

Theory of x-ray absorption spectroscopy for ferrites

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The theoretical calculation of the interaction of electromagnetic radiation with matter remains a challenging problem for contemporary *ab initio* electronic structure methods, in particular, for x-ray spectroscopies. This is not only due to the strong interaction between the core hole and the photoexcited electron, but also due to the elusive multiplet effects that arise from the Coulomb interaction among the valence electrons. In this work we report a method based on density functional theory in conjunction with multiplet ligand-field theory to investigate various core-level spectroscopies, in particular, x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD). The developed computational scheme is applied to the $L_{2,3}$ XAS and XMCD edges of magnetite (Fe_3O_4) as well as cobalt ferrite (CoFe_2O_4) and nickel ferrite (NiFe_2O_4). The results are in overall good agreement with experimental observations, both regarding the XAS $L_{2,3}$ branching ratio, the peak positions, as well as the relative intensities. The agreement between theory and experiment is equally good for XAS and the XMCD spectra, for all studied systems. The results are analyzed in terms of σ_{ij} and t_{2g} orbital contribution, and the robustness of the spectra with regard to the uncertainties of the Slater parameters is investigated. The analysis also highlights the strong effect of the $2p$ - $3d$ interaction in x-ray spectroscopy.

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I. INTRODUCTION

Core-level spectroscopy is one of the most powerful techniques for probing the electronic structure of strongly correlated electron systems, as the interaction between x-rays and matter gives element-specific insights into the magnetic and electronic properties of materials, as well as their structural and chemical properties. In particular, the development of synchrotron radiation facilities and recently, x-ray free-electron lasers, have made the analysis efficient and the resolution very high in energy, and in specific cases, in time [1]. This increase in the experimental resolution has made it possible to detect fine features and structures in the x-ray spectra [2,3]. This development of experimental tools and techniques calls for a novel theoretical approach to compute x-ray absorption spectra, most notably, for systems with complex or correlated electronic structures, like magnetites as well as cobalt and nickel ferrites. In this work we focus specifically on these materials and on insights for the theoretical description of x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD).

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For the theoretical modeling of core-level and x-ray absorption spectroscopy, there have been two main distinct methodologies that are primarily used today. The first one is based on density functional theory (DFT) and the Kubo formula [4], while the second focuses on multiconfiguration effects [5]. Both methodologies are well reviewed in the literature (see, for example, Refs. [6,7]). The DFT-based methods are essentially parameter-free and provide, as a main advantage, a description of magnetism, structural properties, and chemical bonding in general. These methods are designed for the ground-state properties of materials, and hence their description of the excited states probed with x-ray spectroscopy is very limited. This is due to an incomplete treatment of electronic correlations, in particular, the multiconfigurational effects. Overall, these methods work best for widely dispersive electron systems. In contrast, atomic multiplet theory is essentially intended for a better description of the multiconfigurational effects originating from on-site electron-electron interaction [8]. For practical computational efficiency, the multiplet theory is often implemented for an isolated ion and thus neglects hybridization effects. It is therefore not surprising that this implementation of the theory has been found to work best for localized electronic states, e.g., the $4f$ orbitals of rare-earth elements. However, effects like crystal-field splitting and orbital hybridization can be treated by adding suitable model terms to the Hamiltonian of the system. The resulting theory is often referred to as the multiplet ligand-field theory (MLFT) [8]. This theory is very powerful in reproducing experimental spectra when the

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