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Curie Temperature of Ferromagnetic Metal Mono-layers Studied by First Principle Band Calculation

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Magnetic moments and Curie temperatures of Fe, Co and Ni mono-layers are calculated, in comparison to the bulk. The electronic structures were calculated within the framework of the local density approximation using the LMTO-ASA method. According to Liechtenstein, the exchange integral J_0 and the Curie temperature were evaluated by the first principle calculation. The magnetic moment of the mono-layer is higher than that of the bulk, because the band width of the mono-layer is reduced by the localization of the two dimensional configuration and the increment of the density of states around the fermi-energy enhances the polarization. However, it is estimated that the magnetic moment of the mono-layer is not so high at room temperature, because of a low Curie temperature.
Key words: Curie temperature; Fe; Co; Ni; Mono-layer; First-principle band calculation

1. INTRODUCTION

Transition metal mono-layers have attracted much attention, since the first principal band calculation predicted that they have larger magnetic moments than the bulk metal [1]. However, the experimental results were not clear [2, 3] as yet, because of the difficulty in fabricating the complete mono-layer and measuring the minute magnetic properties. Moreover the absolute zero temperature is assumed in the first principle calculation normally. So, one is not able to obtain physical values at ordinary temperatures. It is also interesting that 2 dimensional atom configuration with 3 dimensional interaction has spontaneous magnetization at finite temperatures.

In this article, Curie temperatures for Fe, Co and Ni mono-layers are evaluated by the first principle band calculation in order to study the possibility of the large magnetic moment at room temperature. Exchange integral is calculated in the first principle calculation, and Curie temperature is estimated in the spin molecular field approximation, using a linear relationship between Curie temperature and exchange integral.

2. METHODS

Magnetic moments and Curie temperatures of Fe, Co and Ni mono-layers are calculated, in comparison to the bulk. Figure 1 shows the atom configuration of the mono-layer, which is supposed to be located on the (001) surface of the fcc-Ag [4]. The mono-layers are free standing and the Ag has no effect on the electronic structure of the mono-layer.

The electronic structures were calculated within the framework of the local density approximation using the LMTO-ASA method [5], which has the advantage of treating large numbers of atoms and being a reliable method. The periodic super-lattice which consists of mono-layer with separation of 5 vacant layers are used for the conventional band calculation method. It was confirmed previously that vacant layers more than four layers are distant enough to ignore the effects from the adjacent mono-layers. In order to calculate the Curie temperature accurately, there needs a large number of neighbor atoms around an

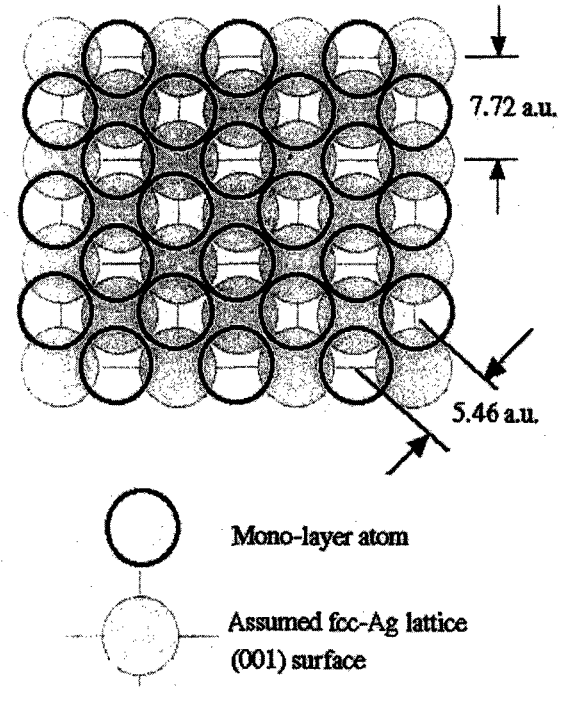

Fig. 1 Atom configuration of the mono-layer, which is supposed to be located on the (001) surface of the fcc-Ag.

atom in the unit cell. The unit cell of the mono-layer with vacant separation used for the calculation is shown in Fig. 2.

