Tailoring the photon beam for non-linear spectroscopy in solids

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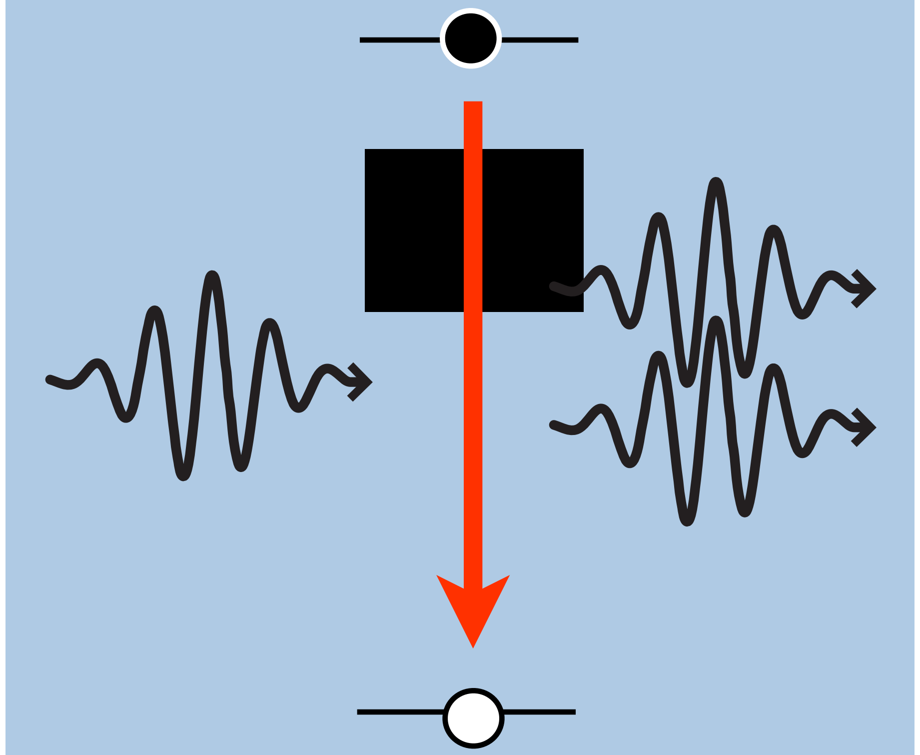
Non-Linear Processes

