Time-Resolved Stimulated X-ray Raman Scattering

Bruce D. Patterson

SwissFEL Project
Paul Scherrer Institut, CH-5232 Villigen, Switzerland

Conventional resonant inelastic X-ray scattering (RIXS) is a powerful photon-in / photon-out method for performing chemically-specific spectroscopy in solids. However, in spite of resonance enhancement, the inefficiency of RIXS implies long measurement times and precludes time-resolved measurements. Like its Raman scattering optical analog, RIXS relies on spontaneous emission to produce the outgoing photon. At sufficiently high incoming fluence, *stimulated* Raman scattering (see Fig. 1) will dominate over spontanteous scattering, dramatically increasing the efficiency and allowing a variety of time-resolved techniques.

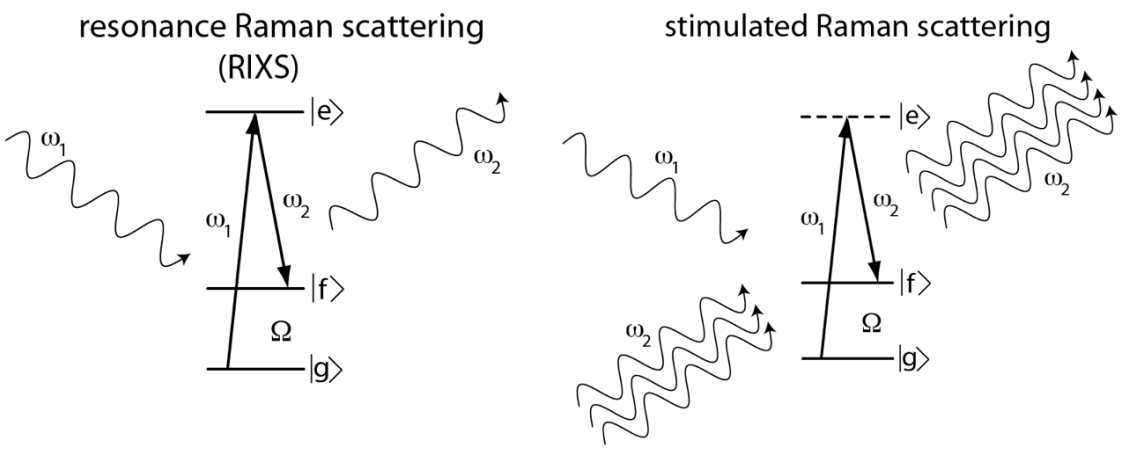

Fig. 1. A comparison of spontaneous and stimulated Raman scattering.

Spontaneous Raman scattering can be considered to be a stimulated process in which the stimulating wavefield ω_2 originates from the zero-point field of the black-body spectrum. The number of photons per mode produced by a light source is given by its degeneracy factor δ. The zero-point field contributes one photon per radiation mode, and stimulated scattering predominates over spontaneous scattering when this contribution is exceed by the incoming ω_2 flux. Whereas for a Hg lamp and a synchrotron undulator, $\delta \approx 10^{-3}$, a He-Ne laser has $\delta \approx 10^{7}$, and an XFEL $\delta \approx 10^{9}$! Just as the optical laser launched non-linear optics, the XFEL has the potential of opening the field of non-linear X-ray spectroscopy.

The cross-section for inelastic stimulated Raman scattering, and for the related elastic four-wave mixing processes, are proportional to the third-order susceptibility $\chi^{(3)}$, which can be estimated at X-ray wavelengths using a simple anharmonic oscillator model. The resulting probabilities for non-linear interactions are encouraging, and evidence exists that stimulated elastic scattering by atoms has already been observed at the LCLS at Stanford.

If a two-color mode of XFEL operation can be realized, it should be possible to perform stimulated scattering in the *transient grating spectroscopy* (TGS) geometry (Fig. 2). Here, a broadband pump pulse, which is split and recombined at the sample, undergoes impulsive stimulated Raman scattering (ISRS) to produce a spatially and temporally periodic modulation of sample excitation. This transient grating is subsequently queried by the diffraction of a delayed probe pulse at a different wavelength.

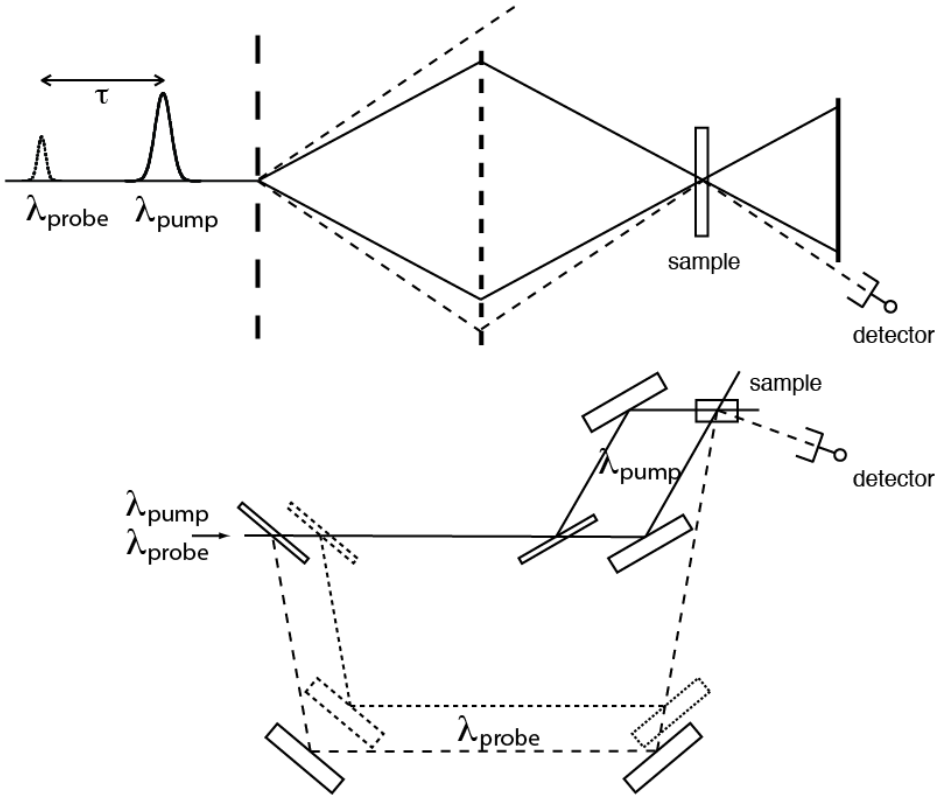

Fig. 2. Proposed geometries for transient grating spectroscopy at a two-color XFEL: diffraction gratings for soft X-rays (top); crystal optics, including an optical delay, for hard X-rays (bottom).