Resonant inelastic x-ray scattering studies of elementary excitations

Ament, L.J.P.

Chapter 8

Outlook – X-ray Free Electron Lasers

RIXS is now beginning to become a mainstream technique of condensed matter physics. Due to the tremendous progress in energy resolution and intensity, the energy scale of excitations accessible to RIXS has moved down from several eV in the late 1990's to several tens of eV now. If this progress continues at the same pace, it will soon be possible to probe excitations at even lower energy scales. This will not only bring more of the same materials within the range of RIXS experiments (for instance, materials with a lower superexchange constant), but it will also enable experimentalists to access excitations not seen before with RIXS. A very interesting prospect would be to map out the dispersion of excitations across the superconducting gap.

Not only the energy resolution is improving. The very recent availability of ultra-short and ultra-bright X-ray pulses generated by free electron lasers such as FLASH at DESY and the Linac Coherent Light Source (LCLS) at SLAC National Accelerator Laboratory offers unique opportunities for X-ray science, in particular time-resolved experiments, see, *e.g.*, Ref. [241]. In the remainder of this chapter, we discuss some implications of these X-ray Free Electron Lasers (XFEL's) for regular X-ray scattering, focussing on the bosonic enhancement of absorption and emission due to the enormous occupation number of the incident mode.

8.1 Bosonic enhancement in an XFEL

A Free Electron Laser populates a certain mode with a huge number of photons. This mode will be in a superposition of states with different, large occupation numbers n. Typically, when the mode is populated with a linear Hamiltonian, meaning $H \propto (a + a^\dagger)$ with a the photon annihilation operator, the result is a coherent state $|z\rangle = e^{-\frac{|z|^2}{2}} \sum_{n=0}^{\infty} \frac{z^n}{\sqrt{n!}} |n\rangle$ with z a complex number with huge modulus. The number of photons in the mode follows a Poisson distribution with mean $|z|^2$. The other modes of the electromagnetic field are, ideally, empty.

In deriving the RIXS cross section in chapter 2, we assumed that there is a single photon in the incident mode, which is reasonable for synchrotrons. The outlandish XFEL field state however, has enormous occupation numbers. When the field operator $\mathbf{A}$ acts on this state, it generates large boson factors $\sqrt{n}$. Processes involving multiple $\mathbf{A}$'s are therefore enhanced in an XFEL, in contrast to synchrotrons. Note that these multiple $\mathbf{A}$'s should pick up the incident mode to get the Boson enhancement factor: that does not happen for the empty outgoing modes.

As an example of this enhancement, we consider the absorption of two photons from the heavily populated incident mode of momentum $\mathbf{k}$ and polarization $\boldsymbol{\epsilon}$ through the $\mathbf{A}^2$ term of Eq. (2.22). From Eq. (2.24), we retain only the two-photon absorption and emission for the beam mode, and get

$$\mathbf{A}(\mathbf{r})^2 \sim \frac{\hbar}{2\mathcal{V}\epsilon_0\omega_{\mathbf{k}}} \left(\boldsymbol{\epsilon} \cdot \boldsymbol{\epsilon} \; a^2_{\mathbf{k},\boldsymbol{\epsilon}} e^{i2\mathbf{k}\cdot\mathbf{r}} + \text{h.c.} \right). \tag{8.1}$$

For a highly populated incident mode, $a^2_{\mathbf{k},\boldsymbol{\epsilon}}$ produces the average occupation number, which is projected to be of the order of 10^9 for the LCLS [31]. Two things can be concluded here. First, for linear polarization, the $\mathbf{A}^2$ term is independent of polarization because $\boldsymbol{\epsilon} \cdot \boldsymbol{\epsilon} = 1$. For circular polarization, $\boldsymbol{\epsilon} \cdot \boldsymbol{\epsilon} = 0$, and the process is forbidden: these two-photon processes cannot transfer more than one unit of angular momentum to the solid. Second, these two-photon processes are (mainly) dipole transitions. This can be seen by expanding the exponential $e^{i2\mathbf{k}\cdot\mathbf{r}}$ for small $\mathbf{k} \cdot \mathbf{r}$. To zeroth order, no transitions can be made. To first order, dipole transitions appear.