Absorption

$$\left\langle e, N-1|\vec{d}|g, N\right\rangle$$

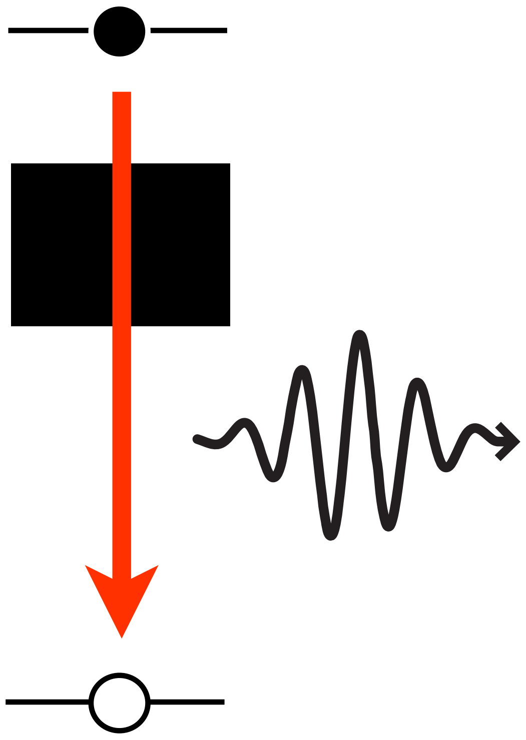
$$P_{abs} = N\sigma\rho_g d$$



Stimulated Emission

$$\left\langle g, N|\vec{d}|e, N-1\right\rangle$$

$$P_{stim} = N\sigma\rho_e d$$



Spontaneous Emission

$$\left\langle g, 1|\vec{d}|e, 0\right\rangle$$

