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Development of fully relativistic pseudopotentials and application to a thin magnetic film on the metal

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Abstract

We have developed a fully relativistic pseudopotential in the planewave method. The pseudopotential shows the sufficient transferability that the error is much smaller than the energy splitting of spin-orbit coupling. This pseudopotential scheme has been implemented for bulk systems, surfaces, and the thin magnetic film on the metal.

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1. Introduction

Pseudopotential (PP) in the first-principles approach is efficiently used in the applications which have nano-scale structures of materials. In the system having heavy elements or magnetic systems, it is important to include a fully relativistic effect into the PP. The spin-orbit splitting in the system of nano-wires, ultrathin films, etc. is observed and concentrates attention in developing the element of electronic devices[1].

In the present paper, we describe the PPs, and, to demonstrate our approach on surfaces, present the application to a thin magnetic film on the metal of heavy element, namely, a monolayer of Fe on the Pt(111).

2. Pseudopotential

We have developed a fully relativistic PP for typical heavy elements. The relativistic effect was included by solving a Dirac-type equation instead of a Schrödinger-type equation in the generation processes of PPs. In this process, the formalism is parallel as the previous one[2], but, for example, the non-local potential $V_{NL} = \sum_{i,j} |\beta_i\rangle D_{ij} \langle \beta_j|$ is constructed from the relativistic atomic orbitals. The projector function β_i has the form of spin-angular harmonic

function which diagonalize the total angular momentum operator.

The transferability test for the PP of Pt is presented in Table 1. The reference state having the valence $(5d)^9(6s)^1(6p)^0$ configuration for making the PP was used, and by using this potential the two cation states were tested. The energy differences between the all-electron and PP approaches are much smaller than the spin-orbit splittings in $5d$ and $6p$ orbitals, which are 0.0984 and 0.0404 Ry, respectively, for the reference state.

To demonstrate efficiency of PPs, we have calculated the electronic structure in the several systems. In the

Table 1

The pseudopotential transferability test (in Ry) for the energy level and the total energy (ΔE_{tot} , difference from the energy of respective reference state) in Pt^+ and Pt^{2+} . The AE and PP represent the relativistic result for all-electron and pseudopotential, respectively.

state	level	AE	PP	level	AE	PP
$\text{Pt}^+ (5d^9 6s^0 6p^0)$	$5d_{3/2}$	-1.1354	-1.1338	$5d_{5/2}$	-1.0339	-1.0327
	$6s_{1/2}$	-0.9750	-0.9771			
	$6p_{1/2}$	-0.5591	-0.5609	$6p_{3/2}$	-0.4834	-0.4850
	ΔE_{tot}	+0.7025	+0.7034			
$\text{Pt}^{2+} (5d^8 6s^0 6p^0)$	$5d_{3/2}$	-1.9907	-1.9968	$5d_{5/2}$	-1.8793	-1.8850
	$6s_{1/2}$	-1.6560	-1.6571			
	$6p_{1/2}$	-1.1545	-1.1550	$6p_{3/2}$	-1.0389	-1.0394
	ΔE_{tot}	+2.1927	+2.1949			

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Fig. 1. Band dispersions in the fcc metals of Au and Pt. The thick and thin curves represent dispersions of fully relativistic and scalar relativistic cases, respectively. The Fermi energies are adjusted to the energy of zero. The experimental lattice constants are used.

calculations of extended systems, we used the scheme of two-component-spinor wavefunctions with planewave basis[3,4]. Relativistic effects are fully included and the self-consistent treatment for wavefunctions enables us to evaluate atomic forces and static pressures easily. Figure 1 shows the band dispersions for the bulks of Au and Pt. As in the previous work[5], we used the local spin density approximation[6]. We used the energy cutoffs of 30 and 300 Ry for wavefunctions and charge densities. Our result agrees well with that of all-electron approaches[7]. The splittings due to spin-orbit interaction are observed at the bands which have *d* character in vicinity of -5 eV and/or around the wave vectors of Γ , X, and L. Additionally, it is reproduced that in Pt the spin-orbit splittings reduce the density of states at the Fermi level[8].

Further, we checked the atomic relaxation of Pt(111) surface. Using 13 monolayer model, the relaxation ratio of surface layer, which is defined as $\Delta d_{12} = (d_{12} - d_0)/d_0$ (d_{12} and d_0 are the real and ideal distances between 1st and 2nd monolayers), was estimated to be 1.12 %, comparable to the experimental value (1.1 %) [9] and the all-electron value (1.3 %) [10].

3. Fe monolayer on Pt(111)

In the previous work[5], for the magnetic bulk, FePt(the Fe and Pt layers are stacked alternatively along the *c*-axis), our approach nicely described the magnetic properties of

magnetic anisotropy, compared with all-electron approach. The fully relativistic scheme enables us to estimate the magnetic anisotropy energy from the total energy difference between the two systems which have different magnetization directions.

We constructed the atomic monolayer of Fe on the Pt(111) surface. The atoms were occupied at the fcc-follow sites. All of atoms were fixed at the atomic positions extracting from the fcc crystal with experimental lattice constant. We used the slab model with 1 monolayer for Fe and 4 monolayers for the Pt(111) surface. The 24×24 mesh for sampling k-points was used.

We have estimated the magnetic anisotropy energies for the magnetic layer on the Pt (111) surface. The total energy of perpendicular magnetization (F_z) is higher than that of in-plane magnetizations (F_x, F_y), where the *x*-axis is parallel to the direction connecting the nearest neighbor in the plane; $F_x - F_z = -0.66$ and $F_y - F_z = -0.61$ meV. Thus, our result indicates that the ideal Fe monolayer on Pt(111) has a magnetic in-plane easy axis, while the bulk of regular alloy FePt the easy axis perpendicular to the planes of Fe layers.

4. Summary

The prepared relativistic PP of Pt shows a good agreement with the properties obtained from the all-electron approach and a good transferability. Using the PPs, we successfully included the effect of spin-orbit coupling in the planewave method. This would be feasible for improving the theoretical quality for the systems which contain heavy elements with a complex atomic geometry.

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