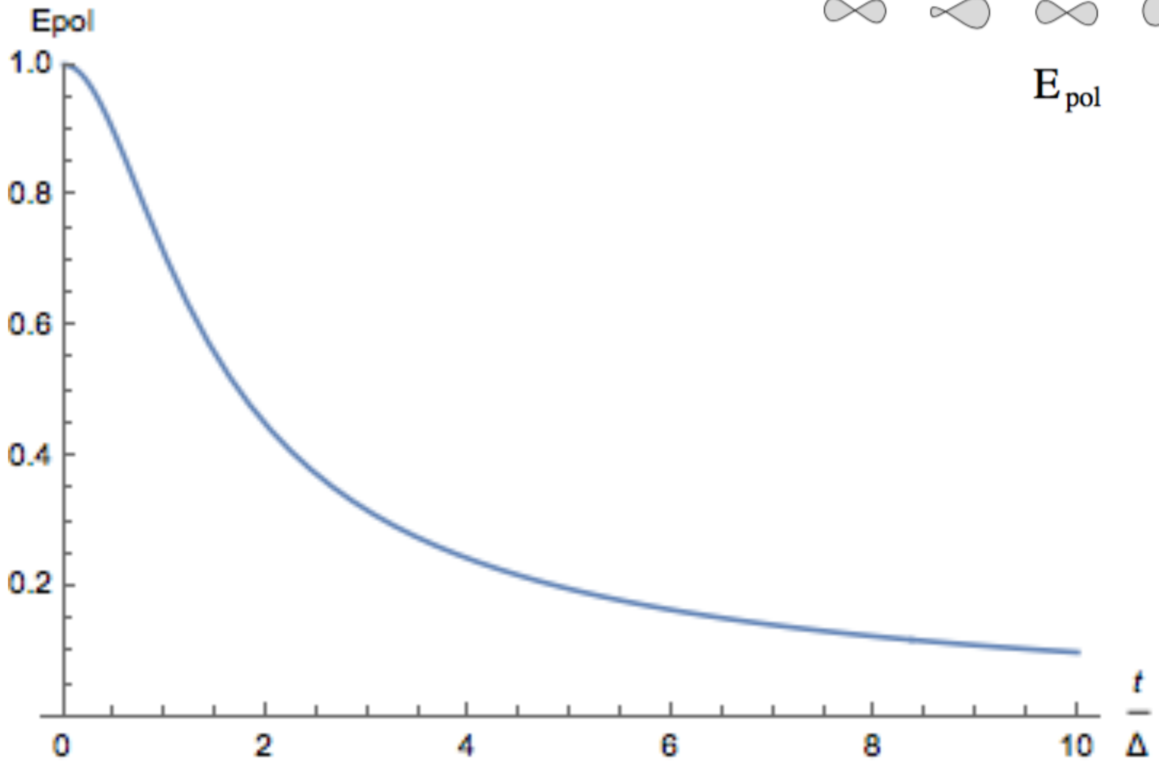
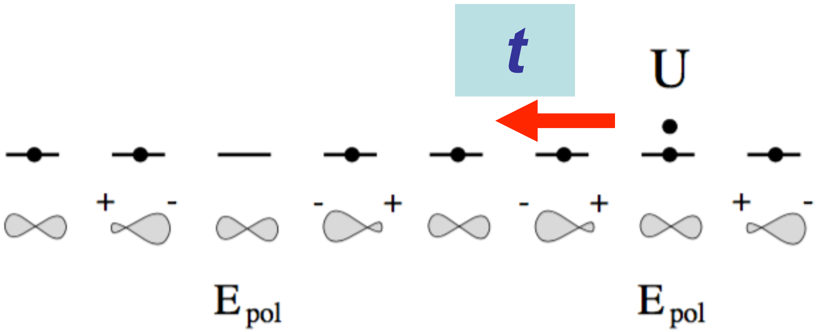
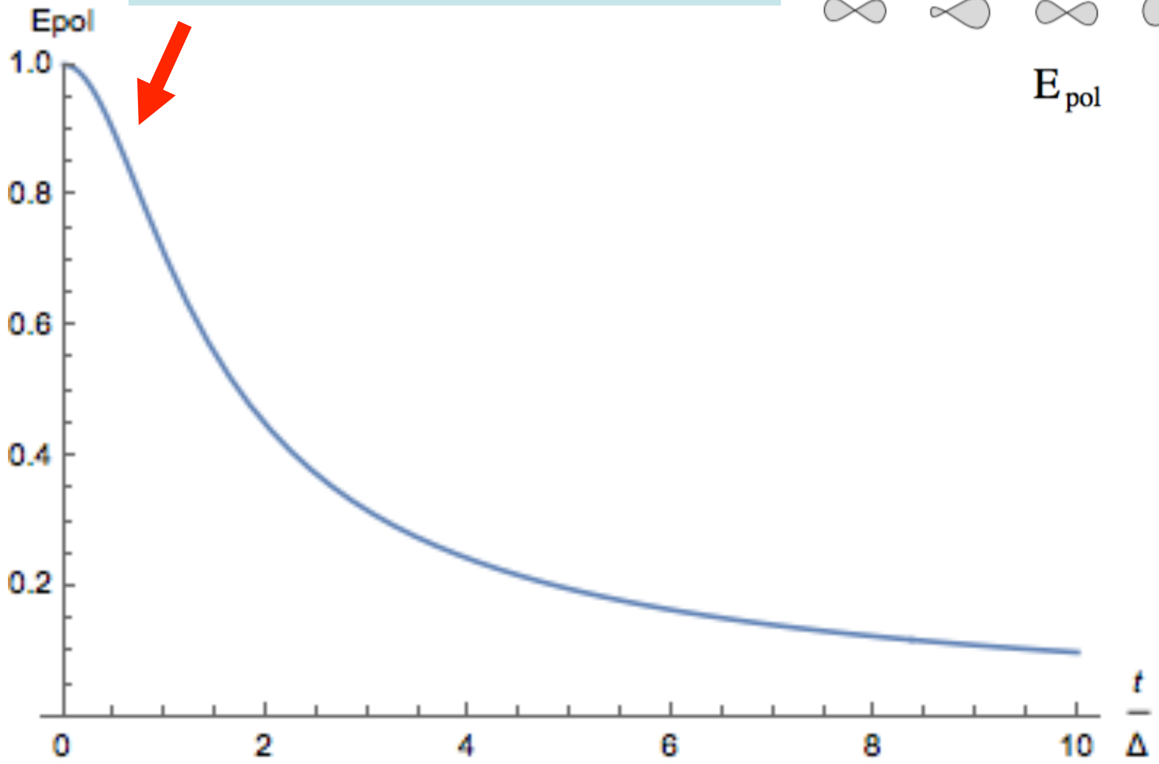
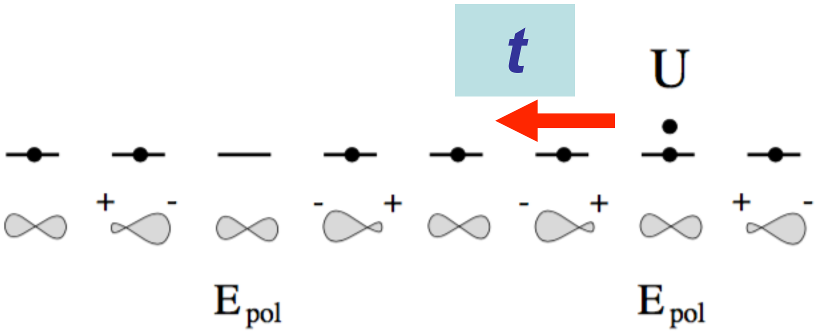




Perturbative dependence of Epol on t/Δ

t << Δ → electrons slow, polarization fast → static limit





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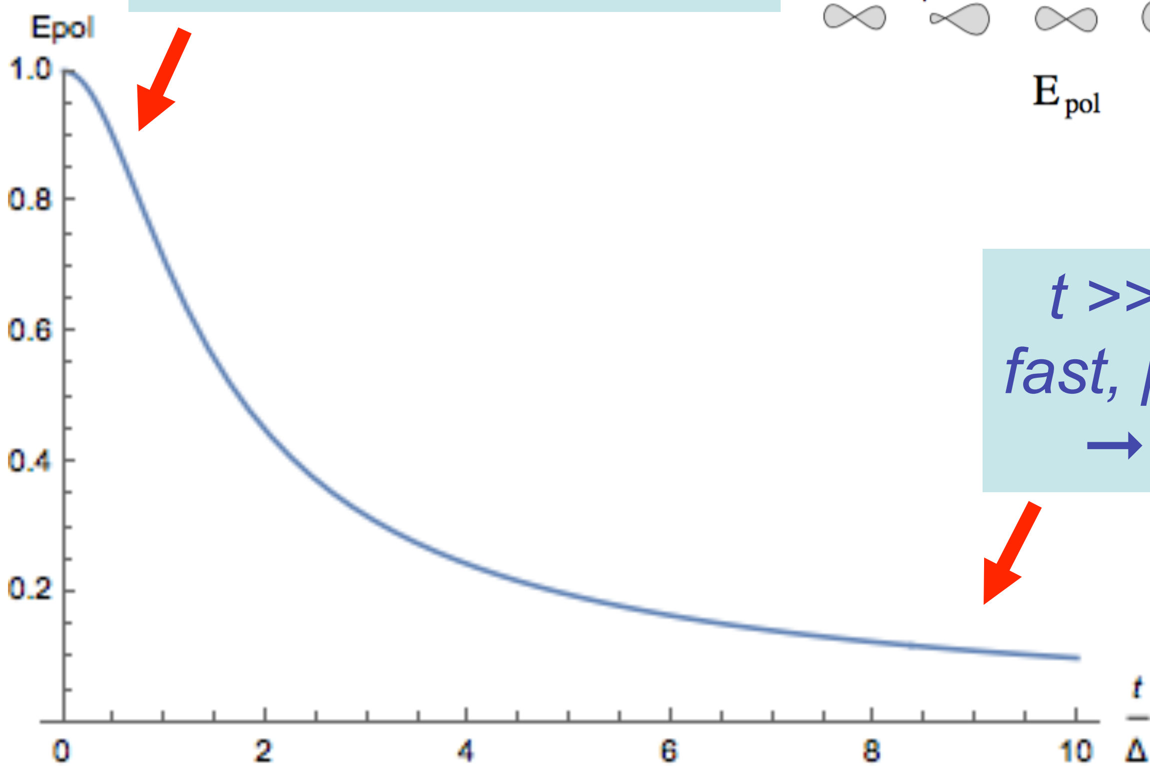
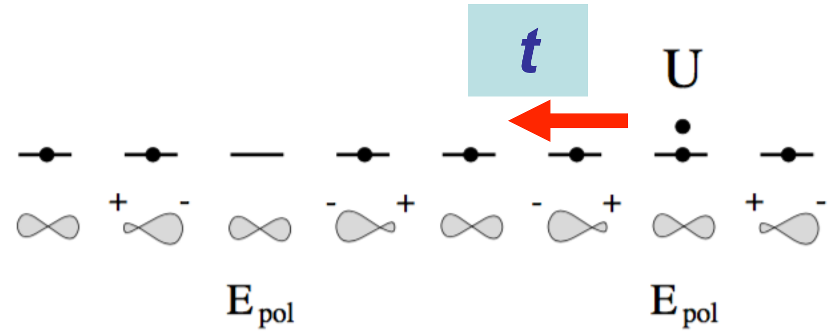
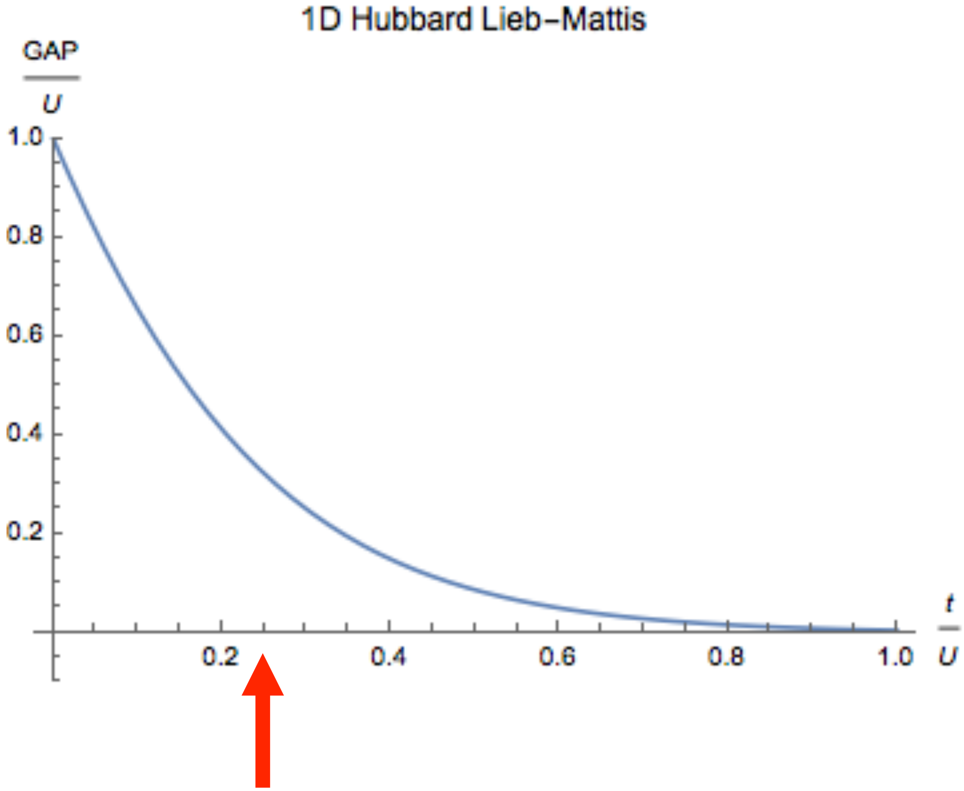




