Ultrafast Structural Dynamics in Strongly Correlated Electron Systems: Timing Specifications

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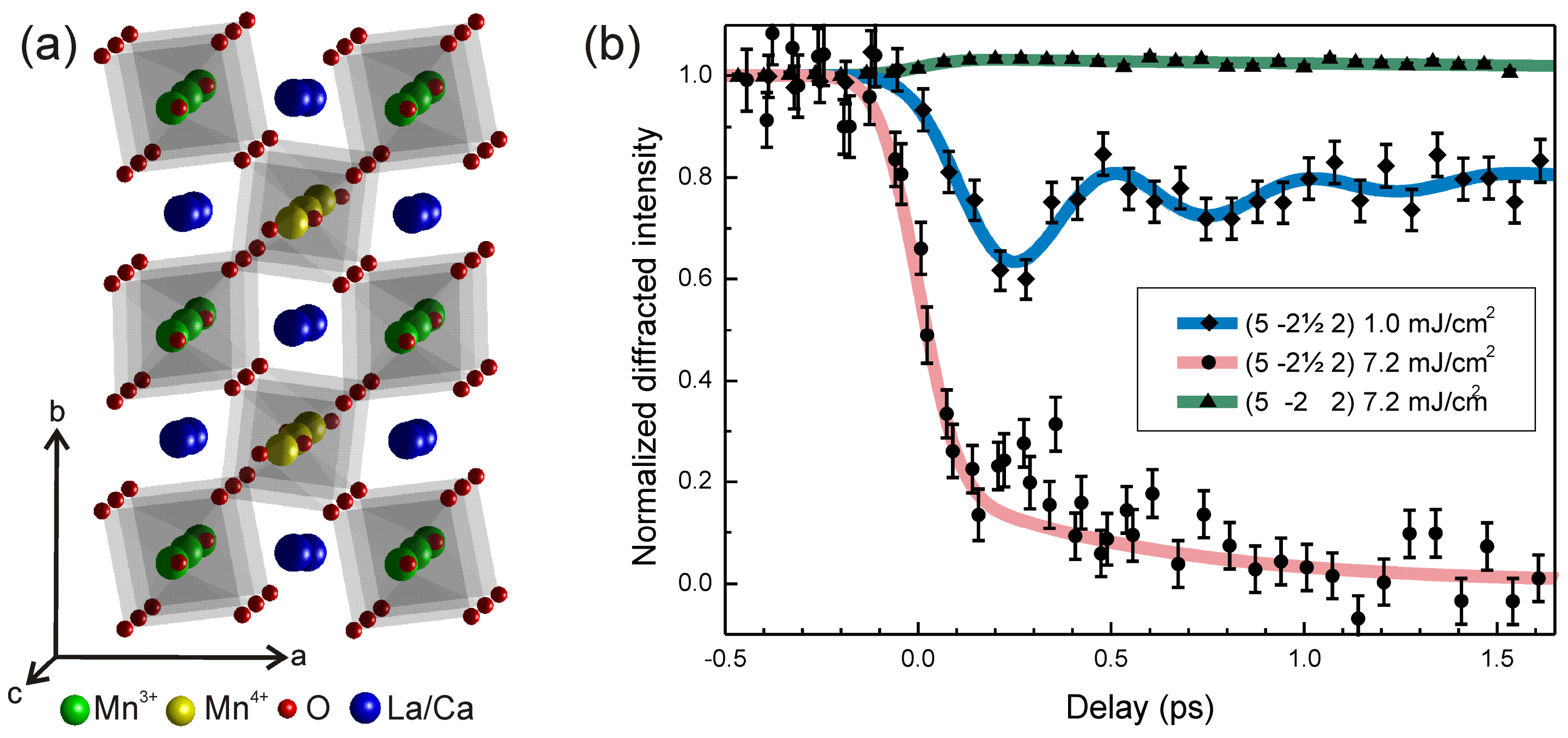
Science

