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Impact of Fe/NaCl (001) interface structure on electronic, magnetic and spin-polarized transport properties of Fe/NaCl/Fe (001) heterojunctions: An ab initio study

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ABSTRACT

Structural, electronic and spin-dependent transport properties of Fe/NaCl/Fe (001) tunnel junctions with c(2 × 2) structure are studied by means of first-principles calculations. Several Fe/NaCl (001) interfaces are considered and their stability is analyzed. The interfacial charge transfer has an important role in stabilizing Fe/NaCl (001) interfaces. Interfacial iron magnetic moments are enhanced over the bulk value. Fe induced gap states are observed in barrier layers on both Na and Cl ions. Small exchange couplings with exponential decays as function of the barriers thickness are evidenced. Resonant tunnelling plays a major role on the spin-polarized transport properties. High magnetoresistance ratios comparable to Fe/MgO/Fe junctions are predicted. Total energy calculations show that interfacial interdiffusion is not favourable but the disorder at interfaces may strongly affect the electronic transport properties. Absence of interfacial interdiffusion together with the small lattice mismatch between Fe and NaCl layers are important advantages of Fe/NaCl/Fe (001) junctions for the purpose of spintronic applications.

1. Introduction

Over the past years theoretical and experimental studies have shown that the most promising tunnel junctions are epitaxial ones with crystalline MgO barriers. First principles calculations in the linear response limit predicted extremely large TMR values for epitaxial Fe/MgO/Fe (001) MTJs [1–3] as well as for junctions with bcc Co (001) and CsCl-type FeCo (001) electrodes [4]. Ab initio predictions have been only in part experimentally confirmed for Fe(Co, FeCo)/MgO/Fe(Co, FeCo) (001) crystalline MTJs elaborated either by molecular beam epitaxy (MBE) or sputtering techniques [5–8]. TMR values beyond 400% at room temperature (RT) were reported for Co/MgO/Co (001) junctions [8]. The experimental TMR values are smaller at least by one order of magnitude than theoretical predictions. The origin of this discrepancy has been related with Fe/MgO interfaces and barriers quality. Meanwhile extensive studies were performed in order to improve the junctions quality and performances, leading to TMR values beyond 600% at RT for MgO based single barrier junctions with CoFeB electrodes [9] and beyond 1000% for double barrier junctions, respectively [10]. However, rather large lattice mismatch at interfaces, i.e., ≈3.8% for Fe/MgO ones, leads to imperfections that in turn limit the experimental TMR values.

It was demonstrated that high magnetoresistance effect observed for epitaxial MTJs with MgO barriers, Fe(Co, FeCo) electrodes and ideal Fe(Co, FeCo)/MgO (001) interfaces is a consequence of a strong spin filter tunnelling effect that results from the band structure characteristics of 3d transition metal electrodes, wave function symmetry conservation and the complex band structure of MgO spacer [1,2,11]. The MgO barrier has its minimum band gap at $\bar{\Gamma}$ point and the lowest decay rate is expected for Δ_1 ($s - p_z - d_{z^2}$) states. The Δ_1 Bloch states with 100% spin polarization (existing at the Fermi level only for majority spins due to the strong exchange splitting between $\Delta_1^{\uparrow}$ and $\Delta_1^{\downarrow}$ sub-bands) in Fe, bcc Co and FeCo electrodes couple with MgO Δ_1 states with lowest decay rate in barrier and high spin polarizations of the currents are obtained in the ferromagnetic (FM) configuration of magnetic electrodes. The conductance in the antiferromagnetic (AFM) state is low due to symmetry considerations. Since of large difference in conductance between ferromagnetic and antiferromagnetic states of the MgO based tunnel junctions, very high TMR ratios are expected. Conservation of symmetry and spin of electrons tunnelling across the barrier in Fe/MgO/Fe (001) junctions had been proved by inserting a Cr layer at Fe/MgO (001) interfaces. Due to the absence of Δ_1 states at the Fermi level in chromium, the

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interfacial Cr spacer was found to act as a spin filter for Δ_1 electrons [12]. The role of the electrode–barrier interfaces on the magnetoresistive properties of MTJs [13–16] and the importance of interface resonance states on the spin-dependent transport properties were, also, predicted [3,17].