t >> Δ → electrons fast, polarization slow → retarded limit

Apply to 1D Hubbard model

$$GAP = U - 4 - 8\sum_{n=1}^{\infty}(-1)^n\{\sqrt{1+n^2U^2/4} - nU/2\}$$



U= 4t = bandwidth

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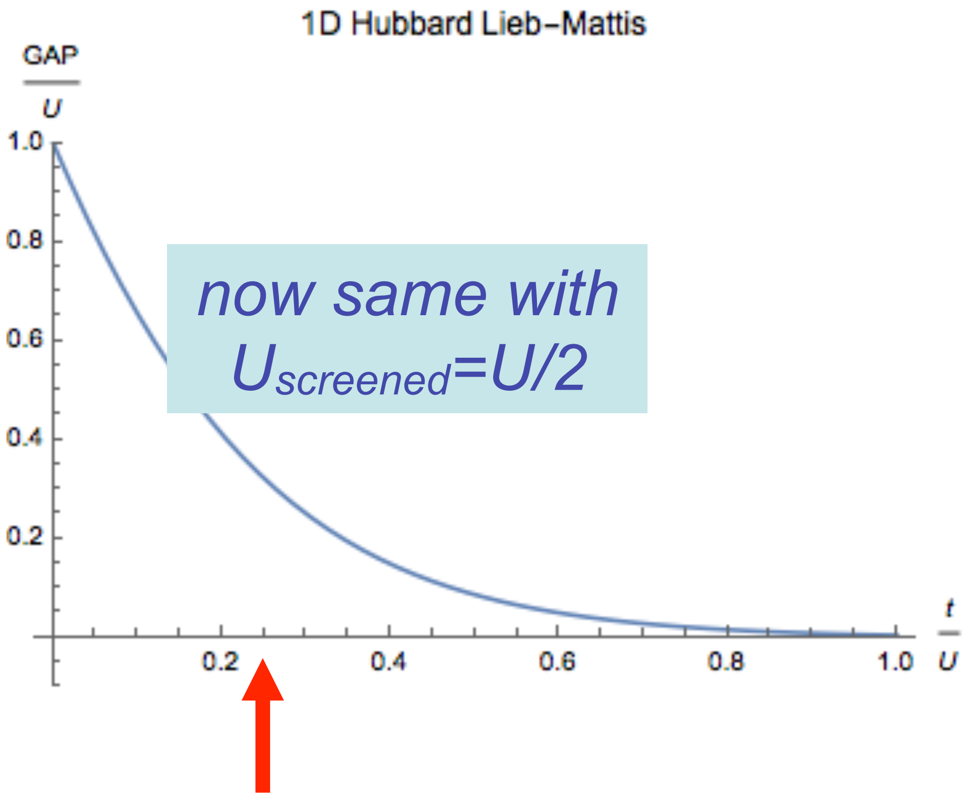

now same with $U_{screened}$=U/2

U= 4t = bandwidth

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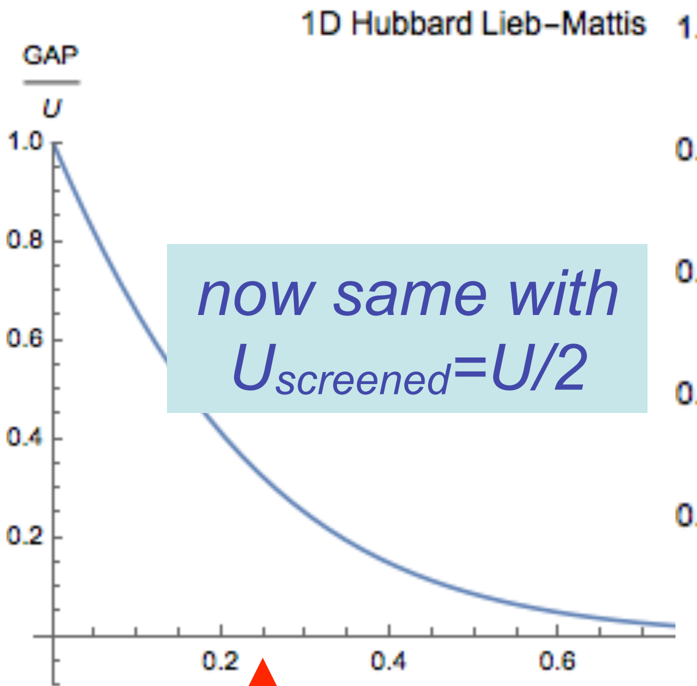
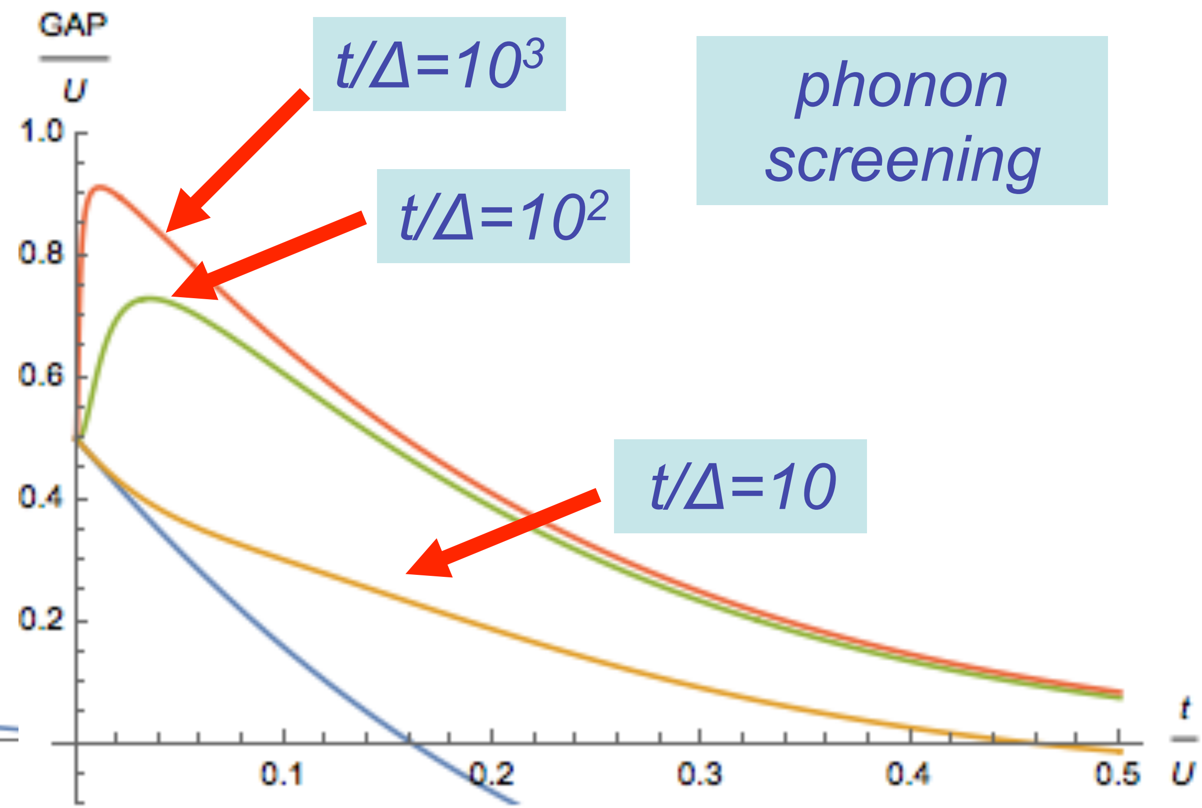


$U = 4t$ = bandwidth



$t/\Delta=1$

static limit

electronic screening

Relation to experiment: RIXS on Li_2CuO_2

ARTICLE

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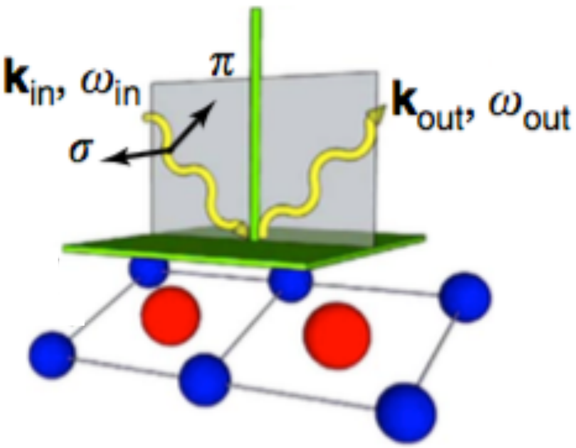
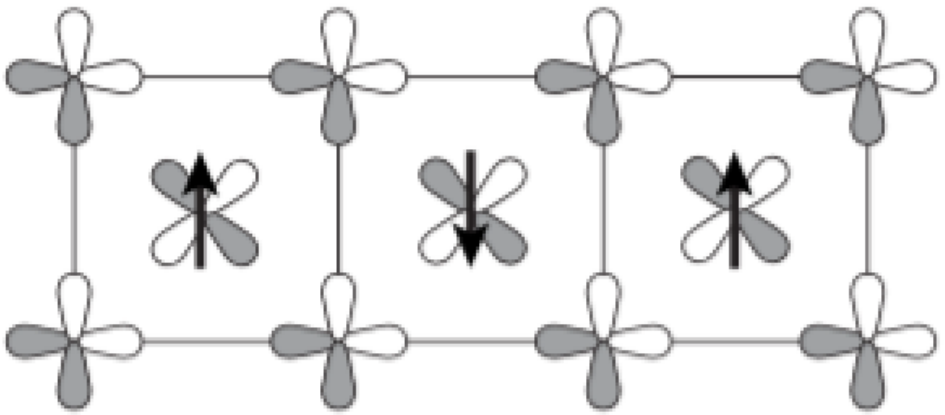
Electron-lattice interactions strongly renormalize the charge-transfer energy in the spin-chain cuprate Li_2CuO_2

Steve Johnston[1], Claude Monney[2,3], Valentina Bisogni[4,5], Ke-Jin Zhou[2,6], Roberto Kraus[4], Günter Behr[4], Vladimir N. Strocov[2], Jiři Málek[7], Stefan-Ludwig Drechsler[4], Jochen Geck[4], Thorsten Schmitt[2] & Jeroen van den Brink[4,8]





What is

Resonant

Inelastic

X-ray scattering

RIXS

What is *Resonant* *Inelastic* *X-ray scattering* } *RIXS*

X-ray scattering: photon in→ solid → photon out

inelastic: $\omega_{out} < \omega_{in}$

resonant: tune ω_{in} *to an atomic absorption edge*

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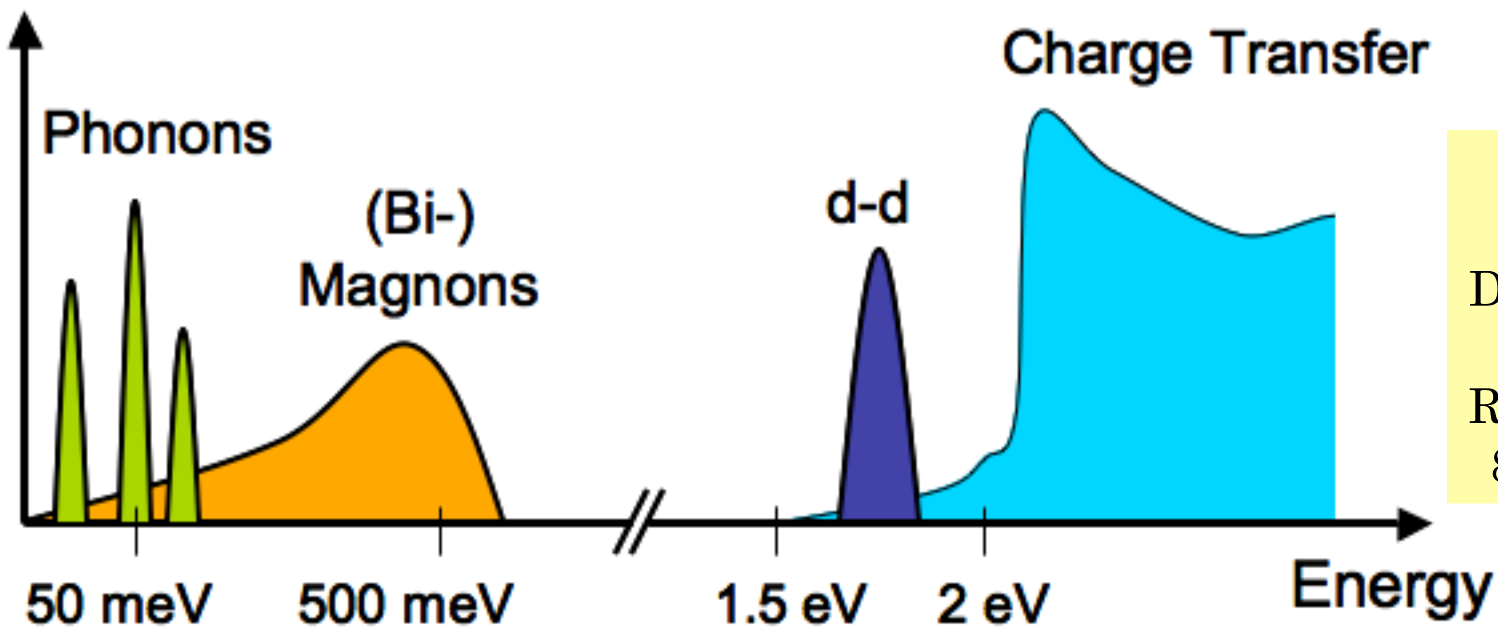