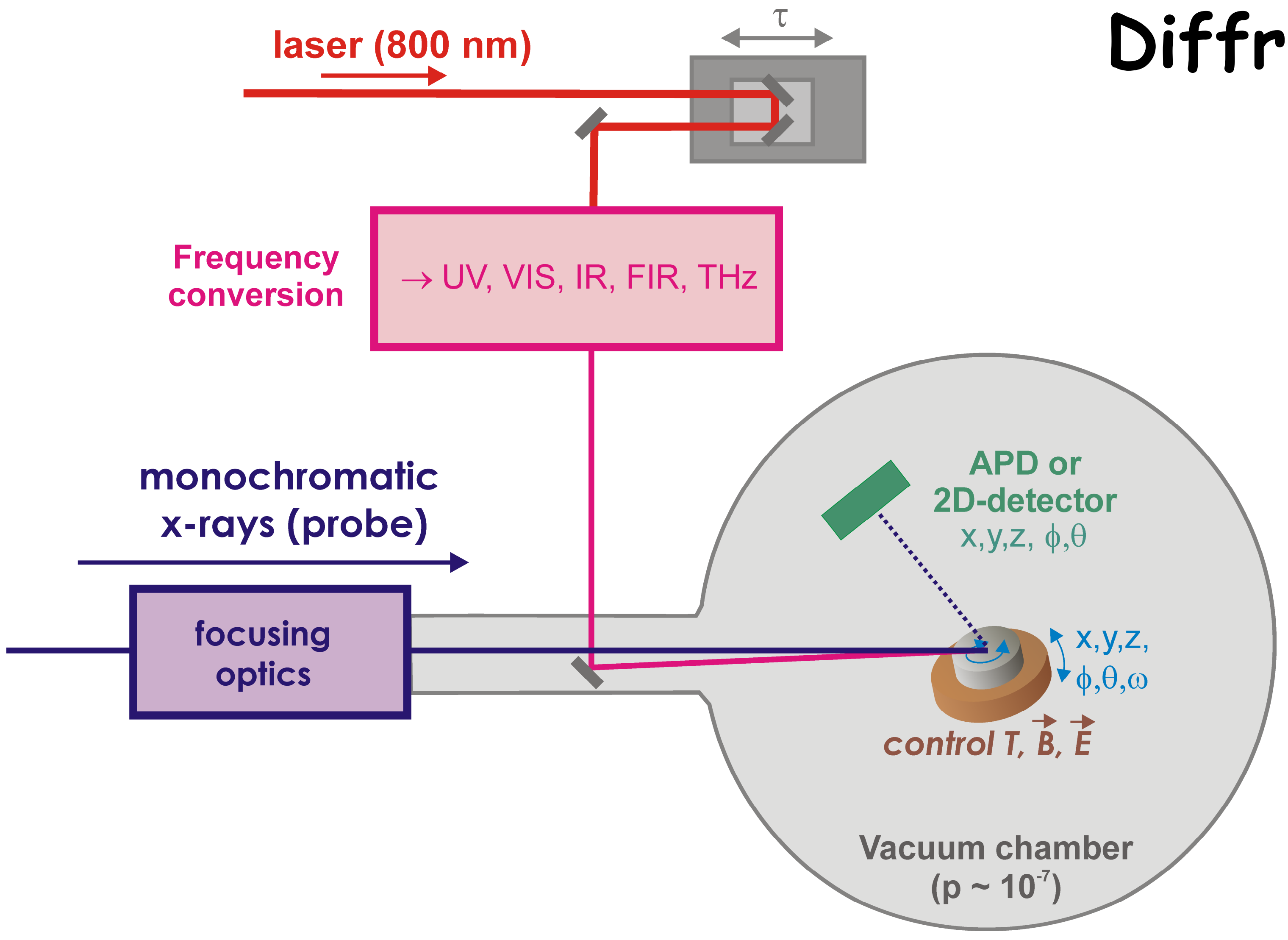
Goal: Understand complex interactions between lattice and electronic degrees of freedom in strongly correlated electron systems which often lead to exotic electronic and magnetic properties, *such as High-Tc superconductivity, colossal magnetoresistance, multiferroicity, ...*

Experiments at FEMTO on a manganite [1] and on a charge density wave system [2] demonstrated that photo-doping can induce non-thermal phase transitions as evidenced by the disappearance of a superlattice reflection. Initial dynamics are significantly faster than the FEMTO time resolution of 200 fs. Optical data on a magneto-resistive manganite indicate [3] relevant dynamics up to 30 THz.

To resolve these dynamics in greater detail and to disentangle the atomic motions within the unit cell requires measurement of as many Bragg reflections as possible with sufficient time resolution. Only an FEL can provide the required time resolution of < 20 fs with sufficient flux to efficiently measure the relevant but often weak superlattice peaks.