As an attempt to identify alternative junctions, several barrier materials such as $SrTiO_3$ [18], $Mg_3B_2O_6$ [19], NaCl [20,21] or CaS [22] have been studied. The experimental data revealed the possibility of fabrication of novel tunnel junctions with epitaxial NaCl (001) barriers using a vacuum deposition technique [20]. First-principles calculations predicted high TMR values for Fe/NaCl/Fe (001) magnetic junctions with hypothetical Fe (001) [100]||[100] NaCl (001) epitaxial relation at Fe/NaCl (001) interfaces (corresponding to c(2 × 2) structure with Fe (001) and NaCl (001) layers orientated parallel) [21]. The growth of Fe film on NaCl [23] as well as the fabrication of single crystalline Fe/NaCl/Fe (001) tunnel junctions is hindered by the large difference in the surface free energy between Fe and NaCl layers. Recent experimental data of Tekiel et al. [24] showed that growth of NaCl on Fe (001) surface in c(2 × 2)-type structure is difficult due to chemical and elastic properties of Fe (001) surface. They have shown that the addition of oxygen make possible layer-by-layer growth of ultra-thin and atomically flat NaCl films on Fe (001)-p(1 × 1)O surface with Fe (001)–O [100]||[110] NaCl (001) orientation, leading to a (4 × 4) structure.

NaCl or common salt crystallize in ionic B1 (rock salt)-type structure having a lattice parameter of 5.64 Å that differ only by 1.6% of the c(2 × 2) Fe structure lattice spacing. Sodium chloride has a direct sp Γ–Γ band gap of 0.625 Ry that is only 8.9% larger than the sp direct band gap of MgO but otherwise having band structures along (001) direction rather close. NaCl (001) stoichiometric surface is the lowest in energy, electrically neutral and double layer of NaCl films show almost bulk like electronic structure characteristics. Therefore Fe/NaCl/Fe (001) heterostructures may represent good candidates for theoretical investigations as well as for potential technological applications.

The purpose of the present work is the ab initio investigation of the relative structural stability of c(2 × 2) interfaces, the influence of the interfacial morphology on electronic and magnetic properties of Fe/NaCl/Fe (001) junctions as well as the nature of the electronic transport in such devices.

2. Models and computational details

The present calculations have been performed for epitaxial multilayer structures composed of two semi-infinite left and right Fe (001) leads sandwiching an active (central) region consisting of two FM Fe (001) electrode layers separated by a B1-type NaCl (001) barrier, *n*Fe (001)/*m*NaCl (001)/(*n* + *1*)Fe (001) where *n* and *m* are integers representing the number of Fe and NaCl monolayers (MLs), respectively (Fig. 1). The bcc-like structure is considered for Fe electrodes, B1-type structure for NaCl barriers and a perfect epitaxial lattice matching of bcc Fe with B1 NaCl is assumed, i.e., $a_{NaCl} = 2a_{Fe}$. Each atomic monolayer contains two non-equivalent atomic positions in the two dimensional unit cell and all atomic monolayers are counted from Fe/NaCl interfaces. In the magnetic slabs, from the interface, they are labelled as I, I − 1, I − 2, …, while in barrier they are denoted as I, I + 1, I + 2, …, respectively. Among the continuum of Fe/NaCl (001) interface geometries we assume only ideal (atomically flat) and high symmetry ones. Three model interface geometries have been considered in present calculations: (1) Fe atoms sitting atop Na and Cl positions labelled as IC_1; (2) Fe atoms located above hollow sites between Na and Cl ions named IC_2, and (3) Fe atoms above the middle of the bridge linking first order Na and Cl neighbours denoted as IC_3, respectively (Fig. 2).

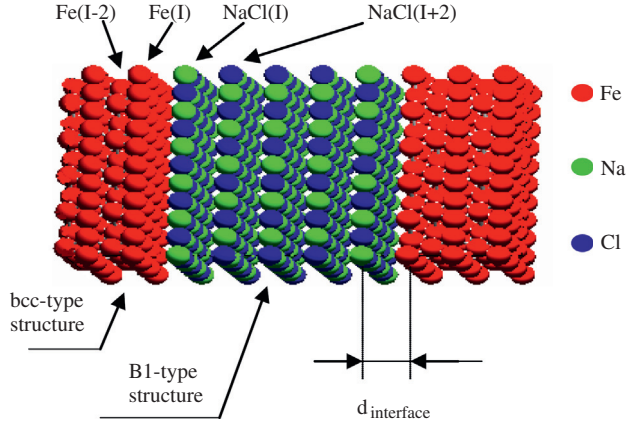

Fig. 1. Active region of Fe/NaCl/Fe (001) magnetic tunnel junctions (asymmetric Fe electrodes are considered in the present calculations due to ASA approximation and the presence of the empty sphere planes in barriers).

