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Magnetization on rough ferromagnetic surfaces

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Using Ising-model Monte Carlo simulations, we show a strong dependence of surface magnetization on surface roughness. On ferromagnetic surfaces with spin-exchange coupling larger than that of the bulk, the surface magnetic ordering temperature decreases toward the bulk Curie temperature with increasing roughness. For surfaces with spin-exchange coupling smaller than that of the bulk, a crossover behavior occurs: at low temperature, the surface magnetization decreases with increasing roughness; at high temperature, the reverse is true.

The magnetic properties of ferromagnetic surfaces and interfaces have been extensively investigated because of their potential impact on magnetic recording devices.[1] A large enhancement of magnetic moments at the surface of ferromagnetic materials is predicted by band structure theory.[2–4] The enhancement is attributed to the reduced dimensionality and coordination of surface atoms.[2–4] At ordinary temperatures, however, the fluctuations in the surface magnetization can be large enough to mask the possible differences in the magnetic moments of bulk and surface.[5,6] Although a direct measurement of surface magnetic moments remains a challenging problem, recent experiments have shown that the surface magnetization is different from that of the bulk. For example, in $4f$ rare-earth films, an enhanced surface magnetic ordering temperature has been observed,[7–9] confirming the earlier theoretical predictions.[10]

Most earlier studies on surface magnetism are based on the assumption that ferromagnetic surface is morphologically perfectly smooth (ideal bulk termination). Real films, however, have a rough surface. The atomic heights of surface atoms can differ by a few atomic spacings because of the formation of a variety of surface defects (steps, islands, vacancies, etc.). Such surface roughness are expected to affect magnetism. Therefore, establishing the relationship of surface/interface magnetic properties to surface/interface roughness is not only of fundamental interest but is also essential for development of new magnetic devices using magnetic multilayers.

There is an increasing recent interest in understanding the effect of surface/interface roughness on magnetic properties. Experimentally, it has been shown that interface roughness may destroy interlayer magnetic coupling between thin films in a multilayer structure.[11] The dynamic response of a surface/interface to an external field can also be altered by roughness. Surface roughness changes the shape of hysteresis curves.[12] Spin-polarized element-specific diffuse x-ray diffraction from ferromagnetic surfaces/interfaces shows that the magnetization at a surface/interface is modified by the surface/interface roughness.[13] Theoretical modeling and simulations[14] show that the critical behavior of edges is different from that of surfaces. Although these studies have begun to recognize the importance of roughness in surface magnetism, a quantitative understanding of their relationship is still far from complete. In this Brief Report, we present a systematic theoretical study to establish the relationship of surface roughness to surface magnetization and its temperature dependence, by introducing the surface roughness in a systematic manner. We find that surface roughness strongly affects surface magnetization, not only changing the surface magnetic ordering temperature but also modifying the magnitude of magnetization in a complex fashion.

We simulate the magnetization at different temperatures for surfaces with controlled morphologies, using the Ising-model Monte Carlo method. The simulations are performed on a simple-cubic spin lattice slab with two free surfaces (see Fig. 1). Periodic boundary conditions are applied in the x and y directions. We use a sample size of $20\times20\times20$ sites, which shows good convergence with respect to sample size.[15] Extra layers are added to create surface morphologies with different degrees of roughness. We use the nearest-neighbor Ising model to represent the interactions between the localized spins. The surface spin-exchange coupling can be chosen to be the same as or different from the bulk spin-exchange coupling. We used 18 000 Monte Carlo steps in each simulation and the results are averaged over 5000 steps after equilibration.

In incorporating surface roughness, surface layers are systematically modified by either introducing steps (vicinal surface with smooth terraces separated by equally spaced monatomic steps) or displacing surface atoms to lattice sites at random heights (one large rough terrace). Figure 1 shows schematically these two typical situations for a rough surface. The first surface contains steps as the only roughness features; the second surface corresponds to a diffusion-limited growth with the resultant rough growth front following a Poisson distribution.