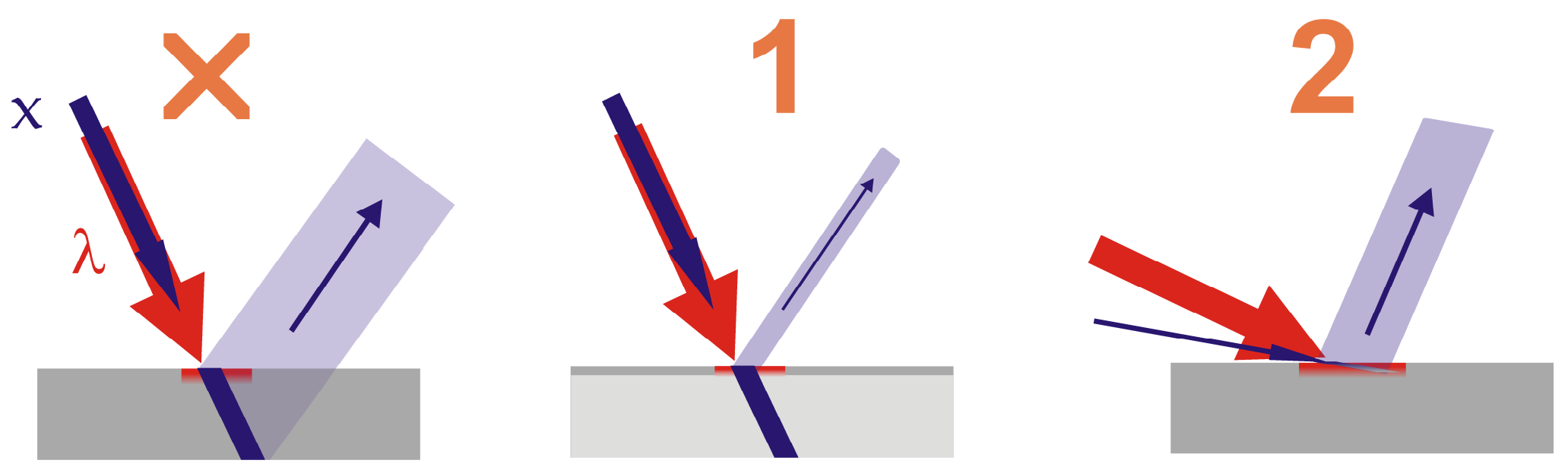


Diffraction setup



Sample environment:
Temperature: 5 – 500 K
Static magnetic field: 0 - 10 T
Static electric field: 0 – 5 kV

Matching X-ray probe depth to excitation depth (often < 50 nm):
- solution 1: use thin films
- solution 2: use grazing incidence diffraction to match probe to excitation depth, requires severe focusing of incoming x-rays in the plane of incidence [4].



Requirements at SwissFEL for pump-probe diffraction experiments

X-rays

Parameter		Unit	Requirement	Motivation /Remarks
Energy		keV	2 – 12	High q-space coverage; select edges for resonant diffraction
	stability	%	0.002	Monochromator required
Bandwidth		%	0.002	Monochromator required
	stability	% bw	< 10	
Beam position	stability	µm	< 1	
Beam size		µm	1 – 100	Grazing incidence requires ~1 µm focus (horizontal dimension only)
Photons per pulse			$10^8 – 10^{12}$	Attenuator needed to avoid sample damage
	stability		-	Shot-to-shot normalization with I_0
Pulse length		fs (rms)	2	Short pulse mode
	stability	%	10	
Pulse arrival time	stability	fs (rms)	< 5	With respect to pump laser
Beam parameter changes during experiment				
Energy	range	eV	±100	Required for resonant diffraction
	step	eV		Required for resonant diffraction
	scan	eV / sec	0.2	Required for resonant diffraction
Beam size			-	Only adjusted during setup
Pulse length			-	Only adjusted during setup
Beam geometry				
Beam slope	maximum	µrad	200	
Working distance	minimum	mm	500	UHV vacuum chamber; sample rotation, translation, cooling

Diagnostics

Parameter	Resolution	Range	Single shot
Intensity	10^{-3}	0 - 10^{12} ph	yes (after monochromator)
Beam position	1 µm	± 100 µm	yes
Beam width	5 um	0 - 300 µm	yes
Pulse duration (rms)	1 fs	0 - 200 fs	nice to have
Arrival time, coarse (rms)	200 fs	± 500 ps	yes
Arrival time, fine (rms)	2 fs	± 500 fs	yes
Mean energy	10^{-3}		no (since mono is used)
Energy spectral width	10^{-3}		no (since mono is used)
Longitudinal source point	1 m	± 20 m	no

Excitation pulse (derived from laser or THz accelerator)

Parameter		Unit	Requirement	Motivation /Remarks
Optical	wavelength	µm	0.25 – 2	
	pulse width	fs (FWHM)	< 20	Pulse shaping, multiple pump pulses
	jitter	fs (rms)	< 5	With respect to x-ray pulses
Far IR / THz multi cycle pulses	wavelength	µm (THz	15 – 150	
	pulse width	fs (FWHM)	500 – 20000	Amplitude phase stable
	jitter	fs (rms) % cycle period	5 10	Whichever is larger
	energy/pulse	µJ	> 10	
THz half cycle	frequency	THz	0.1 – 10	
	field strength	MV / cm	0.1 – 10	

[1] P. Beaud S. L. Johnson, E. Vorobeva, U. Staub, R. A. De Souza, C. J. Milne, Q. X. Jia, and G. Ingold, *Phys. Rev. Lett.* **103**, 155702 (2009)
[2] E. Möhr-Vorobeva, S. L. Johnson, P. Beaud, U. Staub, R. De Souza, C. Milne, G. Ingold, J. Demsar, H. Schaefer, A. Titov, *Phys. Rev. Lett.* **107**, 036403 (2011)
[3] D. Polli, M. Rini, S. Wall, R. W. Schoenlein, Y. Tomioka, Y. Tokura, G. Cerullo, A. Cavalleri, *Nature Materials* **6**, 643 (2007)
[4] S.L. Johnson, P. Beaud, C. J. Milne, F. S. Krasniqi, E. S. Zijlstra, M. E. Garcia, M. Kaiser, R. Abela, and G. Ingoldet, *Phys. Rev. Lett.* **102**, 175503 (2009)