Ament, van Veenendaal, Devereaux, Hill & JvdB
Rev. Mod. Phys. 83, 705 (2011)

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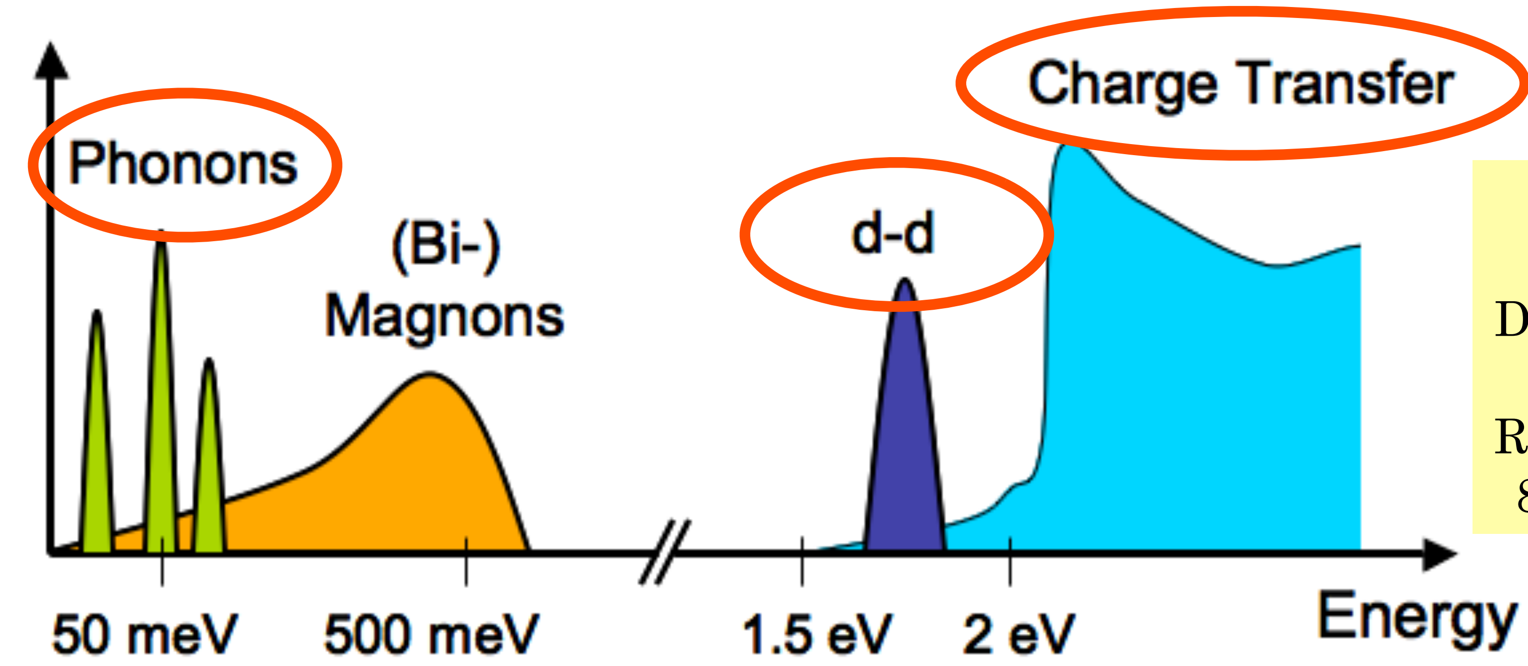

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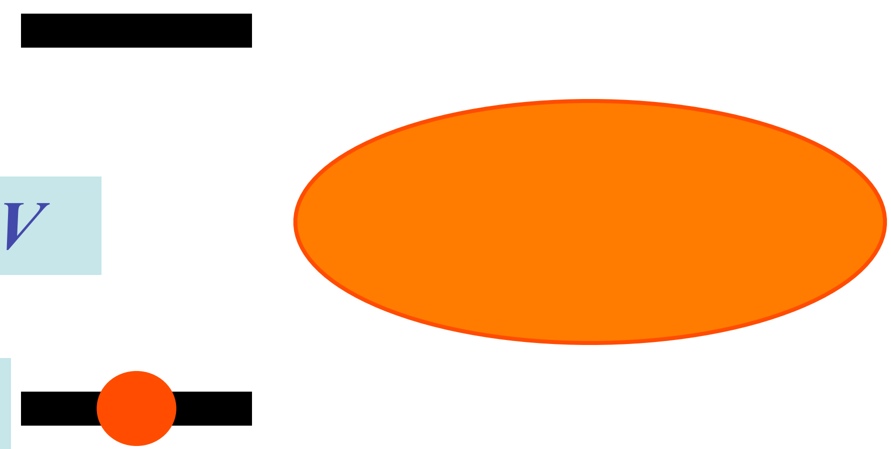
Cu

K-edge

4 p

~9 KeV

1 s



What is *Resonant* *Inelastic* *X-ray scattering* — *RIXS*

X-ray scattering: photon in → solid → photon out

inelastic: $\omega_{out} < \omega_{in}$

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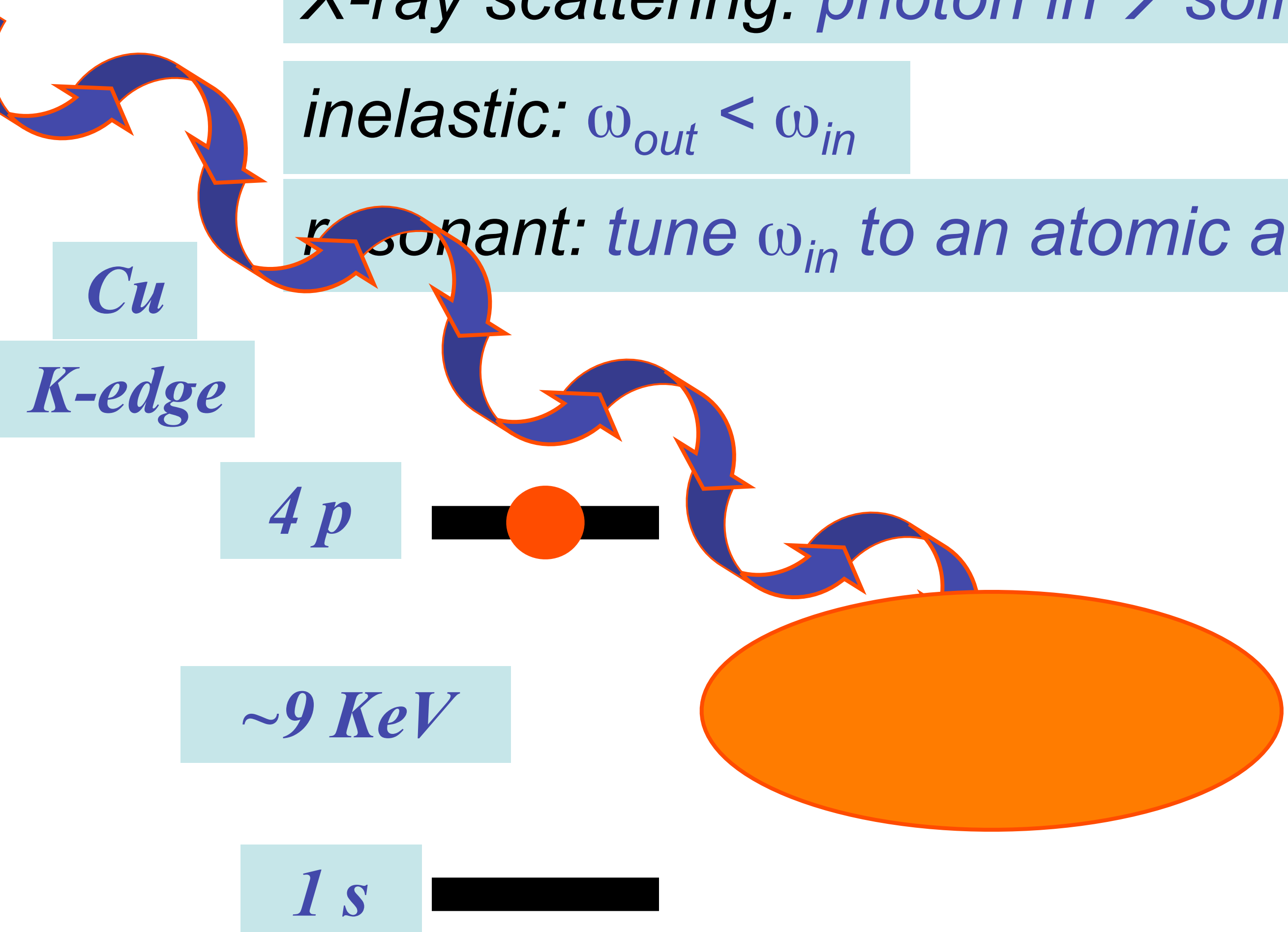

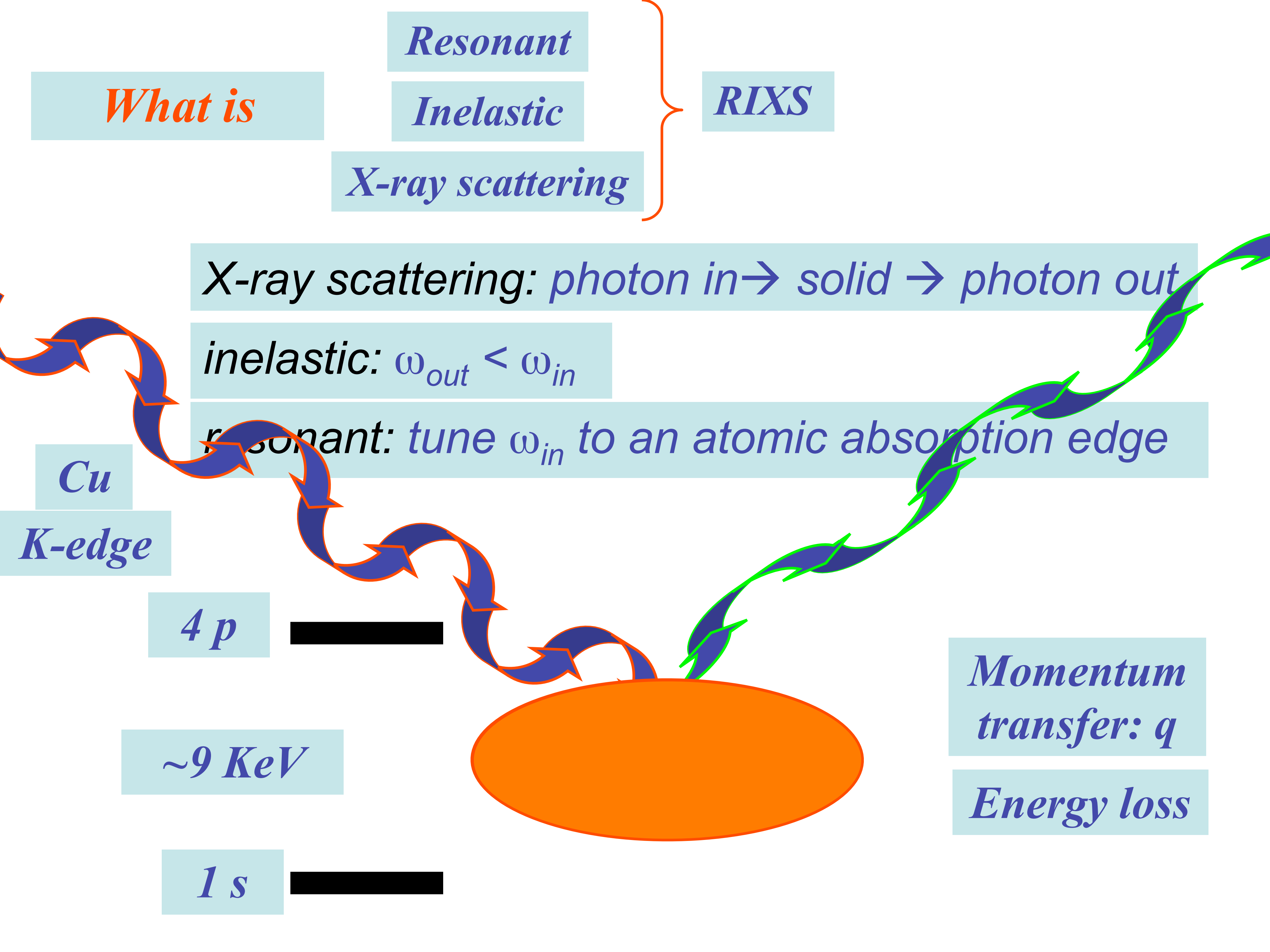
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K-edge
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Momentum transfer: q
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INDIRECT

