Anomalous Total Scattering: Deconstructing Pair Distribution Functions

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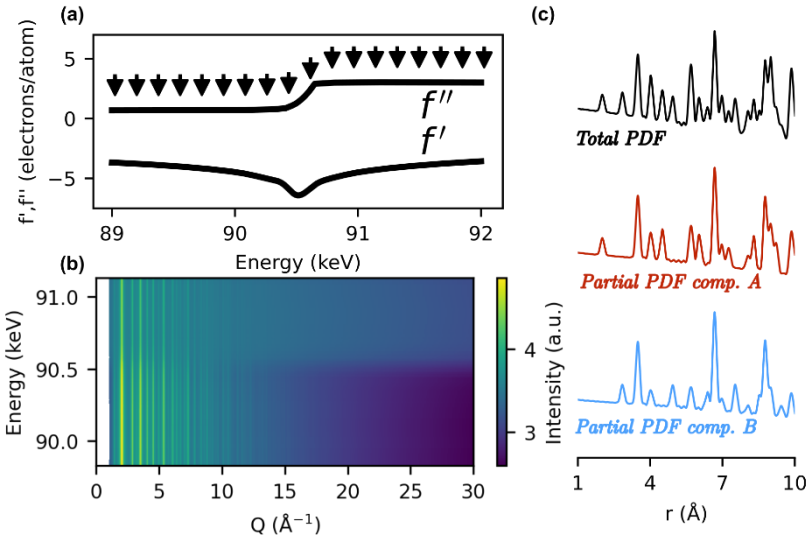


Figure 1. *(a) Calculated dispersive scattering terms f'' and f' of Bi across the Bi-edge. Arrows indicated a simplified measurement grid for total-scattering experiments along the edge. (b) Measured total scattering data of $BaBiO_3$ across the absorption edge of Bi. The intensities are plotted on a logarithmic scale to better highlight incoherent changes across the edge. (c) Simulated total and partial PDFs of a Perovskite structure (ABO_3).*

Pair distribution function (PDF) analysis and X-ray absorption spectroscopy (XAS) are essential tools for determining structure-property relations in defective, amorphous, or nanoscale materials [1], but each has its unique pros and cons. XAS provides rich information about the electronic and atomic structure in the immediate vicinity of a specific element (absorber), whereas a PDF provides structural information beyond that but lacks element-specific contrast. Anomalous diffraction bridges this gap. Elemental contrast in diffraction experiments can be amplified by tuning the incident X-ray energies to the resonant energies of target components in the material [2]. This makes use of the fact that the scattering term *f* of elements is energy dependent (**Fig. 1a**). Near an absorption edge, the dispersive terms of f become strong and change rapidly as a function of energy, but so do other effects like sample absorption and fluorescence---best visible in the incoherent intensity change at high-*Q* in **Fig. 1b**. These latter effects are often larger than the changes in the scattering terms, and have to be rigorously corrected for to extract the pure coherent structure function needed for PDF analysis. If done correctly, partial anomalous differential PDFs (adPDFs) can be extracted from coherent differential structure functions [3], providing element-specific information as shown in **Fig. 1c**.

Inspired by the success of PDFgetX3, we present an *ad hoc* data correction algorithm that takes care of the necessary corrections by approximating the effects of absorption and fluorescence using parametrized polynomial functions. Correct parameters can be found by conducting many total scattering measurements across an absorption edge and establishing a correlation between the correction functions and the anomalous scattering term f''. This approach is fundamentally different from the conventional way of extracting an adPDF; it takes two isolated measurements (near and far from the edge) and explicitly calculates corrections using known physics [3]. As a proof of concept, the workflow was tested on data measured across the Bi-edge of $BiBaO_3$. To test the method's limits and boundaries, we extended the study to perovskites composed of transition metals with lower-energy edges and to more complex materials, including alloys and solid solution metal oxides. We discuss what added benefits the technique provides to conventional PDF and XAS analysis of these complex materials. The work provides a valuable benchmark for the community that could transform anomalous diffraction into a highly practical, high-throughput tool for the broader field.

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