

Calculation of the electronic properties of Mo in a first-principles nonlocal-pseudopotential approach

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The first-principles nonlocal atomic pseudopotentials previously derived from the density-functional formalism are applied in a self-consistent study of the electronic properties of bulk Mo. Using a mixed-basis representation for the crystalline wave functions, both the localized and the extended features of the electronic states are efficiently described. We show that by use of a completely nonempirical approach the basic electronic properties of the system, such as the band structure, density of states, charge density, and Fermi surface, are favorably reproduced within the pseudopotential formalism.

I. INTRODUCTION

The pseudopotential method which has been successfully applied to the study of electronic and structural properties of many nontransition-metal systems,¹ has recently been extended to transition metals.²⁻⁷ This case is more difficult in general because of the inadequacy of perturbative approaches⁸ to the strong transition-metal core pseudopotentials, the inefficiency of a plane-wave basis set for describing the localized features of the *d*-wave functions, and the pronounced orbital nonlocality of the pseudopotentials. In addition, an empirical parametrization of the bare-ion potential⁹ has been largely inhibited by the incompleteness of the measured single-valence electron term value spectra of transition metals. Despite the difficulties, non-self-consistent empirical pseudopotential calculations have been carried out on a number of transition metals²⁻⁷ by fitting the observed optical reflectivity of the bulk solids.

Although generally good agreement is obtained with the experimental spectra and Fermi surface, the underlying pseudopotentials are only partially successful when transferred to study the electronic properties of phases which are substantially different from the reference system used for fitting the potential (i.e., the bulk elemental solid). This results from the fact that in the empirical approach both the core pseudopotential and the (dynamic) valence screening fields (interelectronic Coulomb and exchange) are combined together to a charge-density (independent form $V_{\text{core}}(\vec{r})$) which in turn is adjusted to fit some bulk properties. As no attempt is made to feed back the variational crystal charge density into $V_{\text{core}}(\vec{r})$, the latter cannot effectively describe the charge relaxation in systems different from the reference one.

Recently,¹⁰⁻¹² we have presented a new class of nonlocal atomic pseudopotentials for the first five rows of the Periodic Table, which are self-consistently derived from the density-functional

representation¹³ to the all-electron system. These pseudopotentials involve no (prefixed or adjustable) parameters other than the atomic number. In their derivation, no fitting to either atomic or solid-state experimental data is needed. Instead, they are obtained by a direct inversion of the density-functional effective single-particle equation pertaining to a pseudo atom characterized by local-density Coulomb and exchange-correlation interaction and the (yet unspecified) nonlocal static external field $V_l(r)$. The pseudopotential $V_p(r)$ is then solved for under the constraints that: (i) the ground-state valence energy eigenvalue spectra of the pseudo atom be identical to that of the all-electron atom characterized by the same form of density functionals and (ii) the wavefunction pseudizing process is described by a unitary rotation in the subspace of occupied all-electron ("true") orbitals, $\psi_{\mu}(r)$, i.e., the pseudo wave functions $\chi_{\mu}(r)$ are given as a linear combination of both core and valence functions $\phi_{\nu}(r)$ with coefficients $C_{\nu\mu}^{(i)}$. These rotation coefficients are then fixed by requiring that the pseudo wave functions be (a) nodeless for each of the lowest solutions of every symmetry, (b) normalized, and (c) have the maximum similarity possible under conditions (a) and (b) to the "exact" all-electron density-functional wave functions $\psi_{\mu}(r)$ in the valence region. When additional degrees of freedom are available (e.g., systems having more than one core orbital) an additional minimum radial kinetic energy constraint is imposed. The reader is referred to previous articles^{10,11} for the detailed discussion of the choice of this physical model.

This approach not only assures that both the energies and the wave functions of the pseudo Hamiltonian closely resemble the corresponding all-electron results for the reference ground-state atom, but also results in a very small energy dependence of the pseudopotential when a ground-state first-principles pseudopotential is applied to calculate electronic properties of excited species (having very different wave functions

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