Time Resolved photoemission of correlated electronic systems

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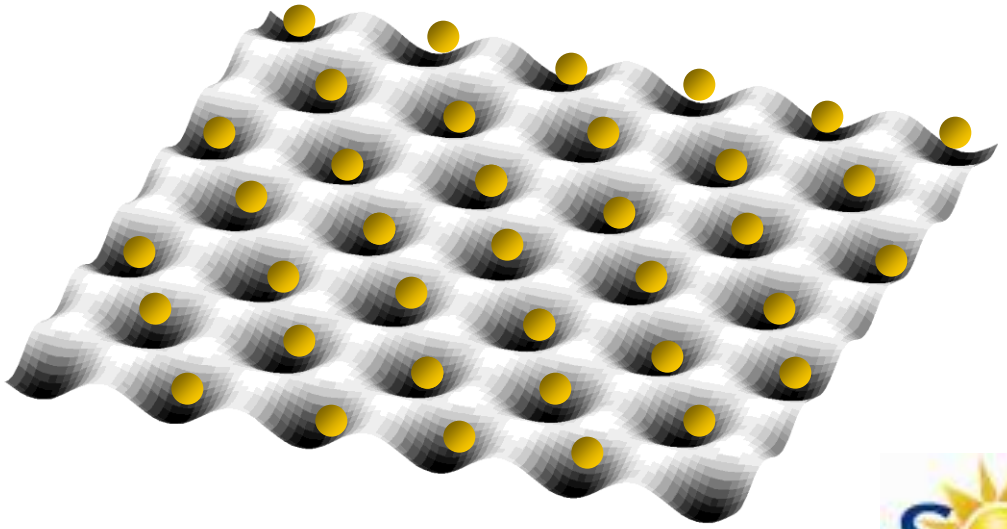


ÉCOLE POLYTECHNIQUE

SOLEIL SYNCHROTRON

Outline

Photoelectron Spectroscopy

Basic concepts of solid state physics

Introdution to strongly interacting materials

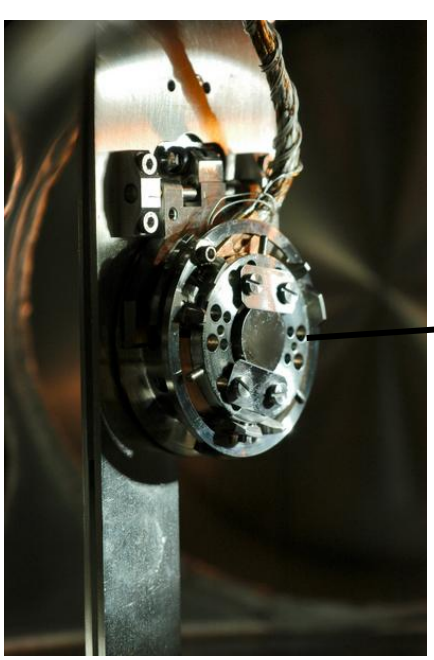
Structural distortion and Mott transition in 1T-TaS_2

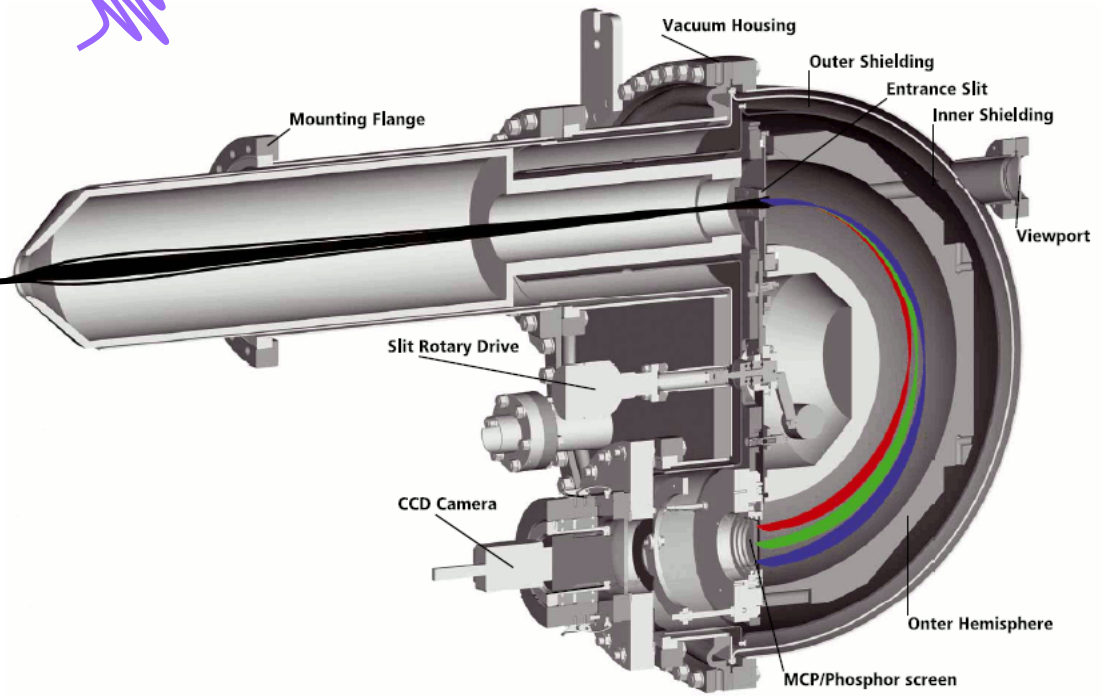
Time resolved photoemission with lasers

New sources and first results

Phonon pumping

Photoelectron Spectroscopy

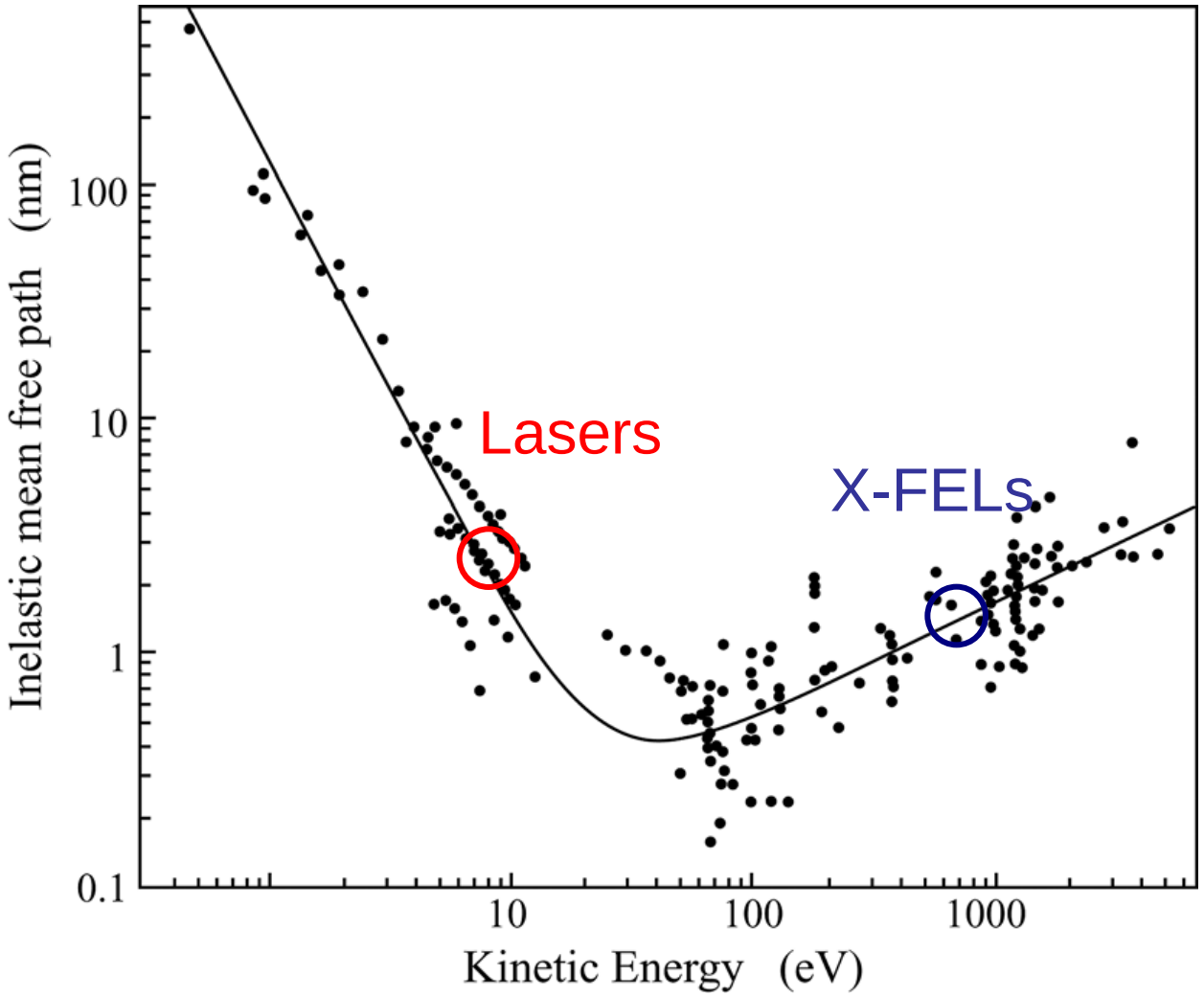




Sample in UHV environment

Strong inelastic collisions of photoelectrons in the solid

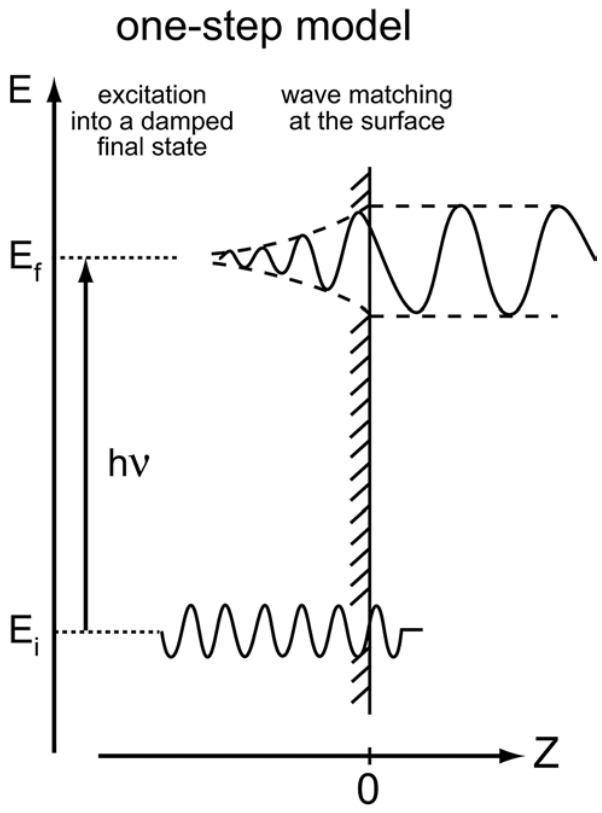
Sudden approximation M. P. Seah, Surf. Interface Anal. 1979

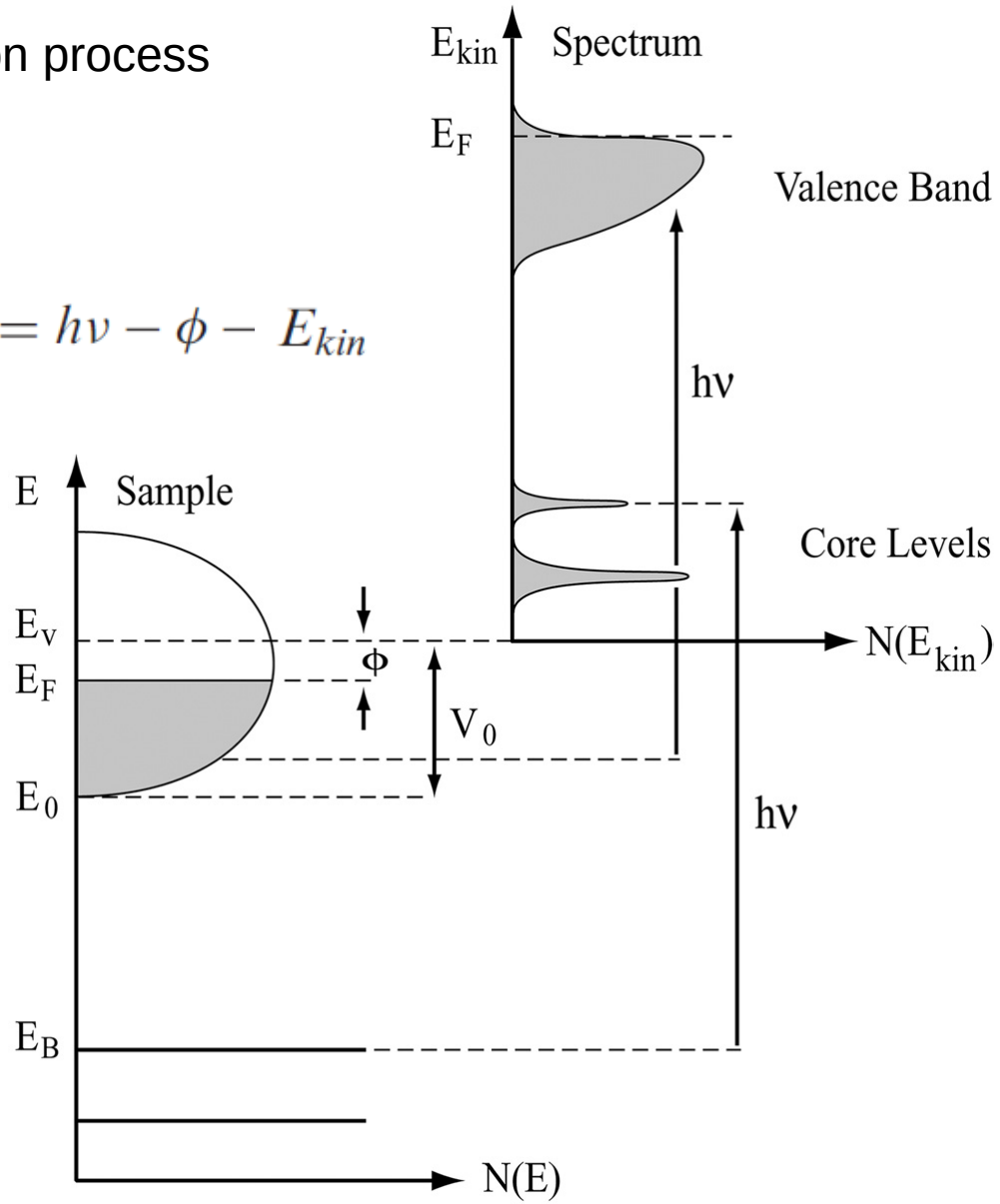


Energetics of the photoemission process

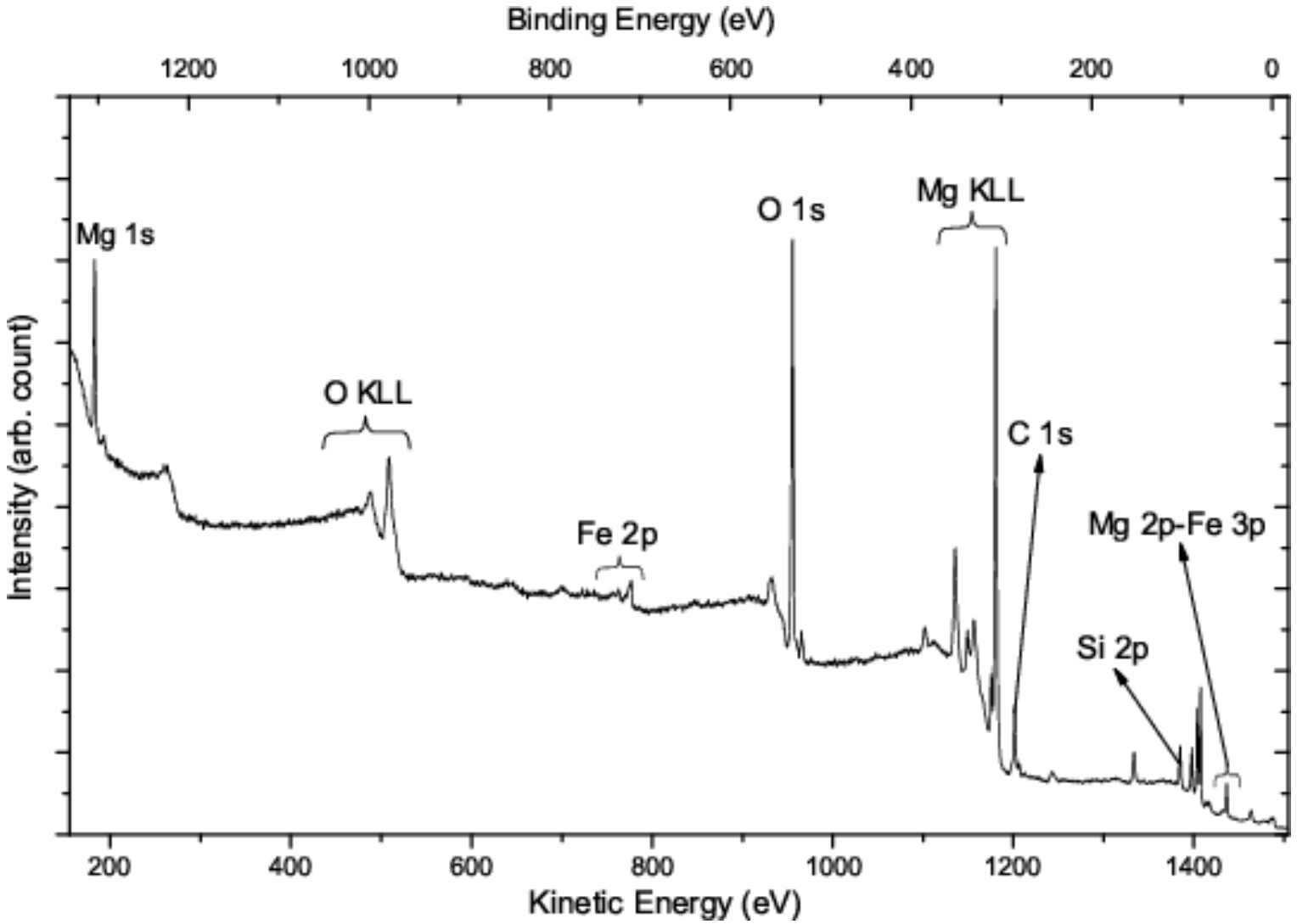
Valence band and core levels

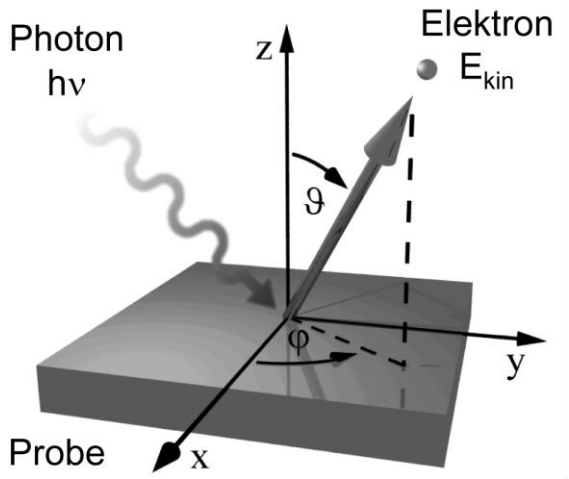
$$E = h\nu - \phi - E_{kin}$$





Core level spectroscopy





$$E = h\nu - \phi - E_{kin}$$

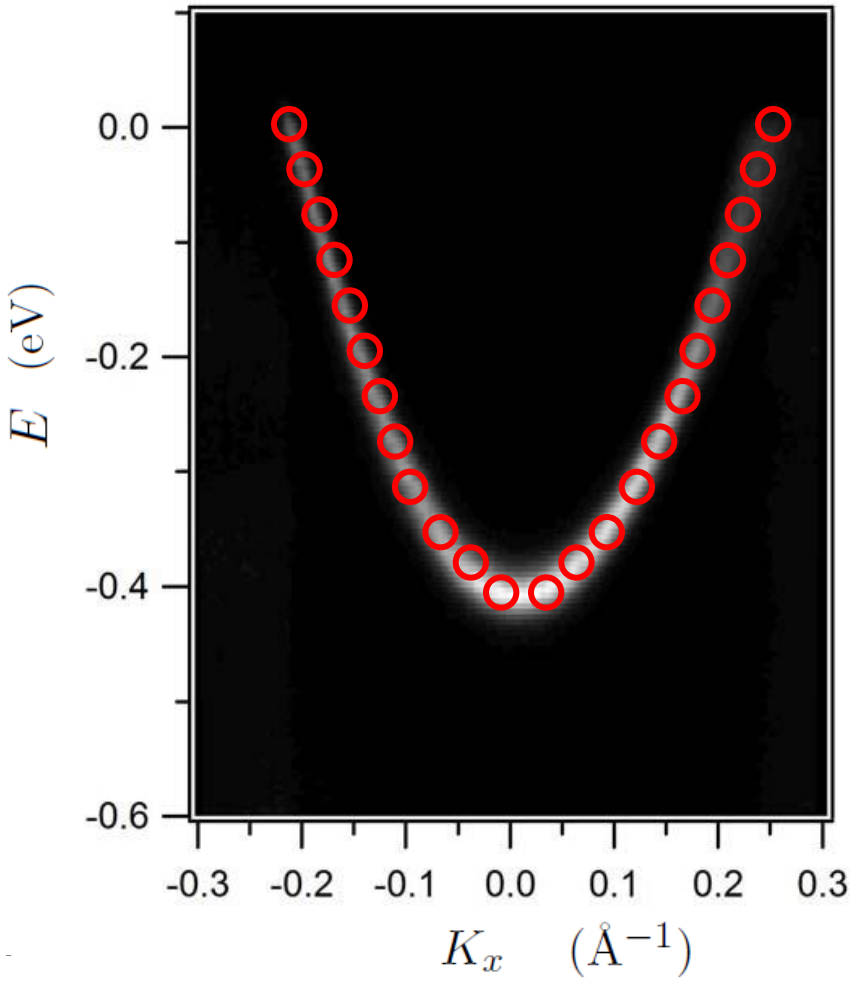
$$K_x = \frac{1}{\hbar}\sqrt{2mE_{kin}}\sin\vartheta\cos\varphi,$$

$$K_y = \frac{1}{\hbar}\sqrt{2mE_{kin}}\sin\vartheta\sin\varphi,$$

$$E = \frac{\hbar^2(K_x^2 + K_y^2)}{2m_e} - E_0$$

Angle Resolved Photoelectron spectroscopy

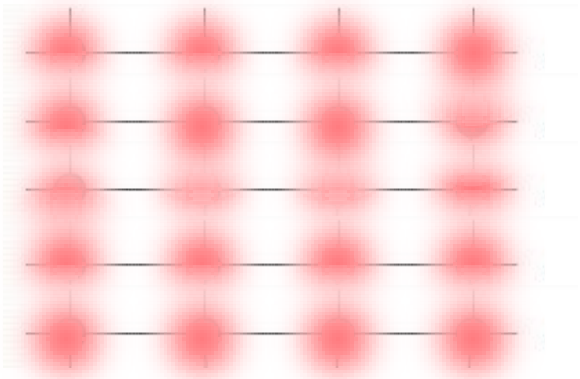
Cu 111



Electrons in a periodic potential

Nuclei + core electrons make the ionic lattice

Valence electrons are delocalized and behave as nearly independent particles

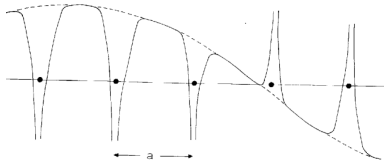


Mean field theory: effective interaction of nearly independent particles

$$\left[-\frac{1}{2}\nabla^2 + v_{ext}(\mathbf{r}) + \int \frac{\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} d\mathbf{r}' + v_{xc}(\mathbf{r})\right]\phi_i(\mathbf{r}) = \varepsilon_i \phi_i(\mathbf{r}) \qquad \rho(\mathbf{r}) = \sum_i^N |\phi_i|^2$$

