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DFT and Plasmon-Coupling Models for Optical and Electronic Scattering Properties

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We present calculations and applications of optical energy loss data for use in studies of inelastic electron scattering in condensed matter systems. A new model of plasmon coupling and excitation broadening is implemented along with high-precision density functional theory to evaluate fundamental material properties critical to many areas of spectroscopic analysis. Recent developments in x-ray and electron spectroscopies have demonstrated critical dependence on low-energy electron scattering and optical loss properties, and significant discrepancies between theoretical and experimental scattering values [1]. Resolution of these discrepancies is required to validate experimental studies of material structures, and is particularly relevant to the characterization of small molecules and organometallic systems for which electron scattering data is often sparse or highly uncertain [2]. We have devised a new theoretical approach linking the optical dielectric function and energy loss spectrum of a material with its electron scattering properties and characteristic plasmon excitations. For the first time we present a model inclusive of plasmon coupling, allowing us to move beyond the longstanding statistical approximation and explicitly demonstrate the effects of band structure on the detailed behavior of bulk electron excitations in a solid or small molecule. This is a novel generalization of the optical response of the material, which we obtain using density functional theory [3]. We find that our developments improve agreement with experimental electron scattering results in the low-energy region (<~100 eV) where plasmon excitations are dominant; a region that is particularly crucial for structural investigations using x-ray absorption fine structure and electron diffraction. This work is further relevant to several commissions of the IUCr including the commissions on XAFS, International Tables, and Electron Crystallography.

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