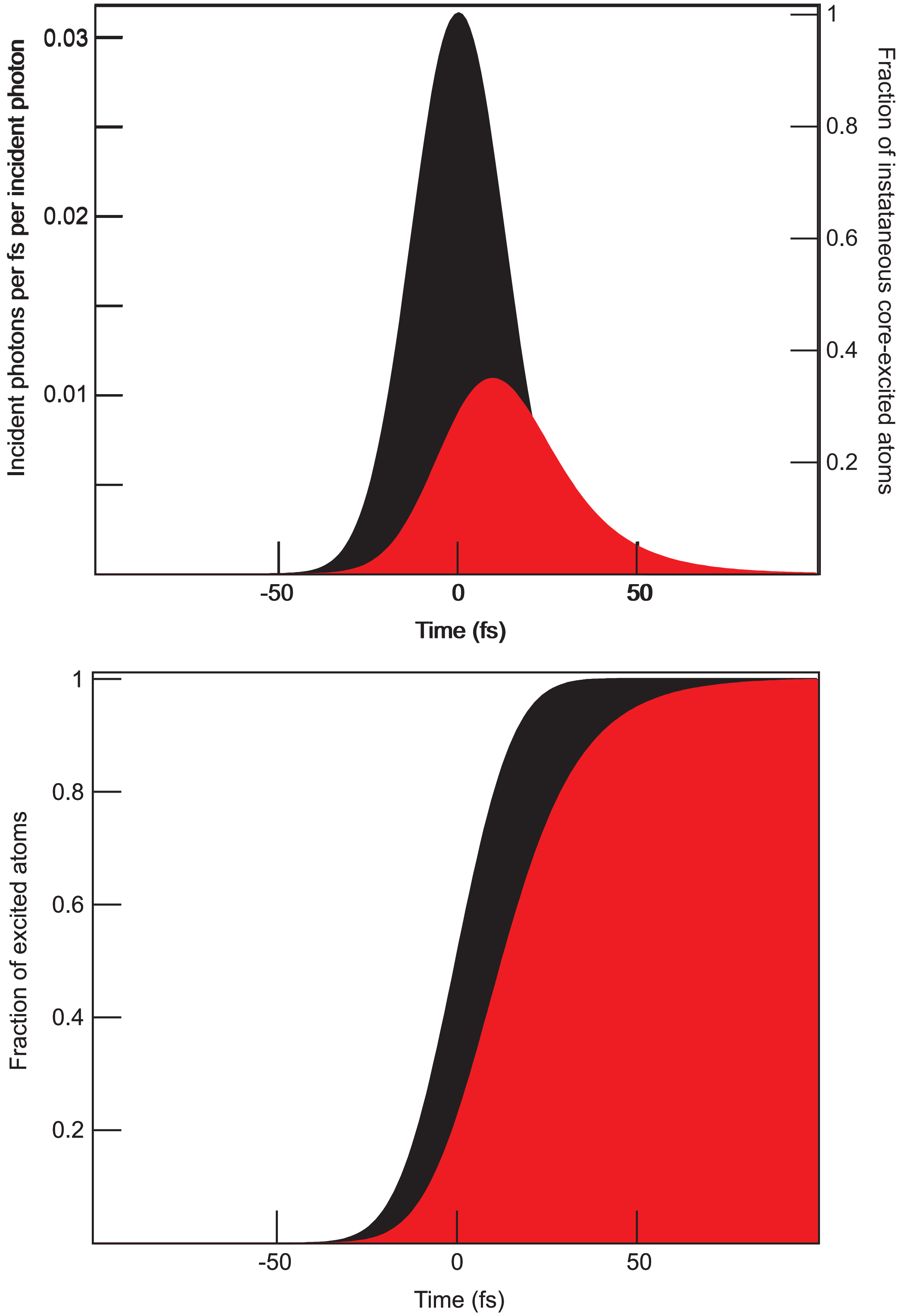
$$P_{spon} = N_{vac}\sigma\rho_e d$$

Tailoring the photon beam for non-linear spectroscopy in solids

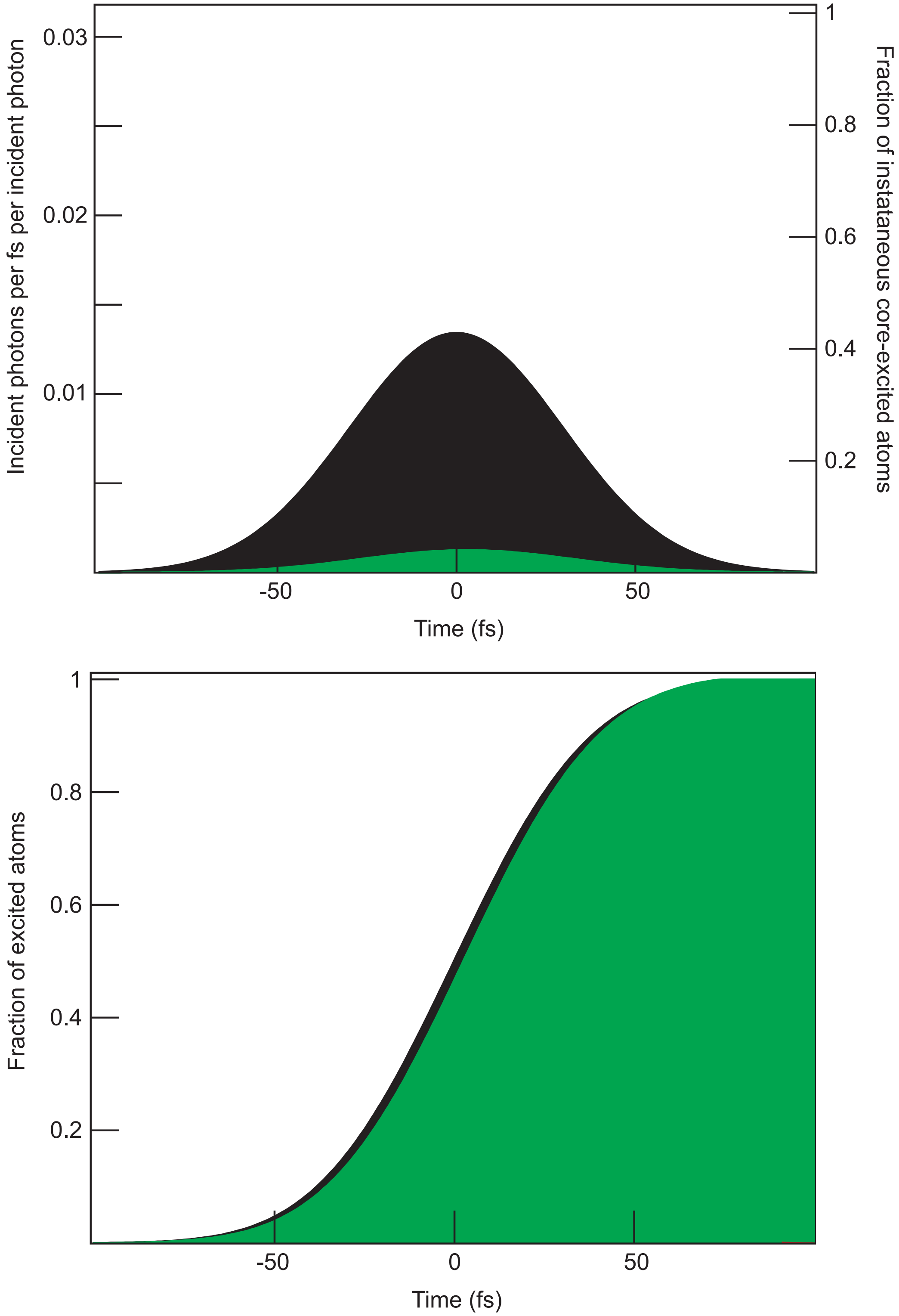
- Needed Photon Parameters
- How to get them from a SASE source
- Possible Experiments