The periodicity of the lattice implies the Bloch theorem

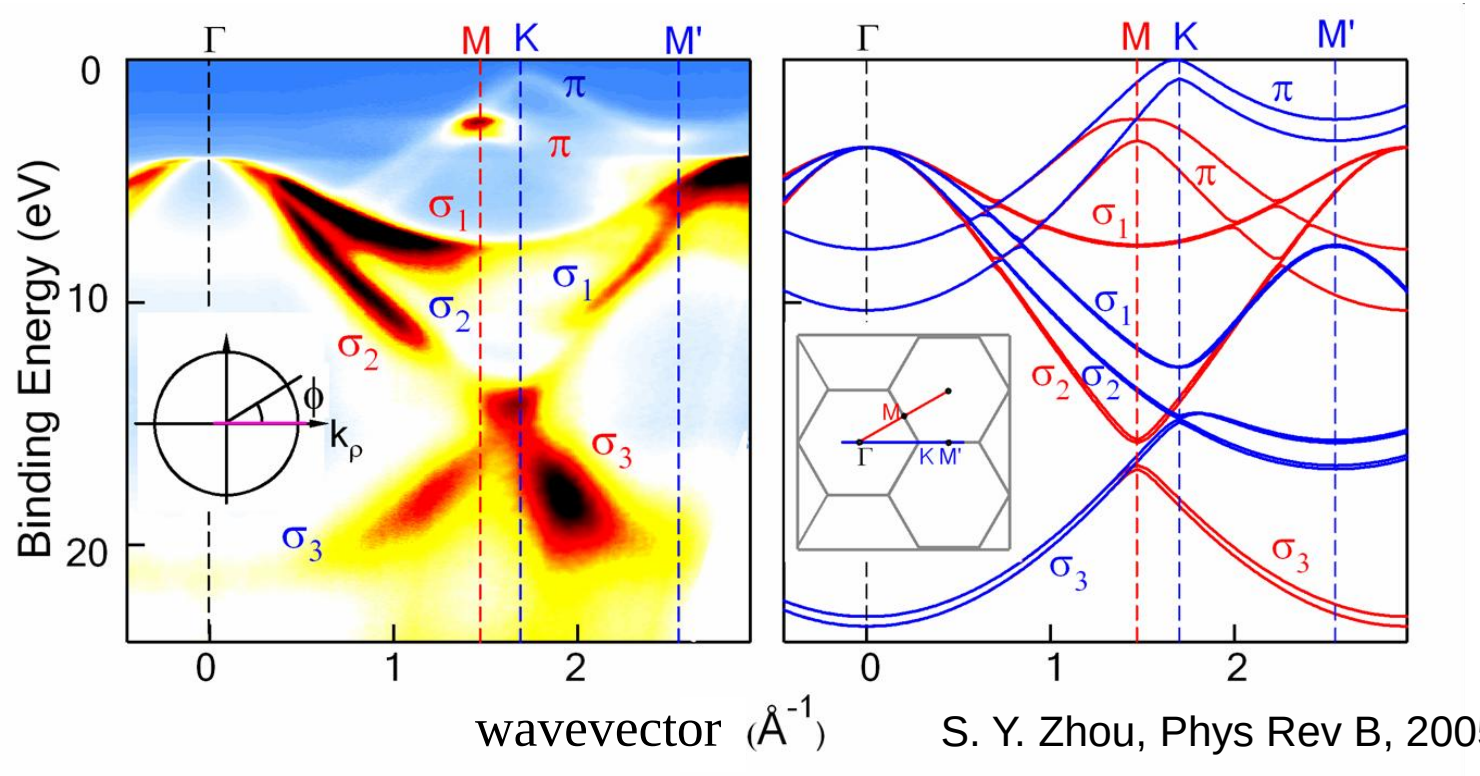
$$\psi_{\mathbf{k}}(\mathbf{r}) = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}} e^{i\mathbf{k}\cdot\mathbf{R}} \phi_{\mathbf{R}}(\mathbf{r})$$



$E(k)$

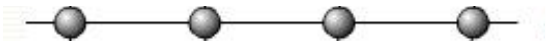
ARPES

Density Functional Theory



S. Y. Zhou, Phys Rev B, 2005

Phonons: the low energy excitations of the lattice



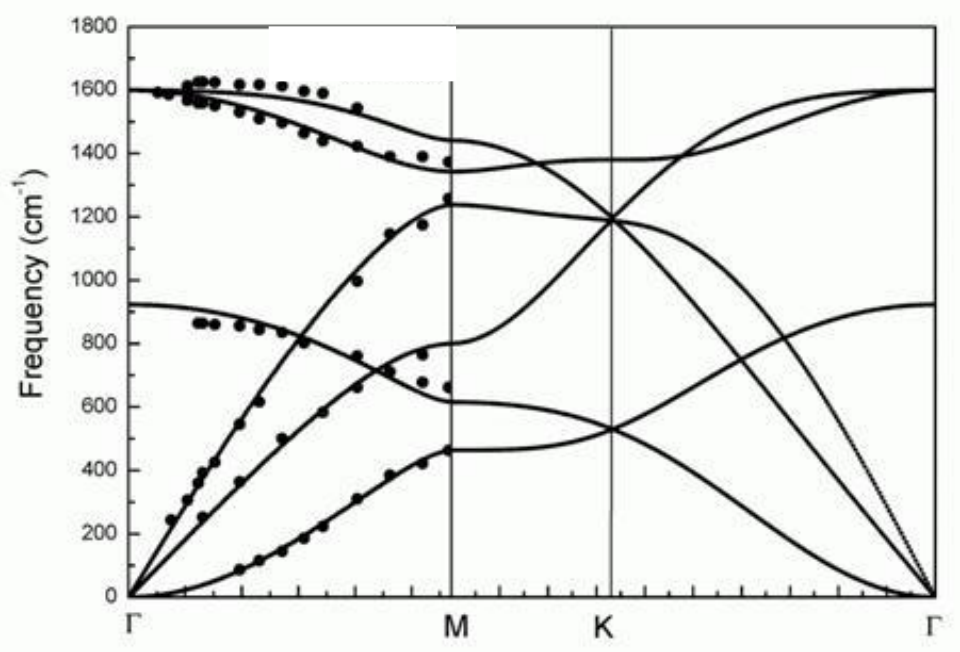
$$-2Cu_n + C(u_{n+1} + u_{n-1}) = m\frac{\mathrm{d}^2 u_n}{\mathrm{d}t^2}$$

$$U_k = \frac{1}{\sqrt{N}} \sum_n e^{ikna} u_n$$

$$2C(\cos kd - 1)U_k = m\frac{\mathrm{d}^2 U_k}{\mathrm{d}t^2}$$

$$\Omega_k = \sqrt{2C(1 - \cos kd)}$$

Eigenmodes of coupled harmonic oscillators



Phonon dispersion in Graphene

Inelastic X-ray Scattering

Structural distortion in $SrTiO_3$

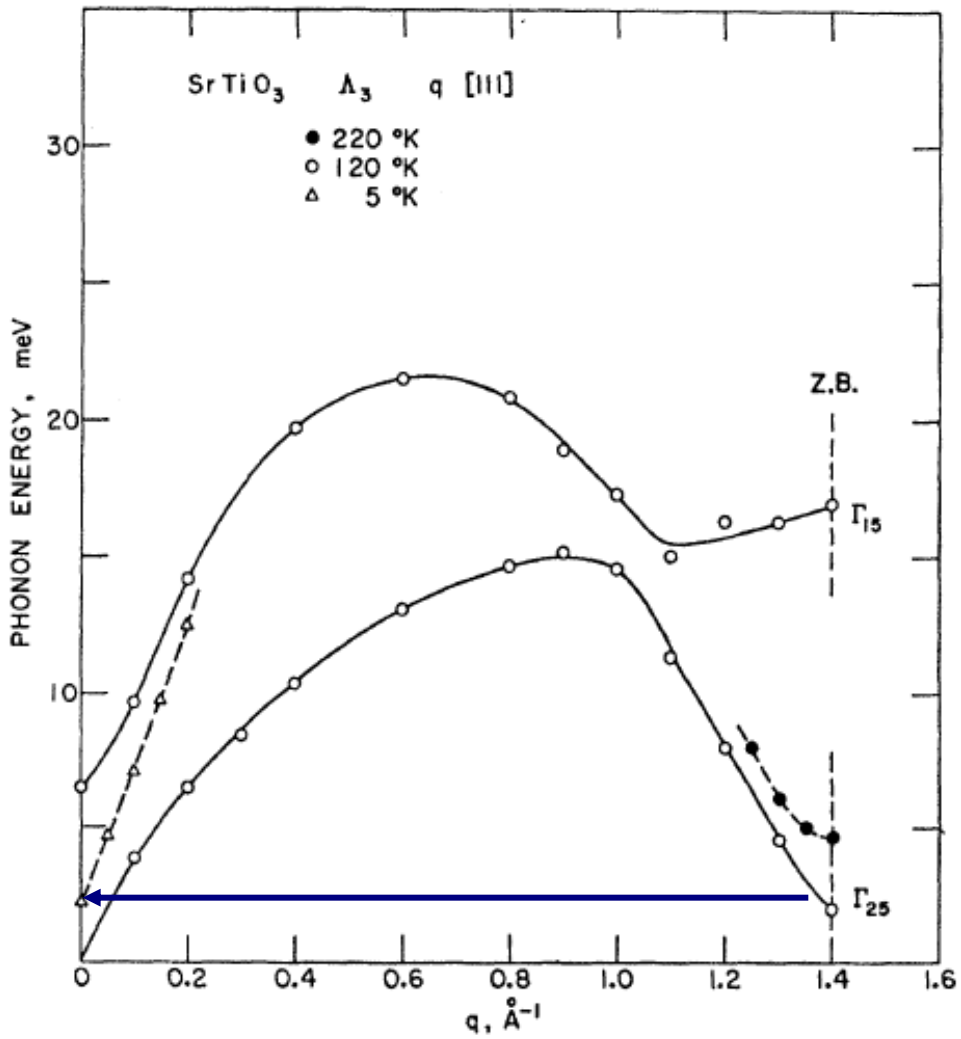
G. Shirane, Phys Rev , 2005

Softening of zone edge mode

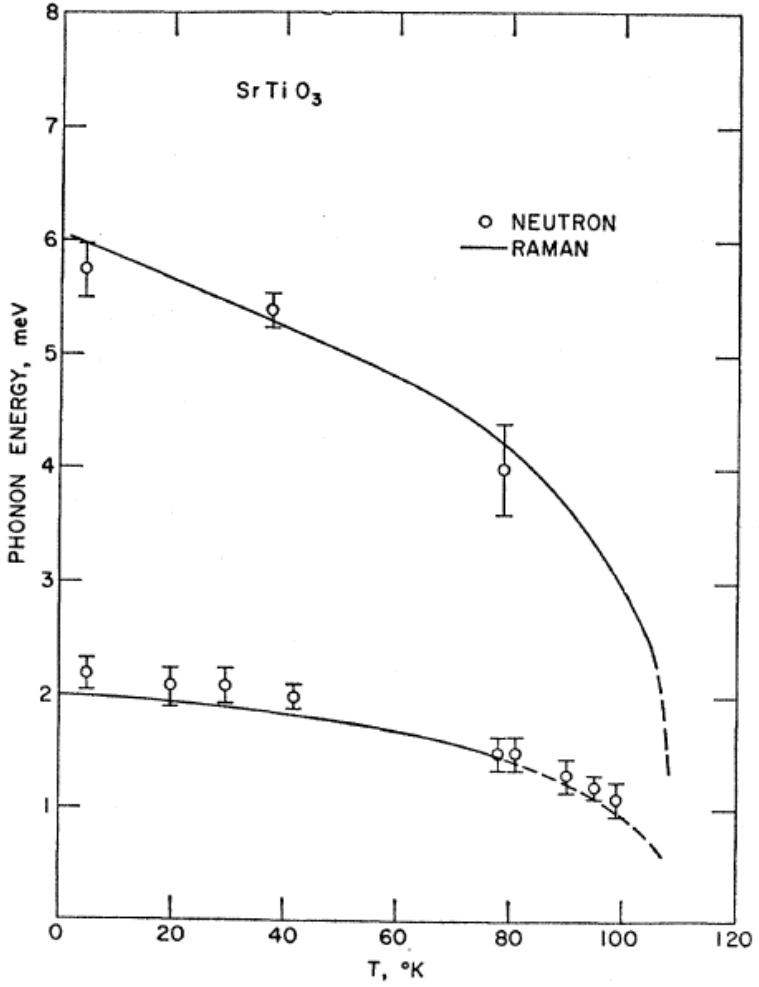
Structural instability

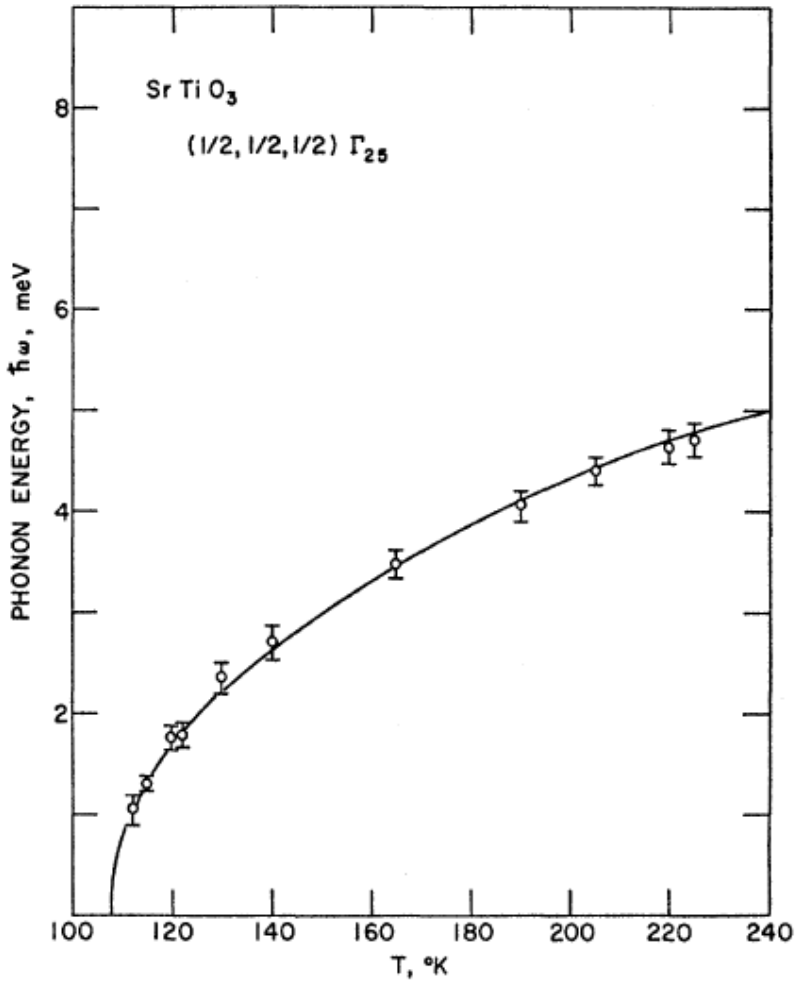
Doubling of the unit cell

The edge mode becomes
a center mode



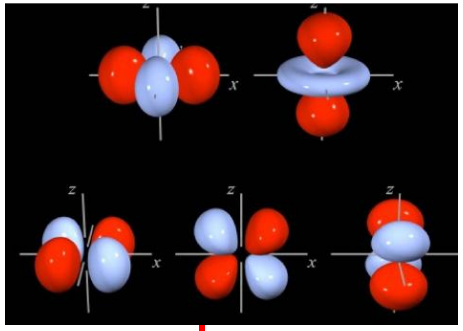
Temperature dependence of the soft phonon





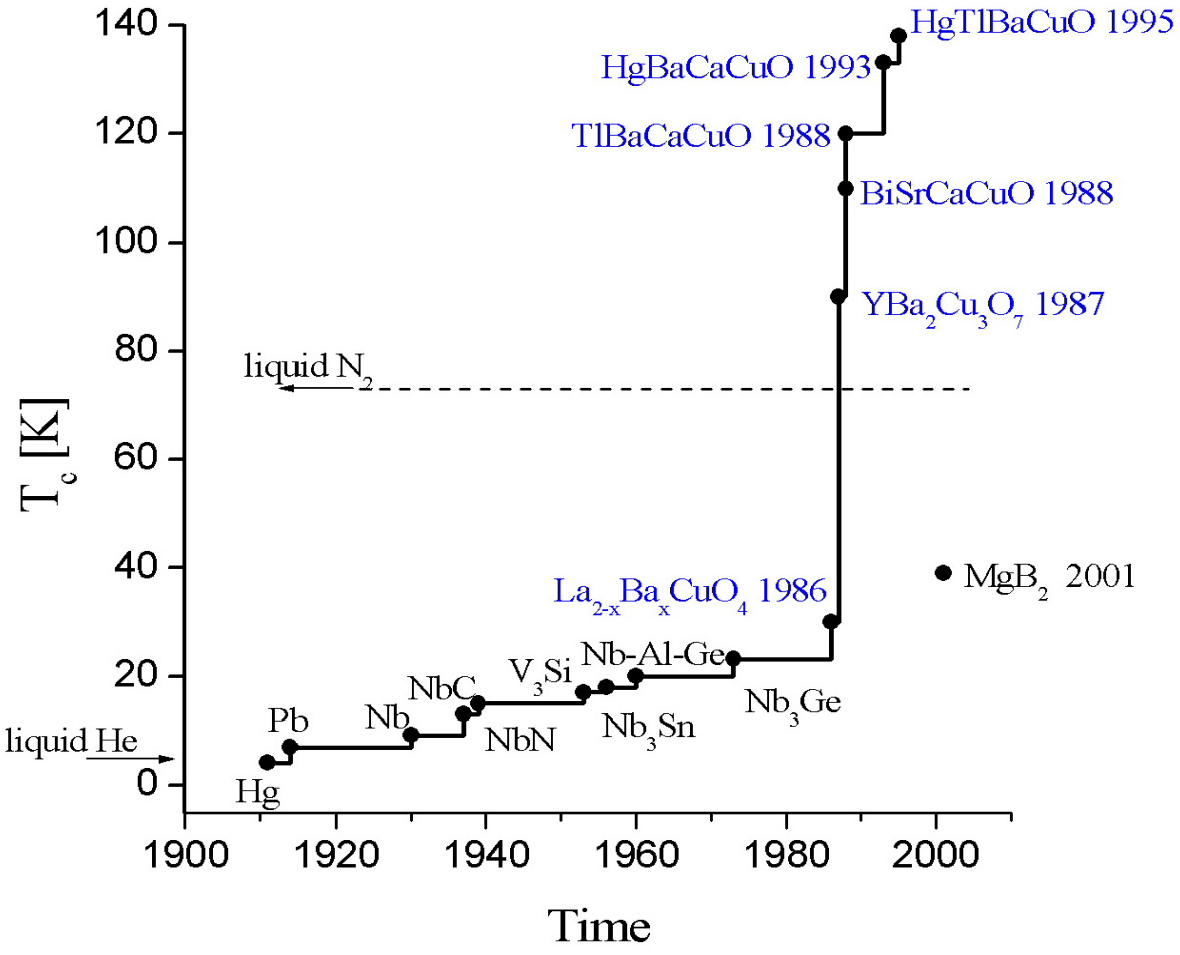
Introdution to strongly interacting materials