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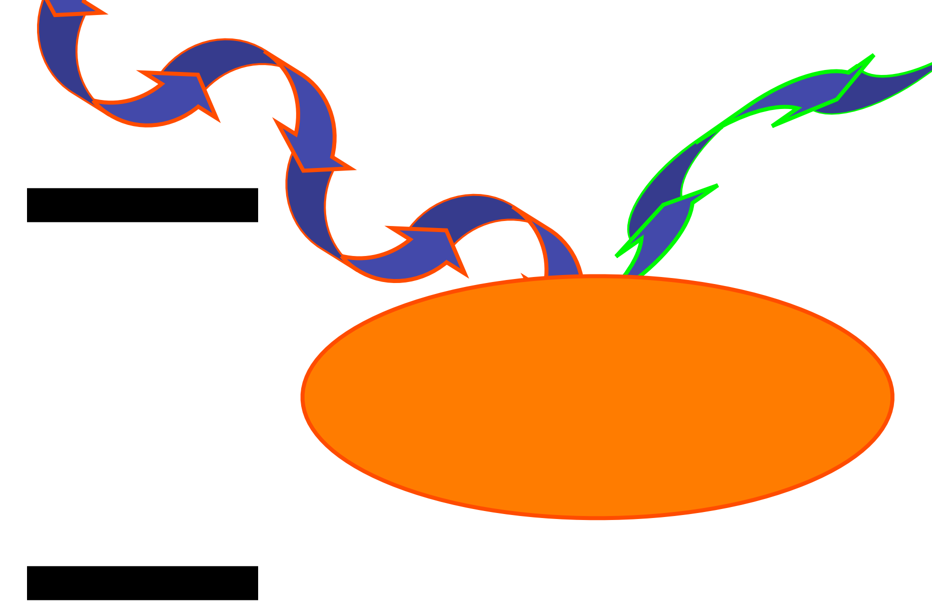
Cu

L-edge



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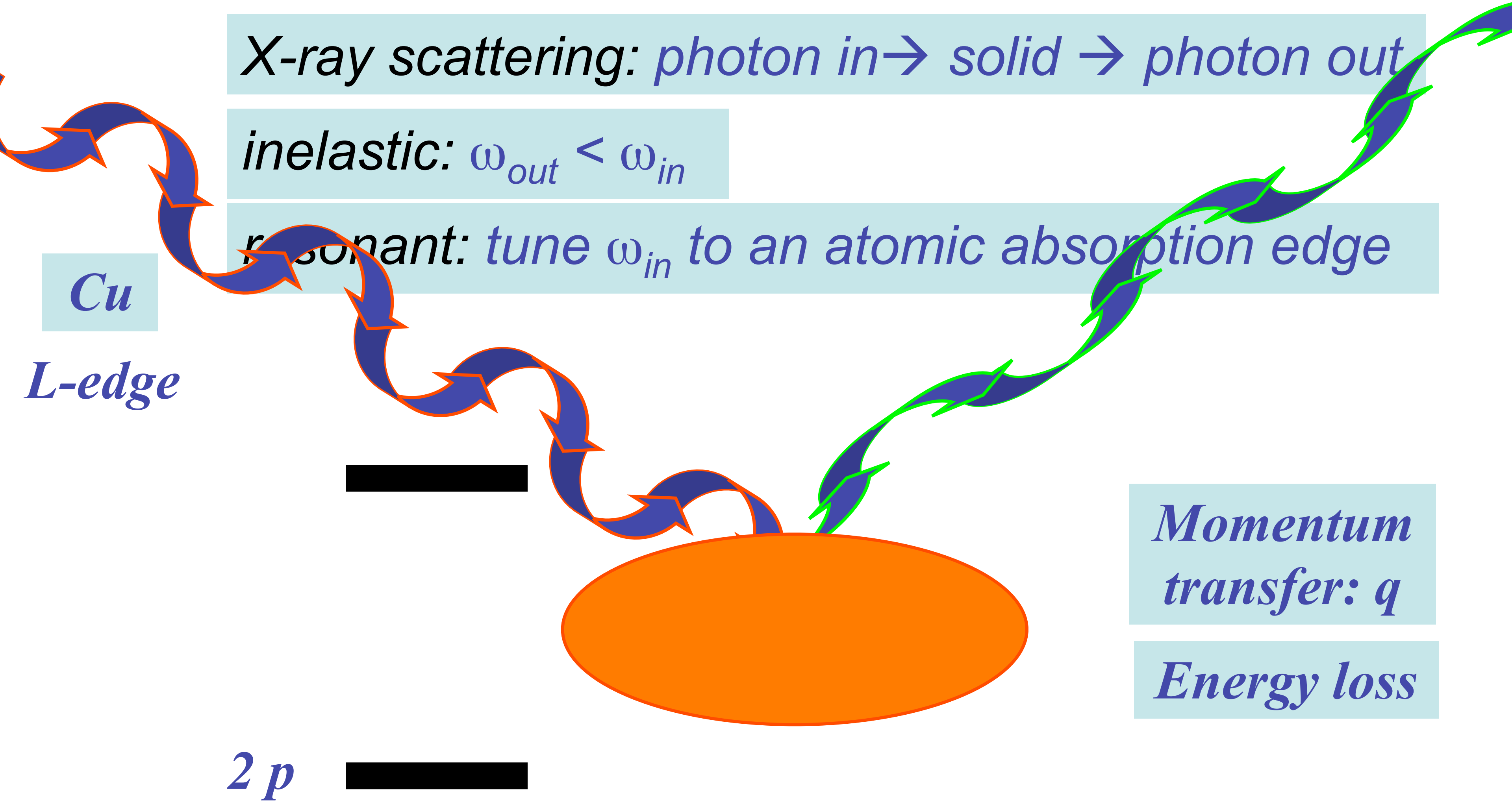

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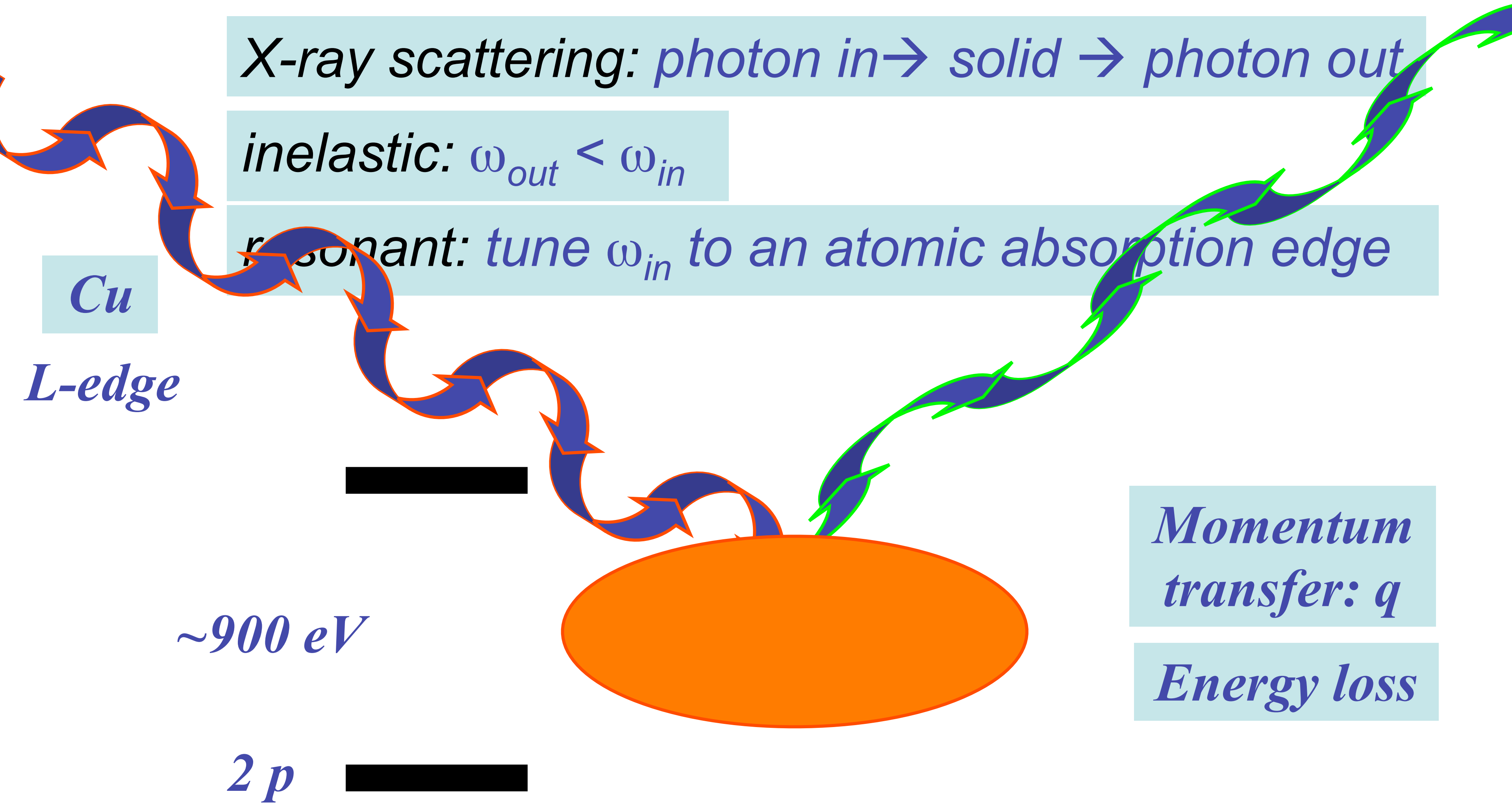

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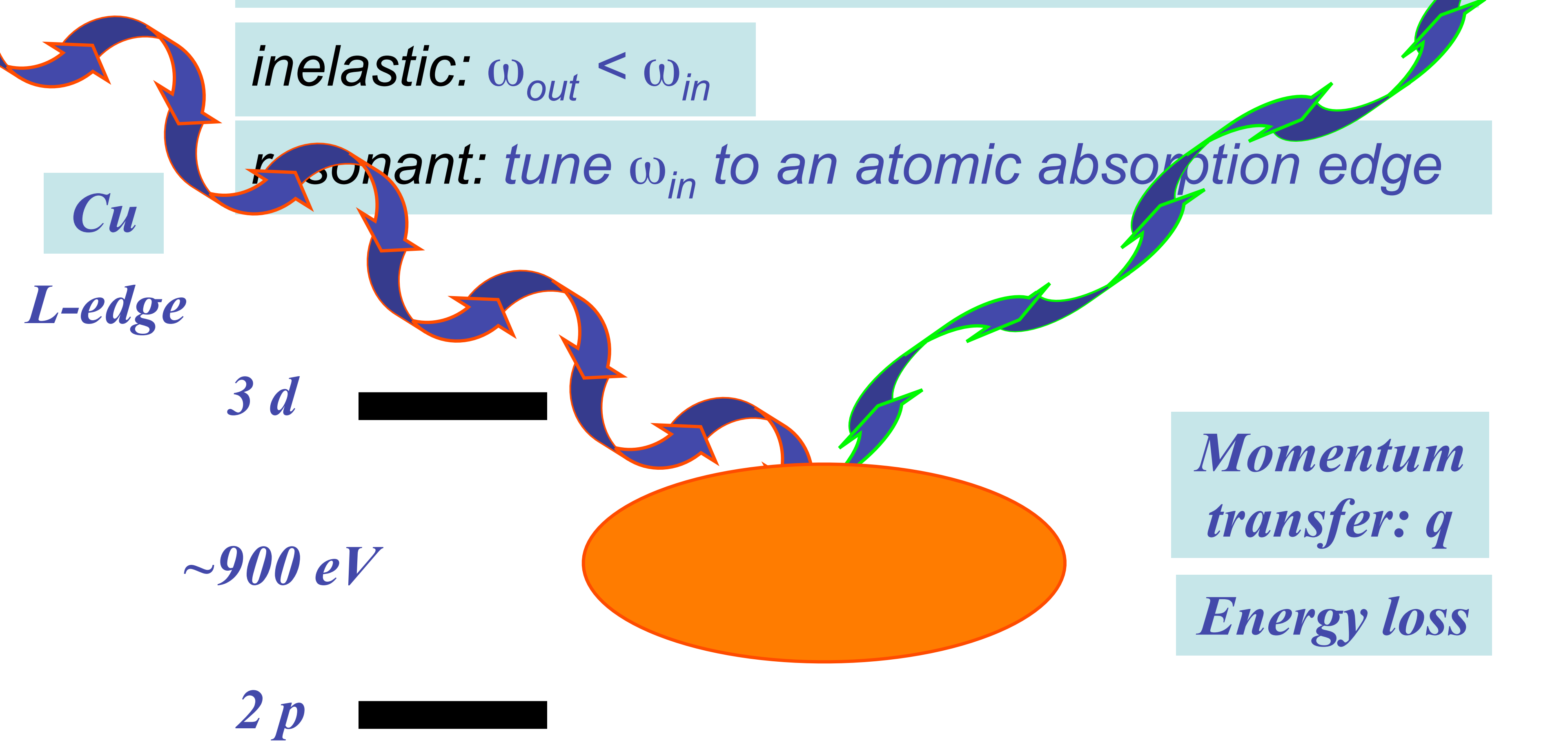

