An *ab initio* study of the magnetic and electronic properties of Fe, Co, and Ni nanowires on Cu(001) surface

J. C. Tung⁹, and G. Y. Guo⁹,⁸

⁹Department of Physics and Center for Theoretical Sciences, National Taiwan University, Taipei 106, Taiwan
⁸Graduate Institute of Applied Physics, National Chengchi University, Taipei 116, Taiwan

Abstract

Magnetism at the nanoscale has been a very active research area in the past decades, because of its novel fundamental physics and exciting potential applications. We have recently performed an *ab initio* study of the structural, electronic and magnetic properties of all 3d transition metal (TM) freestanding atomic chains and found that Fe and Ni nanowires have a giant magnetic anisotropy energy (MAE), indicating that these nanowires would have applications in high density magnetic data storages. In this paper, we perform density functional calculations for the Fe, Co and Ni linear atomic chains on Cu(001) surface within the generalized gradient approximation, in order to investigate how the substrates would affect the magnetic properties of the nanowires. We find that Fe, Co and Ni linear chains on Cu(001) surface still have a stable or metastable ferromagnetic state. When spin-orbit coupling (SOC) is included, the spin magnetic moments remain almost unchanged, due to the weakness of SOC in 3d TM chains, whilst significant orbital magnetic moments appear and also are direction-dependent. Finally, we find that the MAE for Fe, and Co remains large, i.e., being not much affected by the presence of Cu substrate.

Keywords: magnetocrystalline anisotropy, transition metal nanowires, spin-orbit coupling

1. Introduction

In recent years, nanostructured magnetic materials have received much attention, due to their interesting physical properties and potential applications. For example, finite free-standing gold atomic chains were first reported in 1998⁹¹, and their structural properties, such as the actual length of the chain have been the focus of intensive experiments and theoretical studies since then. However, these free-standing atomic chains are unstable and thus can only exist at low temperatures and on a suitably chosen substrate. Physically stable magnetic nanowires deposited on metallic substrates are one of the most important nanostructures and a variety of techniques have been used to prepare and study them. For example, Gambardella *et al*⁹ [3, 4] succeeded in preparing a high density of parallel atomic chains along steps by growing Co on a high-purity Pt (997) vicinal surface in a narrow temperature range of 10–20 K. The magnetism of the Co wires was also investigated by the x-ray magnetic circular dichroism⁹. Experimentally, copper and tungsten are excellent substrates for growth of Fe thin films⁹,⁵,⁶ because of the small lattice constant mismatch between Fe and Cu (3.61 Å) as well as W (3.61 Å). Thus, in this paper, we perform first principles calculations to study the electronic and magnetic properties of the Fe, Co and Ni nanowires. The spin-orbit coupling (SOC) is included in this study to determine the magnetic anisotropy energy (MAE).

2. Theory and Computational Method

In the present calculations, we use the accurate frozen-core full-potential projector augmented-wave (PAW) method,⁷ as implemented in the Vienna *ab initio* simulation package (VASP)⁸,⁹. The calculations are based on density functional theory with the generalized gradient approximation (GGA)⁹[10]. A large plane-wave cutoff energy of 350 eV is used for all the systems considered. The Fe, Co, and Ni nanowires along the \( x \) direction on the Cu (001) surface are modeled by a nanowire attached to both sides of a seven-layer-thick Cu (001) slab as plotted in Fig. 1. The transition metal (TM) atoms on the nanowires are placed either on the top of surface Cu atoms [denoted here as the atop (A) site] or at the hollow position on the Cu surface [called here as hollow (H) site]. The two-dimensional unit cell is chosen to be of \( p(4 \times 1) \) structure. The nearest in-plane (out of plane) wire-wire distance is larger than 10Å (11Å) which is wide enough to decouple the neighboring wires. The theoretical lattice constant (3.60 Å) of bulk copper, which is in good agreement with experimental Cu lattice constant of 3.61 Å, is used as the fixed in-plane lattice constant of the Cu slab. However, the atoms are allowed to move in the out of plane direction, and the structural relaxations are performed using the conjugate gradient method. The equilibrium structure is obtained when all the forces acting on the atoms and the stress are less than 0.02 eV/Å and 2.0 kBar, respectively. The \( \Gamma \)-centered Monkhorst-Pack scheme with a \( k \)-mesh of \( 20 \times 5 \times 1 \) in the full Brillouin zone (BZ), in conjunction with the Fermi-Dirac-smearing method with \( \sigma = 0.2 \) eV, is used to generate \( k \)-points for the BZ integration.
3. Results and Discussion

Because of its smallness, *ab initio* calculation of the MAE is computationally very demanding and needs to be carefully carried out. Here we use the total energy difference approach rather than the widely used force theorem to determine the MAE, i.e., the MAE is calculated as the difference in the full self-consistent total energies for the two different magnetization directions concerned. The total energy convergence criterion is 10⁻⁸ eV/atom. The same k-point mesh is used for the density of states calculations. The MAEs calculated with a denser 32×6×1 k-point mesh hardly differ from that obtained with the 20×5×1 k-point mesh (within 0.02 meV).