Transition metal Oxides



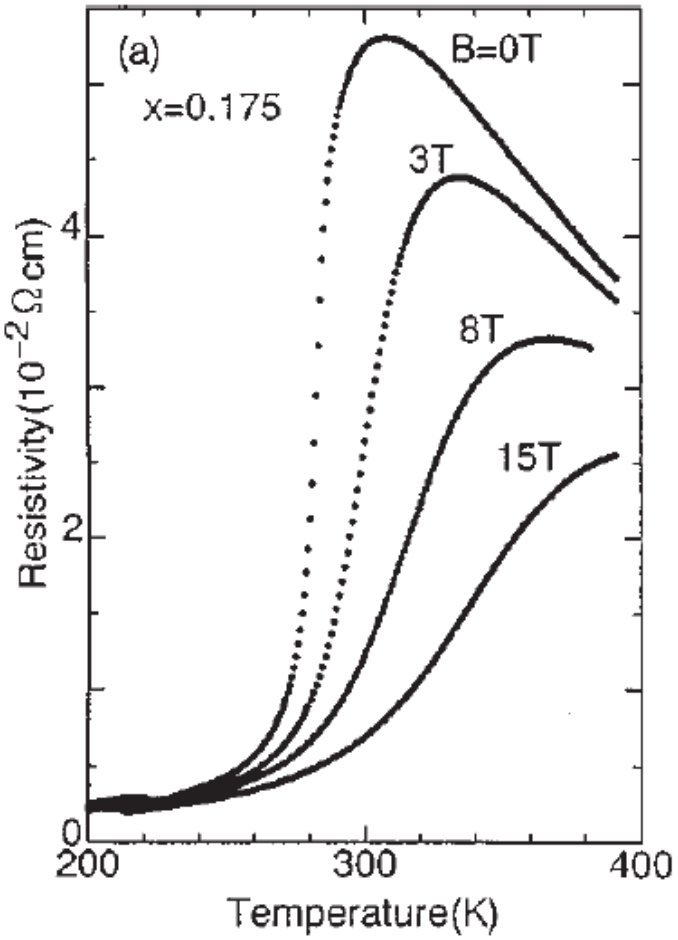
hydrogen 1 H 1.0079																	helium 2 He 4.0026
lithium 3 Li 6.941	beryllium 4 Be 9.0122		Key: element name / atomic number / symbol / atomic weight (mean relative mass)										boron 5 B 10.811	carbon 6 C 12.011	nitrogen 7 N 14.007	oxygen 8 O 15.999	fluorine 9 F 18.998	neon 10 Ne 20.180
sodium 11 Na 22.990	magnesium 12 Mg 24.305												aluminium 13 Al 26.982	silicon 14 Si 28.086	phosphorus 15 P 30.974	sulfur 16 S 32.065	chlorine 17 Cl 35.453	argon 18 Ar 39.948
potassium 19 K 39.098	calcium 20 Ca 40.078		scandium 21 Sc 44.956	titanium 22 Ti 47.867	vanadium 23 V 50.942	chromium 24 Cr 51.996	manganese 25 Mn 54.938	iron 26 Fe 55.845	cobalt 27 Co 58.933	nickel 28 Ni 58.693	copper 29 Cu 63.546	zinc 30 Zn 65.38	gallium 31 Ga 69.723	germanium 32 Ge 72.61	arsenic 33 As 74.922	selenium 34 Se 78.96	bromine 35 Br 79.904	krypton 36 Kr 83.80
rubidium 37 Rb 85.468	strontium 38 Sr 87.62		yttrium 39 Y 88.906	zirconium 40 Zr 91.224	niobium 41 Nb 92.906	molybdenum 42 Mo 95.96	technetium 43 Tc [98]	ruthenium 44 Ru 101.07	rhodium 45 Rh 102.91	palladium 46 Pd 106.42	silver 47 Ag 107.87	cadmium 48 Cd 112.41	indium 49 In 114.82	tin 50 Sn 118.71	antimony 51 Sb 121.76	tellurium 52 Te 127.60	iodine 53 I 126.90	xenon 54 Xe 131.29
caesium 55 Cs 132.91	barium 56 Ba 137.33	57-70 *	lutetium 71 Lu 174.97	hafnium 72 Hf 178.49	tantalum 73 Ta 180.95	tungsten 74 W 183.84	rhenium 75 Re 186.21	osmium 76 Os 190.23	iridium 77 Ir 192.22	platinum 78 Pt 195.08	gold 79 Au 196.97	mercury 80 Hg 200.59	thallium 81 Tl 204.38	lead 82 Pb 207.2	bismuth 83 Bi 208.98	polonium 84 Po [209]	astatine 85 At [210]	radon 86 Rn [222]
francium 87 Fr [223]	radium 88 Ra [226]	89-102 **	lawrencium 103 Lr [262]	rutherfordium 104 Rf [267]	dubnium 105 Db [268]	seaborgium 106 Sg [271]	bohrium 107 Bh [272]	hassium 108 Hs [270]	meitnerium 109 Mt [276]	darmstadtium 110 Ds [281]	roentgenium 111 Rg [280]	ununbium 112 Uub [285]	ununtrium 113 Uut [284]	ununquadium 114 Uuq [289]	ununpentium 115 Uup [288]	ununhexium 116 Uuh [293]	ununseptium 117 Uus —	ununoctium 118 Uuo [294]

High temperature superconductivity



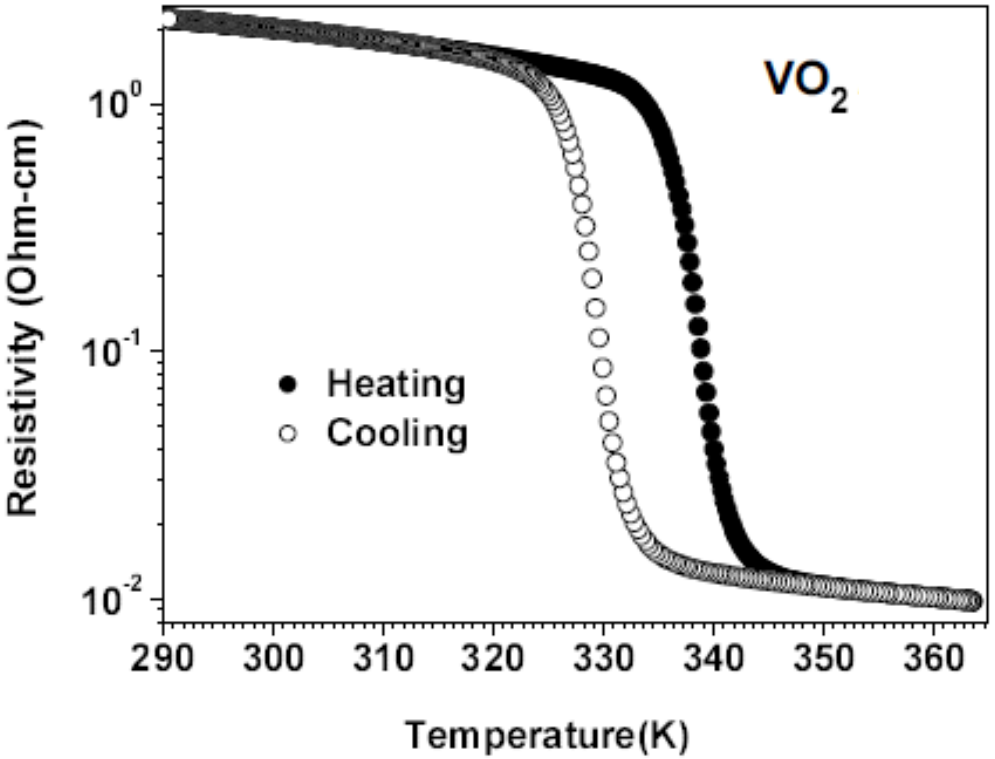
Colossal Magnetoresistance

$La_{1-x}Sr_xMnO_3$



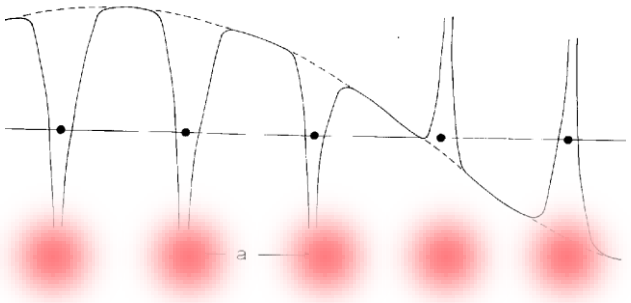
Metal insulator transition

R Von Helmolt, Phys. Rev. Lett, 1993

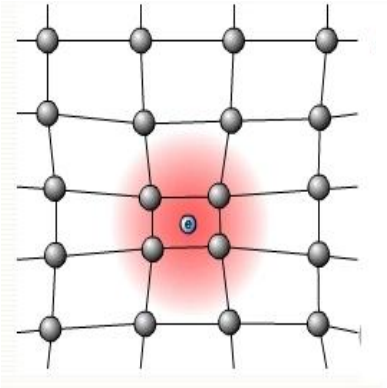


Strongly interacting materials

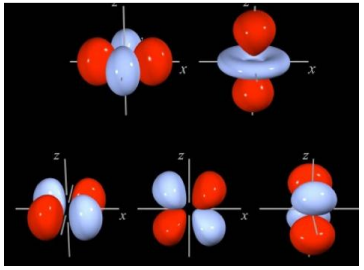
Poor screening



Electron-phonon interaction



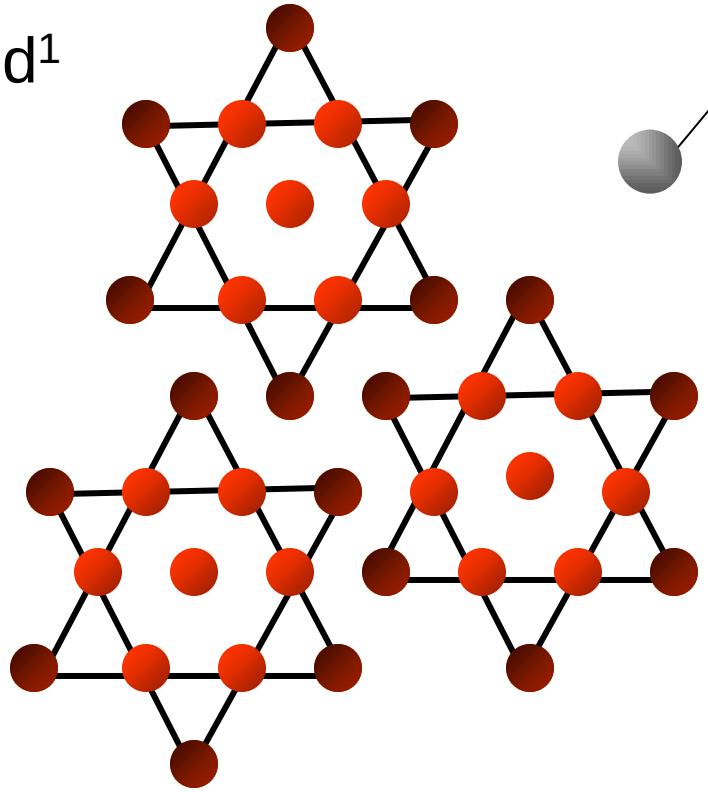
Structural distortion

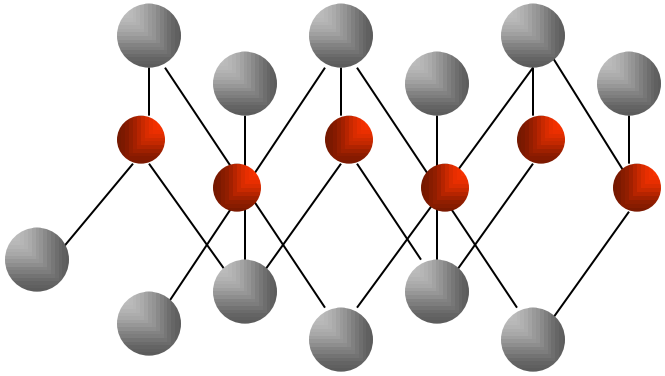


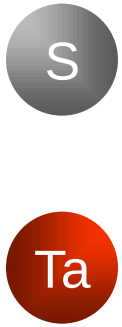
Breakdown of the band picture

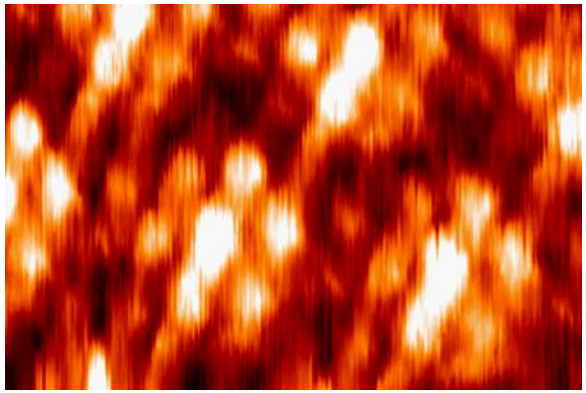
Structural distortion and Mott transition in 1T-TaS_2

$5d^1$



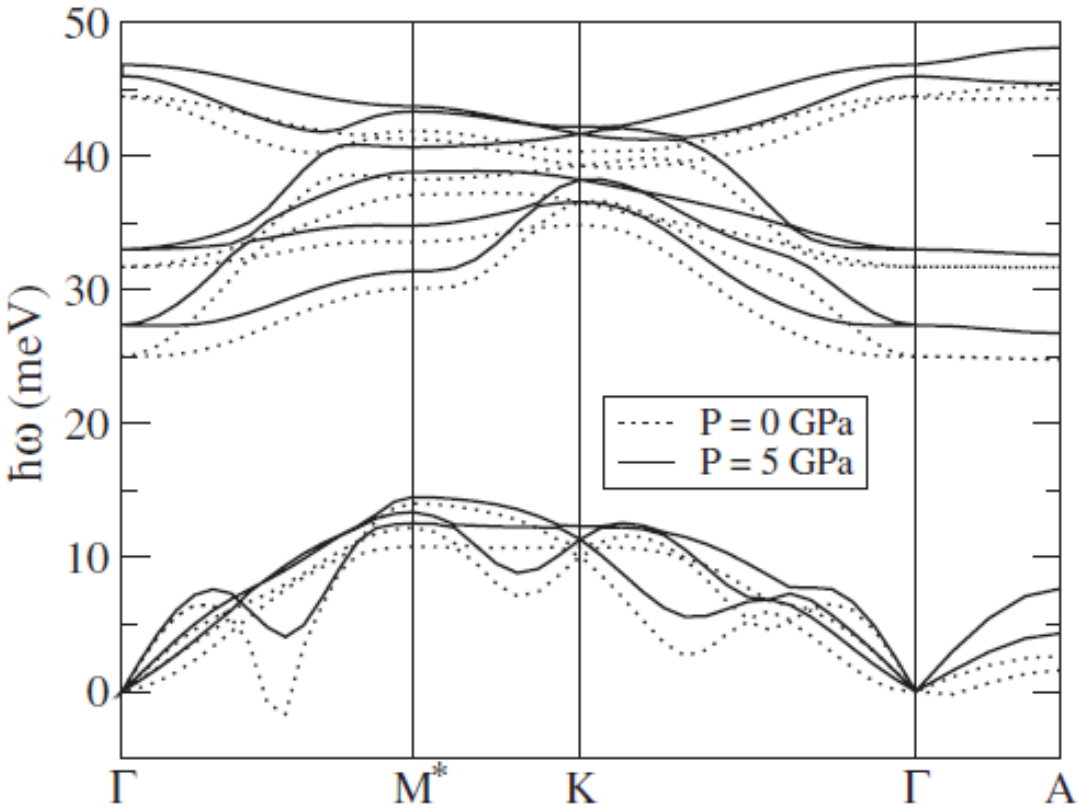




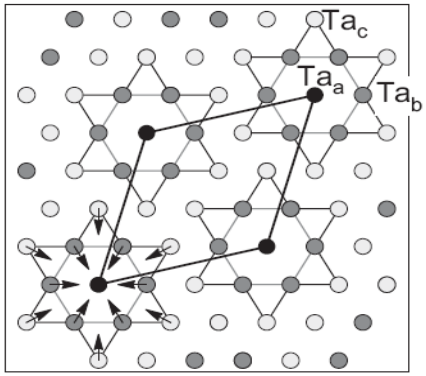


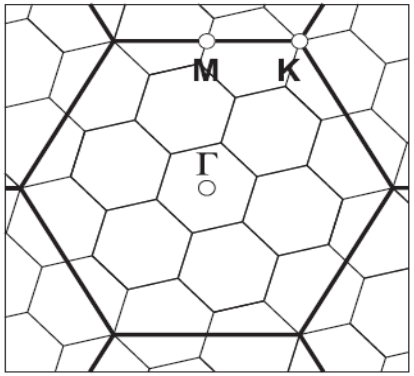
Scanning tunneling microscopy

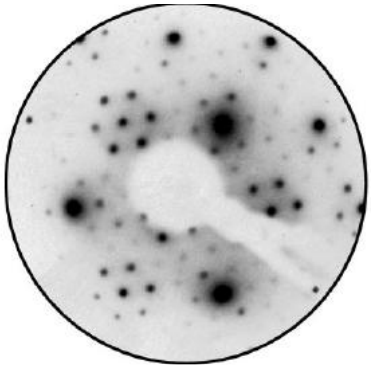
Phonon softening due to large *el-ph* coupling