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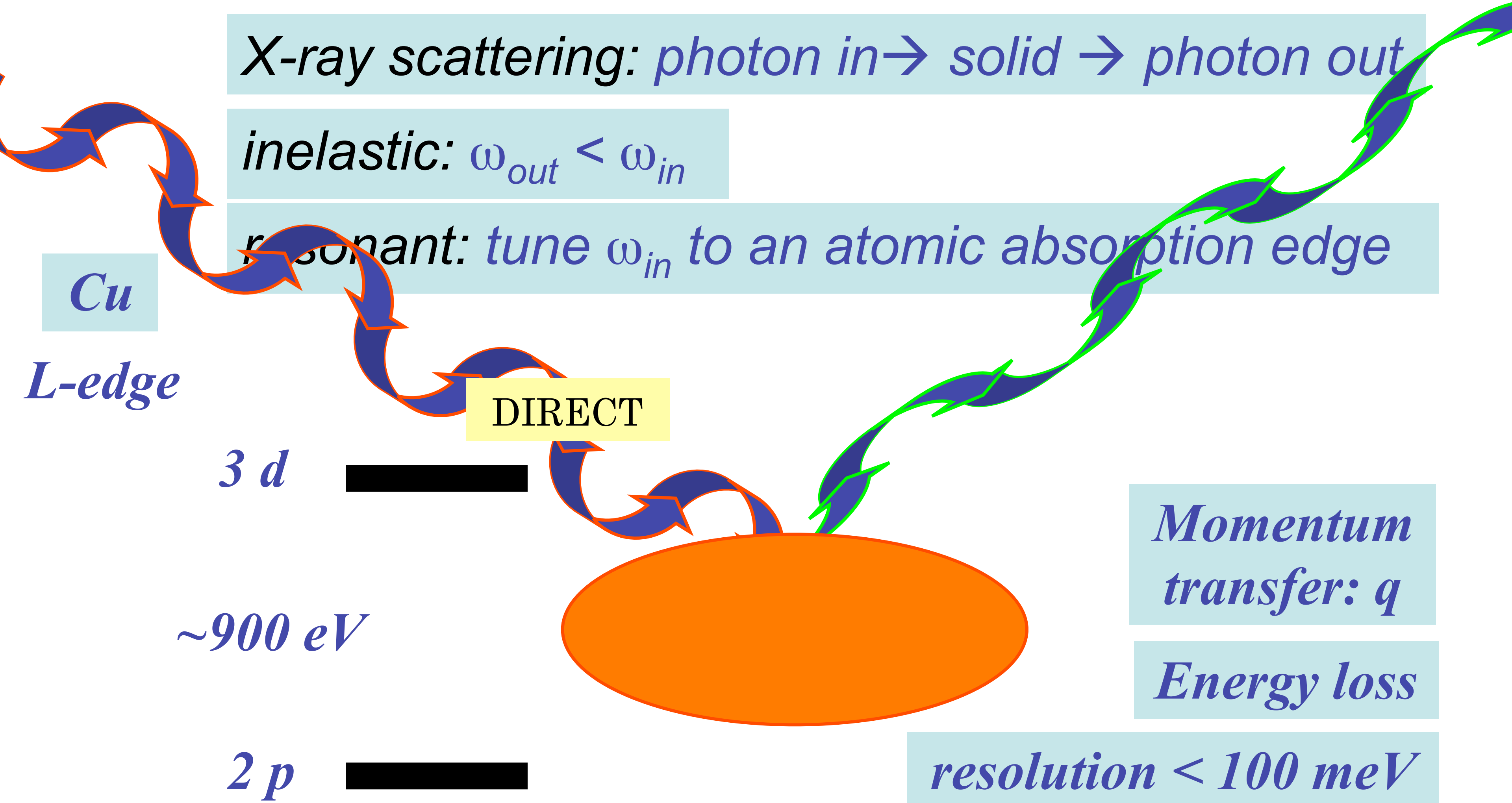


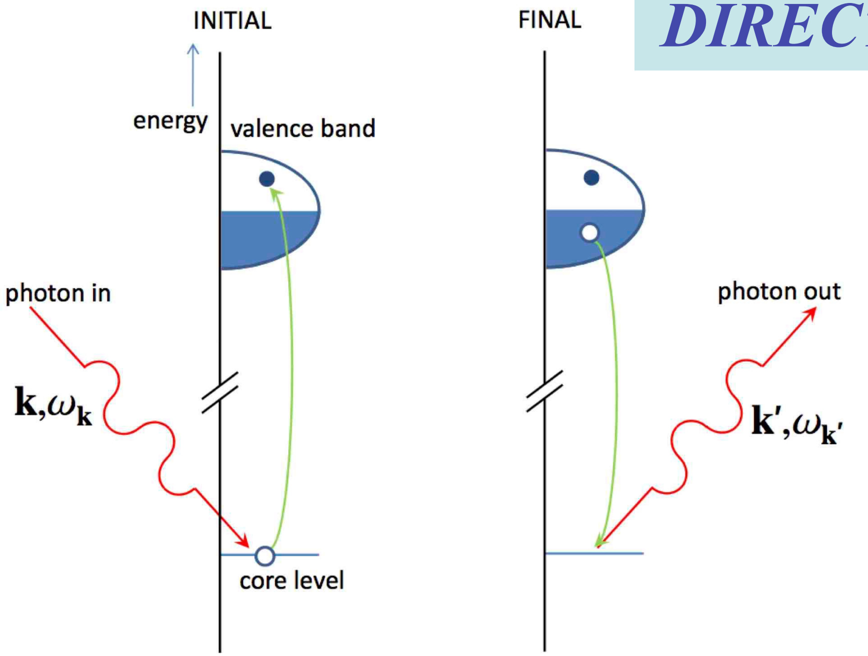
Momentum transfer: q

Energy loss

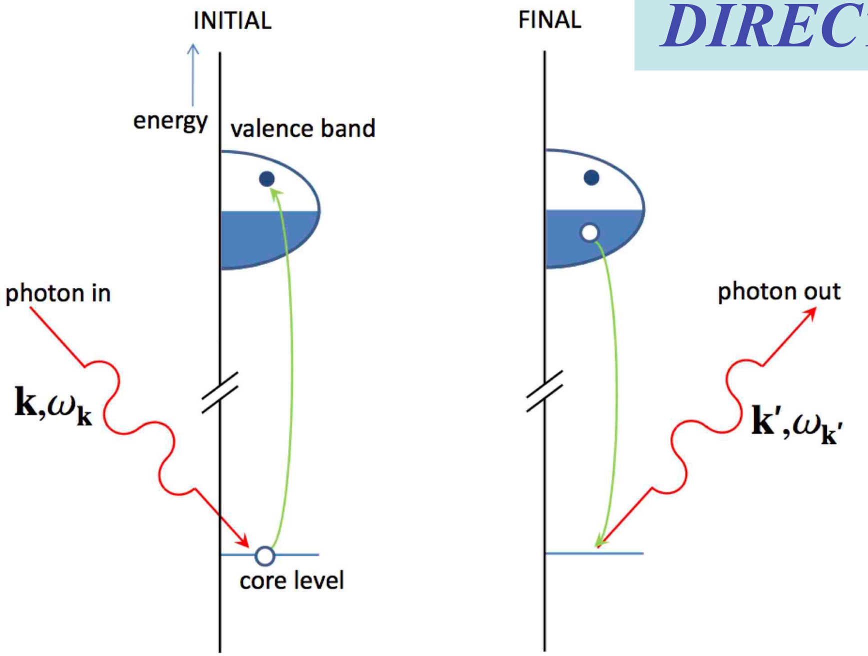
resolution < 100 meV

Direct and Indirect RIXS

DIRECT



Direct and Indirect RIXS



DIRECT

scattering via absorption-emission matrix elements

Direct and Indirect RIXS

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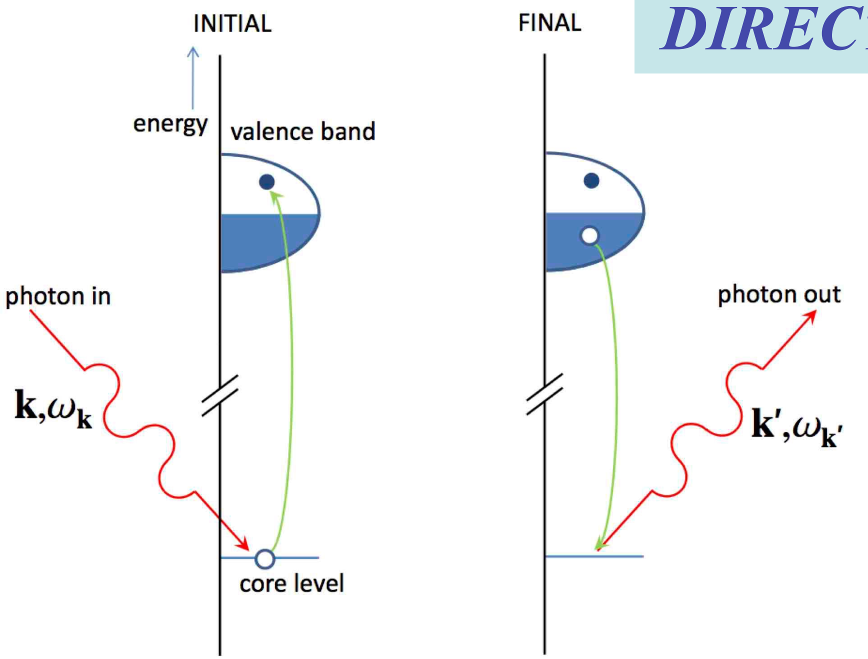

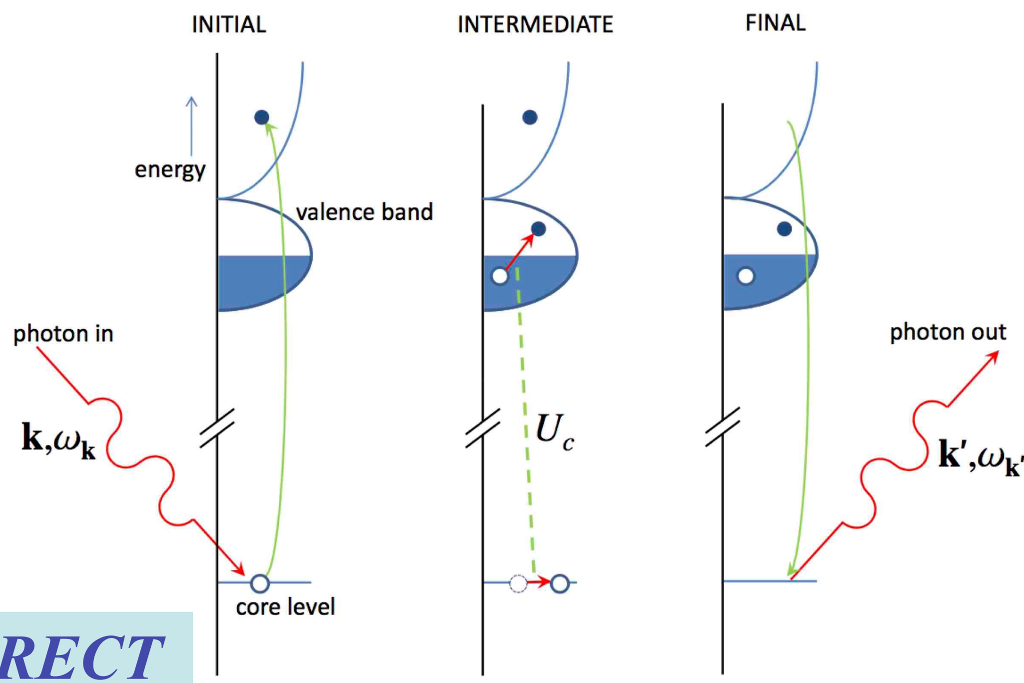

INDIRECT

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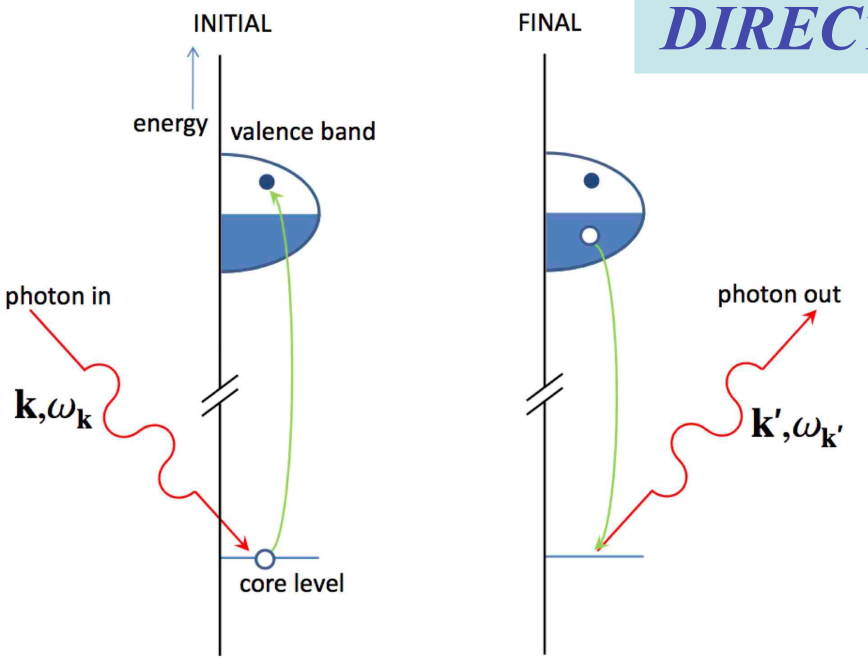
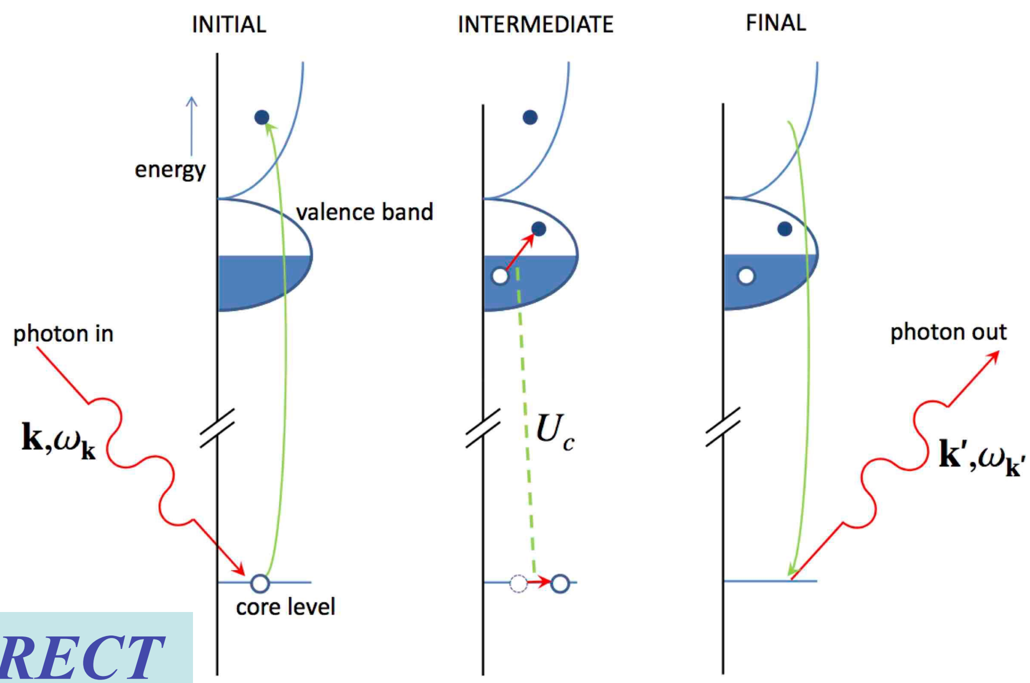


scattering via intermediate state core-hole shake-up



INDIRECT

$$RIXS = |GS\rangle \rightarrow XAS \rightarrow |INTERMEDIATE\rangle \rightarrow XES \rightarrow |FS\rangle$$

