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Large spin–orbit splitting of surface states in ultrathin Au (111) films

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ABSTRACT

Spin–orbit (SO) splitting of surface states in ultrathin Au films was examined on the basis of density-functional theory. Due to the interaction between the top and the bottom Au layers, the SO splitting in the Au (111) films with few atomic layers is usually negligible. The splitting is, however, detectable if one surface of the film is terminated by adatoms, such as Al or H atoms. Similar effect can also be achieved if an external electric field is applied perpendicular to the surface. In addition, an interesting even–odd oscillating behavior in the SO splitting can be observed in a Au/Al interlaced structure. All these can be explained by the inversion-symmetry breaking. Our work provides a deep understanding of the SO effect in ultrathin heavy-metal films, which will be helpful in future all-electric spintronics.

1. Introduction

Spin–orbit (SO) effects have attracted considerable attention recently in the field of spintronics, due to its advantage of manipulation of electron spin without the need of external magnetic field [1]. Many interesting SO effects were observed in experiments or proposed in theories, such as spin Hall effect [2], quantum spin Hall effect [3], and topological insulating behavior [4]. Giant Rashba SO splitting [5] in surface states was detected in several kinds of metal films grown on certain substrates such as Bi/Ag (111) [6,7], Au/W (110) [8], and Ag/Au (111) [9], which would show its importance in the heterostructure physics. Compared with semiconductor heterostructures, these metal films generally have much stronger SO interaction and they may be used to construct tiny spintronic devices with strong SO effect. For example, the Rashba SO strength in the Au (111) surface is one or two orders larger than that in semiconductor heterostructures [6,11,10]. Thus, it is expectable to replace the traditional two-dimensional electron gases (2DEGs) by metal films [11,12].

The flaw to apply the SO splitting of the surface states is that the surface states are usually buried in the tangled bulk states. To extract the surface states, the thickness of the metal films can be reduced to decrease the bulk states. The SO effect of the surface states may, however, be ruined because of the strong interaction of the two (top and bottom) surfaces in the films with reduced thickness [13–15]. For the Au (111) surface, a 23 monolayer (ML) thick film is required to eliminate the interaction between the two surfaces and to achieve large SO splitting of the surface state [14–16]. H termination on one side is a simple and available method for discussing the interfacial bonding, it has been used for a 22 layer Bi film to describe the Rashba effect [17].

In the present work, we explore how to extract the surface states and simultaneously keep their large SO splitting in ultrathin Au films with only few ML. We find that the SO splitting of the Au (111) L-gap surface state [12,15,18,19] can be recovered in the Au films when one surface is terminated by Al or H atoms due to the breakdown of the inversion symmetry. Although symmetry breaking is necessary for the SO splitting as discussed in Ref. [20], it would not always lead to an obvious SO phenomenon. For example, we also calculate the Ag adsorption on Au (111) surface, and find that it cannot recover the Au (111) surface state. The chemical bonding is not strong enough to fully separate the surface states at the undoped surface from the interface states [21]. Besides atomic absorptions, similar effect can also be obtained if an external electric field is applied in the direction perpendicular to the surface of the Au films. These results provide ways to preserve the surface states and their behaviors in ultrathin metal films. It would also help one understand the SO phenomena in the interface structures, such as graphene on metallic substrates [22–25]. An interesting even–odd behavior in the SO splitting is found in the Au/Al interlaced structures by changing the number of ML, which call for detection in experiments.

2. Method and computational details

The electronic structures were calculated by using VASP code at the level of local spin-density approximation (LSDA) [26].

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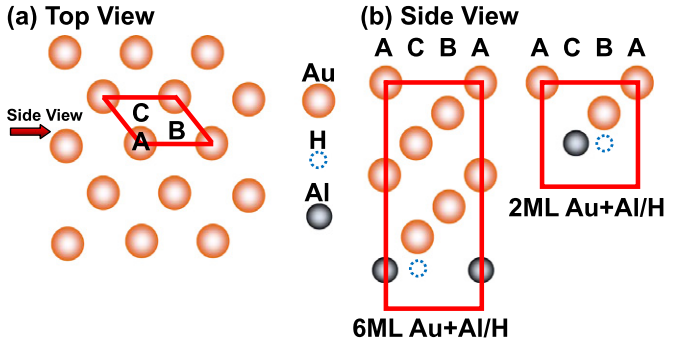

Fig. 1. (Color online.) (a) Top view of the Au (111) film. The red rhombus gives the unit cell along the surface. 'A', 'B' and 'C' represent the three stacking positions in the unit cell. (b) Side view of the ultrathin Au film with 6 ML Au (left) or 2 ML Au (right). The bottom of the Au film is covered by H or Al atoms. The H atoms are just below the Au atoms, while the positions of Al atoms are consistent with the stacking pattern of Au (111).

