Magnetism of One-Dimensional Ni monoatomic chain on Au(110)-(1×2) surface

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Based on the Density Functional Theory, we have calculated the magnetic anisotropy of the supported Ni chains. Our results indicate the closed relation between magnetic anisotropy and the orbital quenching. The easiest magnetized axis changes from parallel to perpendicular to the Ni chains once they absorb on the surafce. PACS: 75.70.Ak, 75.30.Gw, 75.75.+a, 71.15.Mb

I. Introduction

The high-density magnetic recording and the memory devices require the strong magnetic anisotropy to stabilize the magnetism of the basic magnetic structures. The low dimensional materials due to the reduced dimensionality generally have a favorable direction, the physical properties along this direction are generally different from the other directions. Thus we have the most possibilities to find the strong magnetic anisotropy materials from the low dimensional materials. On the other hand, the reduced dimensionality reduces the atomic coordination which can enhances the spin and orbital moments of the materials. Since the pioneering experiments for the magnetism of Fe strips on the W(110) and Cu(111) surface[1−3], the researches along this direction have been expanded to the others quasi-one-dimensional systems and the monatomic magnetic chain on surface[3−12]. The experiments are supported by some calculations of density functional theory and theoretical models[13−19]. Besides the celebrating properties such as the strong magnetic anisotropy, the enhanced spin and orbital moments, some new phenomena have been found, such as the temperature and time dependent magnetization[1,2] which make the realistic applications of these novel experimental devices face new serious problem such as the instability of magnetic structure. Thus it is valuable to investigate theoretically along this direction before the corresponding experiments.

In this work we study the magnetism of mono-atomic Ni chains supported on Au(110)-(1×2) surface with the one-dimensional trough along [1$\bar{1}$0] direction. We have calculated the magnetic anisotropy energies of the supported Ni chains and found an off-plane easiest magnetized axis which is perpendicular to the chains and the surface. The easiest magnetized axis of the free-standing Ni chains are parallel to the chain and change into perpendicular to the chains when supported on the surface. The change of easiest magnetized axis of the supported-surface are also found in the others systems[14]. Our results indicate that the closed relationship between the magnetic anisotropy and the quenching of the orbital moments of the Ni chains.
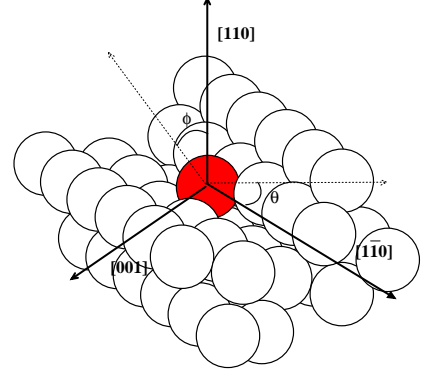

FIG. 1. The schematic diagram of a Ni chain supported on the Au(110)-(1×2) surface. The trough of the surface along the [1$\bar{1}$0] direction. The red sphere is the absorbed Ni atom in the trough. The white spheres are the Au atoms of the surface.

II. Computation Methods

We calculate the magnetism of Ni_n(n=1-5) chains which are supported on the Au(110)-(1×2) surface based on Density Functional Theory[20,21] using the Methods of Projection of Augmentation Wave (PAW)[22] with the plane-wave base set and the Perdew-Wang's exchanging-correlating potential[23,24]. The program is the VASP code written by the computational materials science group at Vinnea university[25−28]. At first step, we prepare the samples of the supported Ni chains on the surface by relaxed the initial structures using the conjugate-gradient Methods with Γ points used as the Brillouin-zone sampling in the corresponding calculations of the electronic structure. The initial structures construct by placing the Ni chains at the place of the missing Au chains with the same atomic nearest-neighbor distances along the [1$\bar{1}$0] direction Fig. 1. The initial surface constructs with 56 Au atoms with 4 atomic layers. The atoms in the bottom layer of the slab are fixed. The sizes of the super-cell are (23.046Å×8.148Å×15.52Å). Based on the relaxed structures, we calculate the electronic structures of the supported Ni chains using RMM-DIIS algorithm[25] with the (4×4×1) Monkhorst-Pack grids sampling the Brillouin zone. The Wigner-Seize radius set to 1.286Å to