$1T$-TaS_2





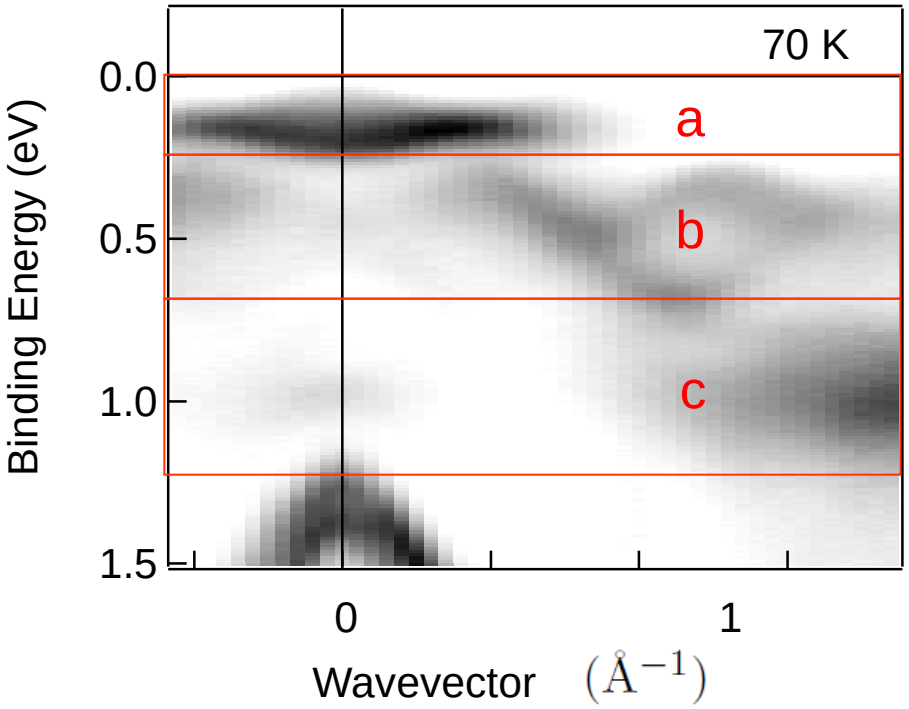


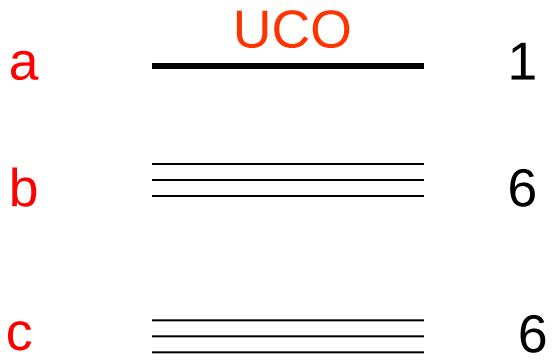
Large structural distortion

Amy Y. Liu, Phys Rev B, 2009

3 many-folds induced by the CDW

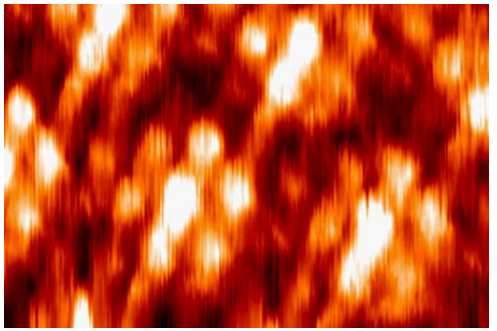
$1T\text{-}TaS_2$



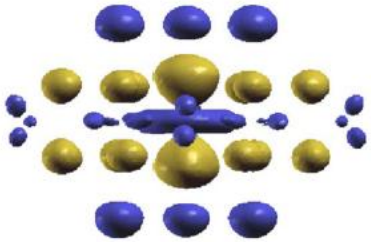


Perfetti, Phys. Rev. B, 2005

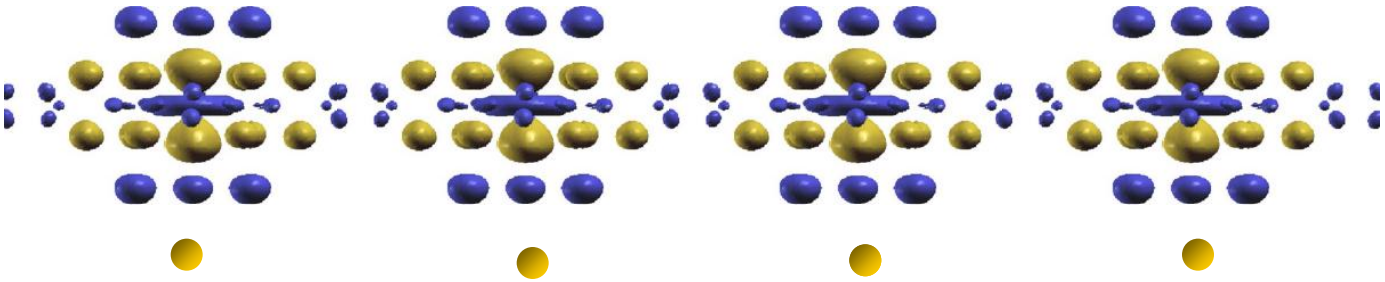
Top view of the UCO



Side view of the UCO



A purely electronic phenomenon: the Mott transition

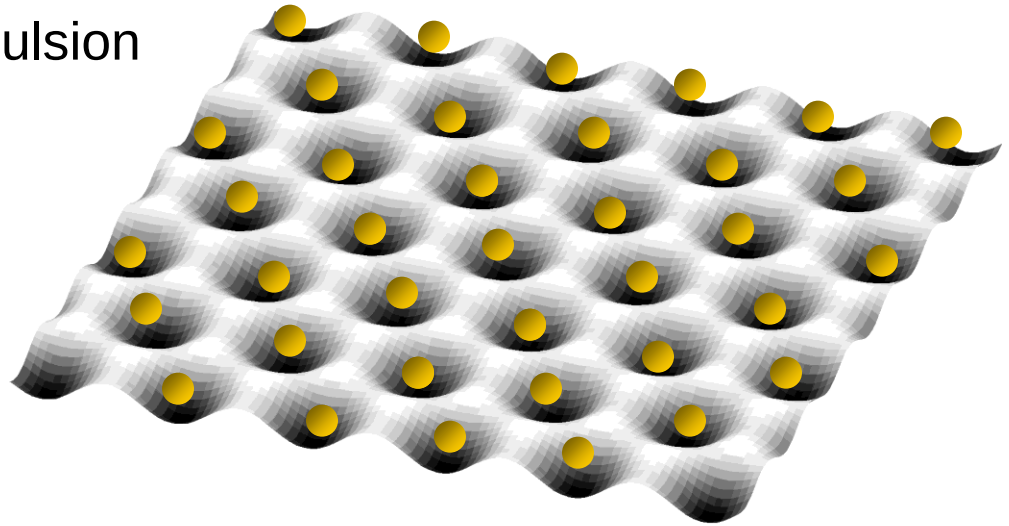


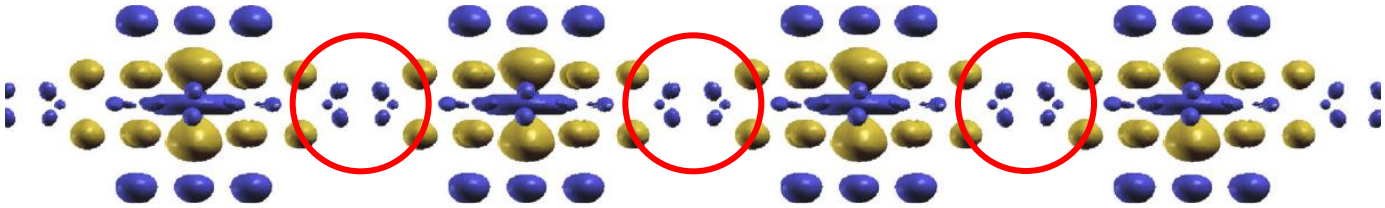
W bandwidth

U Coulomb repulsion

U/W > 1

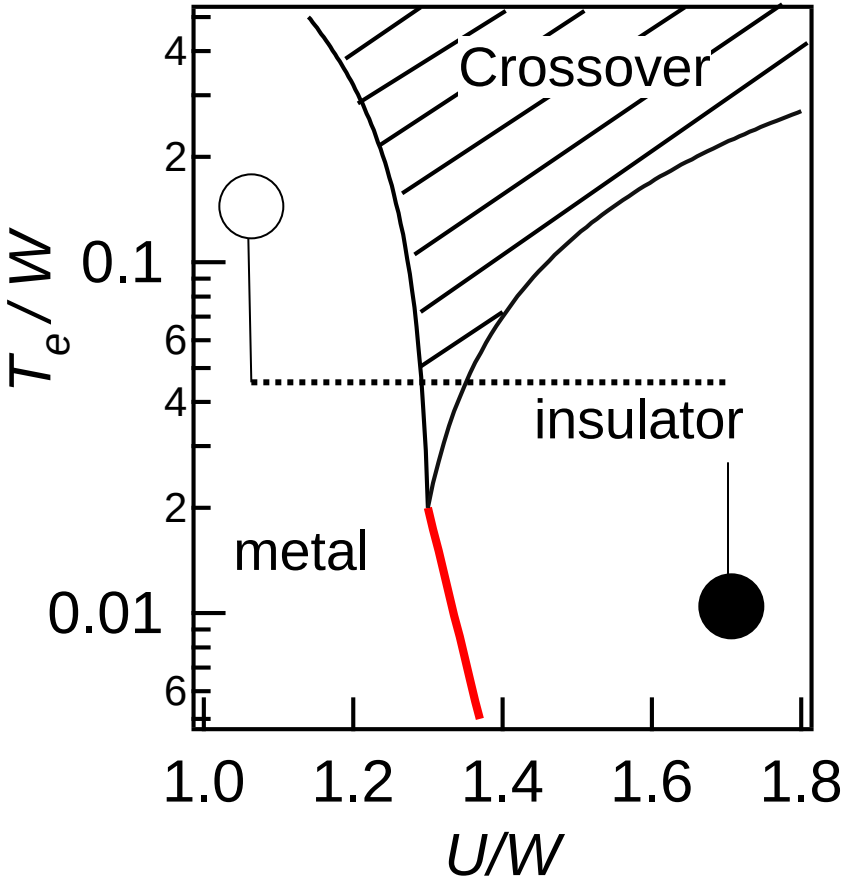
Mott transition

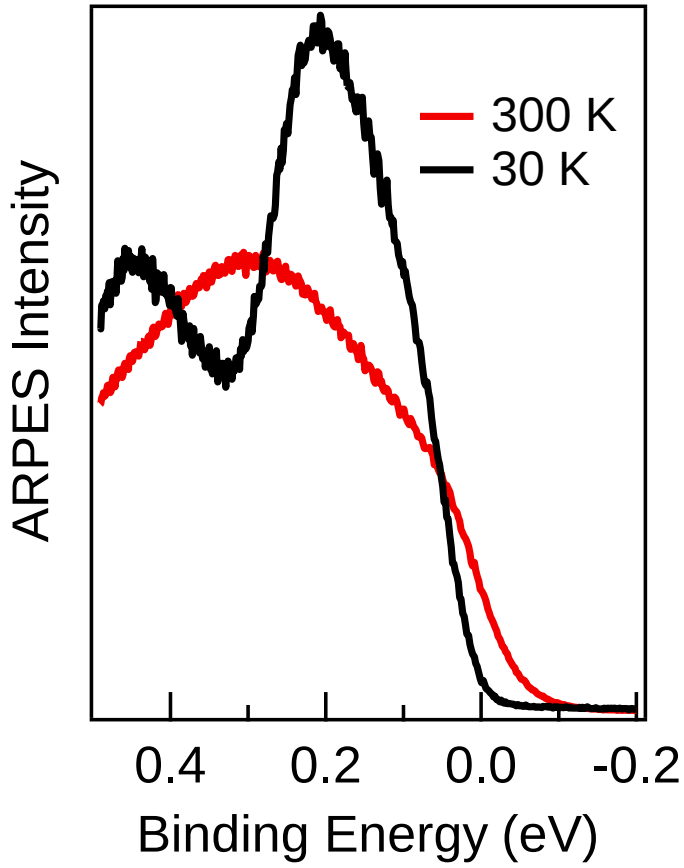




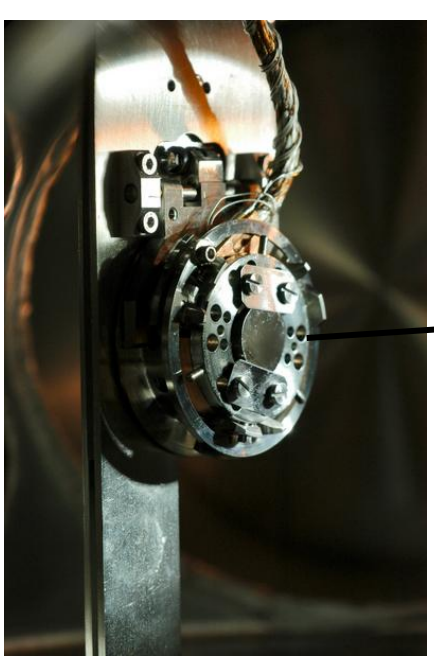
T_l decreases, Larger structural distortion

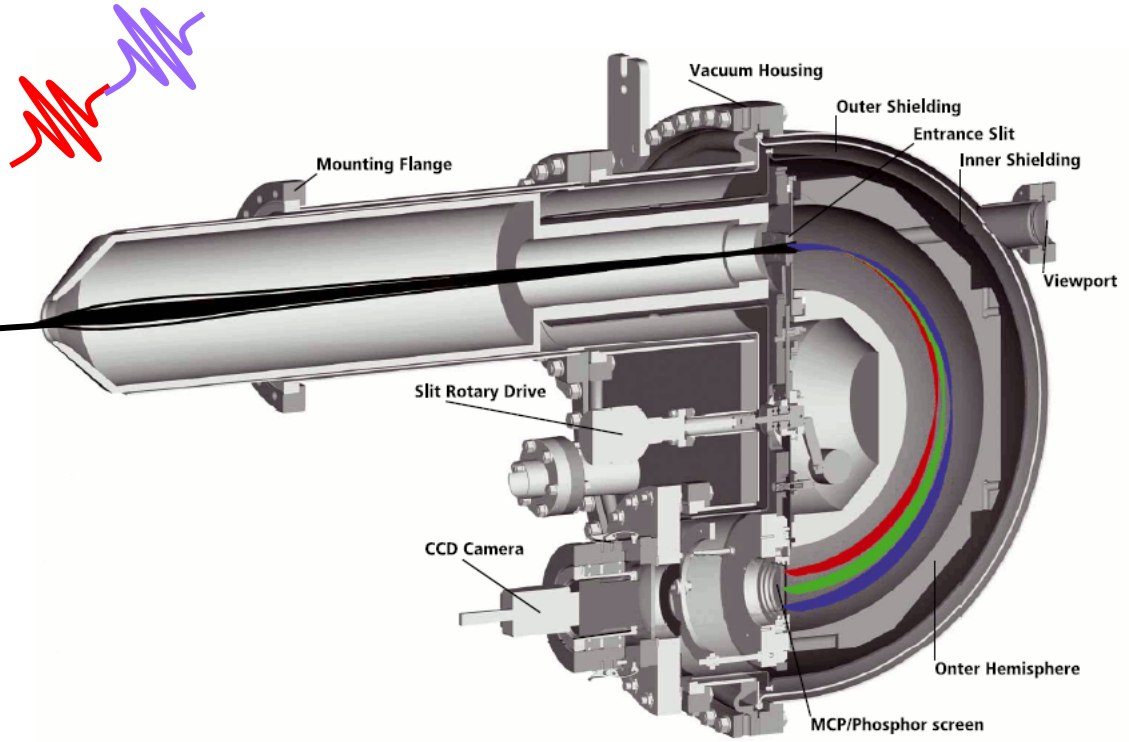
Smaller W and larger U/W



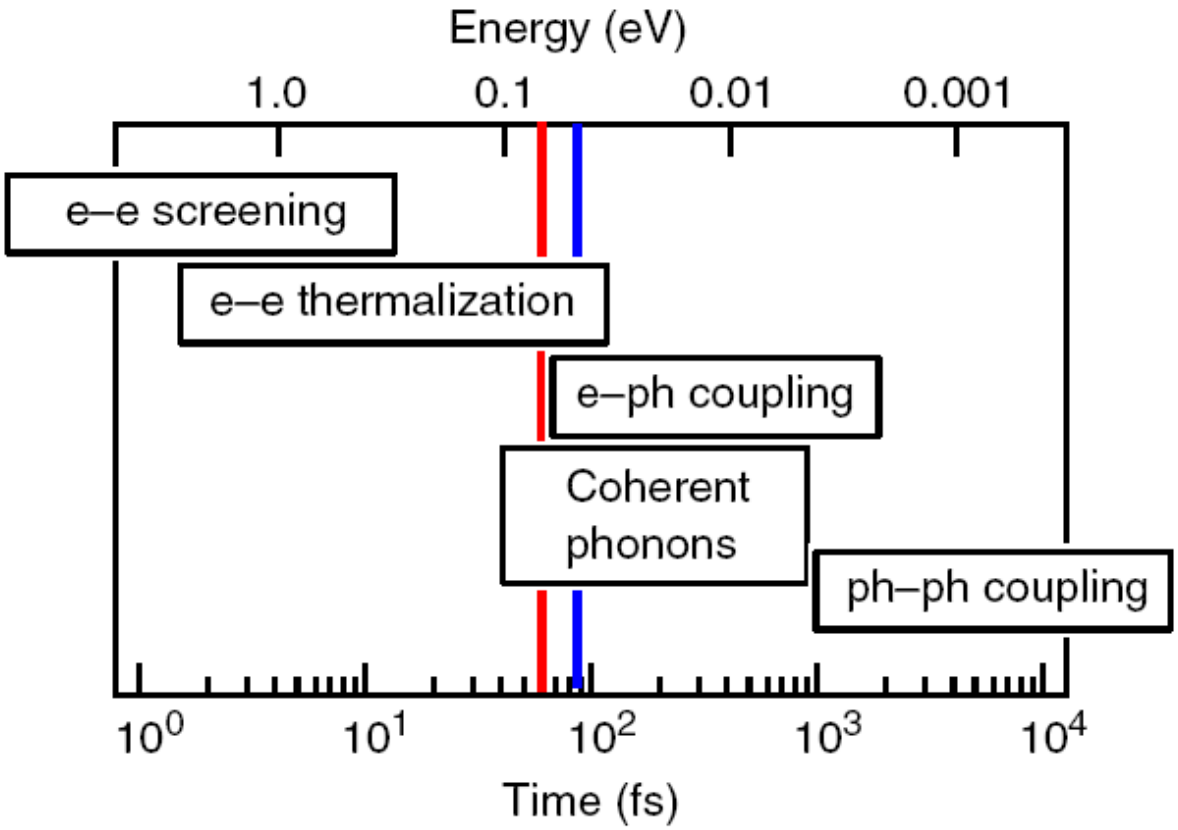


Time resolved photoemission with lasers





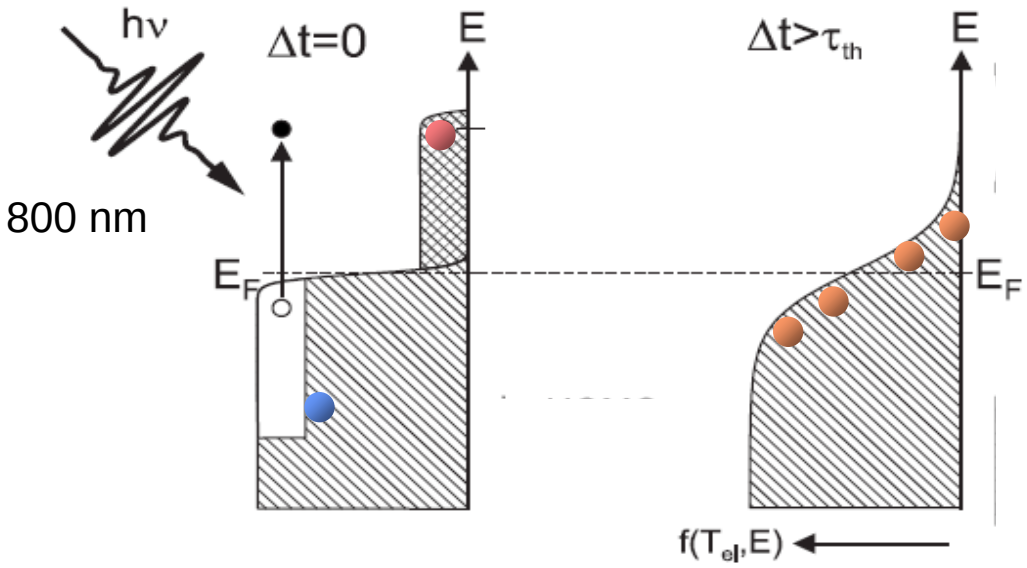
Different timescale for electronic motion and lattice motion



R. P. Feynman: The system is said to be in thermal equilibrium if all the *fast* things have happened and the *slow* things not yet.

Dynamics of the electrons in a metal

Thermalization due to electron-electron interaction

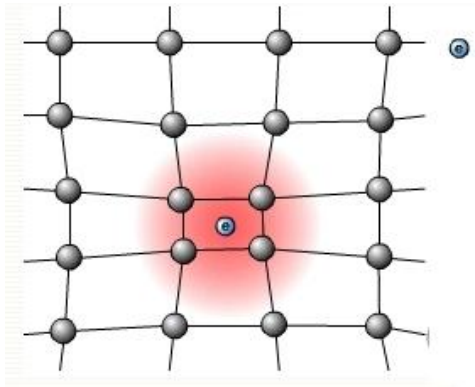


Definition of effective electronic temperature

$$f(E,T_e) = \frac{1}{(e^{\frac{E}{k_b T_e}} + 1)}$$

$$T_e >> T_p$$

P. B. Allen, Phys. Rev. Lett, 1987



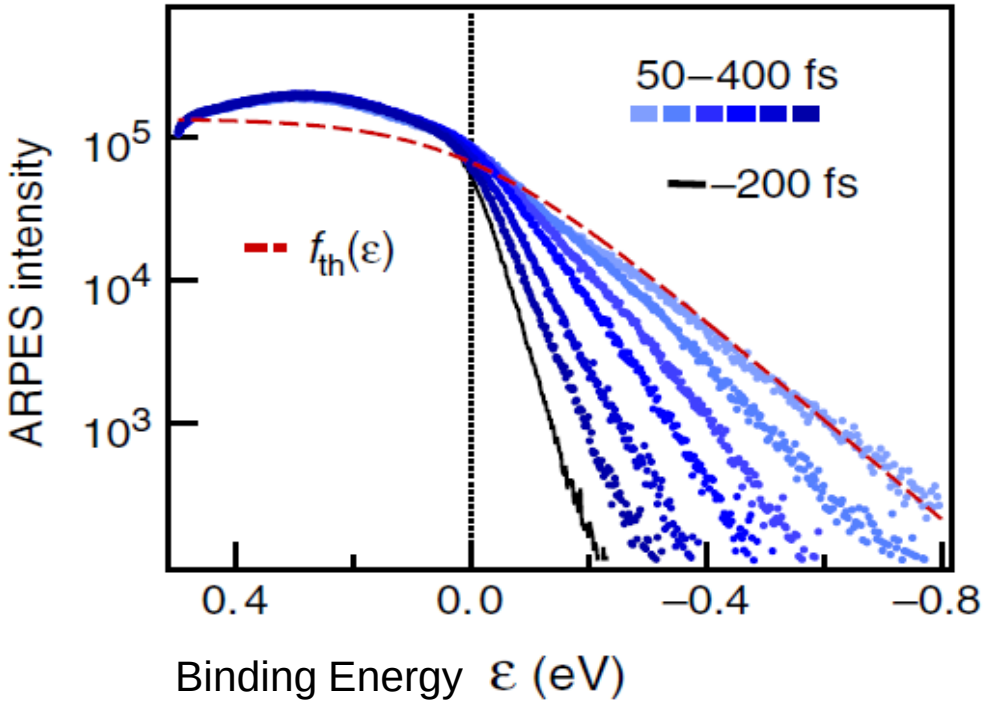
Energy relaxation of excited electrons due to Electron-phonon interaction

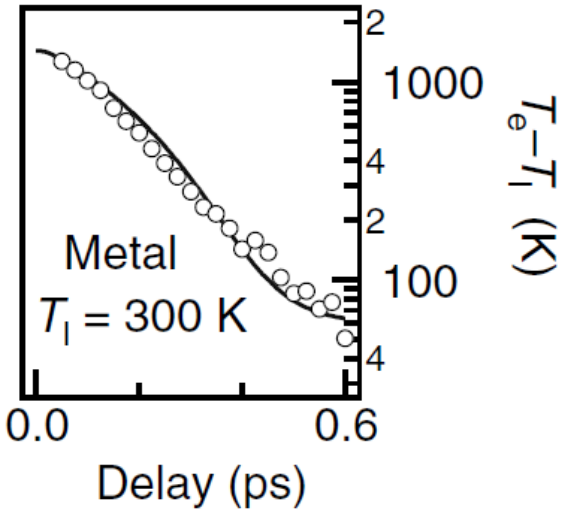
$$\frac{\partial T_e}{\partial \tau} = -\frac{3\hbar}{\pi k_B}\lambda\langle\Omega^2\rangle\frac{T_e - T_p}{T_e}$$