The projector-augmented wave (PAW) pseudopotentials were employed to describe the effect of core electrons. The energy cutoff in the calculations was set to be 400 eV, and the total energy was converged to better than 10^{-5} eV. The equilibrium structures were obtained through structural relaxation until Hellmann–Feynman forces were less than 0.02 eV/Å [22]. The ultrathin films were modeled by a periodic slab geometry, with a vacuum of at least 10 Å between two neighboring slabs. The unit cell and the slab structures are shown in Fig. 1. The length of the lattice vectors in the plane was set to be 2.885 Å. Due to a very small lattice mismatch (<1%), we can use Au and Al ML to construct a special Au/Al interlaced structure. The k-point grids of 31×31 in the plane was employed to sample the Brillouin zone. The SO coupling term considered takes the form of $H_{so} = \frac{\hbar}{4m^2c^2}(\nabla V_{eff} \times \mathbf{P}) \cdot \sigma$, where V_{eff} is the effective potential of electrons, $\mathbf{P}$ is the momentum operator, and $\sigma \equiv (\sigma_x, \sigma_y, \sigma_z)$ are the Pauli matrices. In 2DEGs, due to the structure inversion asymmetry, the SO coupling term can be simplified to the Rashba SO interaction, $H_R = \alpha_R(P_x\sigma_y - P_y\sigma_x)$, where the Rashba strength α_R depends on the gradient of the potential along the direction perpendicular to the 2DEG plane [5]. As a benchmark calculation for the SO effect, we first determined the SO splitting of the surface state of Au (111) near the Fermi level (E_F, set at energy zero) [15]. Our result, $\Delta E_{SO} \sim$100 meV, agrees well with the data in Ref. [15]. For the benefit of discussion, we would focus on the energy bands nearest to the E_F around Γ point, which are the red curves in the following energy-band figures. In the films with SO splitting, these bands are like the Au (111) L-gap surface state and are mostly composed by the Au p_z state. The Au (111) surface state extends several layers into bulk [27], so that the states distribute symmetrically throughout the whole film as quantum well states in a undoped ultrathin Au film. However, in the Au films with Al or H termination on one surface, the states chiefly distribute on the undoped Au surface and quickly decay towards termination. All these states in this work would be called as surface states rather than quantum well states.

3. Results and discussion

The SO interaction, coming from the relativistic effect, is proportional to the potential gradient. For the vacuum/film/vacuum structure, the vertical potential gradient at the two sides of the film is opposite. Hence, the interaction of the top and bottom surfaces would reduce drastically the SO effect [13–15]. As shown in Fig. 2(a), the energy bands of the 6 ML Au film do not have any SO splitting, different to the results of the 23 ML Au film [14,15]. The inversion symmetry of the potential gradient at the two sides of the film should be broken to realize the SO effect [14]. Although

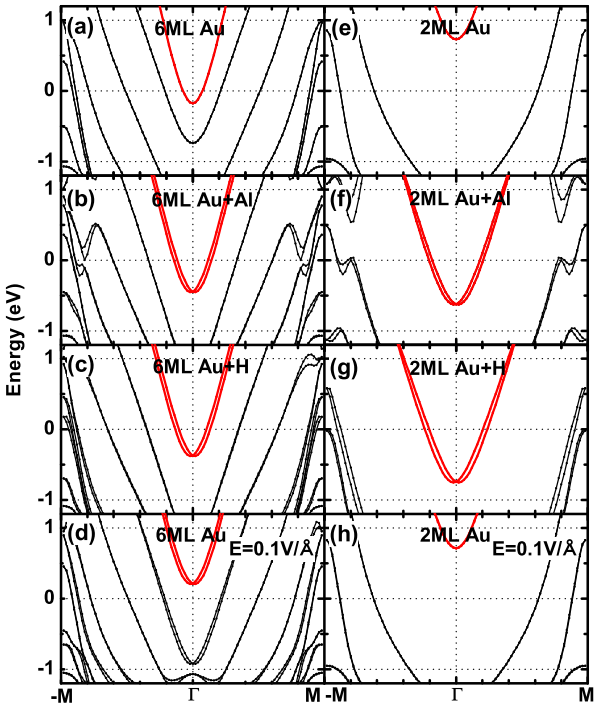

Fig. 2. (Color online.) (a)–(d) The energy bands of the 6 ML Au films. In (b) and (c), the bottoms of the Au films are terminated by Al and H atoms, respectively. An electric field (0.1 V/Å) perpendicular to the Au surface is applied in (d). (e)–(h) are the same as in (a)–(d), respectively, except it is for the 2 ML Au films. The red curves indicate the bands primarily discussed.

there are no dangling bonds clearly formed in metallic systems, we can construct an inversion asymmetry ultrathin film by adsorbing Al or H atoms on the bottom surface. Figs. 2(b) and 2(c) show the energy bands of the 6 ML Au film terminated by Al and H atoms. The binding energy of Al and H atoms is about 1.3 and 2.2 eV respectively. The large binding energy suggests Al or H atoms can be easily adsorbed on Au surface. They would form a stable structure. The adatoms greatly change the energy bands of the 6 ML Au film and recover the Au (111) surface state with $\Delta E_{SO} \sim$ 100 meV. When the Au film is terminated by Al (H) atoms, the interaction between Al (H) and Au in the bottom surface would break the inversion symmetry and separate the top and the bottom surface states. Thus, the large SO splitting returns in spite of weak SO interactions in Al and H atoms. The results also suggest there can be a strong SO effect in Au ultrathin films by depositing them on Al (111), which can be easily carried out in experiments [8,9,28].

Further reducing the thickness from 6 ML to 2 ML, Al or H atoms still have clear effect on the SO splitting as shown in Fig. 2(e)–(g). Clearly, the surface states with large SO splitting are dominantly near the E_F in these films. It's hopeful to use the ultrathin Au films as the 2DEGs with strong SO interaction. We emphasize that the function of the Al or H atoms is to break the inversion symmetry and separate the top and the bottom surface states. Generally, other atoms such as C or S can also lead to a relatively large SO splitting. However, from the benefit of applications, the dopant should make the system have a simple band structure near the E_F. Our results provide hints to understand a relatively large SO splitting (13 meV at Dirac point) of graphene in the structure of graphene/Au/Ni (111), where only one atomic layer of Au was inserted between graphene and Ni substrate [24,29]. Although the breakdown of the inversion symmetry of graphene is essential to obtain the effect [30], the asymmetry Au layer is also necessary. The large SO effect of Au in the asymmetry structure would