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↓

Local atomic transition

$$RIXS = |GS\rangle \rightarrow XAS \rightarrow |INTERMEDIATE\rangle \rightarrow XES \rightarrow |FS\rangle$$

Complicated state with strong core-hole potential

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Contains chemical detail and atom specific physics

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Complicated state with strong core-hole potential

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But:

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Carries low energy, long wavelength, elementary excitations

RIXS can probe universal effective low energy behavior

Resonant Scattering Intensity

RIXS amplitude

$$\mathcal{F}_{fg}(\mathbf{k},\mathbf{k}',\epsilon,\epsilon',\omega_{\mathbf{k}},\omega_{\mathbf{k}'}) = \sum_n \frac{\langle f|\,\mathcal{D}'^{\dagger}\,|n\rangle\,\langle n|\,\mathcal{D}\,|g\rangle}{E_g + \hbar\omega_{\mathbf{k}} - E_n + i\Gamma_n}$$

Kramers-Heisenberg expression

H.A. Kramers and W. Heisenberg, Z. Phys. 31, 681 (1925)

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RIXS transition operator

$$\mathcal{D} = \frac{1}{im\omega_{\mathbf{k}}} \sum_{i=1}^{N} e^{i\mathbf{k}\cdot\mathbf{r}_i}\,\boldsymbol{\epsilon}\cdot\mathbf{p}_i,$$

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RIXS intensity

$$I(\omega,\mathbf{k},\mathbf{k}',\boldsymbol{\epsilon},\boldsymbol{\epsilon}') = \sum_f \left|\mathcal{F}_{fg}(\mathbf{k},\mathbf{k}',\boldsymbol{\epsilon},\boldsymbol{\epsilon}',\omega_{\mathbf{k}})\right|^2 \times \delta(E_f+\hbar\omega_{\mathbf{k}'}-E_g-\hbar\omega_{\mathbf{k}})$$

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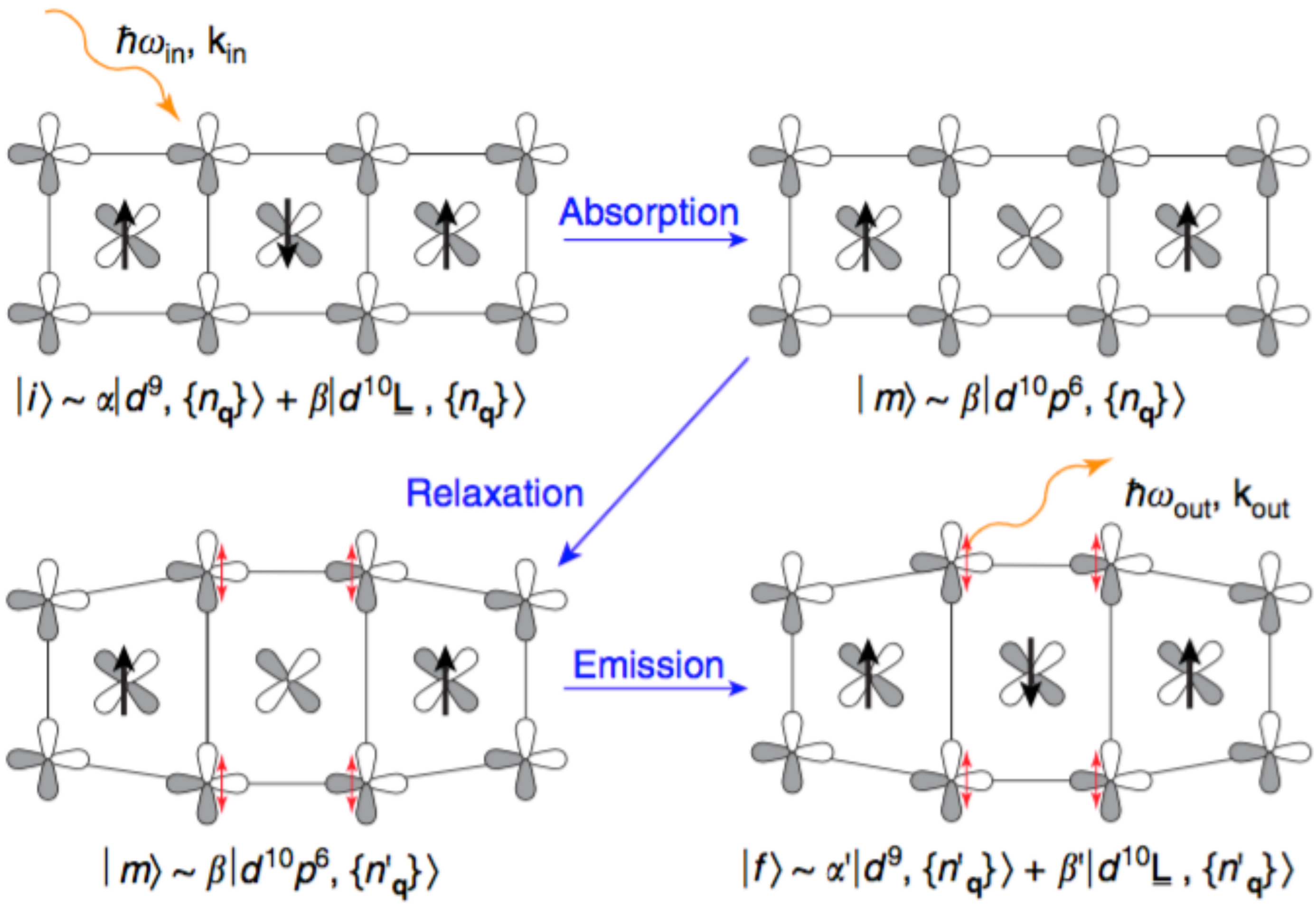
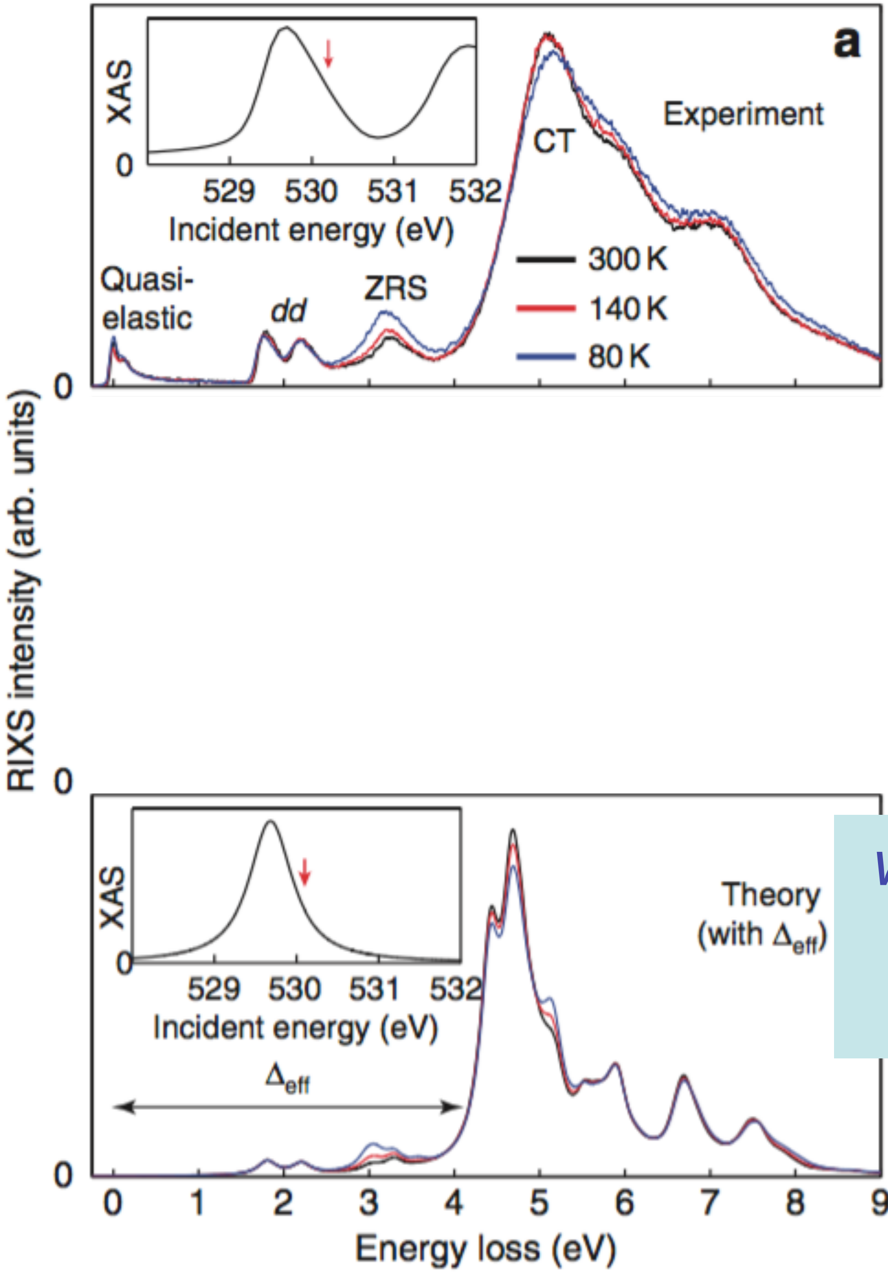
Oxygen K-edge RIXS on Li_2CuO_2