Metallic phase $T_I = 300$ K

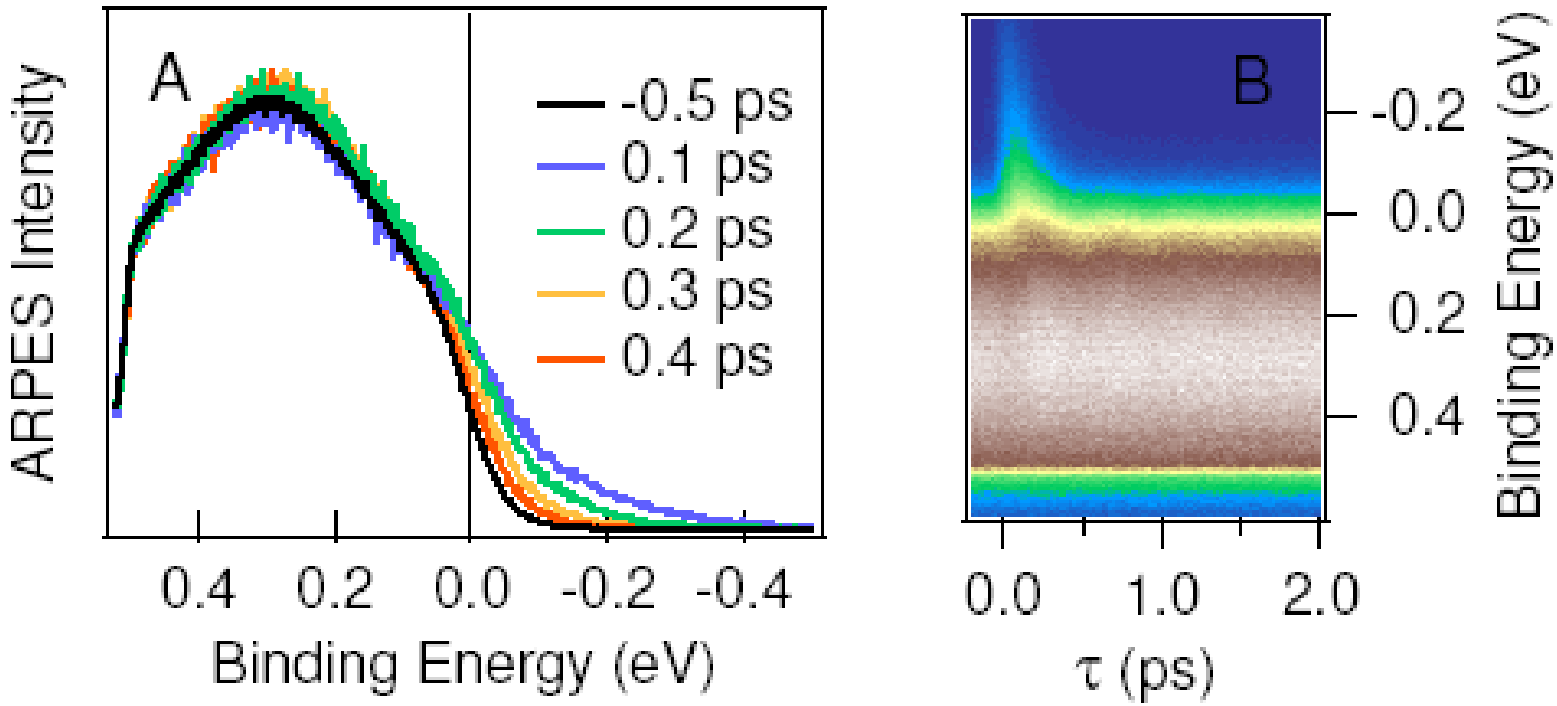
Very fast electronic thermalization

Electrons reach 1100 K and cool down in 150 fs



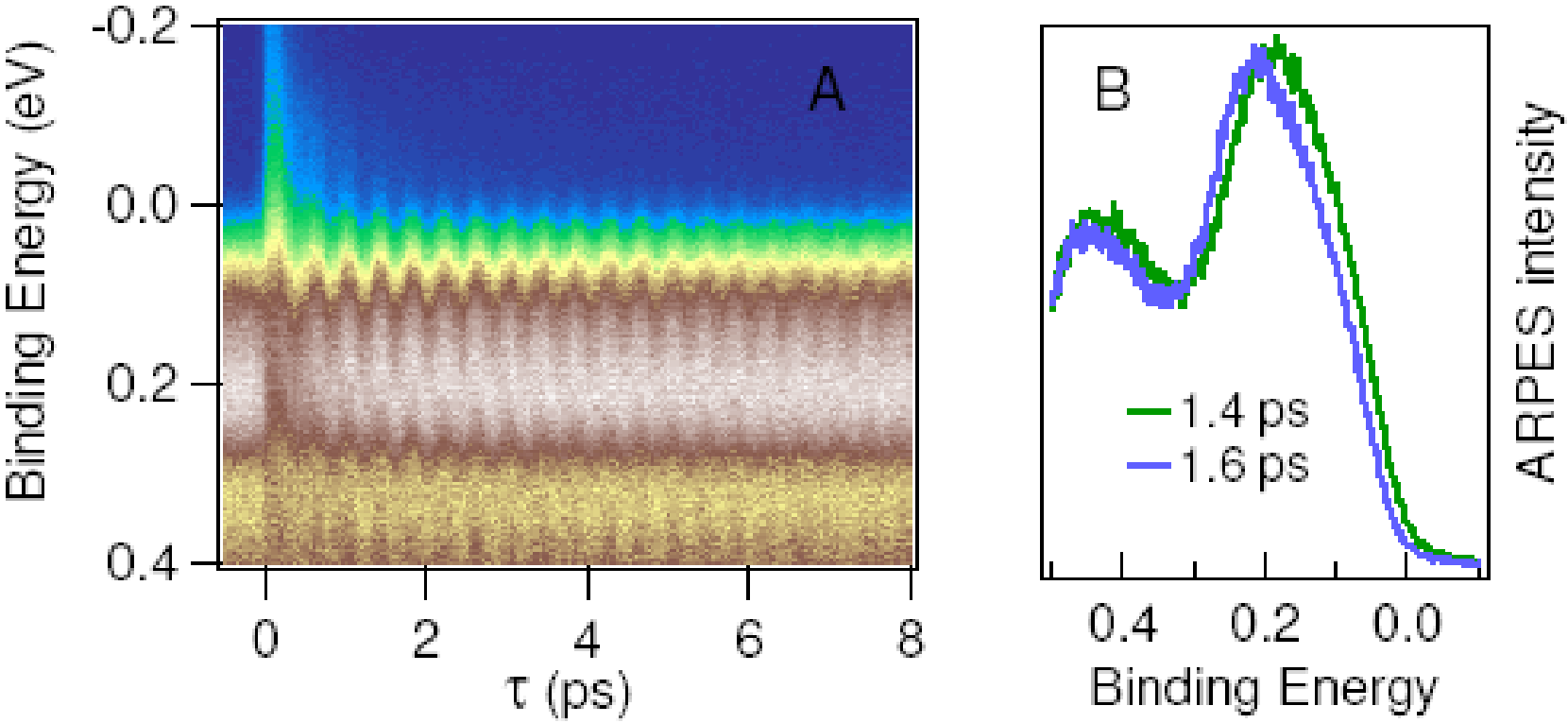


L. Perfetti, Phys. Rev. Lett. 2006



Weak oscillations of the spectrum

Mott phase T_I = 30 K

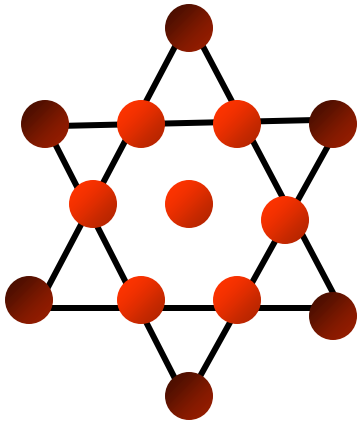


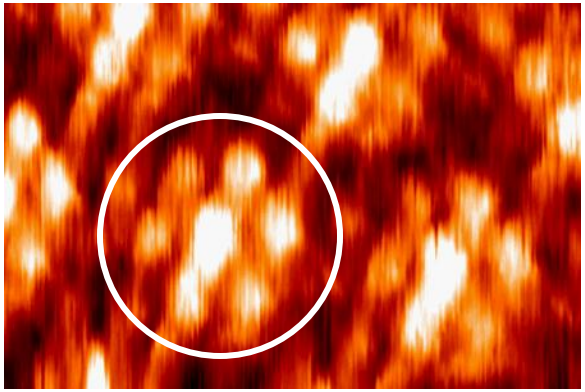
Large oscillations lasting longer than 10 ps

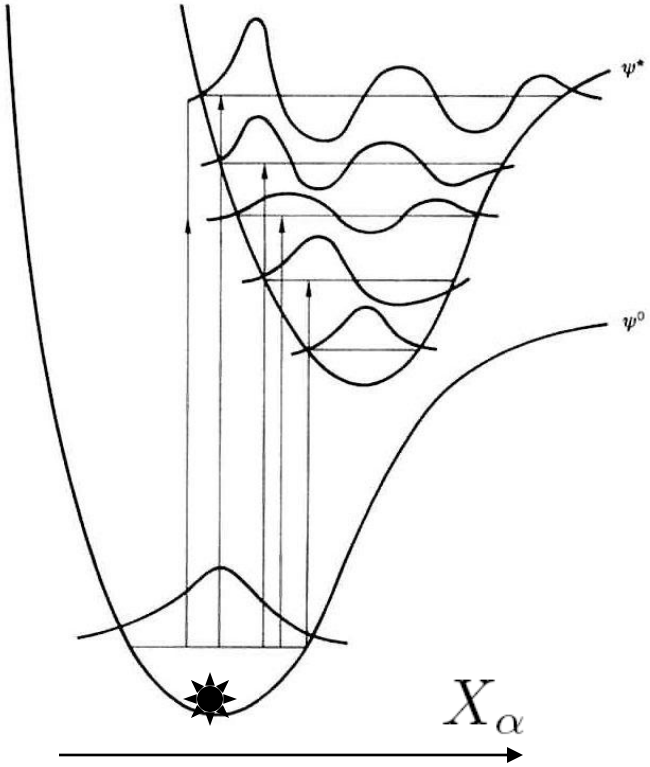
Rigid spectral shift of 18 meV

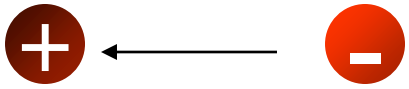
Mott phase stable after t > 1 ps

Coherent phonon excitation







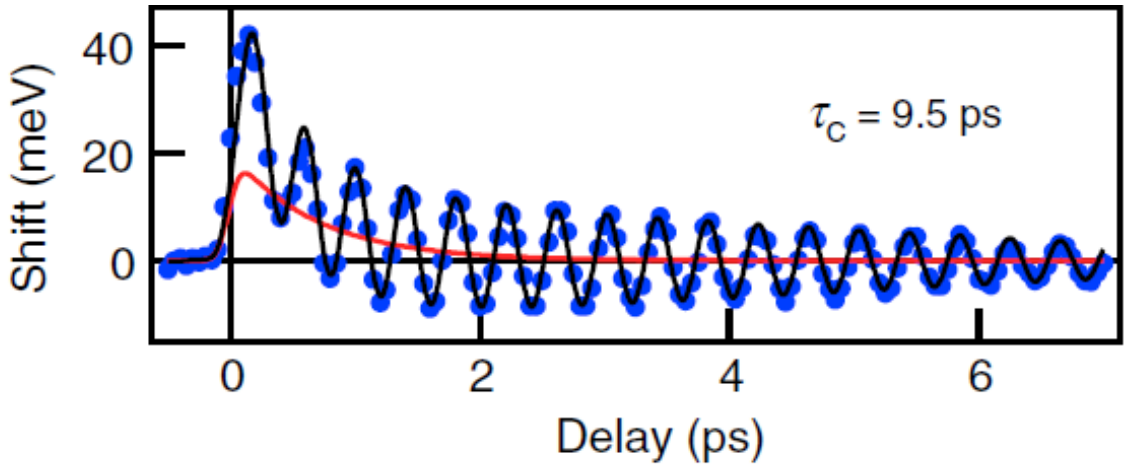


Shift of electronic binding energies

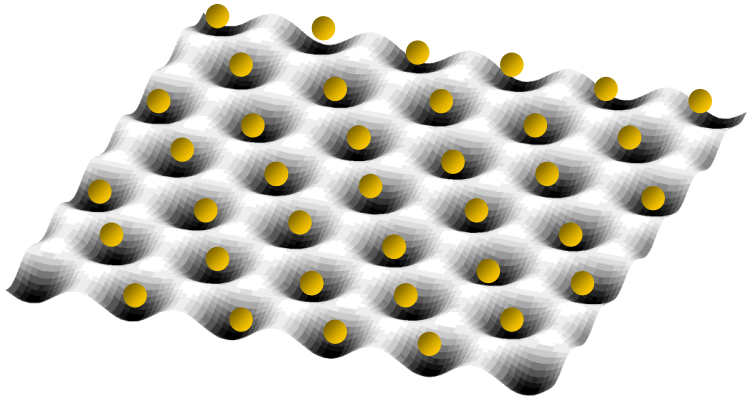
Theory of Coherent Phonons

A.V. Kuznetsov, Phys. Rev. Lett., 1994

$$\frac{\partial^2}{\partial t^2} D_{\mathbf{q}} + \omega_{\mathbf{q}}^2 D_{\mathbf{q}} = -2\omega_{\mathbf{q}} \sum_{\alpha,\mathbf{k}} M_{\mathbf{kq}}^{\alpha} n_{\mathbf{k},\mathbf{k}+\mathbf{q}}^{\alpha} \qquad n_{\mathbf{k},\mathbf{k}'}^{\alpha} = f_{\mathbf{k}}^{\alpha}(t)\delta_{\mathbf{kk}'}$$

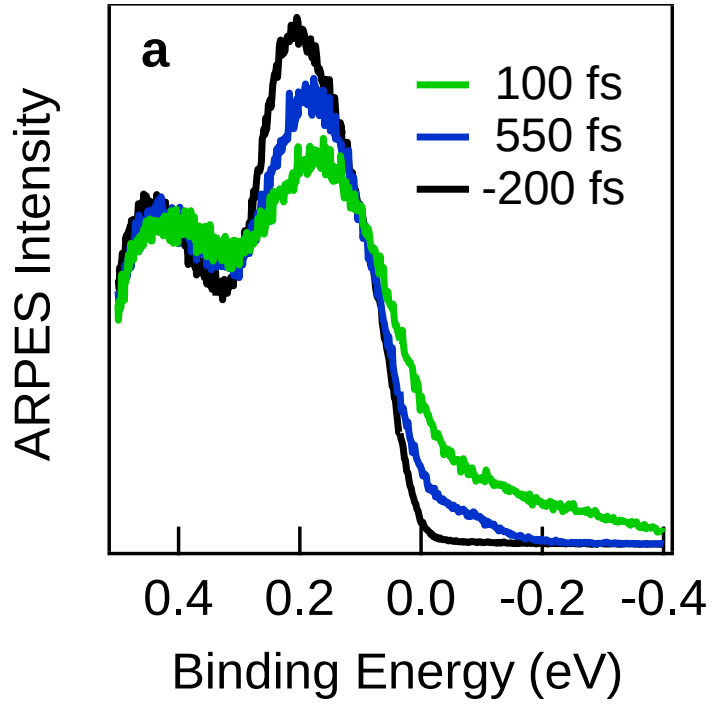


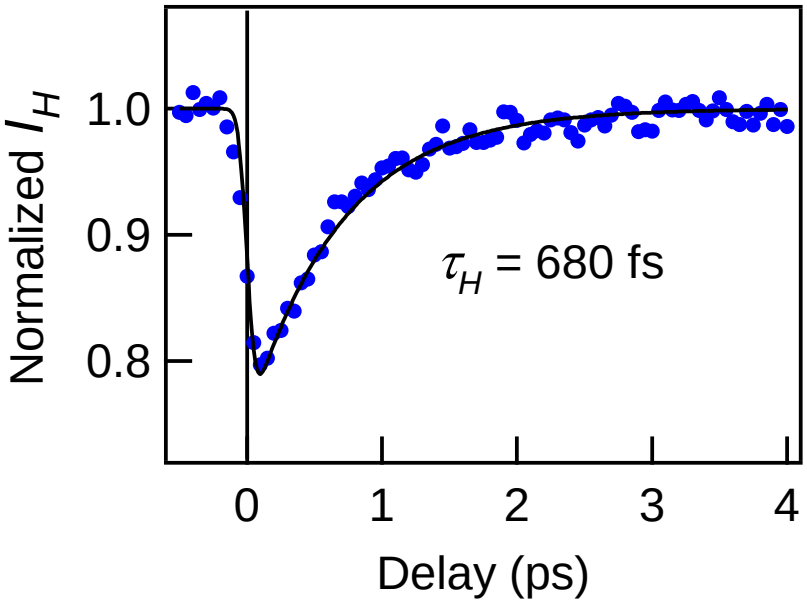
Breakdown of the MOTT phase

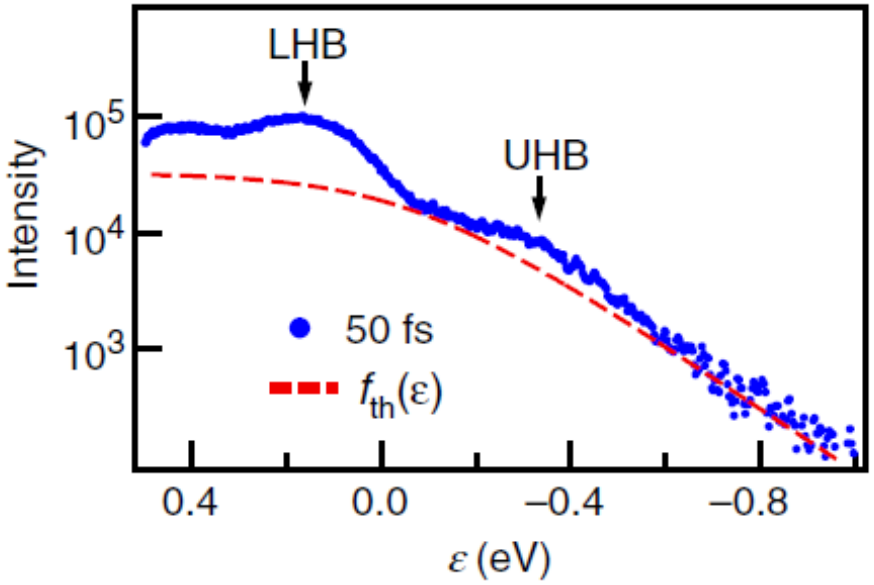


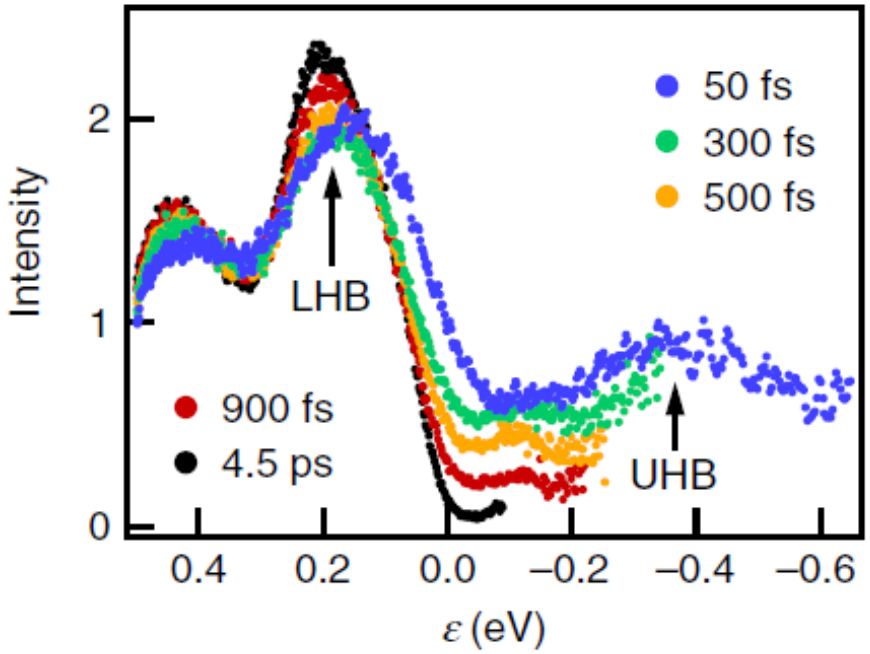
Instantaneous collapse of the charge gap