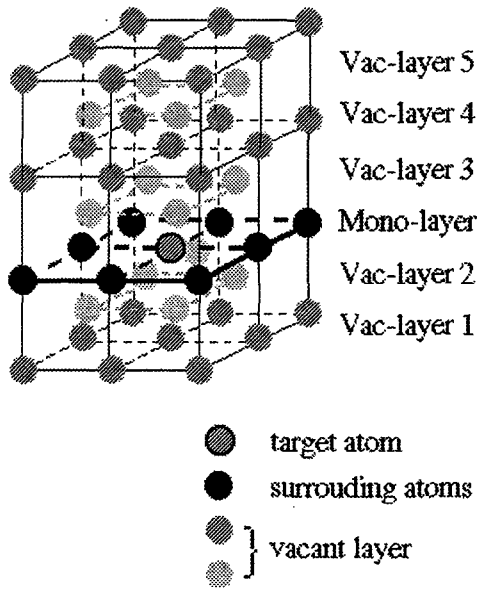

Fig. 2 Unit cell of the mono-layer used for the calculation.

We used the Vosko-Wilk-Nusair form for the exchange-correlation potential [6]. The core electrons for the atom were treated by the frozen-core approximation. The wave functions for the valence electrons were expanded in the basis consisting of *s*, *p*, *d* partial waves. The number of *k*-points was 445 for the mono-layer in the irreducible Brillouin zone, which is equivalent to an eighth of the first Brillouin zone. The *k*-points of 462 and 286 were used for fcc and bcc structure, respectively. The convergence criteria for the energies and magnetic moments at each of the sites were that they are less than 10^{-6}Ry and $10^{-3}\mu_B$ after iteration of the self-consistent calculation.

The calculation leads that the magnetic moments of the mono-layers are larger than those of the bulk as shown in the section 3. However, absolute zero temperature is assumed in the ordinary first principle calculation. So, one is not able to obtain physical values in finite temperatures. In order to investigate the realization of the large magnetic moment at room temperature, Curie temperatures of Fe, Co and Ni mono-layers are calculated. According to Liechtenstein, Curie temperature was evaluated by the first principle calculation. First, ferromagnetic state is assumed to be a ground state. As a perturbation, a slight rotation of the spin at one site denoted as 0 is induced. Exchange integral J_0 can be obtained from energy variation between ground state and perturbed state [7],

$$J_0 = -\frac{1}{4\pi}\int^{E_f} dE \,\mathrm{Im}\, Tr\{\Delta_0(g_\uparrow^{00} - g_\downarrow^{00}) + \Delta_0 g_\uparrow^{00}\Delta_0 g_\uparrow^{00}\}$$
$$\Delta_0(E) = P_{0\uparrow}^{\alpha}(E) - P_{0\downarrow}^{\alpha}(E) \qquad (2.1)$$
$$g(E)_{\sigma} = [P_{\sigma}^{\alpha}(E) - S^{\alpha} - i\varepsilon]^{-1}$$

P^{α} and S^{α} are the potential function P and the structure constant S modified by the screening matrix α [8]. The small factor ε was induced to avoid a divergence in the calculation, although it was not essential. Then Curie temperature T_c is estimated from

$$T_c = 2J_0 / 3k_B , \qquad (2.2)$$

by the spin molecular field approximation.

It is mentioned that more than three nearest neighbor atoms are necessary to calculate the exchange integral accurately in order to obtain a small energy difference between ferromagnetic ground state and its perturbed state. The calculated value is often negative when the first nearest neighbor atoms are few for the calculation. There exists surrounding atoms of the mono-layer and a target atom for the calculation at site 0 in the unit cell as shown in Fig.2.

3. RESULTS

Figure 3 shows the magnetic moments of 3*d* and 4*d* transition metal mono-layers, as well as Fe, Co and Ni. The magnetic moments calculated for the bulk Fe, Co and Ni are also indicated. Generally speaking, the magnetic moment of the mono-layer is higher than that of the bulk. Especially, Ru shows a finite magnetic moment, although the bulk elements of the 4d transition metal are not ferromagnetic. The V and Cr mono-layers also show large magnetic moments in contrast to the nonmagnetic bulk states. The magnetic moment of the Fe mono-layer is $3.02\mu_B$, which is about $1\mu_B$ larger than that of the Fe bulk. The density of

