Multiple Scattering of Electrons in Solids and Molecules: a Novel Cluster-Model Approach for Photoelectron Diffraction

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INTRODUCTION

Core-level photoelectron diffraction (PD) [1-3] is much used to obtain structural information about solid surfaces. Actually, a photoelectron can suffer multiple atomic scattering (MS) events before it leaves the solid and is eventually detected. The photoemission intensity distribution in energy and angle contains information about the relative positions and identities of the atoms surrounding the emitter.

In this context, the present work introduces a new MS calculational scheme, called Electron Diffraction in Atomic Clusters (EDAC), which presents several advantages over previous methods.

The new scheme is easily generalized to treating non-spherical potentials for valence photoelectron diffraction. The resulting EDAC code already is capable of representing electron spin effects, which will be useful to study magnetic materials, such as ultrathin magnetic films.

METHOD

In order to incorporate curved-wave effects, the electron wave function is expressed in spherical harmonics and spherical Bessel functions attached to each atom of the cluster. However, the propagation of these functions between cluster atoms is computationally very demanding [4]. In an attempt to overcome this problem, approximations have been introduced in the past, mostly for the high-energy limit, in which case the electron propagation reduces to plane-wave factors (plane-wave approximation) and each term (i.e., each electron path) in the MS series becomes a product of scattering amplitudes. Expansions that take into consideration the finite size of the atoms have also been developed [5], including full curved-wave formulations of the problem that require dealing with $(l_{\max}+1)^2$ partial wave components per atom, where $l_{\max}$ is the maximum of the significant angular momentum quantum numbers.

Rehr and Albers [6] provided a method based upon a separable representation of the free-electron Green function that allows one to generalize the scattering amplitudes, substituting them by matrices that produce reliable results when keeping only a few of their leading terms. This is particularly suitable to calculate the contribution of different individual electron paths, but it scales like N^3 with the number of atoms N if all paths are included in the calculation. This method is incorporated in our recent MSCD code [7], which is in wide use for interpreting photoelectron diffraction intensities.

In our new EDAC method, the MS expansion is evaluated using an exact representation of the Green function propagator and an iterative procedure whose computational demand scales as $N^2(l_{max}+1)^3$. In addition, the new method of calculation of electron diffraction in atomic clusters (EDAC) prevents divergences in the MS series by using an adaptation of Haydock's recursion method [8].

In EDAC, a muffin-tin description of the atomic potentials and an exact representation of the free-electron Green functions are used. The method relies upon a convenient separation of the Green functions involving rotation matrices to reduce the computation time and storage demand. The multiple scattering expansion is iteratively evaluated using a divergence-free variation of the Haydock recursion method, as modified to allow calculating the far-field electron intensity in an arbitrary number of directions from a single multiple scattering calculation. The computational demand scales as $N^2(l_{max}+1)^3$, where N is the number of atoms in the cluster and l_{max} is the maximum of the relevant angular momentum quantum numbers.

Our new method has been shown to be able to handle clusters of as many as 2000 atoms: this allows calculations with a high precision.

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