Monotonic recovery of electronic correlations in 680 fs



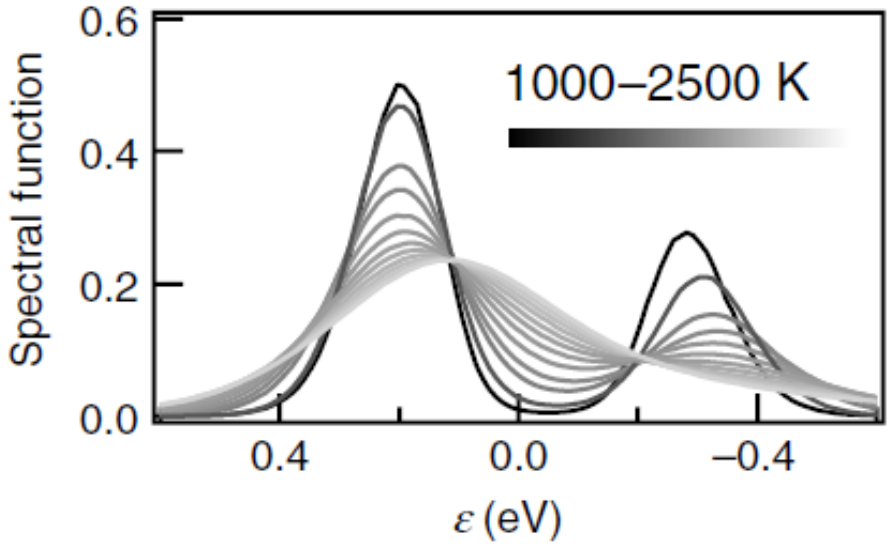


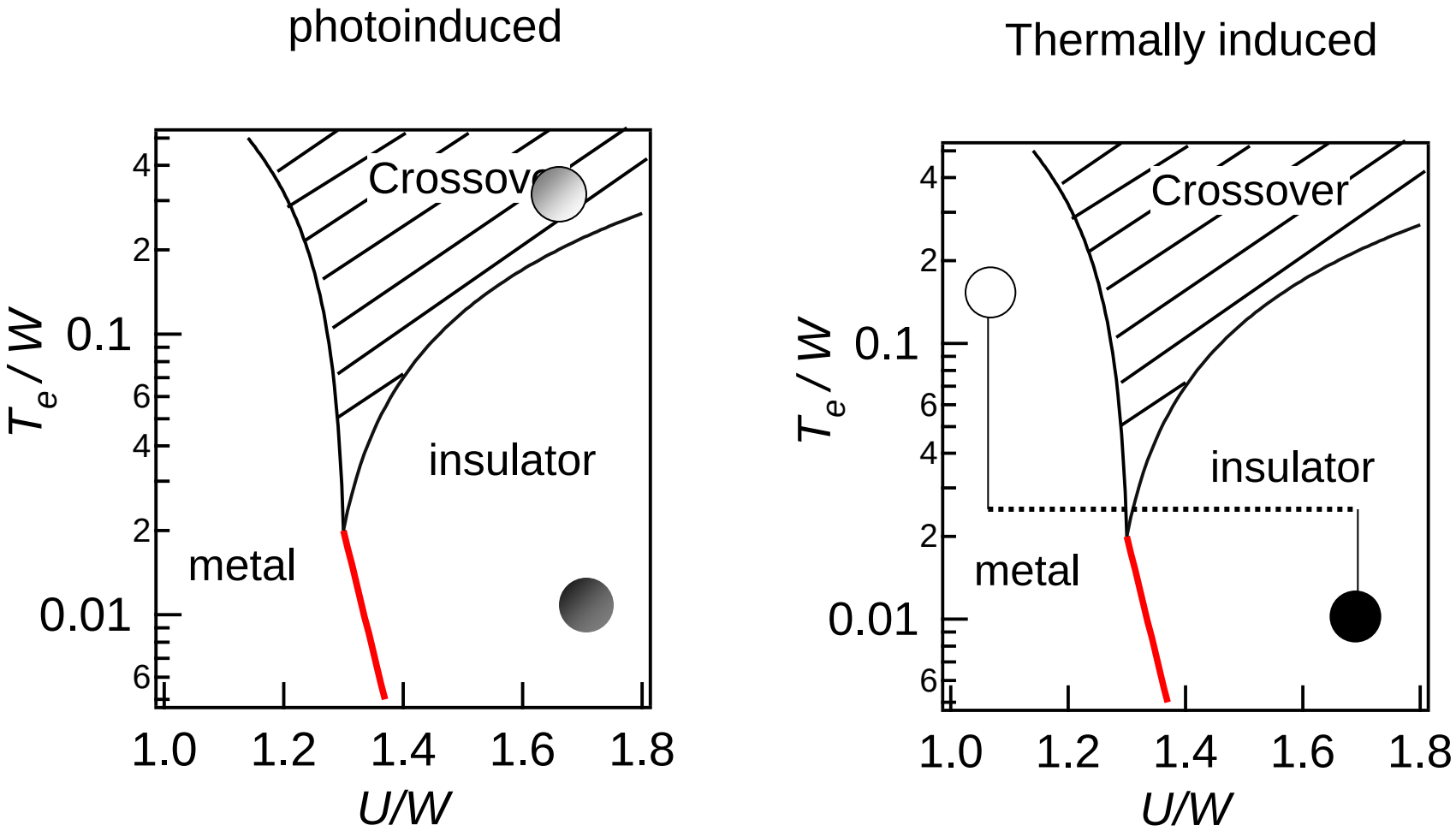




Collapse of the electronic
Gap due to the elevated electronic temperature

Dynamical mean field theory



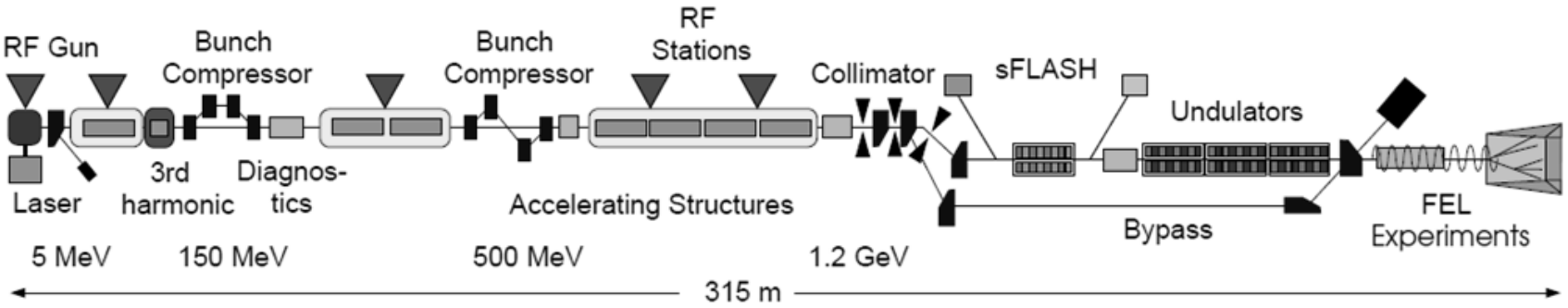

photoinduced
Crossover
insulator
metal
T_e / W
0.1
0.01
4
2
6
4
2
6
1.0
1.2
1.4
1.6
1.8
U/W
Thermally induced
Crossover
insulator
metal
T_e / W
0.1
0.01
4
2
6
4
2
6
1.0
1.2
1.4
1.6
1.8
U/W

New sources and first results

Free Electron Lasers

High flux of photons

High photon energies

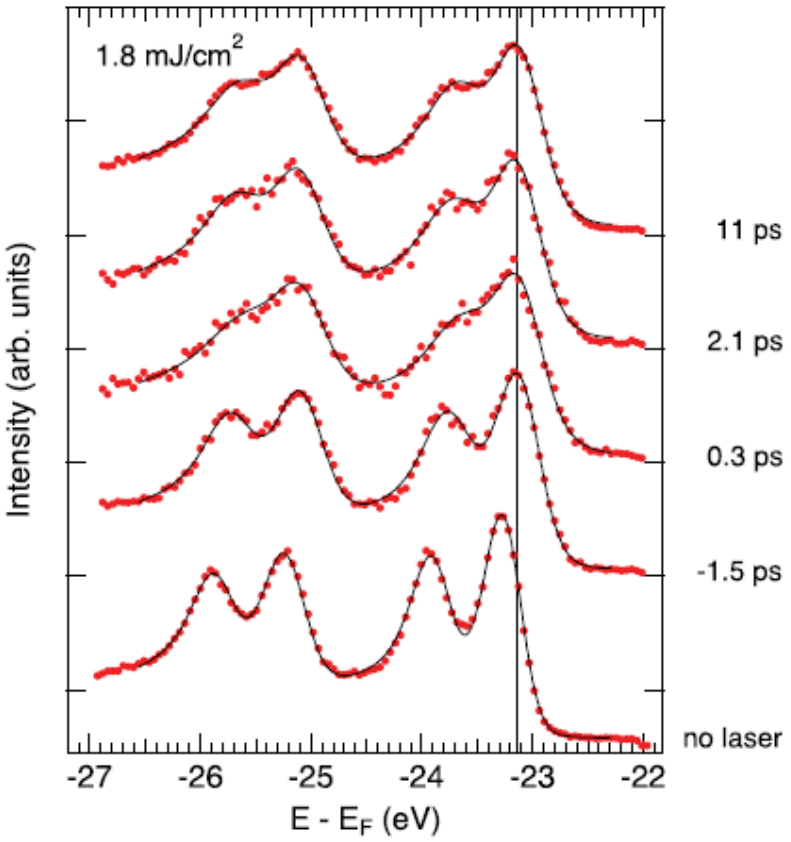


Low repetition rate

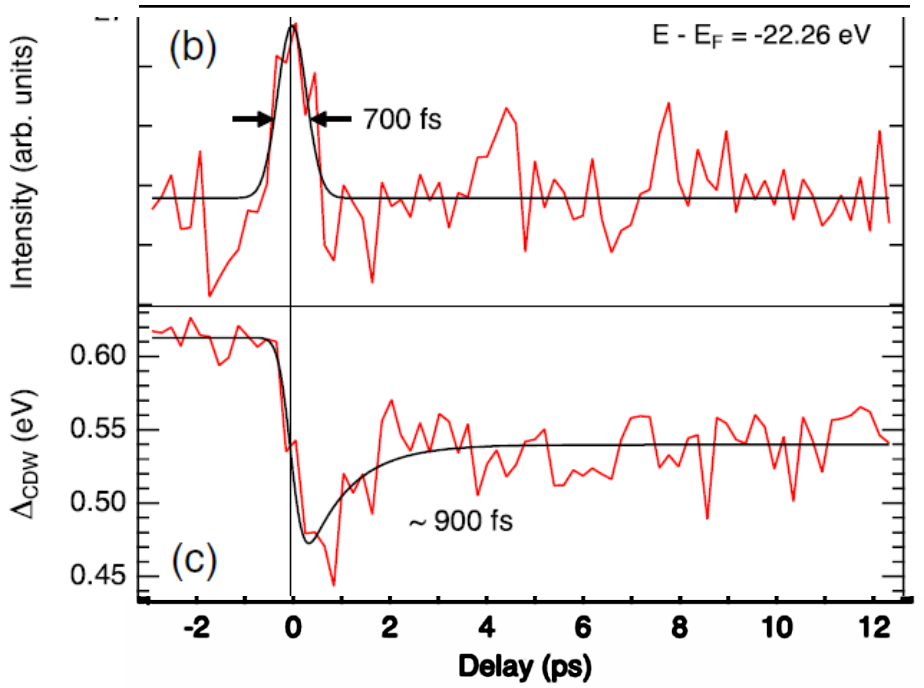
Pulse to pulse fluctuations

Gitter

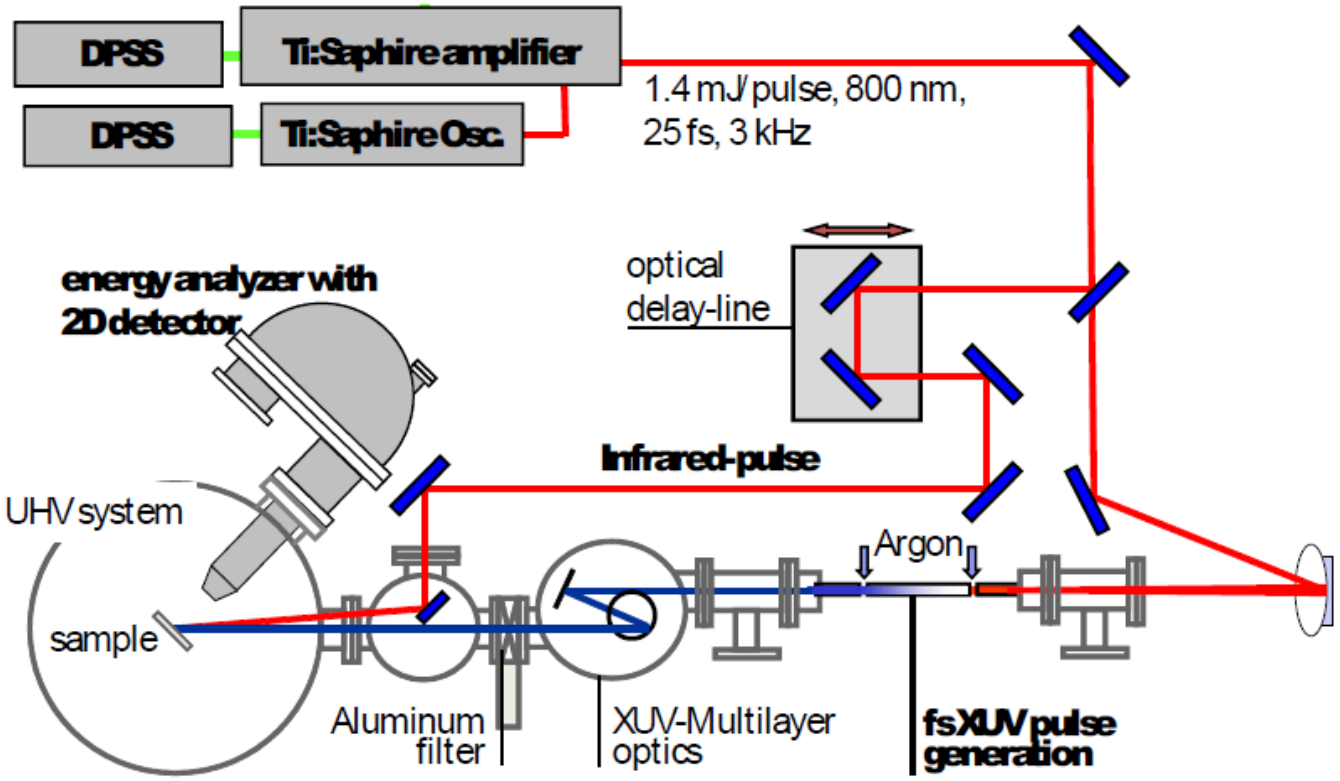
Photoinduced change of lattice structure in 1T-TaS_2