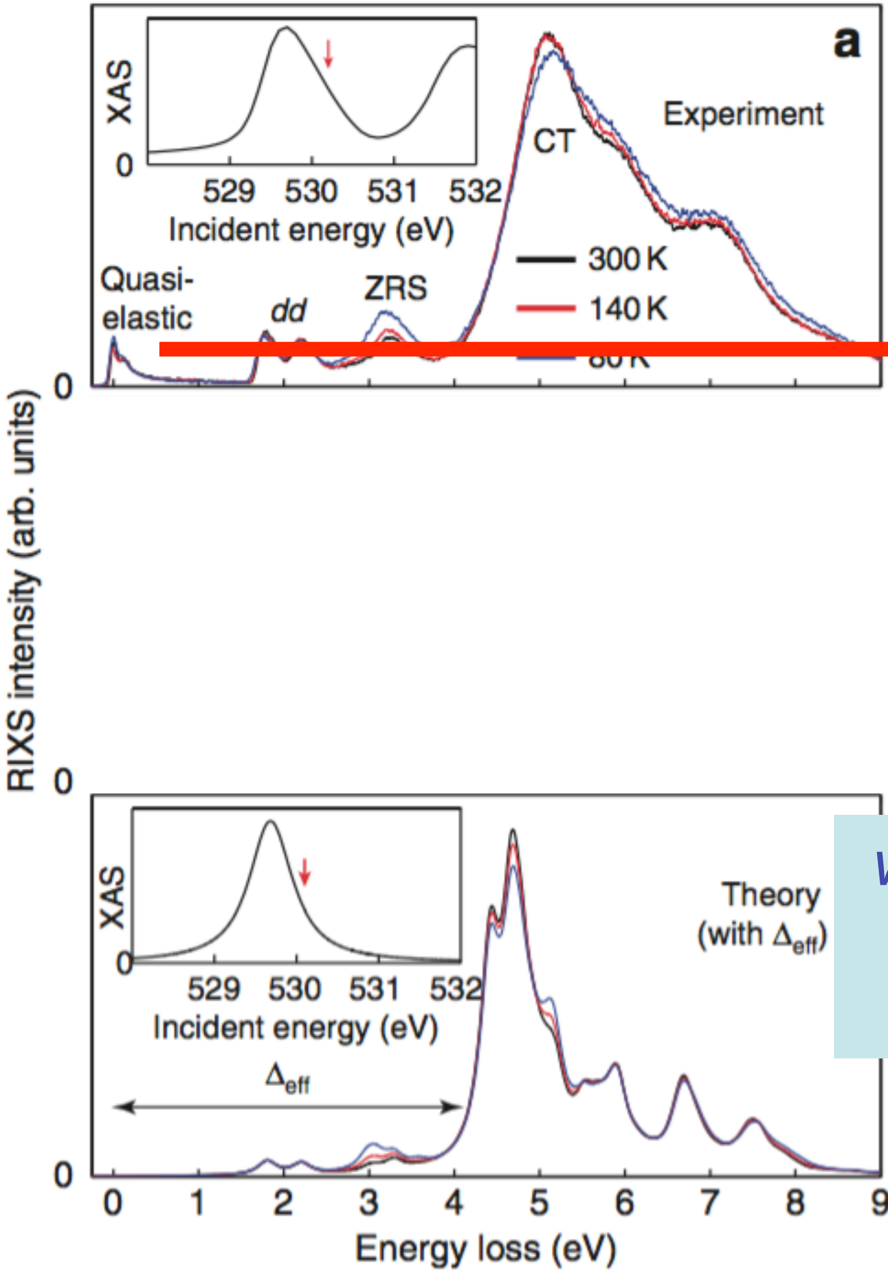


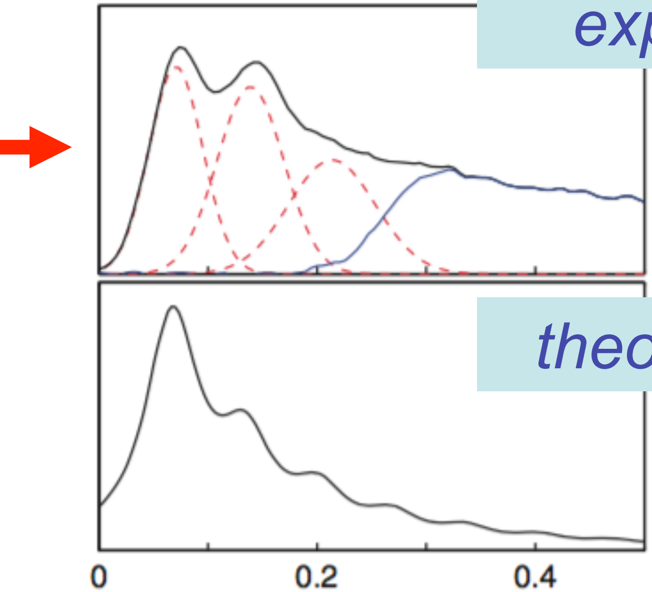
RIXS on Li_2CuO_2



without phonons charge transfer energy 4.6 eV

RIXS on Li_2CuO_2



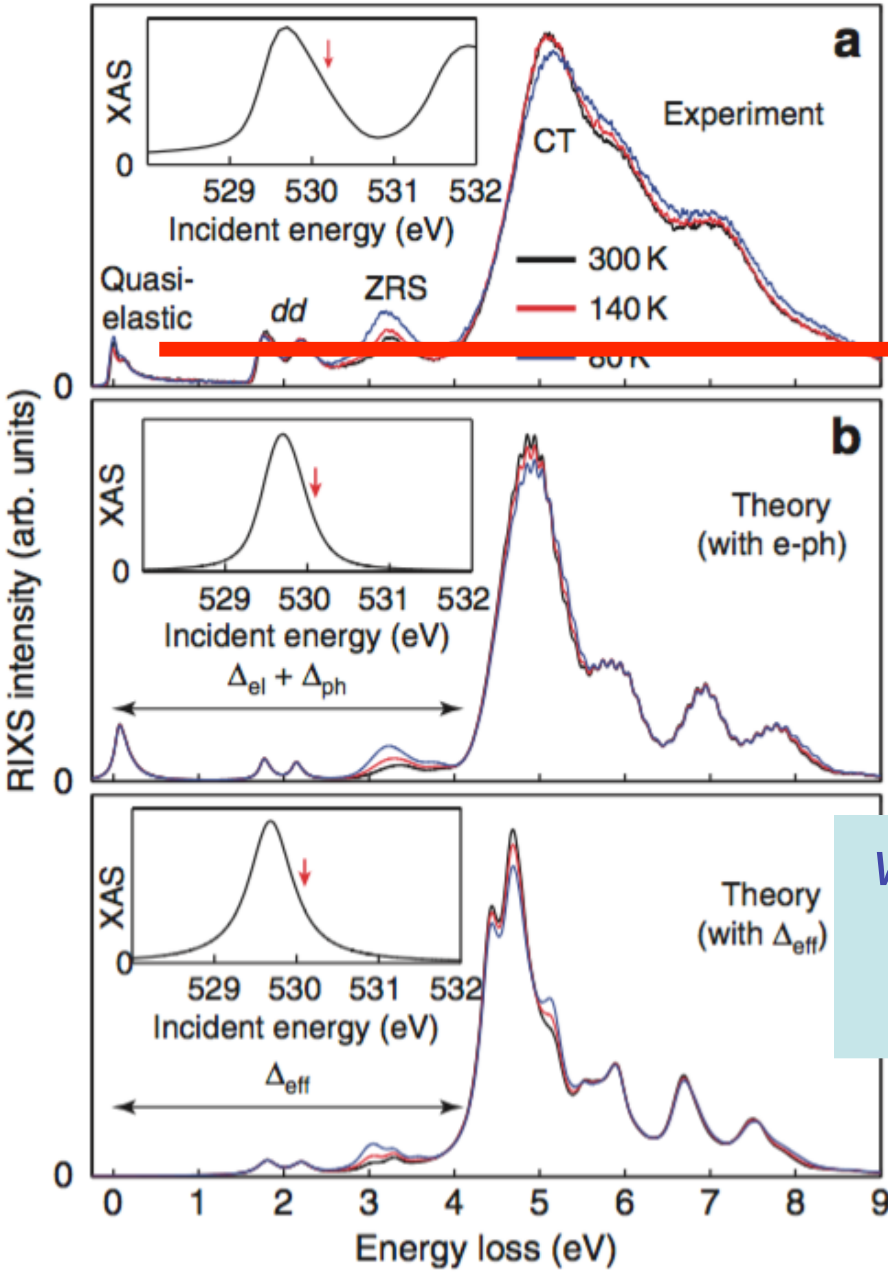


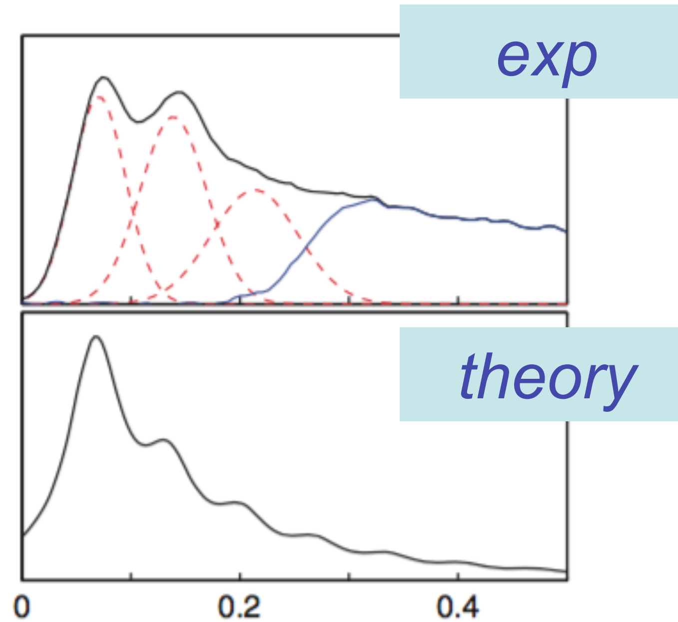
exp

theory

without phonons charge transfer energy 4.6 eV

RIXS on Li_2CuO_2





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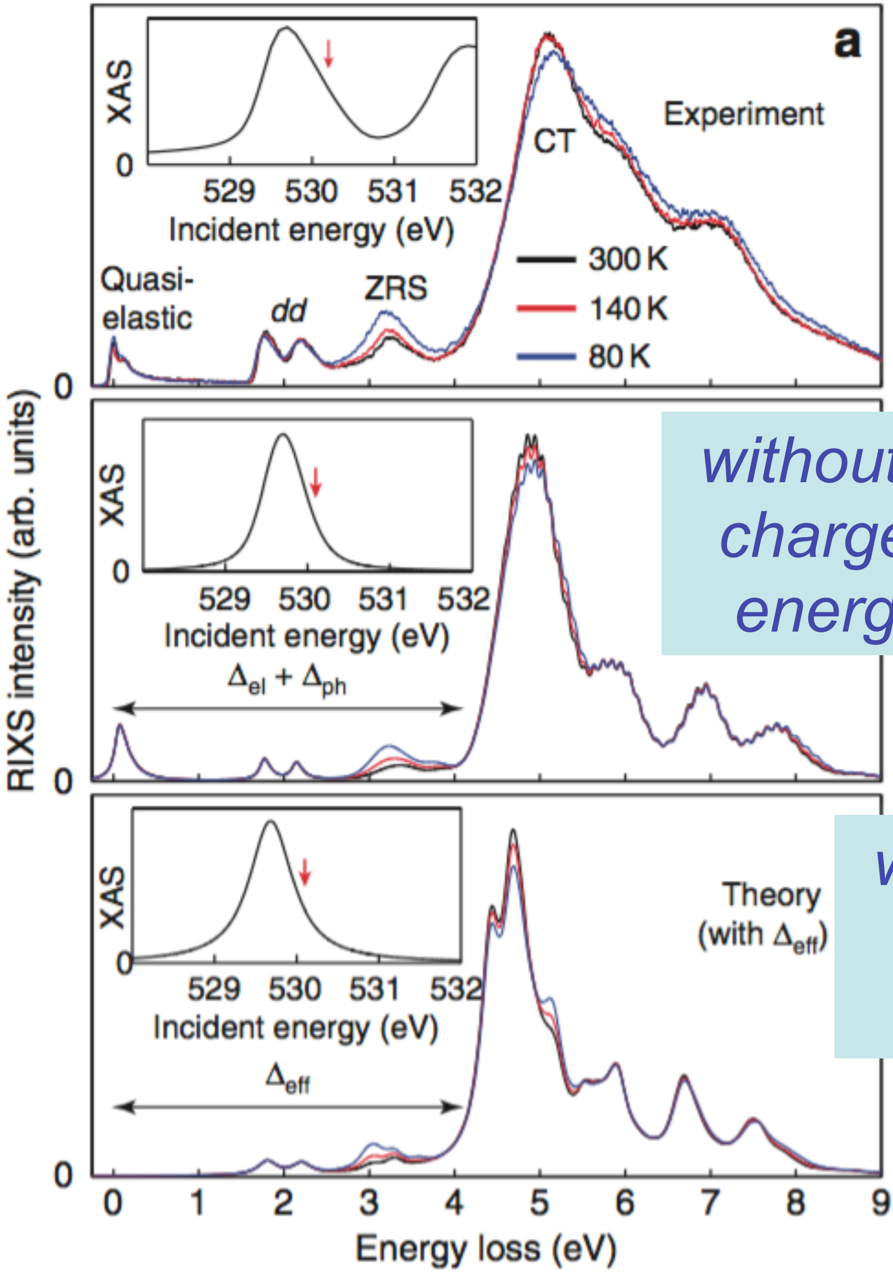
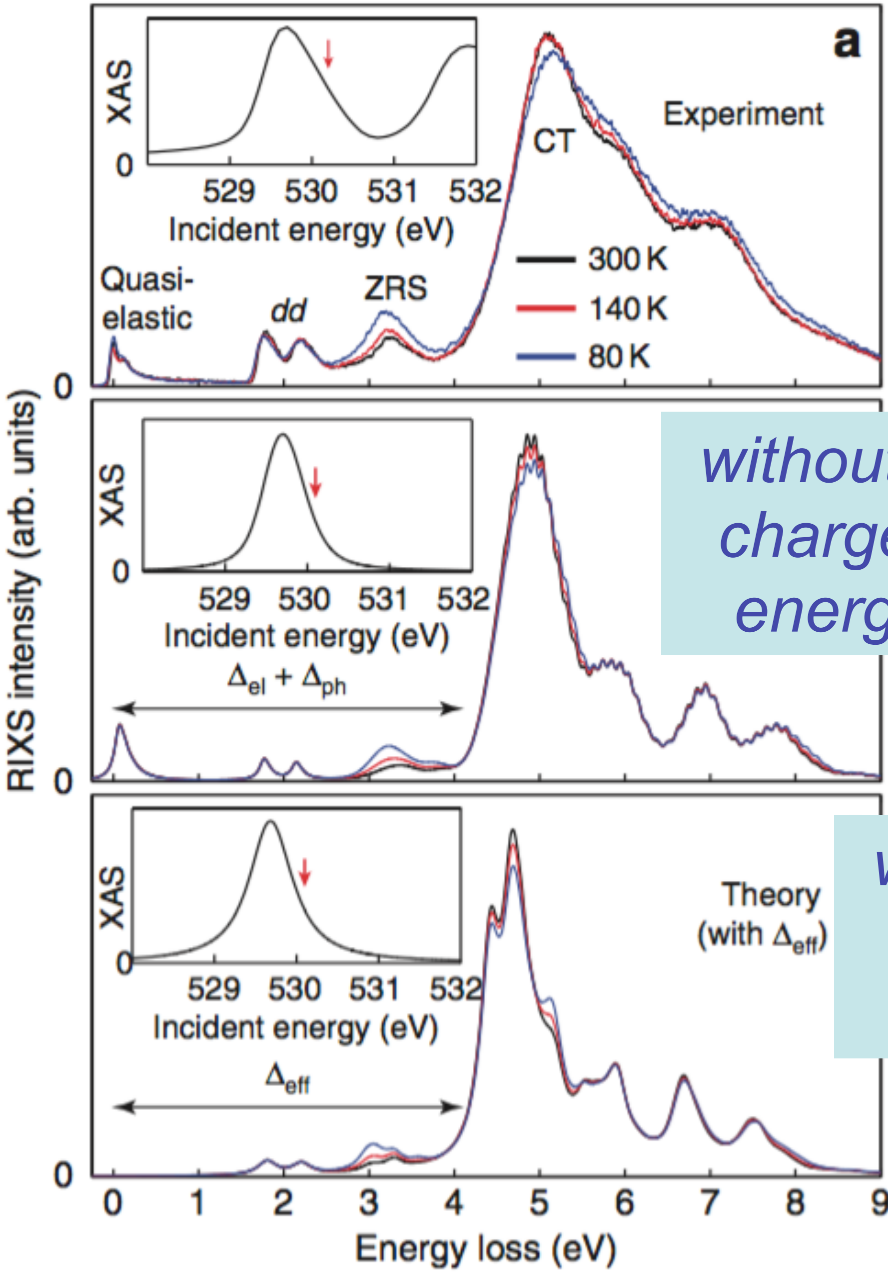