In general, the roughness of a surface (interface) can be quantitatively characterized by its rms roughness (σ), lateral correlation length (ξ), and fractal exponent (h).[17] For a vicinal surface, the rms roughness for a given sample size ($2L$) is given by $\sigma=(\sqrt{3}/6)L\tan\theta$, which increases linearly with the tangent of the miscut angle θ. The larger the miscut angle, the higher the step density, and then the rougher the surface. The roughness is uniformly distributed with an infinite lateral correlation length. For the random surface, the rms roughness is numerically evaluated for each constructed surface. The lateral correlation length is relatively short,

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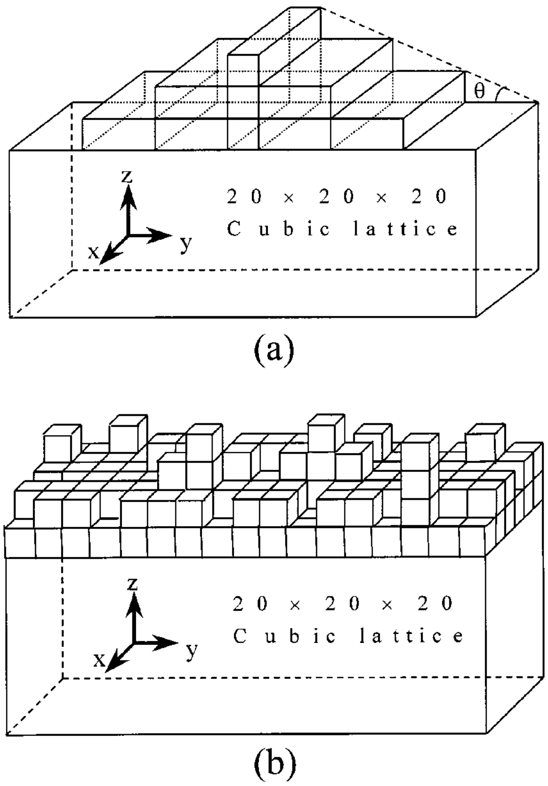

FIG. 1. Schematic views of the two types of lattices used in the Monte Carlo simulations. (a) Vicinal surface, (b) randomly rough surface.

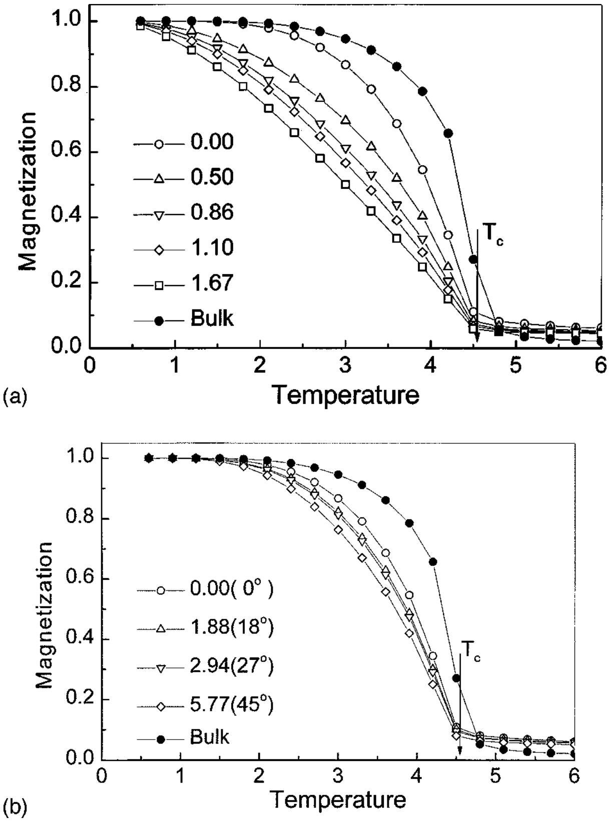

FIG. 2. Surface magnetization as a function of temperature in vicinal surfaces and random surfaces, as shown in Fig. 1, using $J_s = J_b$. (a) Randomly rough surface; the symbols denote different rms roughness values in units of a lattice spacing (a value of 1.67 is equivalent to about one lattice spacing of mean roughness). T_c marks the bulk Curie temperature. (b) vicinal surface; the numbers in brackets are miscut angles. The other notations are the same as in (a).

compared to the miscut surface. These two classes of rough surfaces represent extremes in types of roughness and in lateral correlation.