calculate the magnetic moments of the Ni chains. The energy cutoff of plane waves is 269.561eV. We set the Methfessel-Paxton smearing width equal to 0.20eV to accelerate the speed of convergence. We choose the [110] direction as the quanta axis. If the changes of the total energies are smaller than 0.0001eV between two electronic self-consistent (SC) steps the SC-loops break, and 0.001eV between two ion-steps the programs stop.
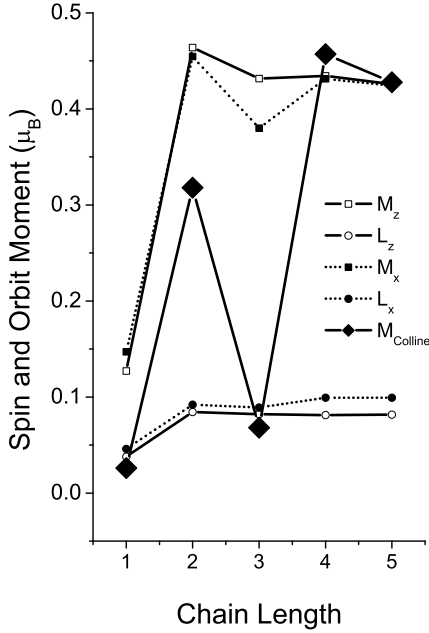
III. The magnetism of Ni_n (n=1-5)

FIG. 2. The spin M_x, M_z and orbital L_x, L_z moments change with the length of Ni chain. x indicate the magnetization along the $[1\bar{1}0]$, y along the [110] direction. $M_{Colline}$ present the spin moment in the collinear calculation.

At first, we calculate the total energies, the spin and orbital magnetic moments of the supported Ni chains when magnetizing the chains perpendicular to the chains ([110] direction) and parallel to the chains ($[1\bar{1}0]$ direction) respectively. The calculations include the non-collinear effects and the spin-orbital coupling which is proved important to the heavy metals such as Gold. As an contrast, we also perform the collinear calculation. By comparing the results with the non-collinear calculations, we can illustrate clearly the influence of both the non-collinear effects and the spin-orbital coupling on the magnetism of the supported Ni chains. The size-dependence magnetic moments are shown in Fig. 2. From this figure we can see that the spin moments $0.147\mu_B$ and $0.127\mu_B$ are small for single atom when magnetizing it along above two directions respectively. The spin moments increase to $0.909\mu_B$ and $0.928\mu_B$ respectively for the Ni_2 chain. When n≥3, the spin moments decrease slightly, except

for the Ni_3 chain when magnetizing it along the chain. From Fig. 2, we can find that the spin moment of the Ni_3 is almost zero for the collinear calculation.
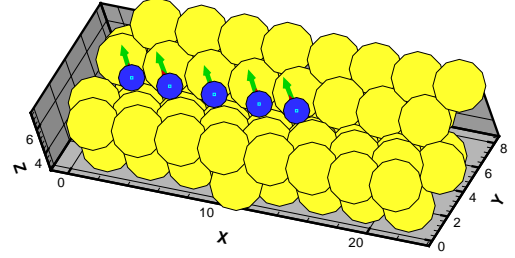
FIG. 3. The vector graph of the spin magnetic moments of the Ni chain which illustrates the ferromagnetic order of the Ni chain

The magnetic order is an important aspect of the magnetism of materials. Experimentally the long ferromagnetic order is found for Co chains on Pt(997) surface which stabilizes above a finite temperature[6]. Above this temperature, the long ferromagnetic order is destroyed and changes into the short ferromagnetic fragments. In our calculations, the short Ni chains are ferromagnetic. Fig.3 shows the ferromagnetic order of the surface supported Ni_5 chains. We also calculate the antiferromagnetic configurations and non-magnetic configurations whose energies are higher than that of the ferromagnetic configuration, thus the ferromagnetic order is energetically favorable.