Interaction with the core-excited state

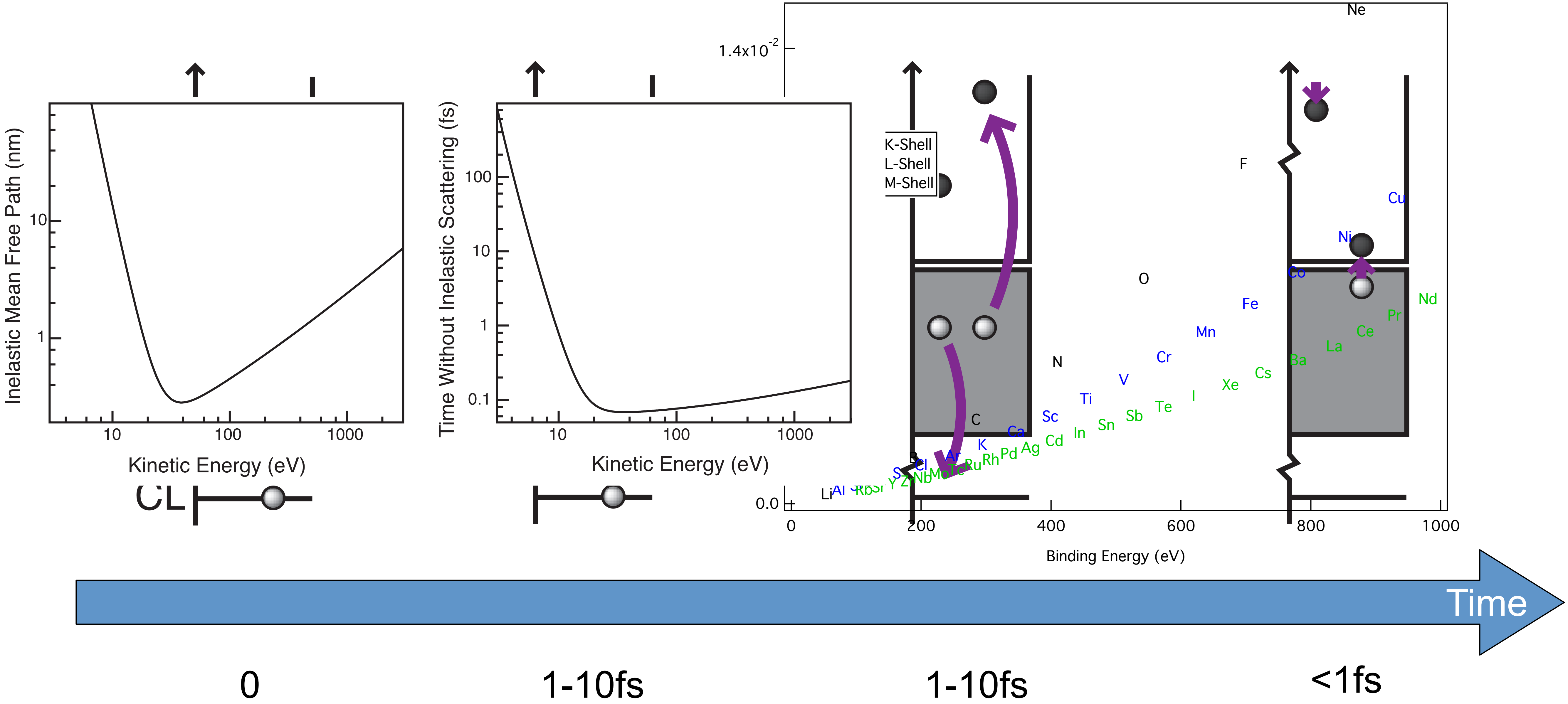
30fs pulse
15fs core hole lifetime



70fs pulse
3fs core hole lifetime



Processes after core excitation

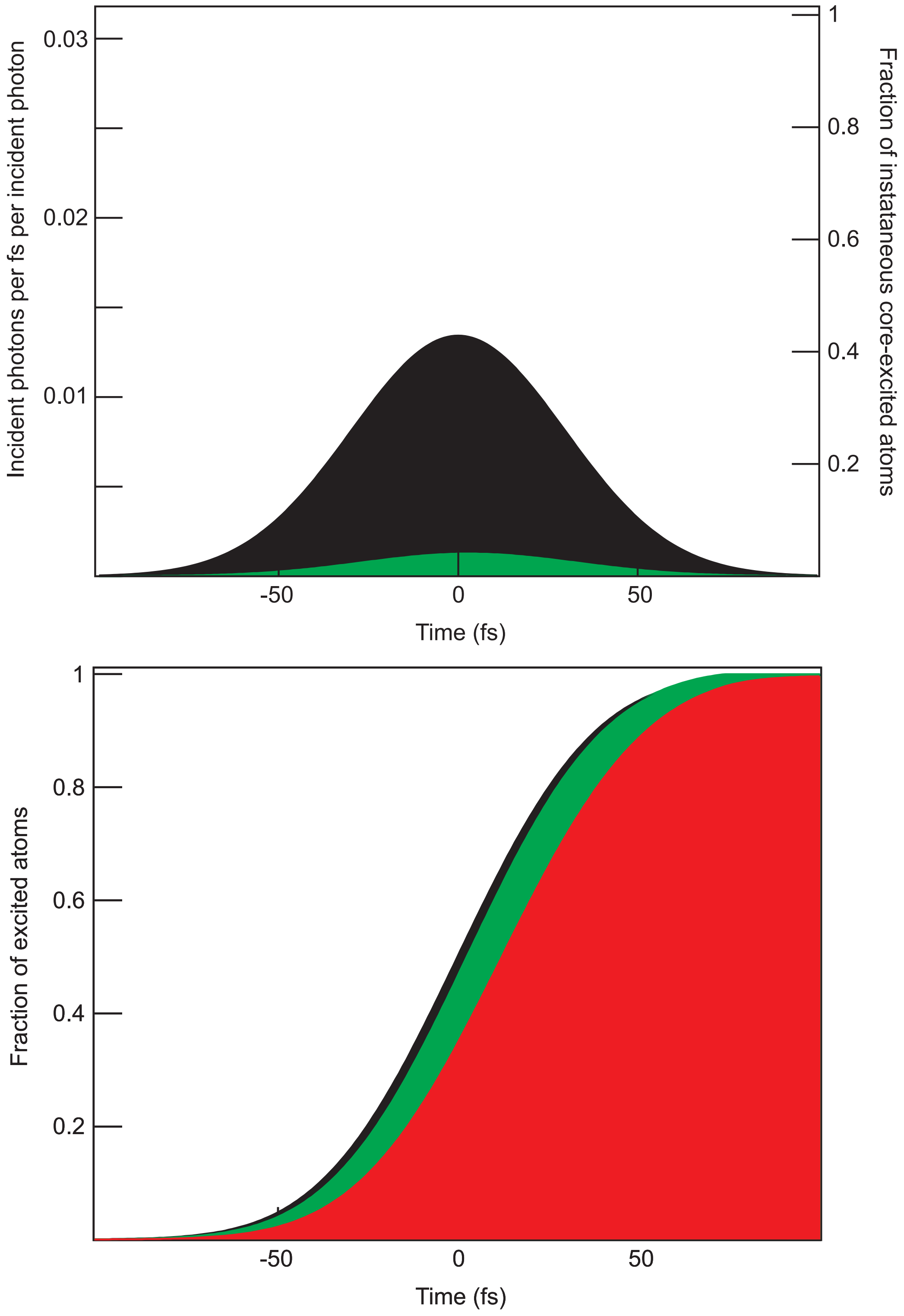