Figure 2 shows the surface magnetization, the average over all spins on the top-layer surface sites, as a function of temperature when the surface spin exchange coupling (J_s) is set equal to the bulk spin exchange coupling (J_b). The surface magnetization decreases with increasing surface roughness for all temperatures. For the random surface [Fig. 2(a)], the surface magnetization decreases as much as 30% at some temperatures, as the surface rms roughness increases from zero to about one lattice spacing (a typical value for an actual film). We believe this amount of change in surface magnetization should be experimentally observable. Recently, the oxygen induced reduction of surface magnetization of Gd(0001) has been measured by spin-resolved photoemission.[19] Similar experiments could be done to verify our prediction by measuring the change of surface magnetization for a variety of vicinal samples or ion-bombarded surfaces.

Qualitatively similar results are obtained for the vicinal surface [Fig. 2(b)], but for the same rms roughness, the magnetization is smaller in the random surface than in the vicinal surface. Because the lateral correlation length of the random surface is much smaller than that of vicinal surface, the random surface is in effect rougher than the vicinal surface for the same rms roughness.

Decreasing magnetization with increasing roughness can be partly understood within the arguments of mean-field theory, in terms of modification of the effective local field of surface spins due to the surface roughness. By assuming $J_s = J_b$, we treat the surface spins and bulk spins the same. The effective local field of a surface spin is simply proportional to its coordination number (see discussion below). As the surface gets rougher, the average coordination of surface spins decreases, and consequently, the surface magnetization decreases. Previous band structure calculations[4] also show that the magnetic moment of a ferromagnetic system displays a strong dependence on the local order of the atomic structure.

When J_s differs from J_b, the behavior of the surface magnetization with changing roughness is much more complex and interesting. J_s may be larger or smaller than J_b. The reduced atomic coordination at a surface produces a narrower band width and hence a larger magnetic moment,[2–4] favoring $J_s > J_b$. On the other hand, the surface lattice spacing can be larger than the bulk lattice spacing, leading to a weaker spin-spin interaction and favoring $J_s < J_b$. In $4f$ rare-earth films, e.g., Gd(0001), a surface magnetic ordering temperature higher than the bulk Curie temperature has been observed,[9] indicating the system has $J_s > J_b$.[16] For $3d$ transition metals, indirect evidence from clusters points to the likelihood of $J_s < J_b$.[6,18] We therefore consider both possibilities.

Figure 3 shows an example of the dependence of magnetization on temperature for various miscuts of the vicinal

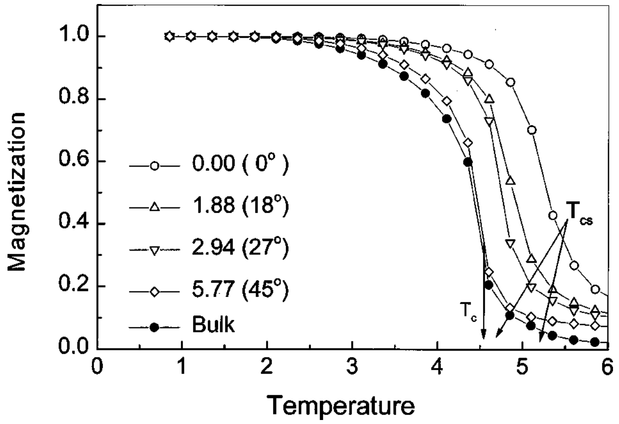

FIG. 3. Surface magnetization as a function of temperature in vicinal surfaces, using $J_s=2J_b$. T_{cs} marks the surface ordering temperature. Other notations are the same as in Fig. 2(a). Randomly rough surfaces show the same behavior.

