Computational investigation of stable formation condition for $\text{Fe}_x\text{Ni}_{1-x}$ alloy films on paramagnetic substrate

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Abstract. The adsorption process of ultrathin ferromagnetic Fe and Ni alloy film on a nonmagnetic metal substrate is studied by the variational spin-density functional method. The conditions for the stable formation of ferromagnetic films with respect to island adsorption are discovered with a change of values of the coverage parameter and alloy component concentration. The equilibrium values of vacuum gap and film thickness are determined from the minimum of the total interfacial energy. The energy characteristics of films $\text{Fe}_x\text{Ni}_{1-x}$ on close-packed surfaces W, Cu and Ag with effects of magnetic ordering are calculated for different temperatures.

1. Introduction
The amazing properties of thin magnetic alloy films are the subject of great research interests [1–3], which are determined by many their applications in microelectronics and computer technology. For example, permalloy $\text{Fe}_{20}\text{Ni}_{80}$ is used in transformer laminations and magnetic recording heads. In this work, the spin density functional method was used for description of the temperature and ferromagnetic ordering influence on the adsorption properties of $\text{Fe}_x\text{Ni}_{1-x}$ alloy films onto the surface of nonmagnetic materials. For theoretical description we applied the method of multi-parameter test functions [4] the parameters of which are determined by the numerical minimization procedure of the interfacial energy functional in the subsurface region.

The experimental investigations of ultrathin magnetic films [5] show that the magnetic properties of system Fe/Cu are characterized by two-dimensional Ising model type, and Fe/Au by two-dimensional XY model type. Therefore, in this work we use for description of temperature dependence for the relative magnetization $m(T)$ the results of renormalization-group finite-size consideration for XY type systems.

2. Model and methods
Consider a semi-infinite metal with an average charge density $n_1$, which is limited by an infinite flat surface and occupies the region $z < -D$. The adsorbate film with a charge density $n_2$ occupies the region $D < z < D + h$, where $h$ is the thickness of the film. Between the adsorbent and adsorbate in this model there is a vacuum gap of width $2D$.

Define the interfacial interaction energy per unit area of contact as an integral over $z$ from the bulk density of the free energy of the electron gas:
The volume density of the free energy of the inhomogeneous electron gas can be represented as a gradient expansion:

\[ f[n(z)] = w_0[n(z)] + w_2[n(z), |\nabla n(z)|^2] + w_4[n(z), |\nabla n(z)|^4] - T(s_{id} + s_{order}), \]

where

\[ w_0[n_{\uparrow\downarrow}(z)] = w_{kin} + w_{cul} + w_{ex} + w_{cor} \]

is the energy density of the homogeneous electron gas, including successively the kinetic, Coulomb, exchange and correlation energy, \( s_{id} \) and \( s_{order} \) are the entropic contributions to the free energy taking into account the temperature changes in the entropy for the electron gas and the effects of magnetic ordering in the electronic subsystem, respectively. Detailed expression for all of this contributions given in [4]. The densities \( n_+ \) and \( n_- \) can be expressed in terms of relative magnetization \( m = M(T)/M(T = 0) \) of electrons as follows:

\[ n_{\uparrow\downarrow}(z) = \frac{n(z)(1 \pm m(z))}{2} \]

We are taking into consideration the inhomogeneous spatial distribution of magnetization in surficial region in the following view:

\[ m(z) = \begin{cases} \frac{1}{2}m_0e^{\beta(z-D)}[1 - e^{-\beta h}], & z < D \\ 0.5m_0[1 - 0.5e^{-\beta(z-D)} - 0.5e^{\beta(z-D-h)}], & D < z < D + h \\ 0.5m_0e^{\beta(z-D)}[e^{\beta h} - 1], & z > D + h \end{cases} \]

where \( m_0 \) which is determined by two-dimensional XY model type.

\[ m_0(T) \approx \left[ \frac{T_c^{(s)}(x) - T}{T_c^{(s)}(x)} \right]^{0.23} \]

The critical temperature \( T_c^{(s)} \) of magnetic ordering of the monatomic film depends on the parameter \( \Theta \):

\[ T_c^{(s)}(\Theta) \approx \Theta T_c^{(b)} \frac{z_{surf}}{z_{bulk}}, \]
Figure 1. The dependence of total interfacial energy for Fe$_x$Ni$_{1-x}$/Cu(111) and Fe$_x$Ni$_{1-x}$/W(110) systems on vacuum gap at T=100K and $x = 80\%$ for different coverage parameter $\Theta$.

where $z_{\text{surf}}$ is the number of nearest neighbors in the ferromagnetic film, and $z_{\text{bulk}}$ in the bulk ferromagnet. In this case, for the critical temperature of magnetic ordering of the bulk ferromagnet alloys we used its experimental dependence on iron concentration [5].

The inclusion of the discreteness in the ion distribution leads to the corrections to the electrostatic interaction energy due to the ion–ion and electron–ion interactions. As a result, the interfacial interaction energy can be written in the form

$$
\sigma = \sigma_0 + \sigma_{ei} + \sigma_{ii},
$$

where $\sigma_0$ is the contribution of the electronic system in the framework of the jellium model, $\sigma_{ii}$ is the correction to the ion–ion electrostatic interaction energy, and $\sigma_{ei}$ is the correction to the energy associated with the difference in electrostatic interactions of electrons with discrete ions and the uniform "jelly" background. The parameters of the discreteness in the ion distribution presented in table 1 as Ashcroft pseudopotential cutoff radius $r_c$, charge of the ions $Z$ and interplanar distance $d$ also depend on the concentration of alloy components:

$$
r_c = \left(\frac{xZ_{Fe}(r_{cFe})^3 + (1-x)Z_{Ni}(r_{cNi})^3}{xZ_{Fe} + (1-x)Z_{Ni}}\right)^{1/3},
$$

$$
Z = xZ_{Fe} + (1-x)Z_{Ni}, \quad d = xd_{Fe} + (1-x)d_{Ni}.
$$

We have developed a multiparameter version of the functional of electron density with test functions which take into account, along with the heterogeneity of the distribution of electron density, the existence of the equilibrium distance between the adatoms and the substrate determined from the condition of the minimum interfacial energy of interaction [3]. Knowing the interfacial energy of the metal contact with the film, as well as the surface energy of each component of a bimetallic system, it is easy to find the energy of adhesion of the system as a work that needs to be carried out to remove the substrate and the film from each other to infinity. We determine the adsorption energy as the specific adhesion energy per adsorbed atom

$$
E_{ads}(\Theta) = \min_{\beta,h,D} \left[ \frac{\sigma(\infty) - \sigma(\beta,h,D,T,\Theta)}{n_s} \right],
$$

where $n_s$ is the surface atomic concentration in the film.
Table 2. Conditions for formation of stable Fe\textsubscript{x}Ni\textsubscript{1-x}/Cu(111) film on Ag substrate.

| T, K | Θ | X, % |
|------|----|------|
| 0    | 0.9 | [40] |
| 100  | 0.7 | [100] |
| 0.8  | 0.8 | [60-100] |
| 0.9  | 1   | [40] |
| 200  | 0.8 | [60-100] |
| 0.9  | 1   | [80] |

Table 3. Conditions for formation of stable Fe\textsubscript{x}Ni\textsubscript{1-x}/Cu(111) film on Cu substrate.

| T, K | Θ | X, % |
|------|----|------|
| 0    | 0.7 | [30-100] |
|       | [0.8-1] | All concentrations |
| 200  | 0.8 | [50-100] |
|       | [0.9-1] | All concentrations |
| 600  | 0.9 | [17-100] |
|       | 1   | All concentrations |

Table 4. Conditions for formation of stable Fe\textsubscript{x}Ni\textsubscript{1-x}/Cu(111) film on W substrate.

| T, K | Θ | X, % |
|------|----|------|
| 0    | 0.5-0.7 | All concentrations |
| 0.8  | 0.9 | [0-60] |
| 1    | [0]  |
| 100  | 0.7-0.8 | All concentrations |
| 0.9  | 1   | [0-20] |
| 200  | 0.7 | [40-100] |
| 0.8  | 1   | [20-100] |
| 0.9  | [0-60] |

3. Results of calculations

The equilibrium vacuum gap $D = D_{\text{min}}$, which simulates the effective influence of the atomic roughness of the substrate and the equilibrium film thickness $h = h_{\text{min}}$ are determined from the minimum of the total interfacial energy $\sigma(\beta, h, D, T, \Theta)$. As shown in Figure 1, minimum of $\sigma(\beta, h, D, T, \Theta)$ is disappears for some values of the parameter $\Theta$ or $\sigma(\beta_{\text{min}}, h_{\text{min}}, D_{\text{min}}, T, \Theta)$ takes negative values. This approach makes it possible to predict the formation of stable Fe\textsubscript{x}Ni\textsubscript{1-x} coatings or to reveal their absence (the case of island adsorption) in depending on concentration $x$. Conditions for formation of stable Fe\textsubscript{x}Ni\textsubscript{1-x}/Cu(111) film on different substrates, obtained from our calculations, are given in Tables 2-4.

We present the results of calculations of adsorption energy for alloy film on Cu, W and Ag substrates in Figures 2, 3. Calculations show that for all substrate the adsorption energy $E_{\text{ads}}$ of film decreases with increasing $\Theta$ in the range of stability of film and with increase of temperature. Also, $E_{\text{ads}}$ significantly increases with growth of iron concentrations $x$ in alloy Fe\textsubscript{x}Ni\textsubscript{1-x}.

Results of calculations, given in Figure 4, demonstrate that the film thickness $h_{\text{min}}$ decreases with an increase in the coverage parameter $\Theta$ in the range of stability of film and keeps a constant value for different iron concentrations.
4. Conclusions
The calculation of the adsorption energy of Fe$_x$Ni$_{1-x}$ alloy film on the close-packed surface faces Cu, W and Ag substrates are carried out in dependence on parameter of coverage $\Theta$ by the spin-density functional method for different temperatures. The stable monolayer film on Cu(111) substrate is formed only for values of $\Theta \geq 0.7$ at $T = 0K$ and for $\Theta \geq 0.8$ at $T \geq 100K$. For samples with threshold values of $\Theta$, the stable films are realized for high concentration of Fe. Stable film Fe$_x$Ni$_{1-x}$ on W(110) substrate is realized for $\Theta > 0.7$($T > 0K$), but for $\Theta = 0.9 - 1.0$ it is formed stable state for low concentrations of Fe only. The behavior of a interfacial energy for Ag is similar to W, that cause losing stability for large values of $\Theta$.

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References
[1] Tryputen L, Guo F, Liu F, et al 2015 Phys. Rev B 91 014407
[2] Ustinov V V, Milyaev M A and Naumova L I 2014 SPIN 04 1440001
[3] Mamonova M V, Morozov N S, Prudnikov V V 2009 Physics of the Solid State. 51 2169
[4] Mamonova M V, Prudnikov V V, and Prudnikova I A 2013 Surface Physics: Theoretical models and experimental methods. (CRC Press: Tailor and Francis Group) p. 384
[5] Schumann F O, Wu S Z, Mankey G J and Willis R F 1997 Phys. Rev. B 56 2668