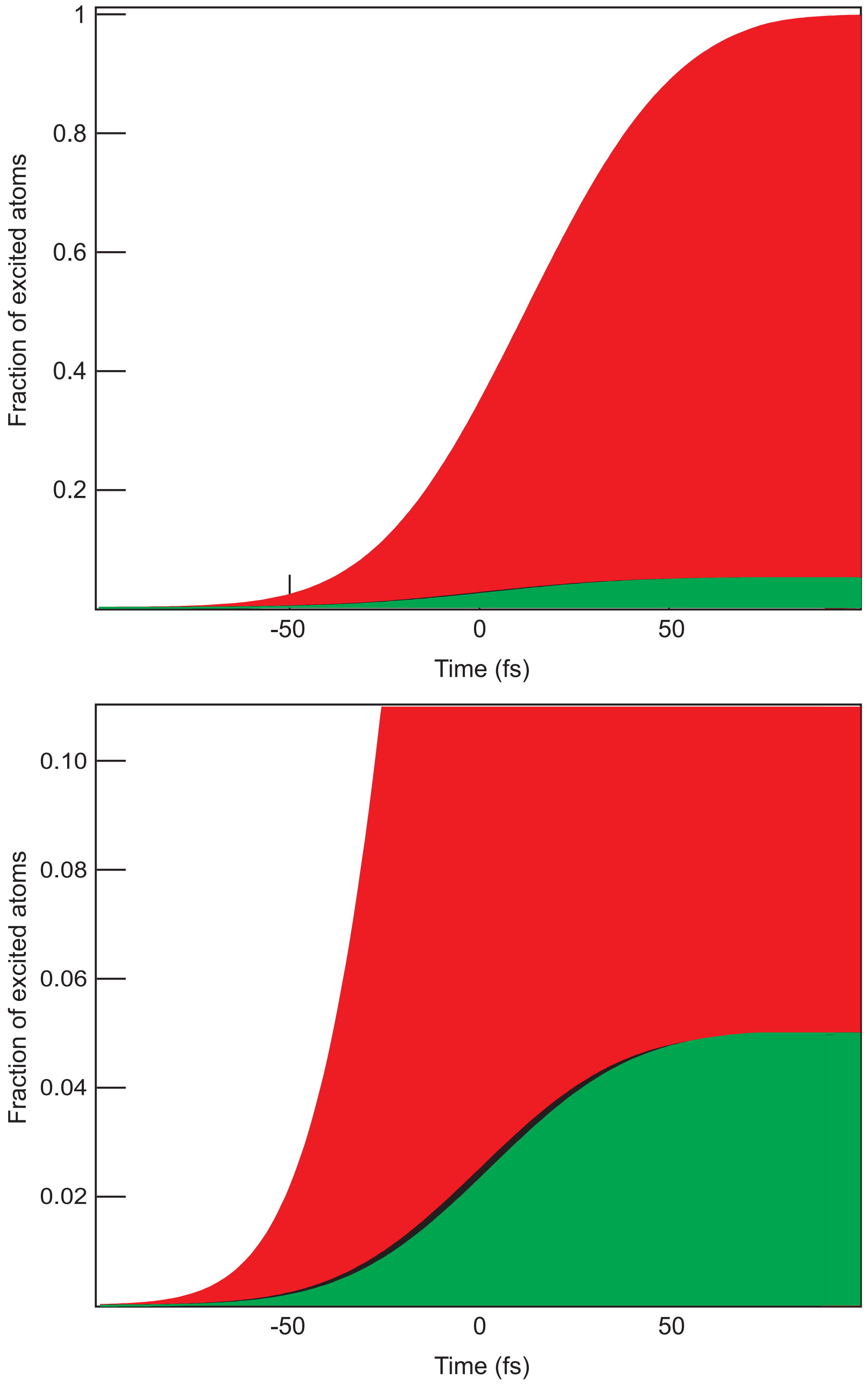
70fs pulse

3fs core hole lifetime

10fs „Energy loss time“ of Auger electron

20 excitations per Auger electron





Short pulses are absolutely crucial:

- to enhance the number of core holes at a given moment
- to avoid probing „damage“ (valence excitations / plasma)
- to have core holes only in an „unexcited“ sample

Fourier-Transform limit:
(Gaussian pulses, FWHM)

$$\Delta E \cdot \Delta t \geq 0.44h \approx 1.8fs \cdot eV$$

Monochromator resolution:
(N illuminated lines)

$$\frac{E}{\Delta E} \leq N \qquad \Delta E \geq \frac{E}{N}$$

Pulse lengthening:
(N illuminated lines)

$$N \cdot \lambda$$

$$\Delta t = N \cdot \frac{\lambda}{c} = \frac{N \cdot h}{E}$$

$$\Delta E \cdot \Delta t \geq \frac{E}{N} \cdot \frac{N \cdot h}{E} = h$$

Monochromatization keeps short pulses,

when a small number of lines (about 1000) is illuminated

-> extremely low line density

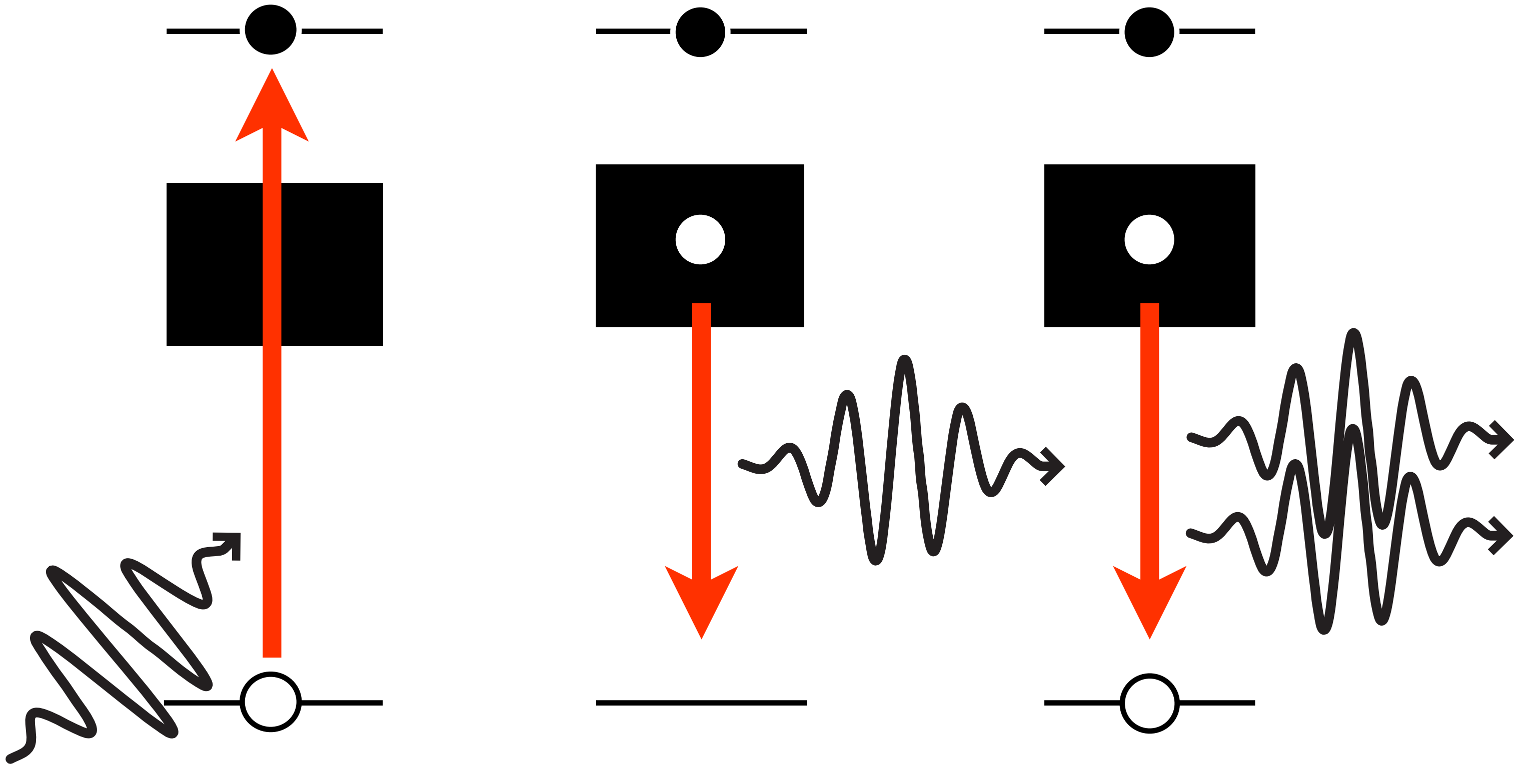
Split-and-Delay works in energy-time phase-space