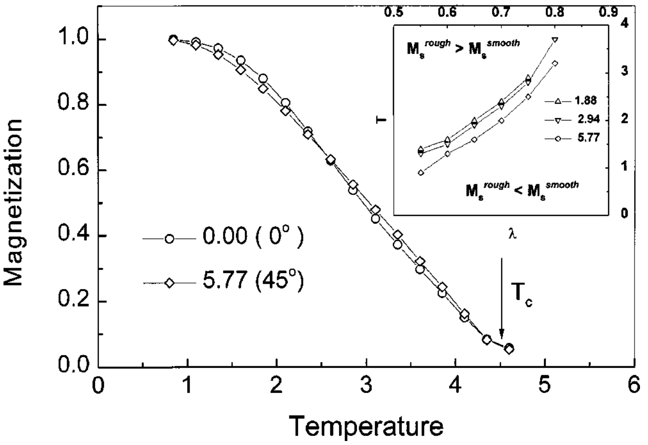

FIG. 4. Surface magnetization as a function of temperature in vicinal surfaces, using $J_s=0.65J_b$ for two values of surface roughness, a smooth surface and a 45° miscut surface. Notations are the same as in Fig. 2(a). Randomly rough surfaces show the same behavior. The inset shows the dependence of the crossover temperature (relative to the smooth surface) as a function of the relative strength of $J_s(\propto\lambda^2)$ for various values of roughness. The crossover temperature increases with increasing J_s and decreasing roughness.

surface for $J_s>J_b$. The surface magnetization always decreases with increasing surface roughness, as for $J_s=J_b$ (Fig. 2). For sufficiently high values of J_s, the surface magnetization can be higher than the bulk magnetization in the smooth surface but becomes smaller than the bulk magnetization as the surface gets rough. In agreement with previous simulations,[16] we find that the surface has an ordering temperature above T_c. Most importantly, however, we are able to show that the surface ordering temperature (T_{cs}) decreases toward T_c as the surface gets rougher. The enhancement of surface ordering temperature has been observed in Gd(0001),[9] and the enhancement is seen only in clean films. The disappearance of the enhancement on contaminated films is speculated to be due to surface roughness.[9] Our theory shows that surface roughness can indeed lower surface ordering temperature without a need for impurities.[19] Experiments using clean samples with different degrees of surface roughness, such as vicinal surfaces with different miscuts, can confirm our prediction.

A more complex behavior occurs when $J_s<J_b$. As an example, Fig. 4 shows the dependence of the surface magnetization on temperature for $J_s=0.5J_b$ in a smooth and a 45° miscut rough surface. Both surfaces start ordering at the bulk Curie temperature because the bulk ordering can induce the surface to order when $J_s<J_b$. The relationship of magnetization to roughness displays a crossover as a function of temperature. At low temperatures, the surface magnetization is *higher* in the smooth surface, while at higher temperatures, the surface magnetization is *lower* in the smooth surface. In Fig. 4, the crossover appears at about $0.5T_c$. In general, it depends on J_s and surface roughness. As shown in the inset of Fig. 4, for a given surface roughness, the crossover temperature increases monotonically with increasing J_s. For a given J_s, the crossover temperature also changes slightly with surface roughness, decreasing with increasing roughness. Although the crossover is not a phase transition, the inset of Fig. 4 resembles a phase diagram: in the upper left region, the surface magnetization *increases* with increasing surface roughness; in the lower right region, the surface magnetization *decreases* with increasing surface roughness. The fact that the crossover appears over a large range of values of J_s makes an experimental observation of this behavior plausible even though the differences in magnetization shown in Fig. 4 are not large.