Before discussing specific scattering processes, we make an estimate of the size of (the matrix elements of) $\mathbf{A}$. The coherent volume $\mathcal{V}$ is projected to be $\sim 3 \cdot 10^{-13}$ m^3 for the LCLS [31]. We thus get

$$\langle \mathbf{A} \rangle \sim \sqrt{\frac{\hbar}{2\mathcal{V}\epsilon_0 c}} \left\langle \sqrt{n}\mathbf{r} \right\rangle \sim (3 \cdot 10^{-10} \text{ Js/Cm}^2) \left\langle \sqrt{n}\mathbf{r} \right\rangle. \tag{8.2}$$

8.2 X-ray scattering with an XFEL

There are several interesting aspects of the bosonic enhancement for X-ray scattering. Multi-photon processes are enhanced, and one could either stimulate the

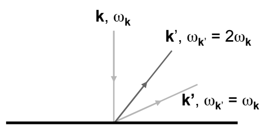

Figure 8.1: If X-rays at half the resonance energy are incident perpendicular to the sample surface (taking a simple cubic lattice, cut perpendicular to one of the cubic axes), the truely elastic Bragg peaks (light gray, $\omega_{\mathbf{k}'} = \omega_{\mathbf{k}}$) are pushed away from the surface normal. The 'elastic' line emitted at twice the incident X-ray energy has different Bragg conditions (dark gray): the first Bragg peak is closer to the surface normal.

absorption or the emission process. Enhancing both would require two XFEL beams.

Enhanced absorption. An interesting way to lose the non-resonant contributions to the spectrum is to excite the system with X-rays of half the resonance energy via the $\mathbf{A}^2$ term. Non-resonant X-ray scattering with matrix elements $\langle f | \mathbf{A}^2 | i \rangle$ produces outgoing X-rays with an energy of about half the resonance energy. Resonant scattering, however, mainly comes from the $\mathbf{A}\cdot\mathbf{p}$ term, creating X-ray photons with double that energy, *i.e.*, close to the resonance energy. The Kramers-Heisenberg equation becomes

$$\mathcal{F}_{fi} \propto \sum_n \frac{\langle f | \mathbf{A}\cdot\mathbf{p} | n \rangle \langle n | \mathbf{A}^2 | i \rangle}{E_i + 2\hbar\omega_{\mathbf{k}} - E_n + i\Gamma}. \tag{8.3}$$

This could be of use for resonant X-ray scattering: one can completely eliminate the non-resonant contributions. They remain at half the energy.

The theoretical treatment of RIXS also simplifies, because the polarization dependence of the absorption is trivial, as shown in Sec. 8.1.

Further, the Bragg conditions are interesting because the wavelength of the outgoing X-rays is halved with respect to the incident ones. Thus, there will be two sets of Bragg peaks: one set of truely elastic peaks at the regular angles, and one set at double the incident energy, arising through the resonant process of Eq. (8.3). The latter peaks are closer together: there are more Bragg peaks, as shown in Fig. 8.1. In resonant X-ray scattering, the elastic peaks arising from some ordering of the solid also appears at twice the incident energy and at outgoing wave vectors where no truely elastic ordering peaks appear.

Enhanced emission. Another interesting effect is stimulated emission. Not only the absorption can be enhanced by the boson factor, but also the consequent emission into the XFEL mode is boosted. This enhances forward scattering ($\mathbf{k}' = \mathbf{k}$): during the core hole's lifetime, which is much shorter than the pulse duration[1], elastic decay into the beam mode is stimulated. Forward scattering is

[1] At the LCLS, the X-ray pulses are projected to be 230 fs long [31], compared to $\hbar/\Gamma \sim 1$ fs.

enhanced by a factor $\langle n \rangle$. Note that this is not a shift of spectral weight away from other Bragg peaks or inelastic features: extra spectral weight is generated because one stimulates extra scattering events.

This effect is interesting because it allows for experimental control of the lifetime of the core hole: it is decreased by stimulated emission. To determine how much this is, we have to know the resonant contribution to the spectral weight of a Bragg peak, multiply it by $\langle n \rangle$ and see if this number is a sizeable fraction of the total number of decay processes (including Auger decay etc.). The resonant contribution to the spectral weight of a Bragg peak can in principle be determined by the resonant X-ray scattering method dicussed above, where the non-resonant contribution is quenched.