3. Results and Discussion

The calculated formation energy $E^f$, equilibrium interlayer distance $d_{eq}$, spin magnetic moment per magnetic atom, $m_s$, and magnetization energy per magnetic atom $E^{mag}=E^f-M_{FM}-N_{NM}$ of the 3d TM nanowires on both the hollow (H) and atop (A) sites of the Cu (001) surface. Here superscripts $FM$ and $NM$ denote the ferromagnetic and nonmagnetic states, respectively.

| site | $E^f$ (eV/u.c.) | $d_{eq}$ (Å) | $m_s$ (µB) | $E^{mag}$ (eV) |
|------|----------------|--------------|------------|----------------|
| Fe H | -2.14          | 1.64         | 3.07       | -0.337         |
| A    | -0.31          | 2.31         | 3.29       | -0.444         |
| Co H | -2.15          | 1.57         | 1.79       | -0.130         |
| A    | -1.19          | 2.27         | 1.99       | -0.205         |
| Ni H | -2.26          | 1.55         | 0.00       | 0.000          |
| A    | -1.32          | 2.26         | 0.65       | -0.032         |

Table 1: Calculated formation energy $E^f$, equilibrium interlayer distance $d_{eq}$, spin magnetic moment per magnetic atom, $m_s$, and magnetization energy per magnetic atom $E^{mag}=E^f-M_{FM}-N_{NM}$ of the 3d TM nanowires on both the hollow (H) and atop (A) sites of the Cu (001) surface. Here superscripts $FM$ and $NM$ denote the ferromagnetic and nonmagnetic states, respectively.

Figure 1: (color online) The slab supercell for modeling Fe, Co and Ni nanowires on the hollow sites of Cu (001) surface.
Figure 2: (Color online) Spin-polarized density of states of the Fe and Co nanowires on Cu (001), for an easy comparison. Clearly, the more dispersive. The reduction in coordination number also decreases the great enhancement in the spin splitting of Fe and Co moments are larger. The splitting of the 3\textit{d}\textit{e}-bands for hollow and atop sites are 2.58 (2.82), and 1.51 (1.75) eV, respectively. The interlayer distance between the TM nanowires and Cu substrate is still very large when compared with that of bulk Fe and Ni.

| m_s (\mu_B) | m_d (\mu_B) | MAE (meV) |
|-------------|-------------|-----------|
| 100         | 010         | 001       |
| H-site      |             |           |
| Fe          | 3.07        | 0.10      | 0.09  | 0.10 | 0.32 | 0.25 |
| Co          | 1.78        | 0.27      | 0.18  | 0.17 | -1.17 | -1.16 |
| Ni          | 0.02        | 0.02      | 0.00  | 0.00 | -0.53 | 0.01 |
| A-site      |             |           |
| Fe          | 3.28        | 0.12      | 0.11  | 0.13 | 0.38  | 0.29 |
| Co          | 1.99        | 0.19      | 0.25  | 0.12 | -0.40 | -1.51 |
| Ni          | 0.64        | 0.13      | 0.27  | 0.11 | -0.05 | -0.32 |

4. Conclusions

We have performed \textit{ab initio} GGA calculations for the Fe, Co and Ni linear atomic chains on Cu (001) surface in order to examine how the substrates would affect the magnetic properties of the nanowires. We found that Fe, Co and Ni linear chains on Cu (001) surface still have a stable or metastable FM state. When the SOC is included, the spin magnetic moments remain almost unchanged, due to the weakness of SOC in 3d TM chains. However, the significant orbital magnetic moments appear and are also direction-dependent, except Fe. We also found that the Fe system has a zero-plane magnetic anisotropy whilst the Co and Ni systems have an in-plane anisotropy.

Acknowledgments

The authors acknowledge support from the National Science Council and the NCTS of Taiwan.

References

[1] H. Ohnishi, Y. Kondo, and K. Takayanagi, Nature (London) \textbf{395}, 780 (1998).
[2] A. I. Yanson, G. R. Bollinger, H. E. van der Brom, N. Agrait, and J. M. van Ruitenbeek, Nature (London) \textbf{395}, 783 (1998).
[3] P. Gambardella, P. Blanc, M. Burgi, L. Kuhne, K. Kern, Surf. Science \textbf{449}, 93 (2000).
[4] P. Gambardella, A. Dallmeyer, K. Maiti, M. C. Malagoli, W. Eberhardt, K. Kern C. Carbone, Nature \textbf{416}, 301 (2002).
[5] J. Hauchschul, H. J. Elmer, and U. Gradmann, Phys. Rev. B \textbf{57} R677 (1998).
[6] D. Tian, F. Jona, and P. M. Marcus, Phys. Rev. B \textbf{45} 11216 (1992).
[7] P. E. Blöchl, Phys. Rev. B \textbf{50}, 17953 (1994); G. Kresse and D. Joubert, \textit{ibid.} \textbf{59}, 1758 (1999).
[8] G. Kresse and J. Hafner, Phys. Rev. B \textbf{48}, 13115 (1993).
[9] G. Kresse and J. Furthmühler, Comp. Mater. Sci \textbf{6}, 15 (1996).
[10] Y. Wang and J. P. Perdew, Phys. Rev. B 44, 13298 (1991); J. P. Perdew and Y. Wang, ibid. 45, 13244 (1992).
[11] G. Y. Guo, W. M. Temmerman, and H. Ebert, Physca B 172, 61 (1991).
[12] R. Mazzarello and E. Tosatti, Phys. Rev. B 79, 134402 (2009).
[13] Y. Mokrousov, A. Thiess and S. Heinze, Phys. Rev. B 80, 195420 (2009).
[14] J. C. Tung and G. Y. Guo, Phys. Rev. B 76, 094413 (2007).