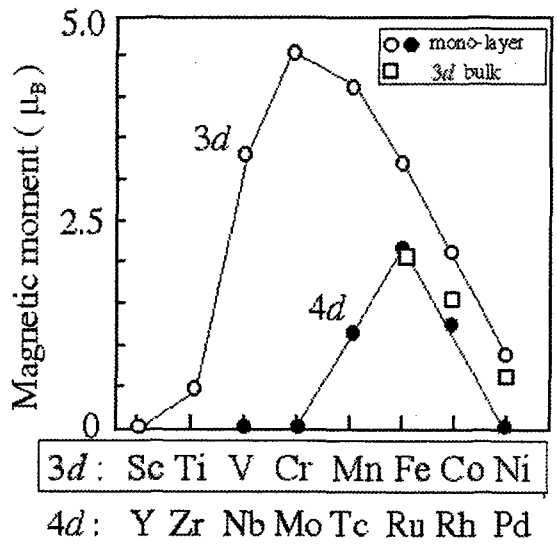

Fig. 3 Magnetic moments of 3d and 4d transition metal mono-layers

states of the Fe mono-layer is shown in Fig. 4, in comparison to the bulk. The band width of the mono-layer is reduced by the localization of the two dimensional configuration for all transition metals typically as indicated for Fe.

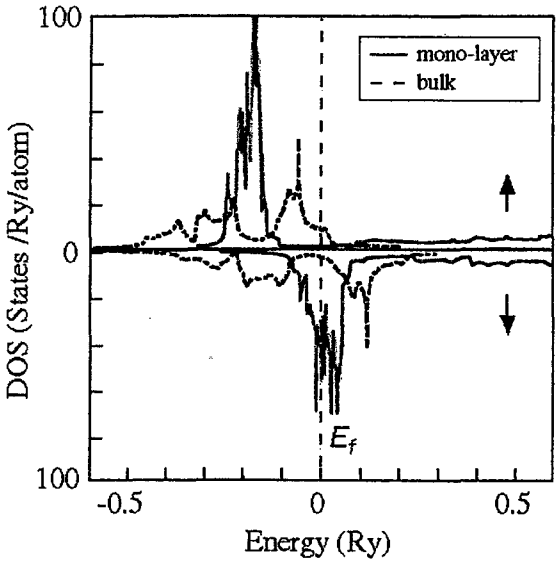

Fig. 4 Density of states of the Fe mono-layer and the bulk Fe.

The Curie temperatures of Fe, Co and Ni were calculated to study the possibility of the large magnetic moments at finite temperatures. The relative relation seems to be reliable between calculated Curie temperatures of the mono-layer and the bulk although the absolute value of the Curie temperature has some ambiguity in the calculation. The calculated value of the bulk can be compared with the experimental value of Fe, Co and Ni. The reason we focused on the 3*d* ferromagnetic elements Fe, Co and Ni is also due to the requirement of the enormous computer power.

Table I. Curie temperatures of the bulk bcc-Fe, fcc-Co and fcc-Ni.

element	Curie temperature (K) this work	exp.
Fe	628	1044
Co	1110	1388
Ni	389	631

The Curie temperatures of the bulk bcc-Fe, fcc-Co and fcc-Ni are shown in Table I. The calculated values are little lower than the experimental values. However, the calculated values seem to be reasonable, because they have the same sign and the same order as the experimental values and a relationship between atomic elements is satisfied. The calculated Curie temperatures of the Fe, Co and Ni mono-layers are shown in Table II in comparison to the experimental value. The calculated Curie temperatures of the mono-layer Fe, Co, and Ni are 211 K, 189 K, 268 K, respectively,

Table II. Curie temperatures of the Fe, Co and Ni mono-layers

mono-layer	Curie temperature (K) this work	exp.
Fe	211	173~303 [9]
Co	189	207 [10]
Ni	268	197 [10]

4. DISCUSSION

Magnetic moments and Curie temperatures of Fe, Co and Ni mono-layers are calculated, in comparison to the bulk. The magnetic moment of the mono-layer is higher than that of the bulk, because the band width of the mono-layer is reduced by the localization of the two dimensional configuration as shown in Fig.4 for Fe. The increment of the density of states around the Fermi-energy enhances the polarization.

However, the Curie temperatures of the mono-layer are lower very much than those of the bulk. The experimental values of the Curie temperature of the Fe very thin layer on Ag [9] and Co, Ni very thin layers on Cu [10] are the same order as our calculated value indicated in Table II. It is estimated that the mono-layer does not have a high magnetic moment at room temperature, even though the mono-layer shows higher magnetic moment than the bulk at zero temperature.

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