In order to understand the origin of this ''crossover'' behavior, we may apply mean-field theory to the miscut surface. Within the nearest-neighbor Ising model, the effective local field of a surface spin M_s can be written as

$$M_s=z_sJ_sm_s+z_bJ_bm_b, \tag{1}$$

where z, J, and m denotes, respectively, the number of nearest-neighbor spins, the exchange coupling, and the average magnetization. Subscripts s and b indicate surface and bulk, respectively. Assuming $J_b=J$ and $J_s=\lambda^2J_b$, the surface magnetization is derived as[20]

$$m_s=\tanh\left[\frac{\lambda J}{k_BT}(\lambda z_sm_s+z_bm_b)\right], \tag{2}$$

where k_B is the Boltzmann constant. Now we compare two extreme surfaces: one with miscut angle 0°, perfectly smooth; one with miscut angle 45°, extremely rough. For the smooth surface, $z_s=4$ and $z_b=1$ in a simple cubic lattice. Equation (2) becomes

$$m_s^{\text{smooth}}=\tanh\left[\frac{\lambda J}{kT}(4\lambda m_s+m_b)\right]. \tag{3}$$

For the 45° miscut surface, $z_s=2$ and $z_b=2$. Equation (2) reduces to

$$m_s^{\text{rough}}=\tanh\left[\frac{2\lambda J}{kT}(\lambda m_s+m_b)\right]. \tag{4}$$

For $0.5<\lambda<1.0$, it is easy to show that as $T\rightarrow 0$, m_s, $m_b\rightarrow 1$. From Eqs. (3) and (4) we have

$$m_s^{\text{smooth}}=1-2\exp\left[-\frac{(4\lambda+1)\lambda J}{3kT}\right] \tag{5}$$

and

$$m_s^{\text{rough}} = 1 - 2\exp\left[-\frac{2(\lambda+1)\lambda J}{3kT}\right]. \tag{6}$$

As $T \to T_c$ ($T_c = 6J/k$ is the mean-field bulk Curie temperature), m_s, $m_b \to 0$; then

$$m_s^{\text{smooth}} = \frac{\lambda m_b}{6-4\lambda^2} \tag{7}$$

and

$$m_s^{\text{rough}} = \frac{\lambda m_b}{3-\lambda^2}. \tag{8}$$

As a result, $m_s^{\text{smooth}} > m_s^{\text{rough}}$ at low temperature but $m_s^{\text{smooth}} < m_s^{\text{rough}}$ at high temperature, leading to the crossover behavior.

In conclusion, we have investigated the behavior of surface magnetization on a rough surface. We show that the surface magnetic properties sensitively depend on the surface roughness, suggesting that earlier results, which have neglected roughness, may need to be reinterpreted. Specifically, we establish that for surfaces with spin exchange coupling larger than the bulk, the surface magnetic ordering temperature (T_{cs}) is higher than the bulk Curie temperature (T_c) if the surface is perfectly smooth, but decreases toward T_c as the surface gets rougher. The surface magnetization decreases at all temperatures with increasing roughness and the change in surface magnetization is large enough so that it should be measurable. These conditions apply also for surfaces with spin exchange coupling equal to the bulk, except that T_{cs} always equals T_c. For surfaces with spin exchange coupling smaller than the bulk, a crossover behavior exists in the relationship of magnetization to roughness: the magnetization decreases at low temperature but increases at high temperature as the surface roughness increases. Measurements on different clean vicinal surfaces could demonstrate a dependence of the magnetization on roughness below T_c, especially for materials with $J_s \geqslant J_b$ and the dependence of T_{cs} on roughness for materials with $J_s > J_b$. The crossover behavior for materials with $J_s < J_b$ is possibly observable in $3d$ metal films or clusters. Finally, although all the simulations are done here with a surface model, we expect similar results at rough interfaces in a multilayer structure.

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