In the environment of crystal with cubic symmetries, the orbital moment of a magnetic atom will be quenched to a small value. There is a simple rule which can estimate the influence of symmetry on the orbital moment of materials. The broken of the local symmetries (Point group) generally quenches the orbital moment efficiently because the lost of the local symmetries can efficiently lift the degeneracy of the ground state. The broken of global symmetry (Space group) generally enhances the orbital moment. When the three-dimensional translation symmetries in the bulk break to the two-dimensional translation symmetries on the surface, the orbital moments can be enhanced due to the uncompleted orbital quenching. Thus we hope that the surface supported atoms have relative large the orbital moments. From the Fig. 2, we can see that the orbital moments of the Ni chains are nonzero except for the single Ni atom. It is interesting to find from this figure that the orbital moments when magnetizing the Ni chains parallel to the chain are larger than that perpendicular to the chain. At the end of the paper, after having studied the magnetic anisotropy of Ni_5 chain in details, we explain the results using the classic concept of orbital moment. It will show that the degrees of the orbital quenching are different when magnetizing

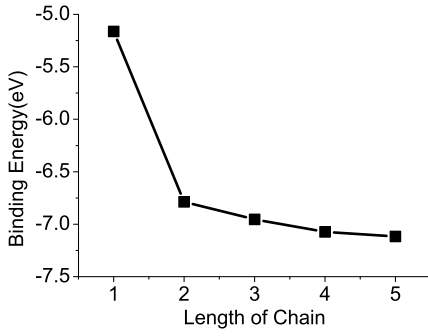the Ni chains along different directions.

FIG. 4. Interaction energy between the Ni chains and the surface. The larger absolute values indicate the strong interaction with the surface.

Generally, the magnetism of materials is closely relative to its structural characteristic. If the atomic nearest neighbor distance is small, the exchange integral J is larger and the magnetic correlation of different atoms is also strong. The distances of atoms at the surface are generally smaller than those in bulk. Thus the magnetism of surface is generally stronger than that in bulk. The environment of the atoms supported on the surface is similar to the surface atoms. Based on the same logic, they possibly have strong magnetism. The degrees of the interactions between the supported Ni_n chains and the surface can be measured with the interaction energies E_I which are equal to the differences of the total energies with the Ni_n chains supported on the surface and the sum of the total energies of the free surface and the free standing Ni_n chains. We generally calculate the interaction energies per atom, which removes the effects of the simple summation. The values of the interaction energies are generally negative. The small absolute values of the interaction energies are indicative of the weak interactions between the Ni chains and the surface and otherwise the strong interactions.
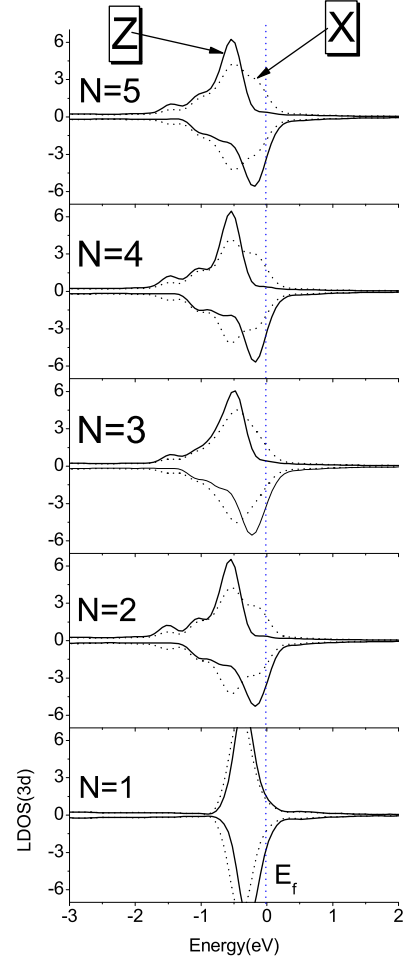

FIG. 5. The densities of 3d states of the supported Ni chains. The solid lines show the 3d DOS of the Ni chains magnetized along the $[1\bar{1}0]$ direction, z along the $[110]$ direction.