-> ideally independent

-> XBSD behind monochromator, separate slits for each arm

-> independent energy / time content

-> spatial overlap of both beams



(Usually)
Unoccupied

Occupied

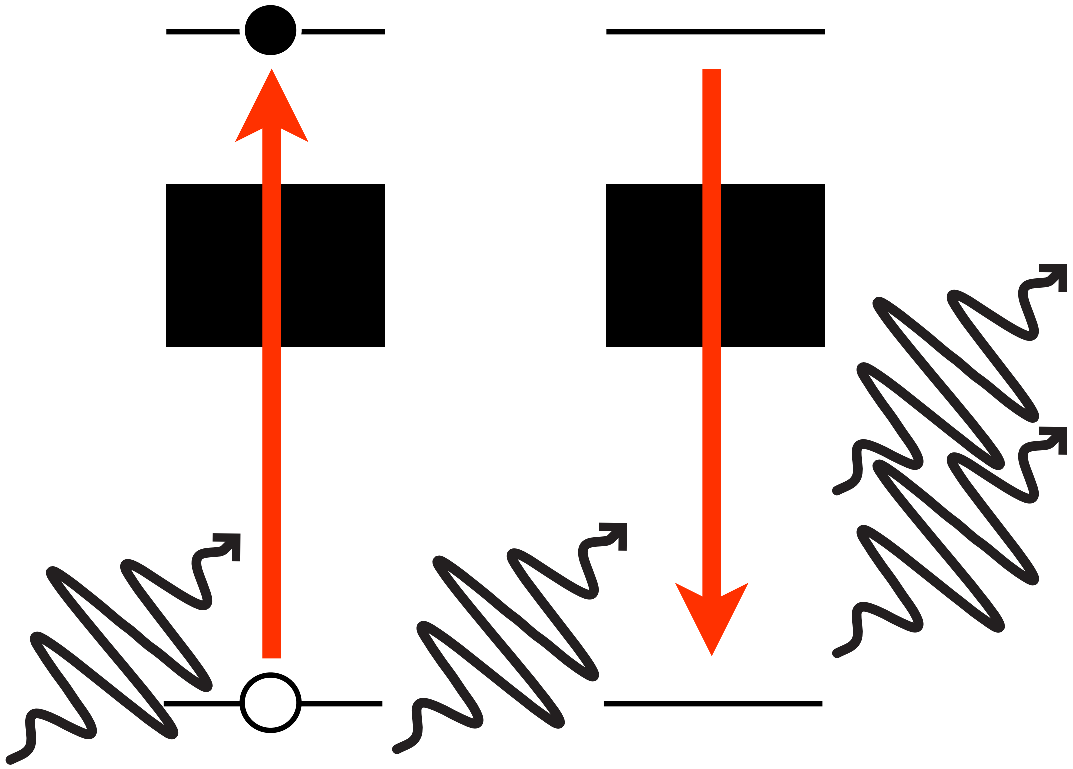
Core level

Fluorescence
stimulates itself:

Amplified
spontaneous
emission (ASE)

Rohringer et al. Nature 481, 488–491 (2012)
Beye et al. Nature 501, 191–194 (2013)