The electronic and magnetic ground state properties of Fe/NaCl/Fe (001) multilayer structures with in plane 2D periodicity and broken translational symmetry perpendicular to interfaces have been determined self consistently by means of a first-principles, scalar-relativistic, and spin-polarized Green's function technique for surface and interfaces, employing principal layer technique, based on the tight-binding linear muffin-tin orbital (TB-LMTO) formalism in its atomic sphere approximation (ASA) [25]. The coherent potential approximation (CPA) has been used to describe the interdiffusion at Fe/NaCl (001) interfaces [25]. The local spin density approximation (LSDA) was used for the exchange–correlation energy functional within Vosko–Wilk–Nusair parameterization scheme [26]. The basis set of linear muffin-tin orbitals includes s-, p- and d-orbitals. Therefore the input electronic configurations were taken as core + $3d^6 4s^2$ for Fe, core + $3s^1$ for Na, and core + $3p^5$ for Cl, respectively. Each principal layer consists of two atomic monolayers. Due to reduced thickness of the active region the Fermi energy (E_F) of the whole system has been approximated by the Fermi energy of semi-infinite Fe (001) bulk-like leads. All calculations are performed for lattice parameters epitaxially fixed at the lattice spacing of bulk iron and neglecting any layer or lattice relaxations. Two empty spheres were introduced on interstitial positions at Fe/NaCl (001) interfaces with IC_1 geometry as well as between nonconducting NaCl layers in all barriers and four empty spheres at Fe/NaCl (001) interfaces with IC_3 interfacial structure. Their radii and positions were adjusted so as to minimize the overlapping, in agreement with ASA space filling requirements, and without breaking 2D symmetry. The distances between interfacial Fe and NaCl layers, d_{int}, are determined from ASA space filling requirement for Fe/NaCl (001) interfaces with IC_2 and IC_3 geometries while for IC_1 interface configuration we assume $d_{int} = a_{Fe}$. A parallel spin alignment is assumed in each magnetic slab while parallel (FM) and antiparallel (AFM) state configurations are considered for the magnetizations of ferromagnetic iron slabs.

The spin-dependent transport properties, in the current-perpendicular-to-plane (CPP) geometry and ballistic limit, at zero-temperature and zero-bias, have been calculated by means of linear response of Kubo–Landauer approach implemented within TB-LMTO-CPA formalism and including vertex corrections [27]. The TMR ratios are expressed by the asymmetry of the conductances for FM and AFM configurations of Fe electrodes normalized to AFM ones, $TMR=(\sigma_{FM} - \sigma_{AFM})/\sigma_{AFM}$.

Electronic structure calculations have been performed by using a 16 × 16 $\mathbf{k}_{||}$ points mesh sampling in 2D surface Brillouin zone (SBZ) and a 60 × 60 $\mathbf{k}_{||}$ points mesh for the transport properties, respectively. The self-consistency is considered to be achieved for a total energy difference between consecutive iterations less than 10^{-6} Ry.

3. Results and discussions

3.1. Structural properties

Due to the small misfit between Fe and NaCl lattices, difficulties in growth of NaCl layers on Fe (001) surface in c(2 × 2) structure and lack of experimental data regarding Fe/NaCl (001) interfaces, we start our study investigating structural properties as well as the relative stability of Fe/NaCl (001) interfaces, and the impact of interfacial morphology on the electronic and magnetic properties.

Structural properties of Fe/NaCl/Fe (001) heterostructures with Fe/NaCl (001) interfaces having IC_1 configuration had been previously reported [21]. Self-consistent total energy calculations as function of the lattice spacing (all values are reported to the bulk Fe lattice spacing, thus in magnetic slabs we have $a = a_{Fe}$ while in barrier $a_{NaCl} = 2a_{Fe}$) performed for heterostructures with IC_2 and IC_3 Fe/NaCl (001) interface configurations (Fig. 3a), for both FM and AFM states of magnetic electrodes, yield an equilibrium lattice constant of ≈2.75 Å, independent on the interface geometry. Calculated equilibrium lattice constants are smaller than either the experimental lattice spacing of Fe or half of experimental lattice