Fig. 4 shows that the interaction energies between the chains and the surface increase with the lengths of the chains. The interactions between the chains and the surface generally deform the structure of the surface. The stronger interaction induces the larger deformation of the surface which generally breaks the translation symmetries of the crystal fields at the region near the absorbed atoms. This will depress the effects of the orbit quenching due to the crystal fields splitting and enhances the orbital moments of the absorbed atoms. From Fig. 2, for the short chains which interact weakly with the surface have small orbital moments, such as the single Ni atom has almost zero orbital moment and the weakest interaction with the surface. The orbital moments increase with the lengths of the Ni chains companying with the expanding of the deformed region and the broken of the translation symmetries along the absorbed Ni chains. From Fig. 4 we can see that the interaction energies pre atom have

a tendency reach a constant and are independent on the sizes of the chains. Our results show that the magnetic-correlated length of the Ni chains on the surface is near the length of the Ni_5 chain, beyond the length the average magnetism of the Ni chains will be independent on the size of the Ni chains. We also calculate the total energies of the Ni chains with the magnetization both parallel and perpendicular to them respectively. Our results show that the total energies with the magnetization parallel to these Ni chains are slightly higher than perpendicular to the surface. This indicates that the easy magnetizing direction is perpendicular to the surface.
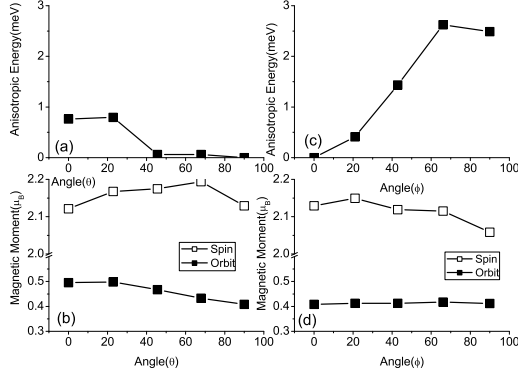

FIG. 7. The anisotropic energies, the spin and orbital moment of the free standing Ni_5 chain change with the magnetization angles θ in the planes S_1. The easiest magnetized direction is parallel to the chain $\theta = 0°$.

IV. The magnetic anisotropy of Ni_5 chain

In this section we study the magnetic anisotropy of the Ni_5 chain. The magnetic anisotropy of one-dimensional structure is more prominent than other compact structures such as surface and bulk. We name the plane with both [110] axis and [1$\bar{1}$0] axis as S_1, the plane with both [110] axis and [001] axis as S_2 and label the rotating angle from any vector in the S_1 to [1$\bar{1}$0] axis as θ, the rotating angle from any vector in the plane S_2 to [110] axis as ϕ (Fig. 1). θ and ϕ are the magnetization angles in the two planes. Fig.6 shows the changes of the total energies, the spin and orbit moments with the two magnetization angles in above two plane. We find from Fig. 6(a,c) that there are weak the magnetic anisotropy in the S_1 plane and the relative strong the magnetic anisotropy in the S_2 plane. The magnetic anisotropy energies are 0.79meV and 2.62meV respectively in the two planes. The easiest magnetizing directions in both S_1 and S_2 planes are all along [110] direction ($\theta = 90°$ and $\phi = 0°$), which is perpendicular to the chain and the surface. From Fig. 6(b) we can find the orbital moments reach the minimum when magnetizing the Ni chain along the easiest magnetization direction. Fig. 6(d) also shows that the orbital moments almost keep a constant when the magnetization angle ϕ changes in S_2 plane.

FIG. 6. The anisotropic energies, the spin and orbital moment of the supported Ni_5 chain change with the magnetization angles θ and ϕ in the planes S_1 and S_2 defined in above. The easiest magnetized direction is perpendicular to the surface $\theta = 90°$ and $\phi = 0°$.

Additionally, we calculate the density of state of the Ni_n (n=1-5)chains which are shown in the Fig. 5. When magnetizing the chains perpendicular to the chains, the spin-up density of state (DOS) have sharp edges at the right side near Fermi energy similar to the DOS of Ni crystal. For the magnetization parallel to the chains, the spin-up DOS of the Ni chains move toward the Fermi energies and the spin-down DOS move away from the Fermi energy. The peaks of DOS become flat compared with the magnetization perpendicular to the chains. This means that the 3d states delocalize when switching the magnetizing directions from perpendicular to parallel to the chain. Our calculations show the larger orbital moments for the magnetization along the chain due to the relative small orbital quenching (Fig. 2).
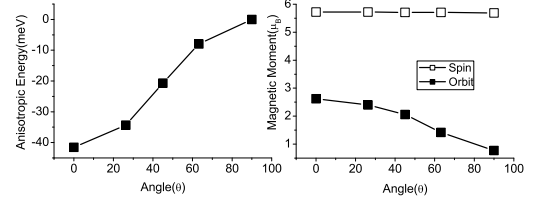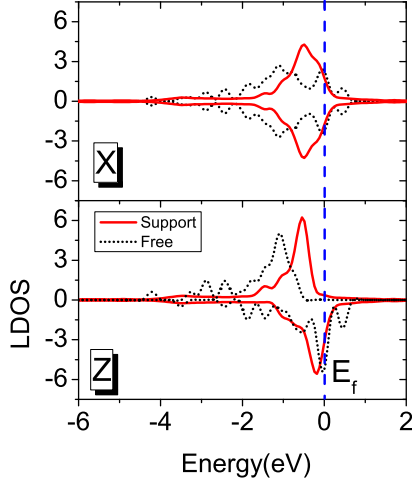
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FIG. 8. The DOS of the supported (solid lines) and free standing (dot lines) Ni_5 chain when magnetizing along x ($[1\bar{1}0]$), z($[111]$) direction.