-gain on top of small signal
-small „background“
-high possible gain

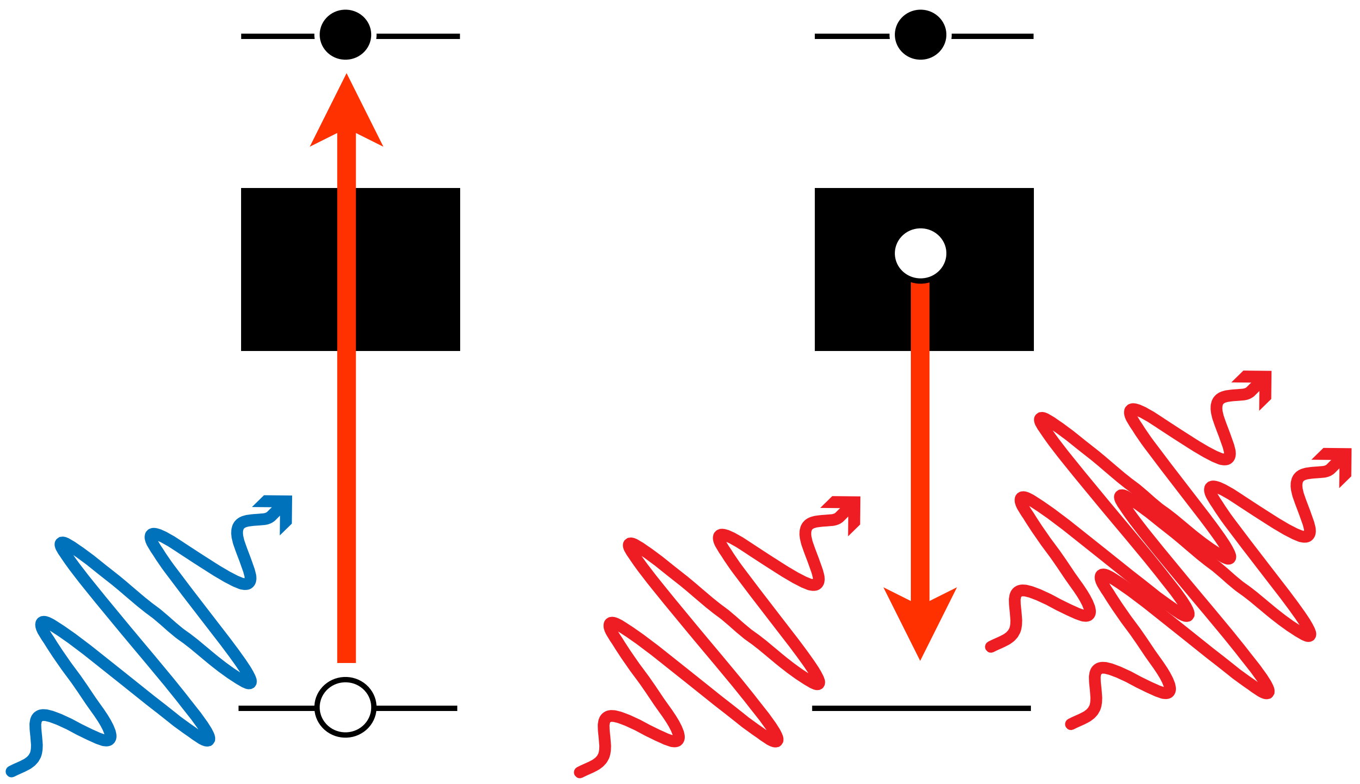


(Usually)
Unoccupied

Occupied

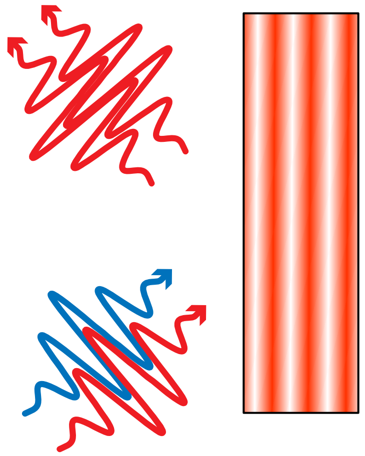
Core level

Same Channel
not observable!
(satured absorption)



Weninger et al. PRL 111, 233902 (2013)

- -gain on top of big signal (FEL beam)
- -maximum gain is splitting ratio
- -long overlap of „dump“ field and „pump“ excited volume restricts geometry
- -q-transfer limited through angular separation of beams
- -reflectivity from multilayer samples can enable q-transfer



	Day 0	Nice to have
Experimental Techniques	Anything with spectral resolution before and behind sample	Same shot-to-shot
Source Properties		
Energy Range	C, N, O + transition metal edges (250-1000eV)	full SASE 3 range
Pulse duration	some femtoseconds	<100 as
Bandwidth	<0.5 eV	Fourier limited, tunable, < 0.05 eV
Device Properties		
Maximum Temporal Delay	twice the pulse duration	some ps
Pulse intensity ratio	4:1 - 1:1 tunable	tunable orders of magnitude
2 Colors	crucial, tunable	also different harmonics
Symmetric delay around t=0	yes, if something else is asymmetric	yes
Spatial separation behind sample	tunable angle for q-transfer / separation on detector	
Suggestion 1	Split after mono to manipulate energy content independently	
Suggestion 2	Measure spectrum / intensity in each arm independently	

Thank you for your attention!