S. Hellmann, Phys. Rev. Lett, 2010

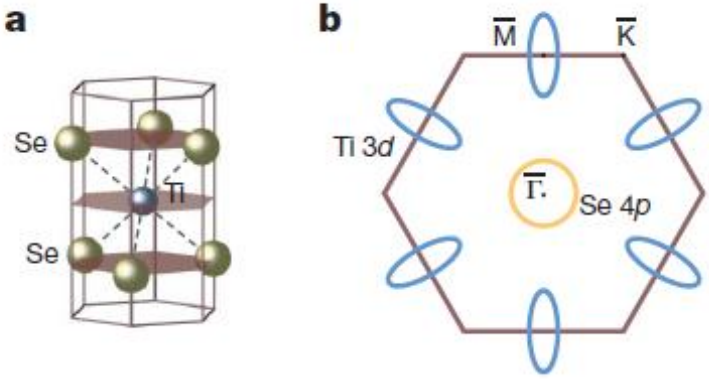


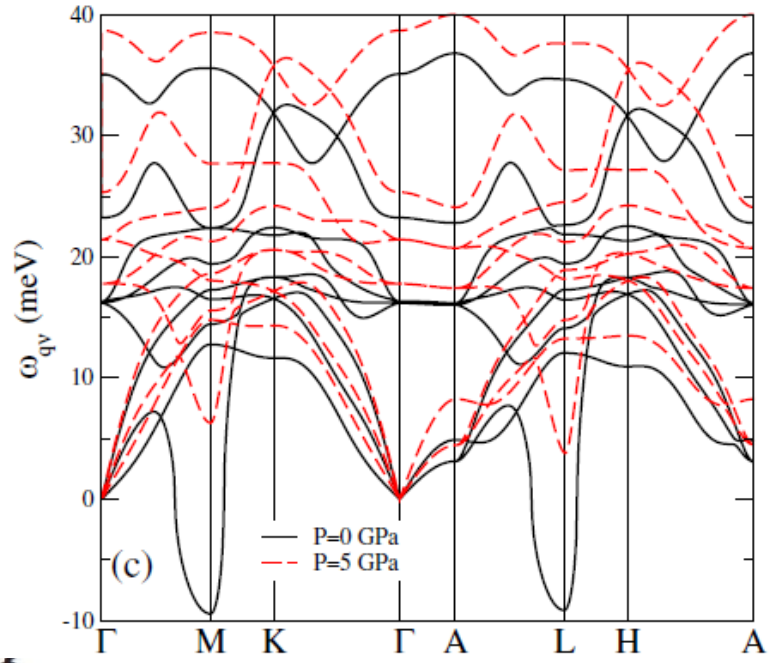
High Harmonics Generation

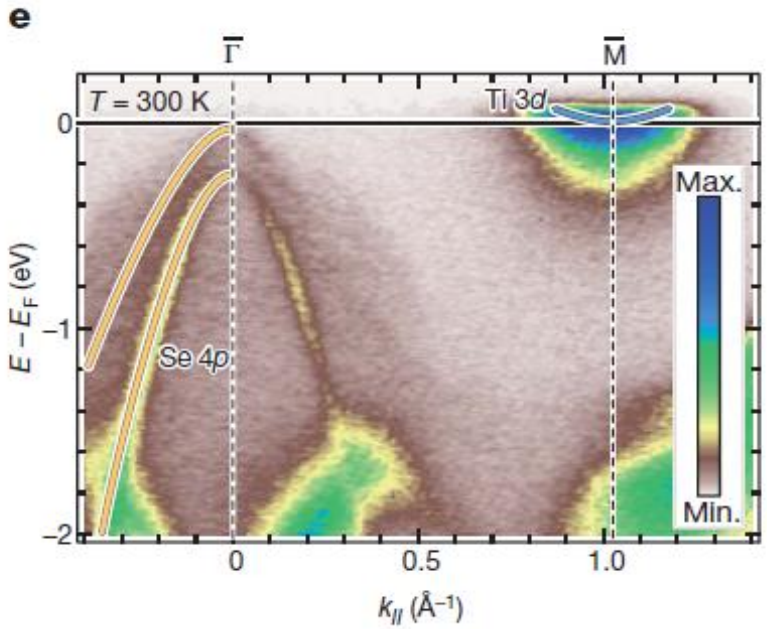


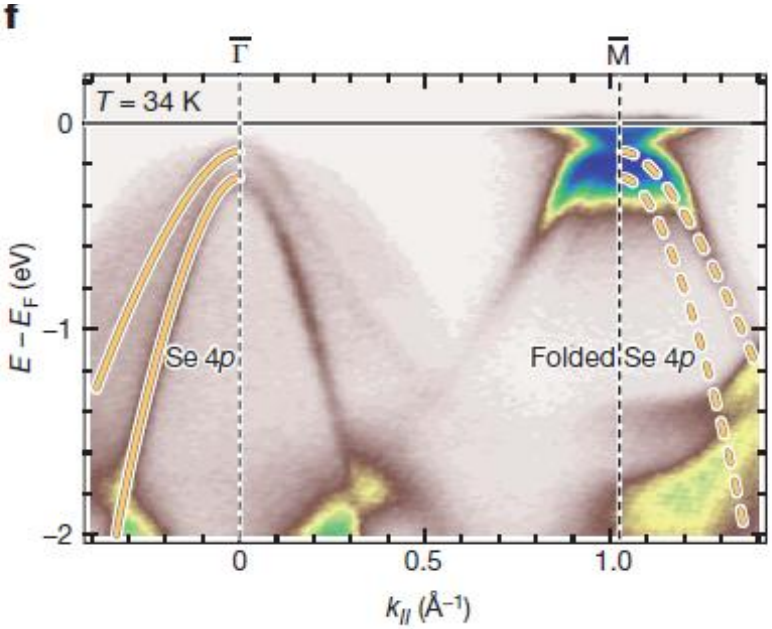
1T-$TiSe_2$

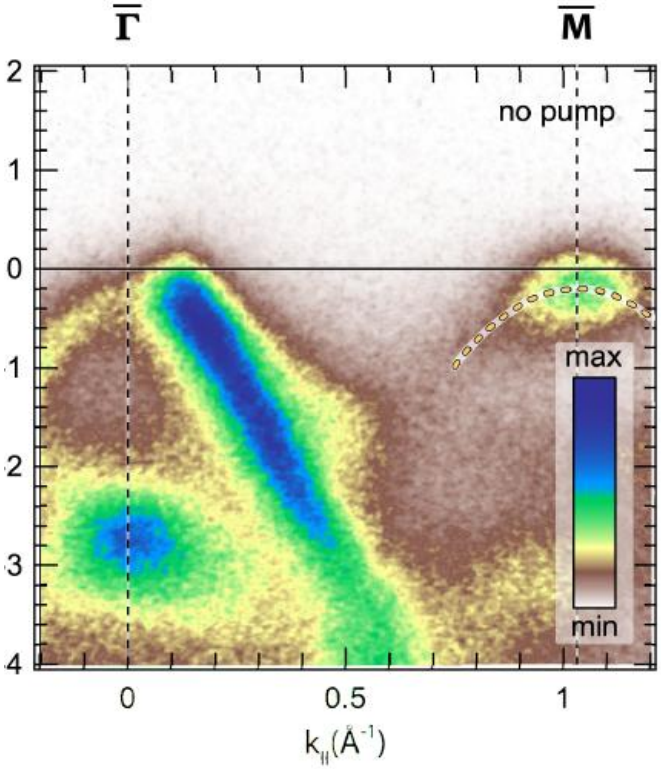
T. Rohwer, Nature 2011

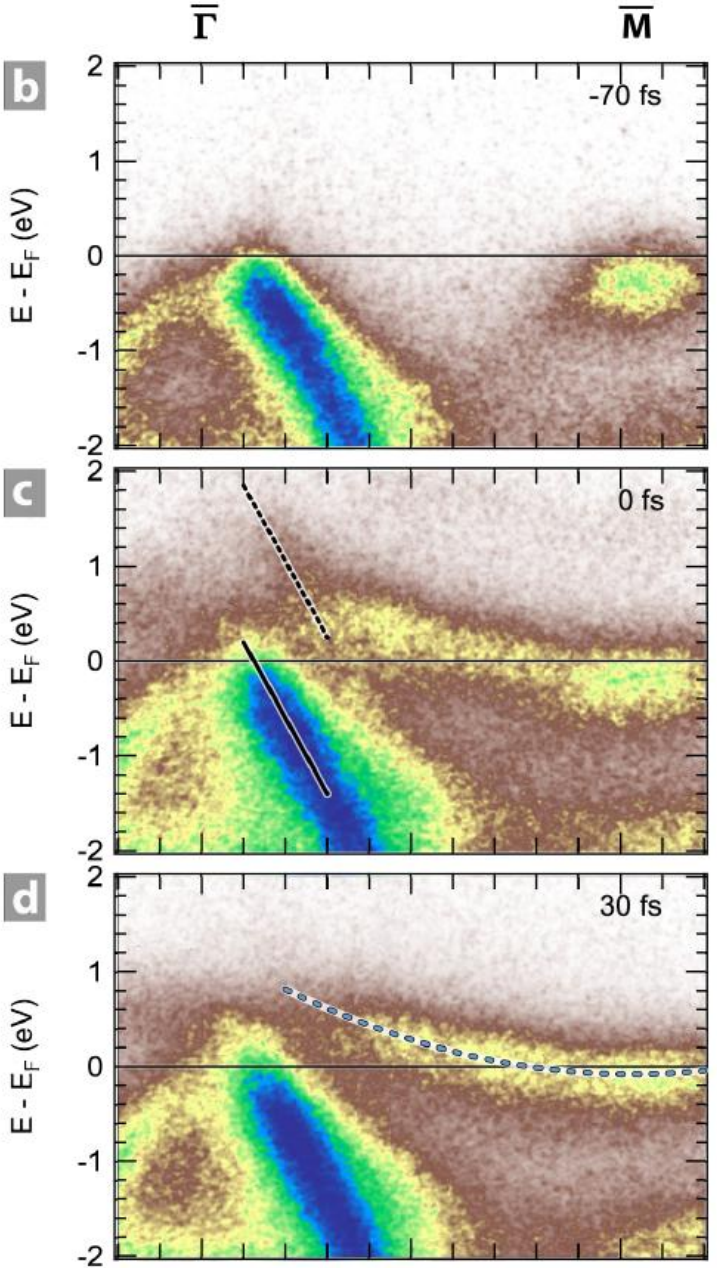








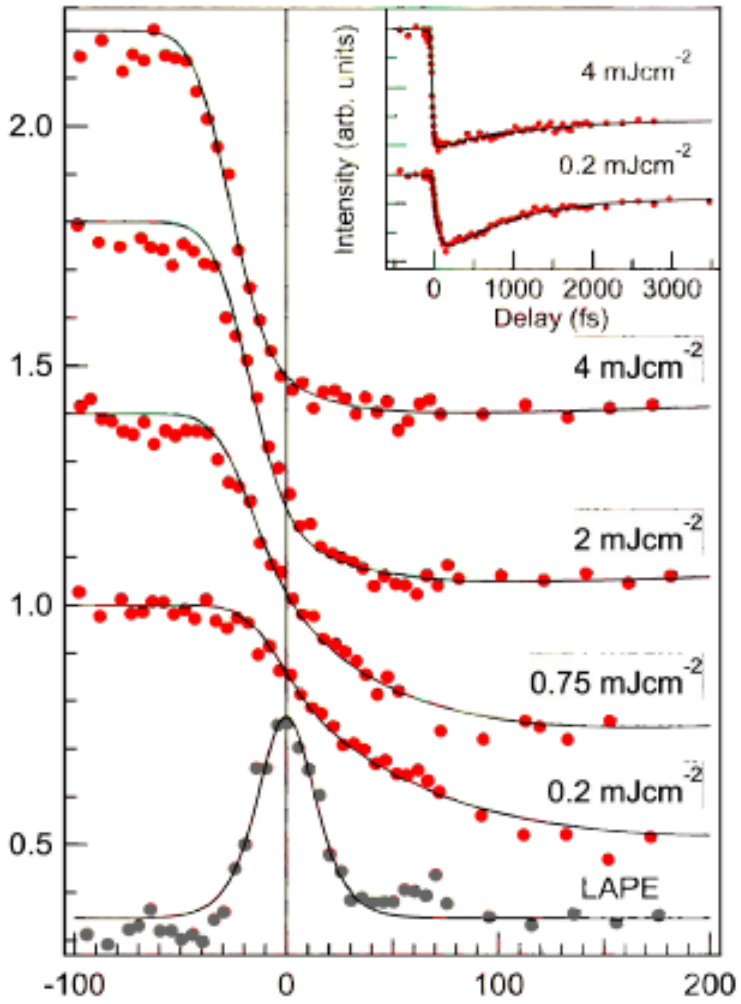




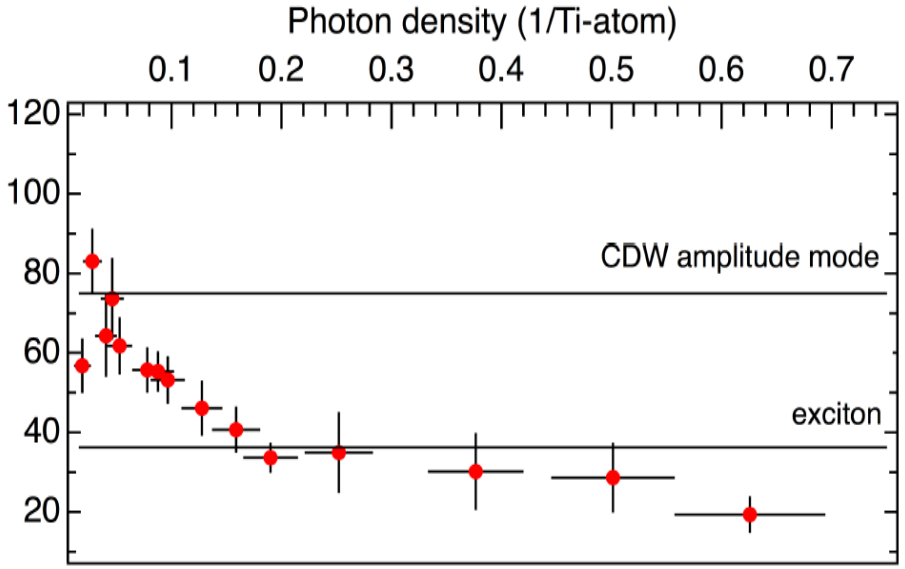
1T-$TiSe_2$

Photoinduced breakdown of the structural distortion

T. Rohwer, Nature 2011



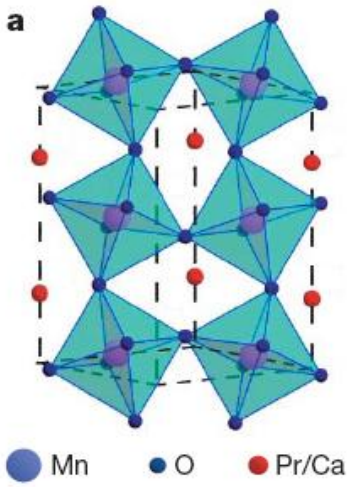
The melting of the ordered state takes place on a timescale that depends on the laser fluence

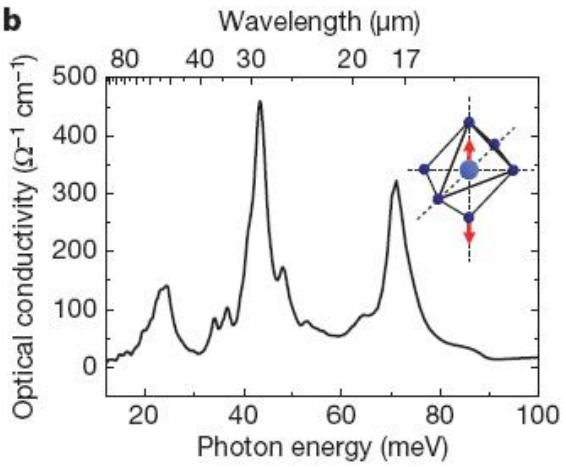


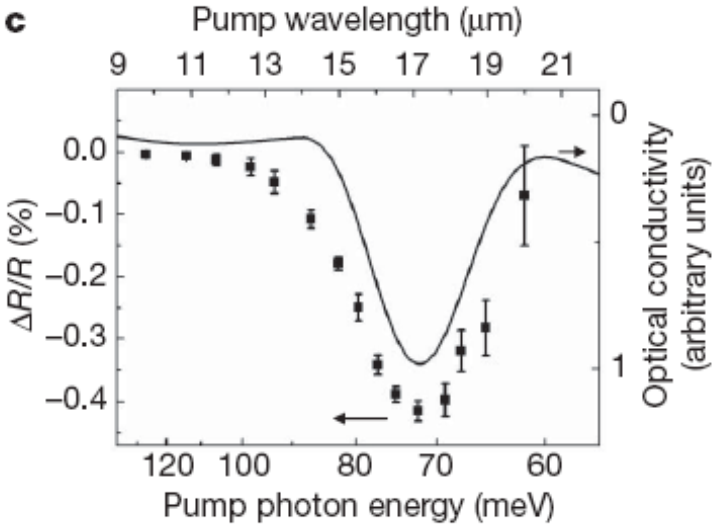
M. Rini, Nature 2007

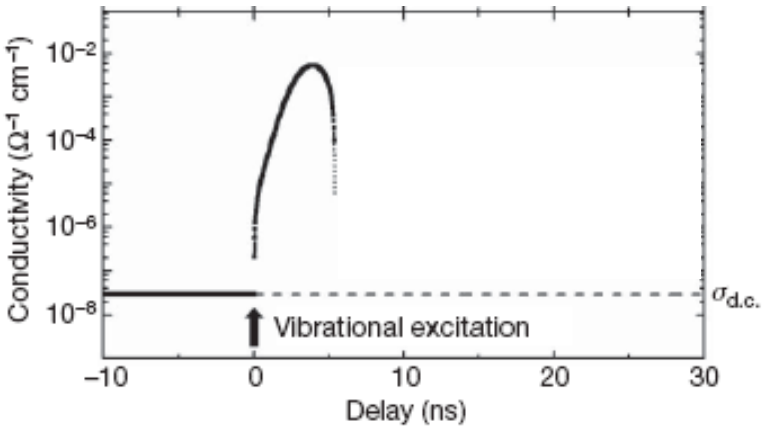
$Pr_{0.7}Ca_{0.3}MnO_3$

Phonon pumping

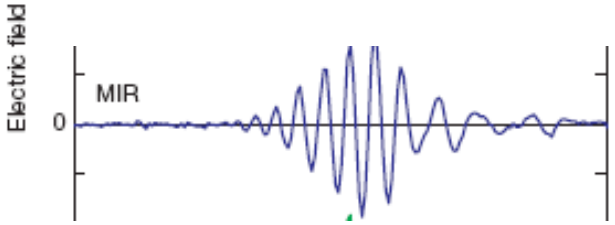








Phase control



Quiz Section

Is La_2CuO_4 expected to be an insulator or a metal ?

La_2CuO_4:

O: $[He]2s^2 2p^4$

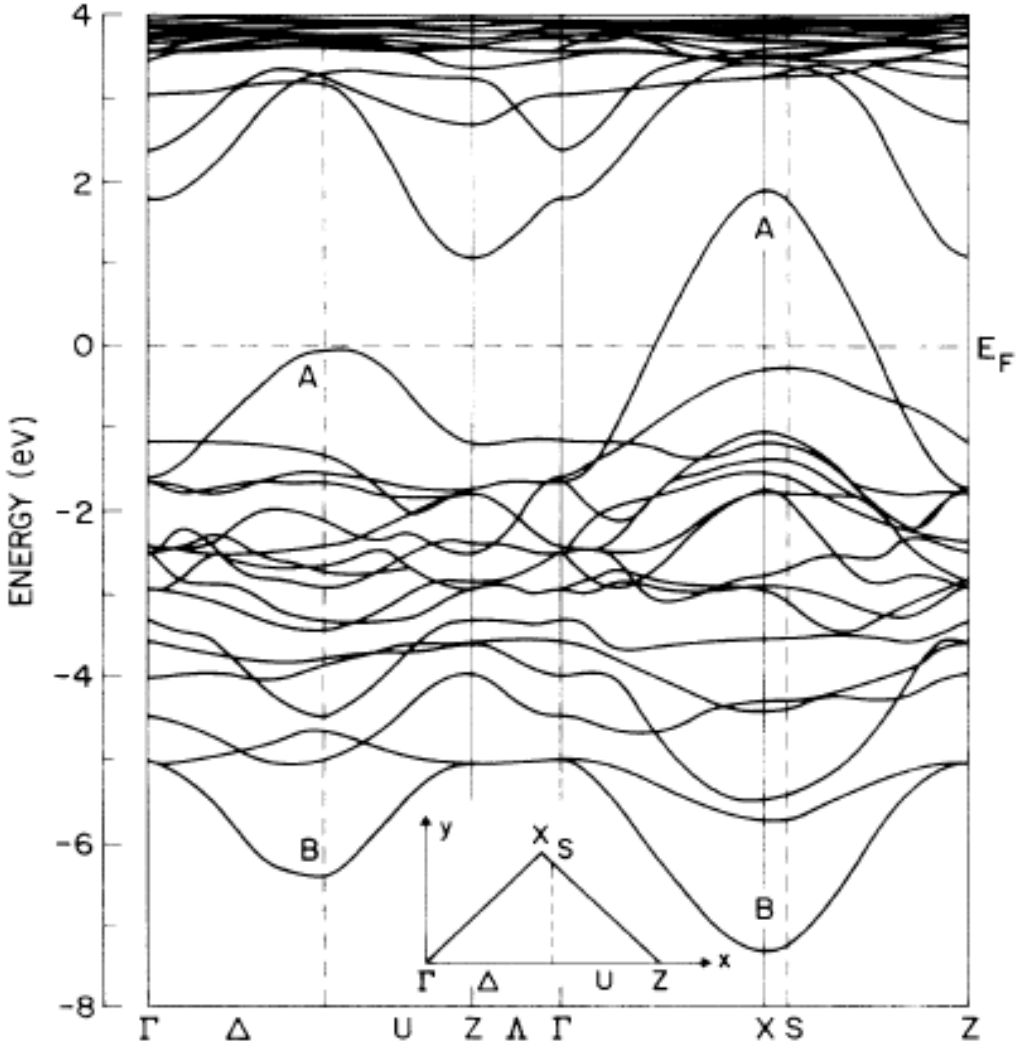
La: $[Xe]6s^2 5d^1$
Cu: $[Ar]4s^1 3d^{10}$

$-2*4+3*2 = -2$

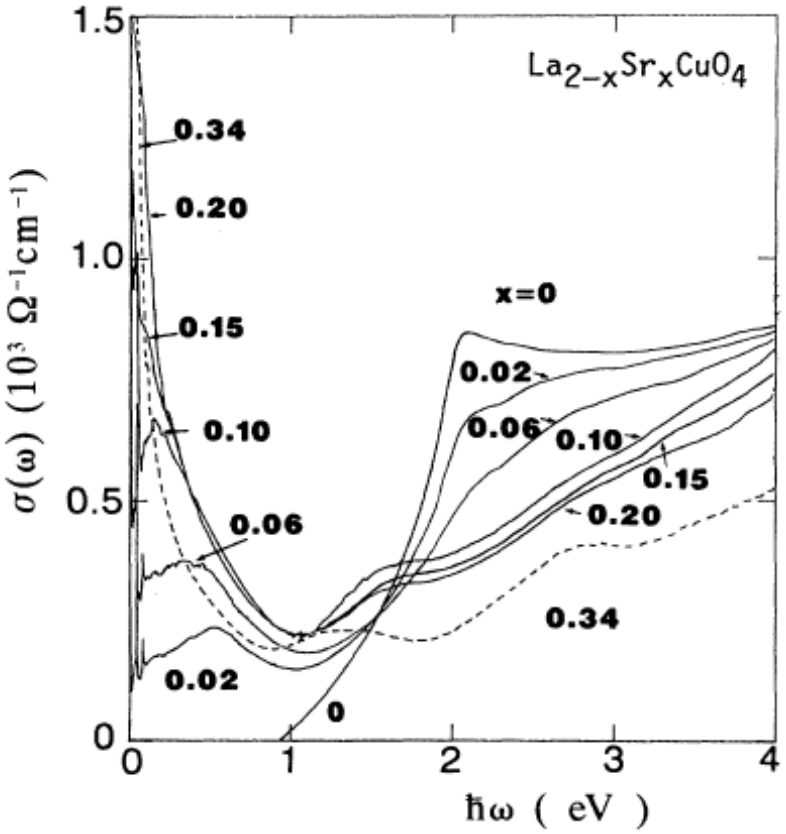
Cu^{2+} : $[Ar]3d^9$

Odd number of electrons
Half filled band

LDA band structure of La_2CuO_4

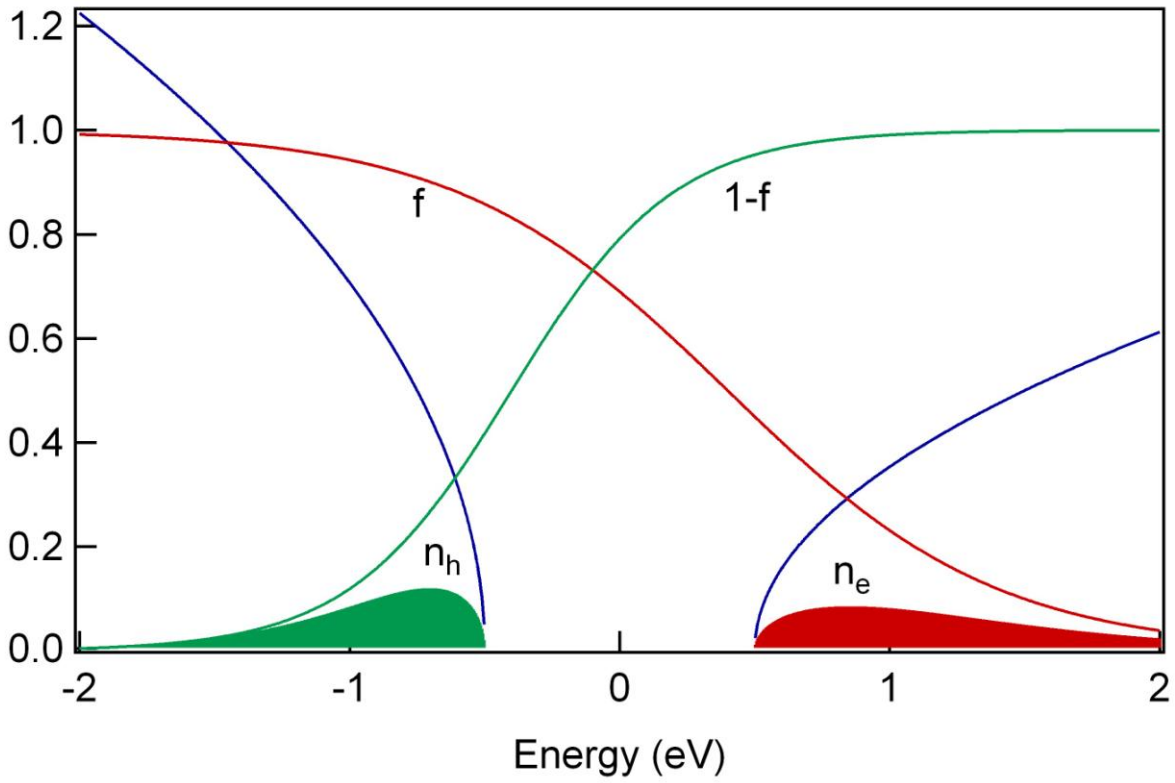


La_2CuO_4 is a Mott insulator

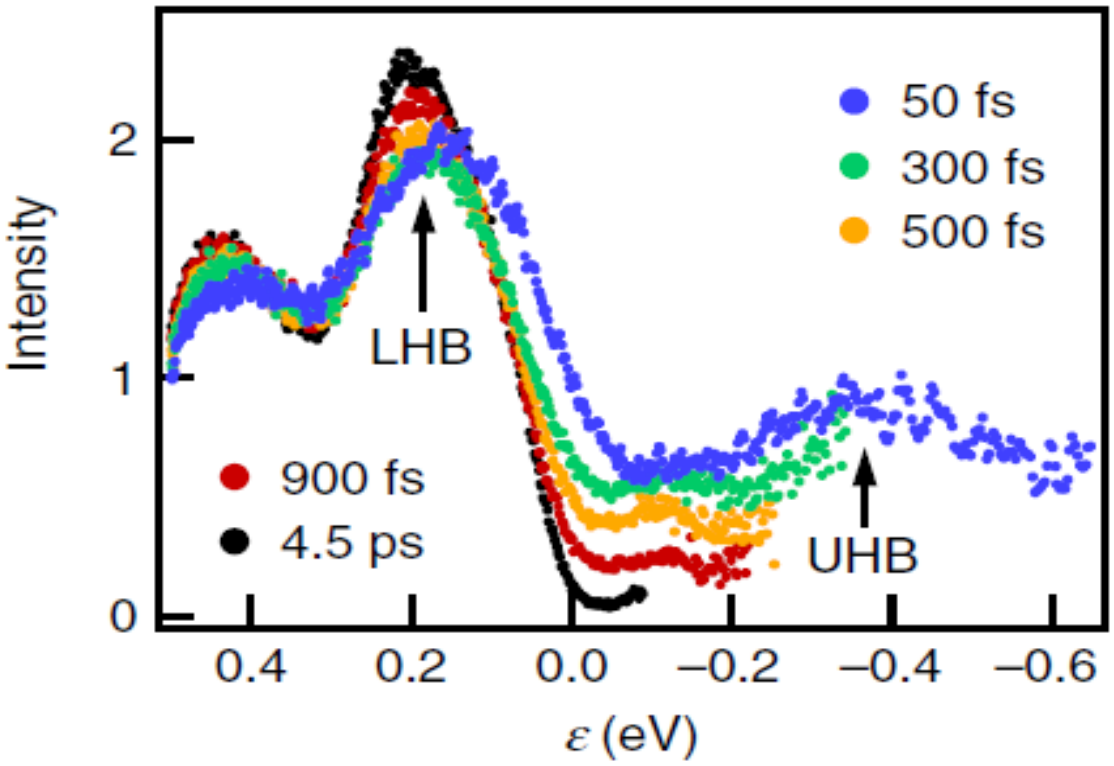


Does photoexcited silicon conduce electricity?
Is photoexcited silicon a metal?

Photoexcited Silicon is a conductor but not a metal?