In order to illustrate the influence of the surface on the magnetic anisotropy of Ni_5 chain we calculate the magnetic anisotropy of free-standing Ni_5 chain. All calculating details are the same as those of the surface-supported Ni_5 chain except for removing the surface atoms. For the free standing Ni_5 chain we can only consider the changes of the anisotropy energies with the angle θ. From the Fig. 7 we can see that the easiest magnetizing direction is now parallel to the chain which is different from perpendicular to the chain for the surface-supported Ni_5 chain. The change of the easiest magnetizing axis had been found in other work[14] for Co chains on Pd(110) surface. We can see from the figure that the anisotropy energy (40meV) is very large for the free standing Ni_5 chain. The spin moments keep almost constant when the magnetizing angle changes from $\theta = ^\circ 0$ to $\theta = 90^\circ$. The behavior of the orbital moment is similar to the surface-supported Ni_5 chain, that is, reaches the minimum when magnetizing the chain perpendicular to the chain. From the figure we also can see that both the spin and orbital magnetic moments of the free-standing Ni_5 chain are larger than those of the surface-supported Ni_5 chain. Thus our results show that the surface effects change the easiest magnetizing direction and decrease both the magnetic anisotropy energy and the magnetic moment. We calculate the density of state (DOS) of the surface-supported Ni_5 chain and the free-standing Ni_5 chain. From Fig. 8 we can see that the surface makes the DOS of the Ni chain accumulate at the energy near Fermi energy. The 3d state will be delocalized when the Ni atoms are embedded in the environment of the conducting electrons of the surface.
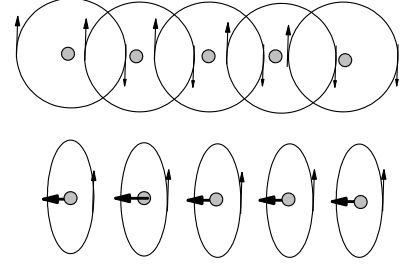

FIG. 9. The schematic diagram for orbital quenching. The diagram can explain the smaller orbital moments for the magnetization perpendicular to the chain x($[110]$) direction for the supported and free standing Ni chains

Our results in the above section have shown that the orbital moments with the magnetization parallel to the chains are larger than that perpendicular to the chains. We obtain the same results for the free standing Ni chains in this section. The results can be explained using the classic theory of orbital moment which is defined as the product of a small electronic current and the closed planar area of the current. The direction of the orbital moment defines as the normal of the area. From Fig. 9, we can see that when we magnetize the chain perpendicular to the chain, the currents of neighbor atoms overlap and cancel each other. This is the classic picture of the orbital quenching. However for the magnetization parallel to the chain, the orbital moment are larger due to the absent of the orbital quenching. The orbital quenching is close relative to the magnetic anisotropy. The degree of the orbital quenching changes with the magnetizing direction, by the spin-orbit coupling, the magnetic anisotropy are imported into magnetic materials[29]. The magneto-crystalline anisotropic energy can be explained as the no-spherical distribution of the charge density due to the spin-orbit coupling[30].

IV. Conclusion

Based on the Density Functional Theory, we have calculated the magnetic anisotropy of the supported Ni chains. The easiest magnetized axis is perpendicular to the chains and the surface. Our results indicate the closed relationship between the magnetic anisotropy and the orbital moments of the Ni chains. We use orbital-counter mechanism to explain the small orbital moments when perpendicular magnetizing the Ni chain.

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