X-ray fluorescence intensity interferometry for atomic resolution structure determination

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X-ray fluorescence is usually considered as an incoherent process, with radiation emitted from atoms with random phases. As such, the angular distribution of fluorescence from a sample is uniform and featureless, unlike that of coherent scattering which gives rise to diffraction patterns used to derive molecular structures. However, light of the same frequency from independent sources, including X-ray fluorescence, will interfere if measured within a duration shorter than the coherence time set by the bandwidth of the light. The coherence time for most K-shell X-ray emission is a few femtoseconds, a time-scale now accessible with X-ray FELs. This is the basis of Hanbury-Brown and Twiss intensity interferometry [1]. The random phases of the interfering photons are dealt with by correlating intensities measured in different pixels of a pixel array detector, resulting in a quantity just like a coherent diffraction pattern, but of just the sub-structure composed of the particular atomic species. Remarkably, true three-dimensional information is encoded for a single orientation of the object [2].

This 3D imaging will be discussed, as well as several more advantages over the conventional crystallographic techniques that have been in use for the last 100 years. The fluorescence emission for an atom such as Fe is about 200 times the probability for scattering from a carbon atom, allowing measurements on single molecules, in solution. The chemical specificity of the emission means systems can be examined in natural environments, and chemical states (such as oxidation) can be discriminated. We expect atomic resolution without suffering from atomic form factors that limit high-resolution coherent scattering measurements. The method can be easily combined with diffraction, and thus may have great impact for understanding the structure, dynamics, and energetics of proteins and other materials.

References

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