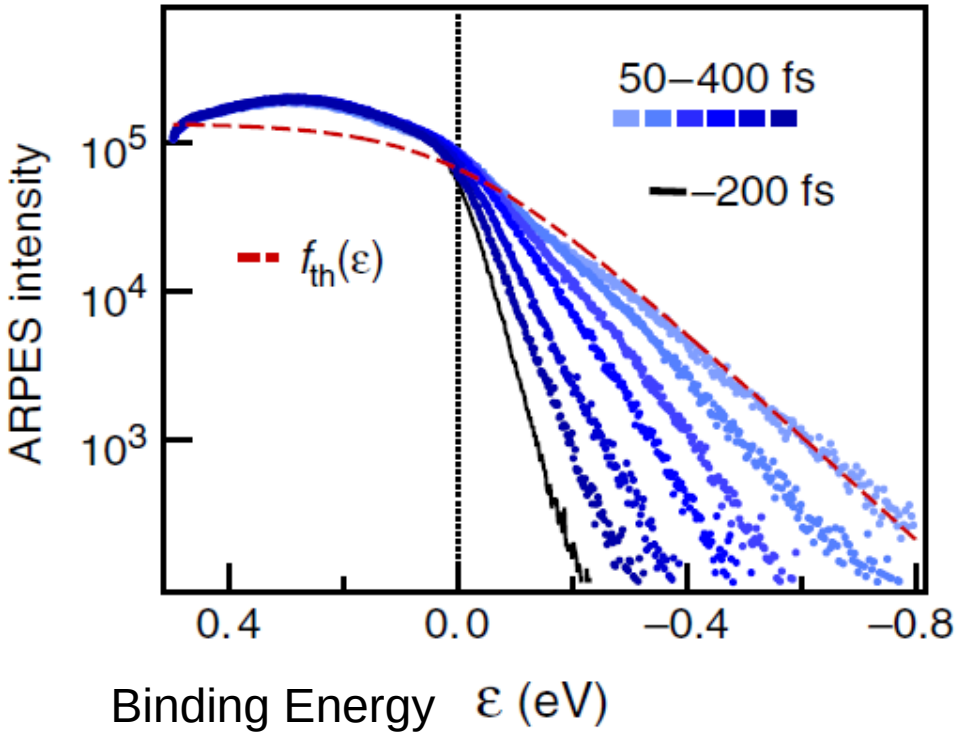


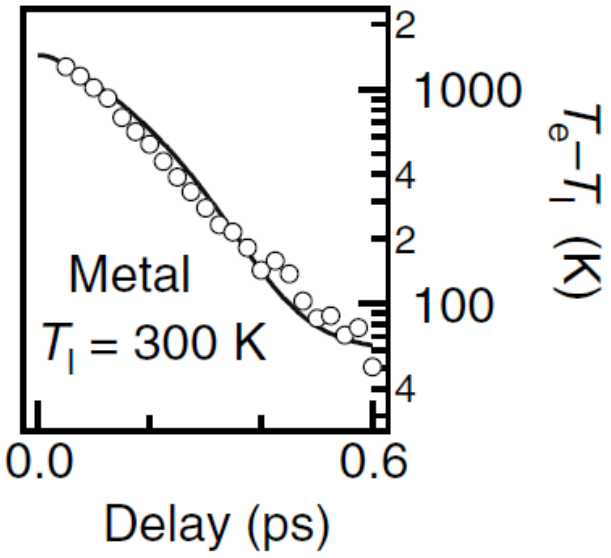
Insulator to metal transition



We indicate C_e and C_l the electronic and lattice specific heat of a metal. Which is the typical ratio C_e / C_l at room temperature?

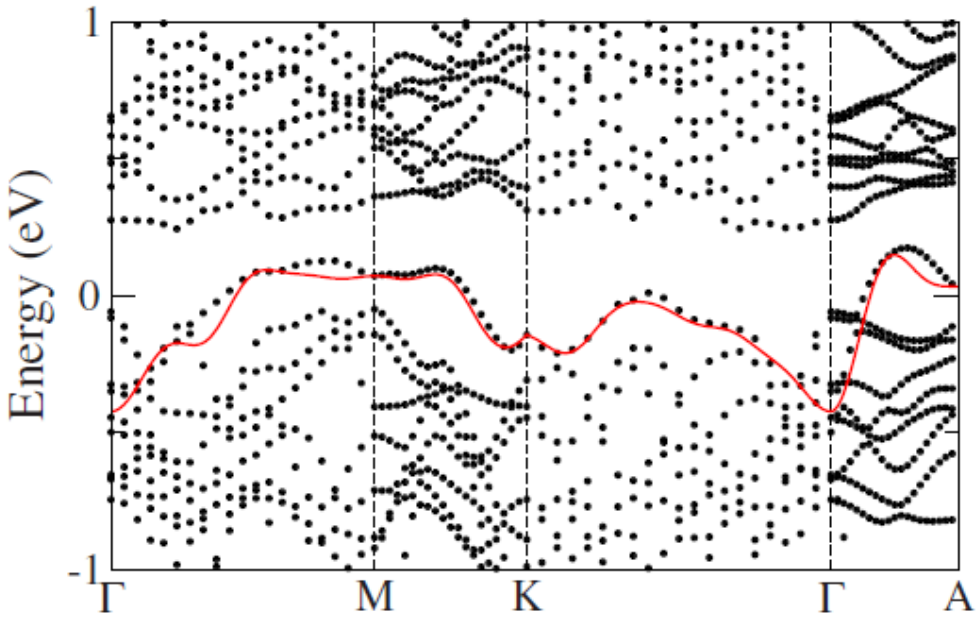
$E_T - E_0$: ~ kT for lattice ~$(kT)^2/W$ for electrons $C_e / C_l \sim kT/W$





On which timescale one expects a breakdown of the Mott insulating phase?

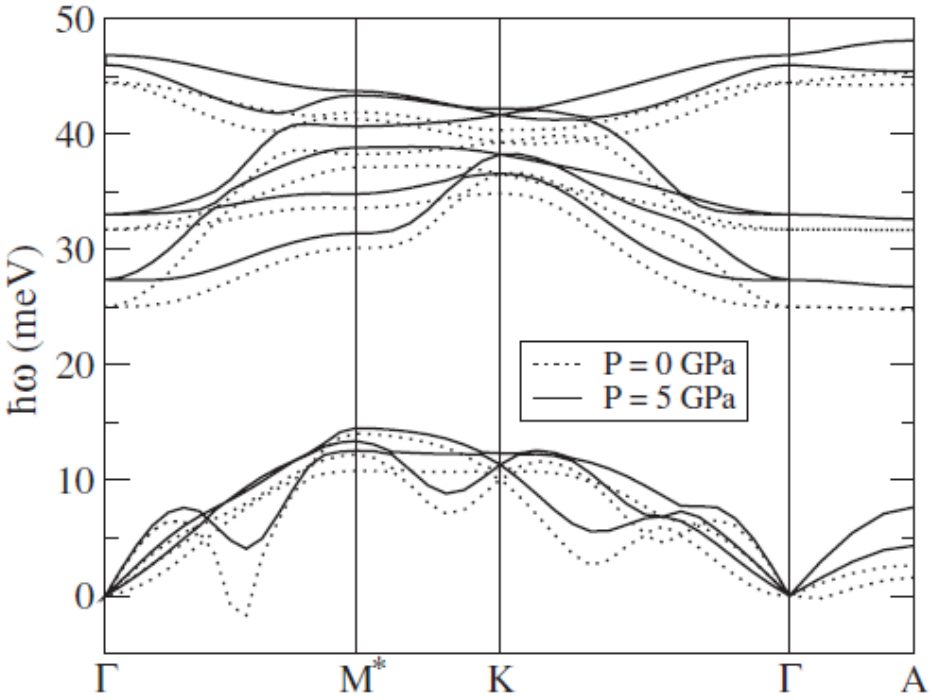
The typical timescale of the electronic motion is ~ħ/W



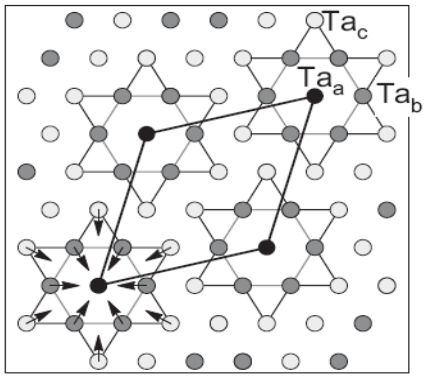
~10 fs in TaS_2

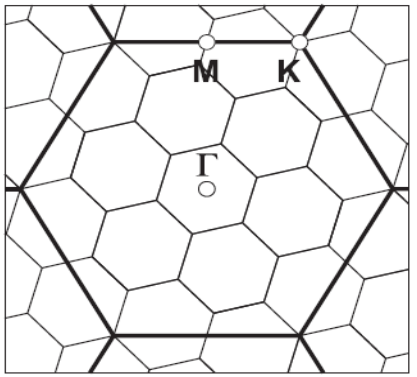
A phonon mode is located at the Brillouin zone boundary of the undistorted phase. Could we coherently excite it in the distorted phase? Under which conditions?

It must be a zone center phonon of the reconstructed BZ.



1*T*-TaS_2

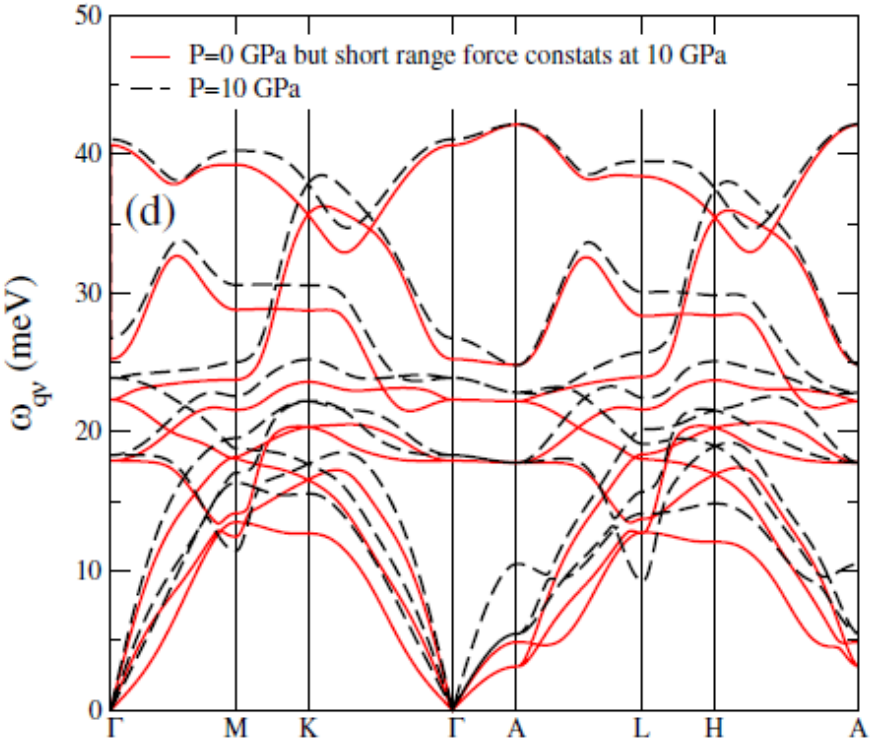


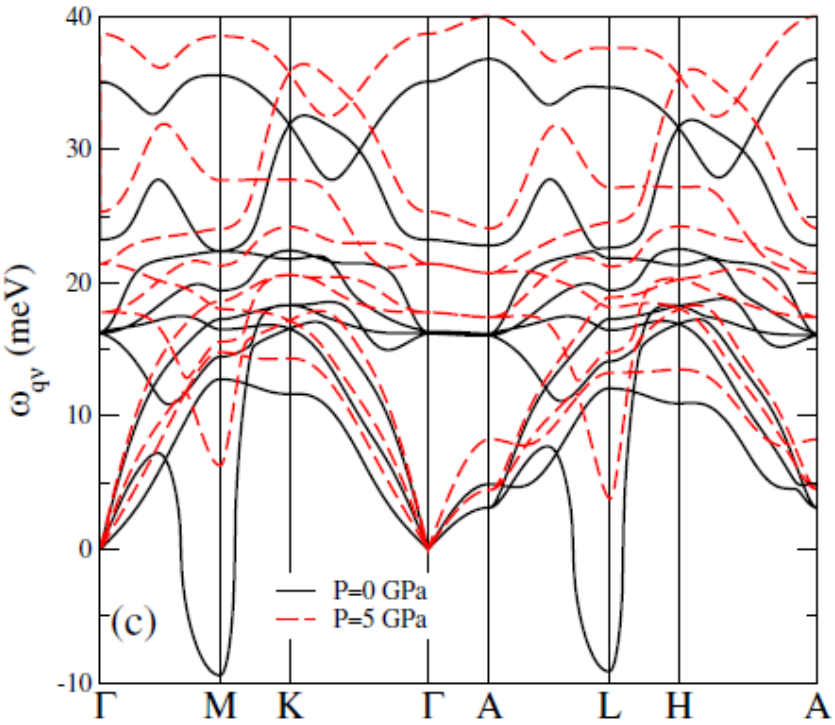


On which timescale one expects a photoinduced transition of the lattice structure?

Period of the soft phonon mode divided by 4

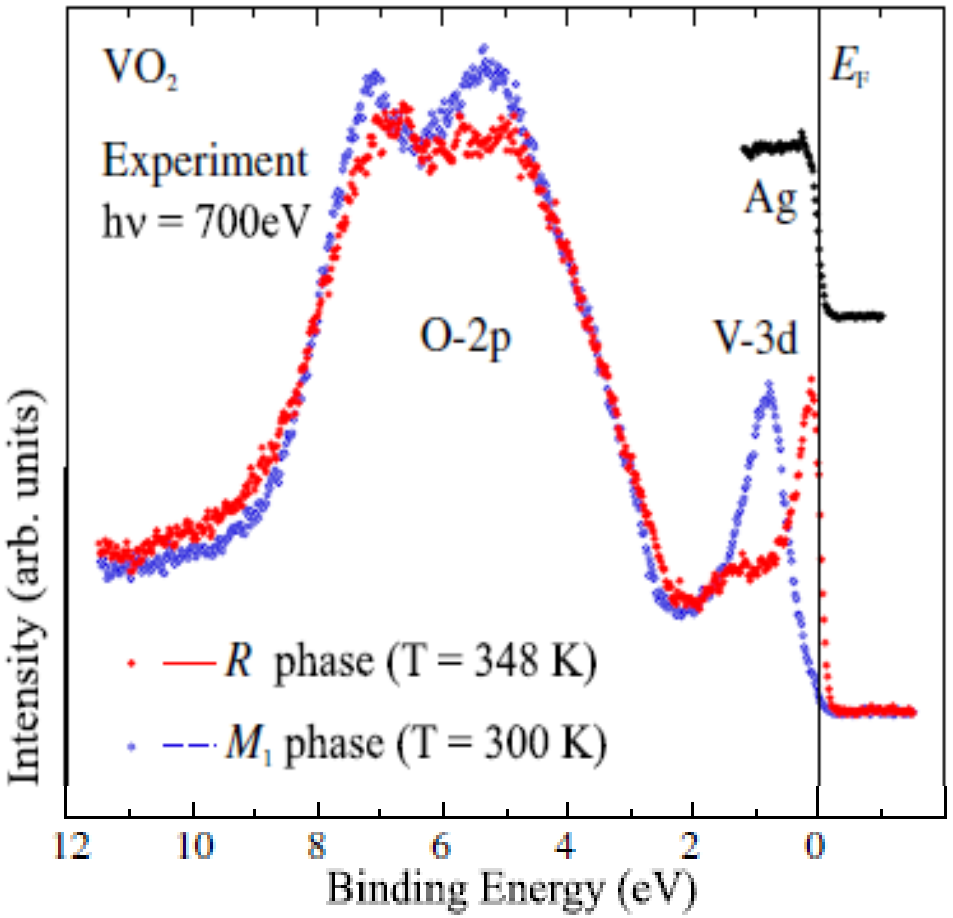
90 fs in $TiSe_2$

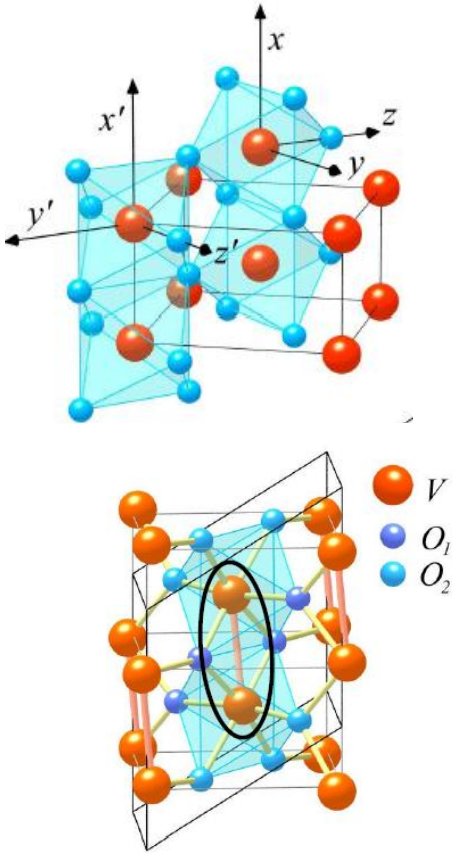




Metal insulator transition in VO_2

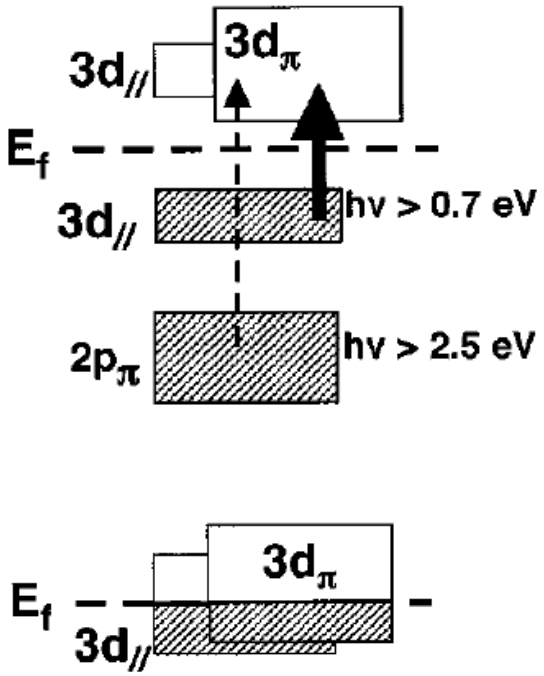
T. C. Koethe, PRL 2006



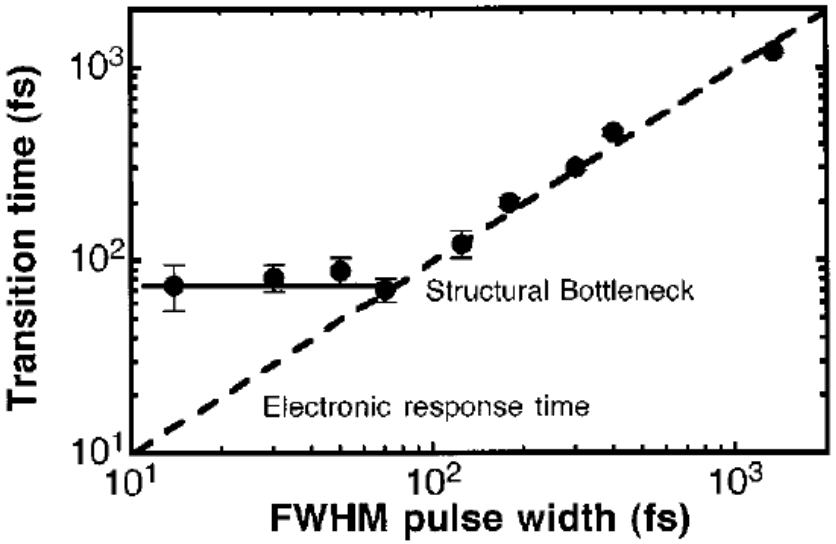


Is the structural transition an effect of the metal-insulator transition ?

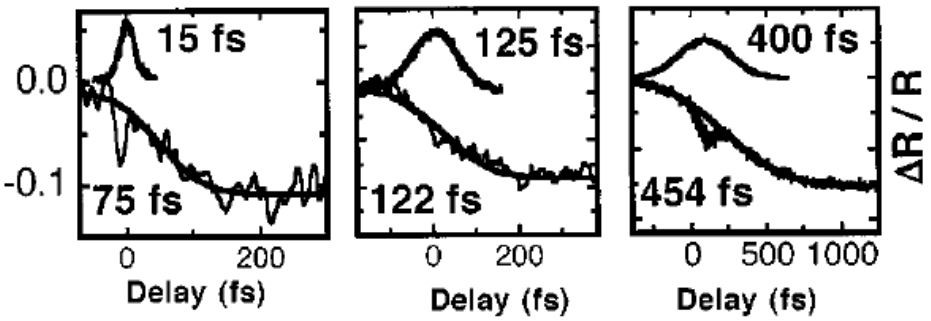
It is not possible to reach a metallic phase without changing the structure



A. Cavalleri, Phys. Rev. B 2004



Threshold fluence corresponding to the latent heat



It has been shown that valence band spectra acquired at high photon energy (1 keV) may differ subtantially from the ones acquired in the range of 20-100 eV.

Could you suggest possible reasons of it ?

Matrix elements

Surface sensitivity

Maiti, Phys. Rev. Lett. 1999

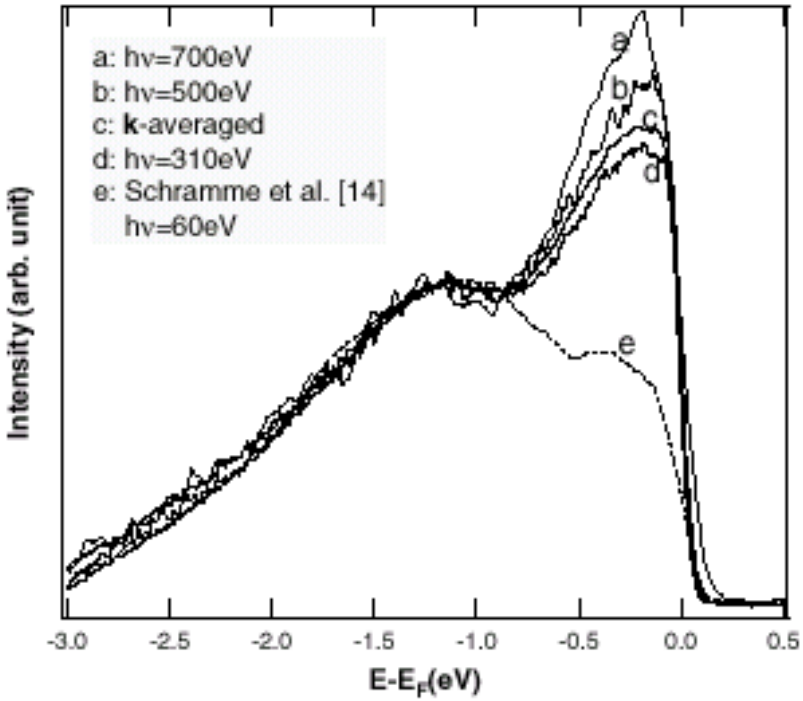


FIG. 2. PES spectra taken with various $h\nu$, the largest of which yields the greatest bulk sensitivity.