without phonons charge transfer energy 2.1 eV

without phonons charge transfer energy 4.6 eV

RIXS on Li_2CuO_2



without phonons
charge transfer
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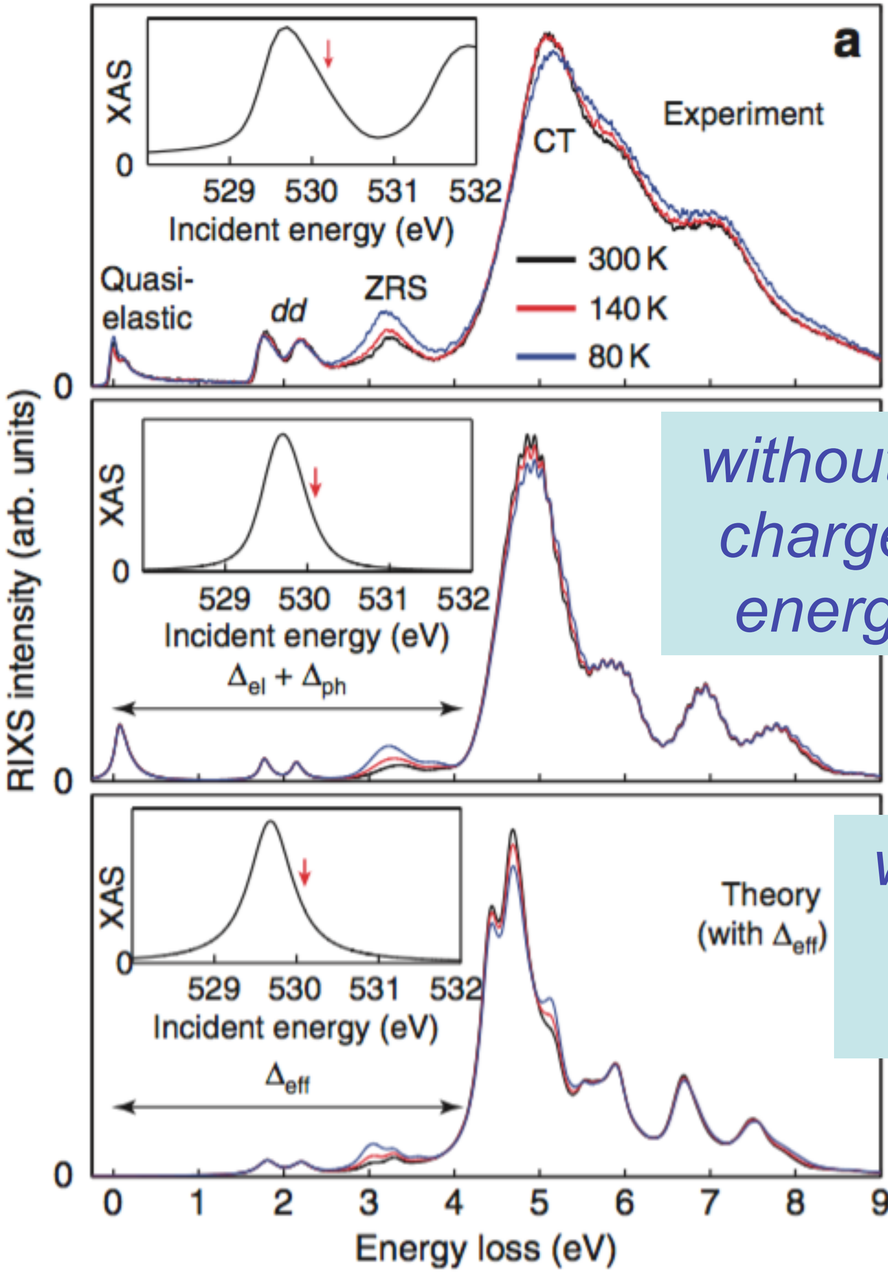
phonon contribution
to "charge transfer"
energy 2.5 eV

without phonons
charge transfer
energy 4.6 eV

RIXS on Li_2CuO_2



phonon induced anti-screening of the CT energy

without phonons charge transfer energy 2.1 eV

phonon contribution to "charge transfer" energy 2.5 eV

without phonons charge transfer energy 4.6 eV

avoid using dielectric screening for short (zero)-range coulomb interactions in correlated electron systems

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important differences for nano-structures, at interfaces, at short distances

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dynamic screening very different for electronic and lattice polarizabilities

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The Hubbard model with orbital degeneracy and in polarizable media

Proefschrift

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The Hubbard model with orbital degeneracy and in polarizable media

Proefschrift

http://www.rug.nl/research/portal/publications/pub(5b75b5ad-8a1b-4ddb-92f9-f73ba8d52479).html

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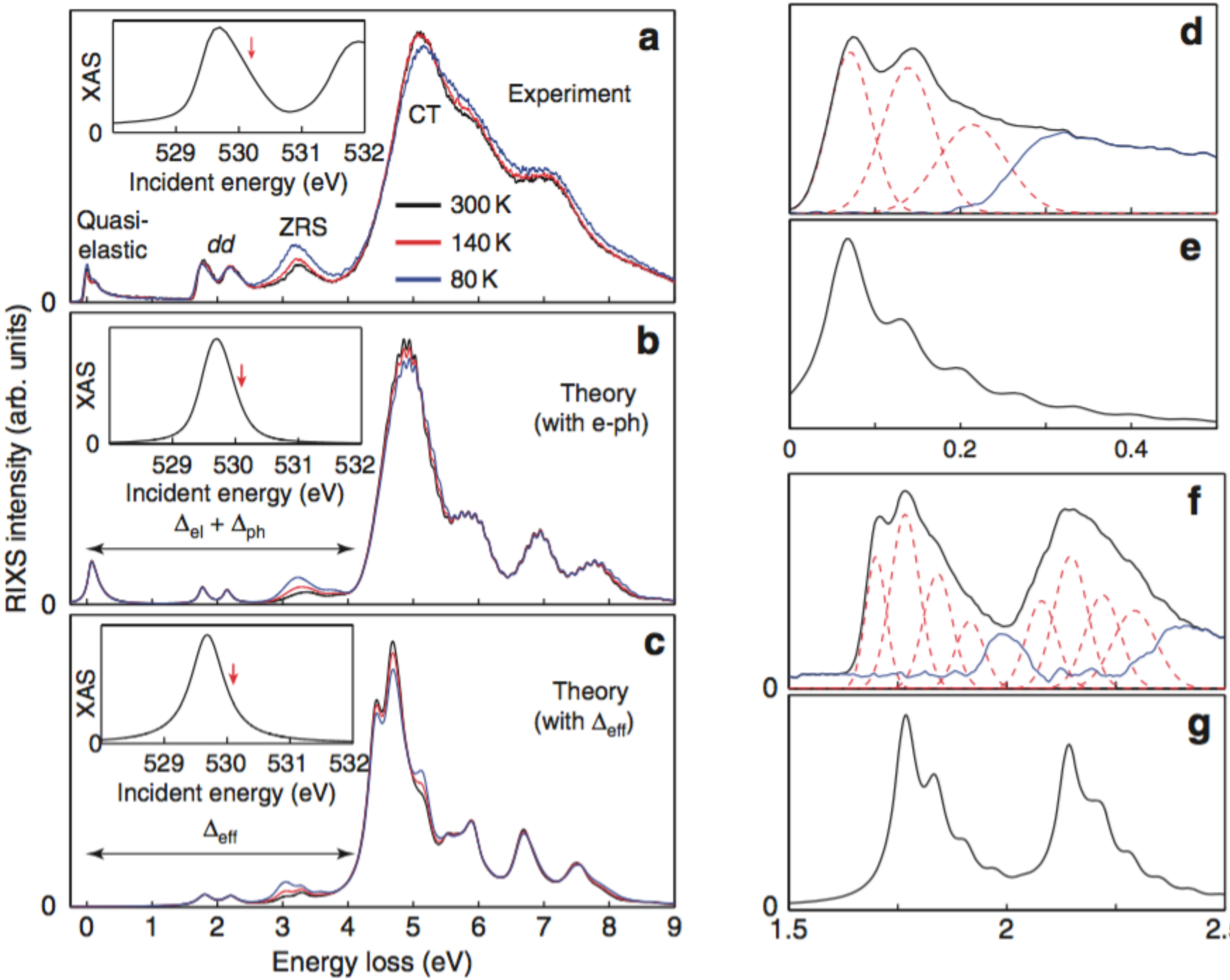

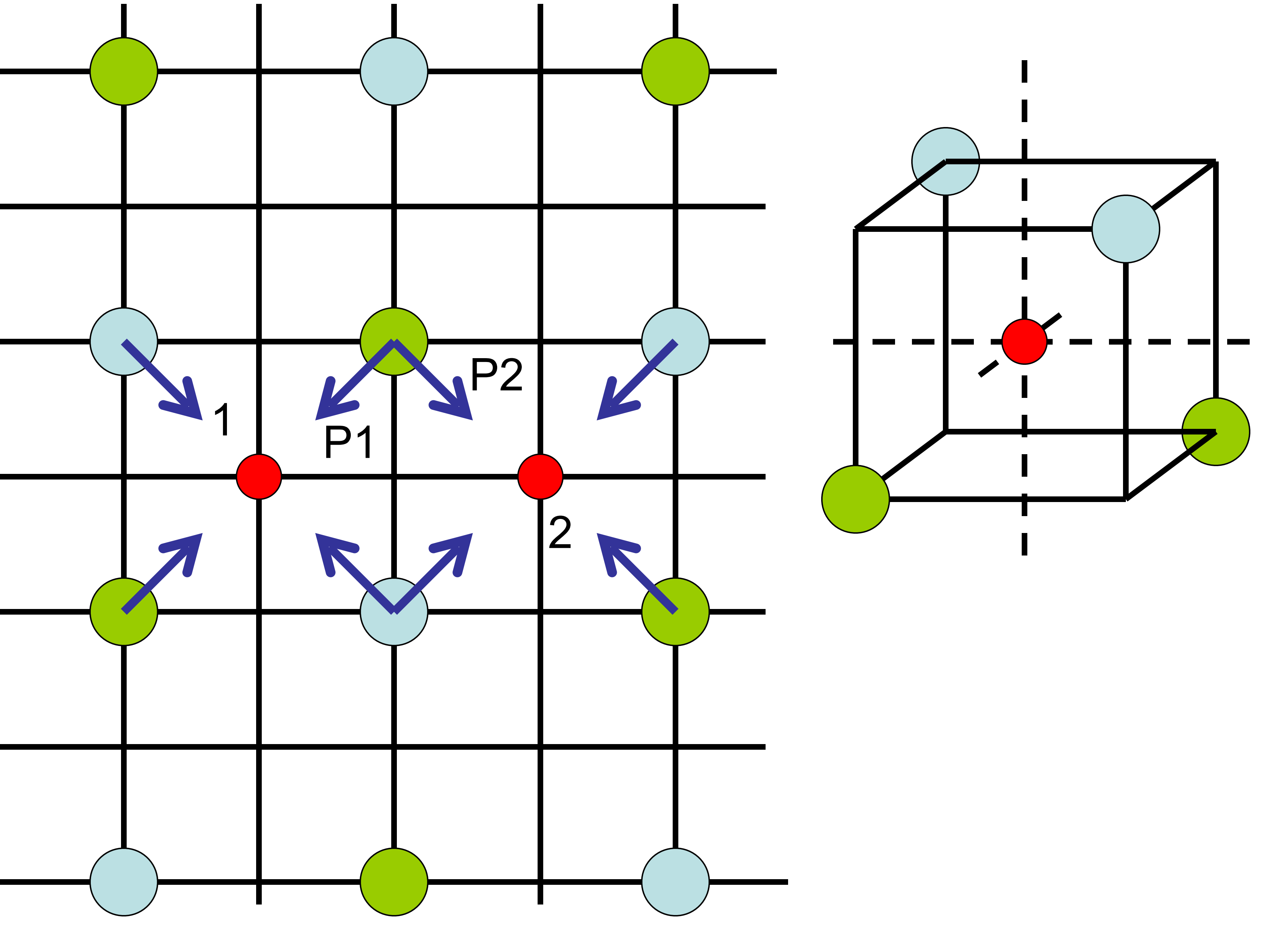

1
P1
P2
2