Monte Carlo study of magnetic order at ferromagnetic and antiferromagnetic surfaces: Implications for spin-polarized photoelectron diffraction

F. Zhang
Department of Physics, University of California at Davis, Davis, California 95616
and Materials Sciences Division, Lawrence Berkeley Laboratory, Berkeley, California 94720

S. Thevuthasan
Environmental Molecular Sciences Laboratory, Pacific Northwest Laboratory, Richland, Washington 99352

R. T. Scalettar and R. R. P. Singh
Department of Physics, University of California at Davis, Davis, California 95616

C. S. Fadley
Department of Physics, University of California at Davis, Davis, California 95616
and Materials Sciences Division, Lawrence Berkeley Laboratory, Berkeley, California 94720
(Received 28 September 1994)

We have used Monte Carlo simulations on simple-cubic Ising lattices with modified surface interaction parameters to model phenomenologically the temperature dependence of magnetic order near ferromagnetic and antiferromagnetic surfaces. These results are also discussed in connection with previous experiments suggesting surface-specific magnetic transition temperatures for semi-infinite systems, with special emphasis on spin-polarized photoelectron diffraction as a probe of short-range magnetic order. The calculated spin-spin correlation functions show no evidence of a high-temperature transition in short-range magnetic order. However, over a plausible range of choices for the surface interaction parameters, these correlation functions do show distinct surface transitions in long-range magnetic order that can be well above $T_{N,\mathrm{bulk}}$ for antiferromagnets (both frustrated and nonfrustrated) and well above $T_{C,\mathrm{bulk}}$ for ferromagnets. Thus, prior spin-polarized photoelectron data from antiferromagnetic $KMnF_3$ and MnO may be explainable via such surface magnetic transitions, although further theoretical and experimental work are necessary to make this connection quantitative and definitive.

I. INTRODUCTION

The nature of magnetic order near surfaces and interfaces is a topic of high current interest, with experimental observations or theoretical predictions of Curie temperatures which may depend either on the thickness of an epitaxial ferromagnetic layer or may vary from the surface layer inward for a semi-infinite sample of homogeneous composition.[1] The temperature dependence of surface magnetic order is thus a key component of surface magnetism, particularly for systems of nanometer scale for which the fraction of surface atoms can become appreciable. Previous theoretical modeling of this temperature dependence has been carried out primarily for the surfaces of ferromagnetic systems using Monte Carlo and cluster variation methods.[2,3] In this paper, we have applied Monte Carlo modeling to both ferromagnetic and antiferromagnetic surfaces, for the later case considered systems without and with frustrated next-nearest-neighbor interactions; explored a broader range of relative interaction parameters; and considered the relationship of these results to both prior theoretical calculations[2,3] and previous experimental data that suggest surface magnetic order behavior different from the bulk.[4–6,8,9,11,12] Special emphasis is placed on prior experimental studies by spin-polarized photoelectron diffraction.[4–6,8,9] We first review the experimental data which appear to show a surface magnetic transition temperature different from the bulk for semi-infinite samples and the previous theoretical modeling of these phenomena, and then turn to our theoretical simulations of such effects.

A. Spin-polarized photoelectron diffraction studies on antiferromagnetic systems

Spin-polarized photoelectron diffraction (SPPD) has been proposed as a probe to study short-range magnetic order in both ferromagnetic and antiferromagnetic materials.[4–10] In its simplest form, SPPD makes use of multiplet-split core-level binding energies as internally referenced sources of spin-polarized electrons, with no external spin detector then being necessary. But if an external spin detector is used with a specimen possessing a net magnetization[9] and/or if spin-orbit-split levels are excited by circularly polarized radiation,[10] SPPD can be related to an external axis of electron spin polarization. Like the much more developed technique of photoelectron diffraction (PD) without spin resolution,[10] SPPD will be primarily sensitive to the first few spheres of neighbors

surrounding an emitter.[9,10] High surface sensitivity is also implied by the fact that SPPD must be carried out at electron kinetic energies of only 50–150 eV, in order to generate sufficiently strong magnetic scattering for detection.[9,10] So magnetic properties in the first few layers below a surface are investigated preferentially by this technique. Thus, the change in orientation of magnetic moments in ferromagnetic or antiferromagnetic materials can, in principle, be studied by analyzing the spin-polarized photoelectron diffraction intensities above a surface as a function of temperature. An experimental quantity which has been used to detect such changes in magnetic order is the so-called spin asymmetry S:[5–9]

$$S=100[(R_{\mathrm{LT}}-R_{\mathrm{HT}})/R_{\mathrm{HT}}]\% \ , \tag{1}$$

where R_{HT} represents the (spin-up):(spin-down) intensity ratio at HT, the highest-temperature data point in the series (assumed to be in the fully disordered or paramagnetic limit); and R_{LT} represents the same ratio for any lower-temperature data point (below HT). The spin asymmetry is thus defined to go to zero at the limit of HT.

The first experimental study of SPPD was reported by Sinkovic and co-workers.[4] They studied the antiferromagnetic surface of $KMnF_3(110)$ through the temperature dependence of the spin-up and spin-down multiplet peaks associated with Mn 3s core emission, and found that the experimental spin asymmetry S_{expt} went through an abrupt transition at a temperature that was about 2.7 times the bulk Néel temperature. They interpreted their results as an abrupt reduction in *short-range* magnetic order (SRMO) at this transition temperature, even though long-range order has completely disappeared well below this temperature. Subsequently Hermsmeier *et al.*[6,8] used SPPD to study another antiferromagnetic surface, MnO(001), and saw a similar effect in the spin asymmetries, but in this case with the transition temperature about 4.5 times the bulk Néel temperature. Figure 1 summarizes some of this experimental data for MnO. The spin asymmetry S_{expt} for this photoelectron emission direction (along the surface normal) exhibits a small but reproducible peak with a value of 33% at 120 K [which is also the bulk Néel temperature ($T_{N,\mathrm{bulk}}$)]. It then decreases monotonically to a value of about 21% at about 540 K—probably due to Debye-Waller effects. Over the narrow range 540–580 K, the spin asymmetry drops rapidly from 21% to about 7%, again a fully reproducible effect.[8] For temperatures above 580 K, S_{expt} again decreases more slowly, probably due to simple Debye-Waller effects.[8] This SPPD study of MnO again concluded that a sharp SRMO transition occurred at a temperature much higher than the bulk Néel temperature. However, it was not possible in either of these prior SPPD studies to fully rule out a surface magnetic transition in long-range order that, through the concomitant abrupt change in short-range order, would also affect the spin asymmetry seen in SPPD. Some sensitivity of SPPD to bulk long-range order is also suggested by the peak near T_N in Fig. 1.

Prior theoretical analyses of SPPD have not dealt specifically with the statistical mechanics of spins near

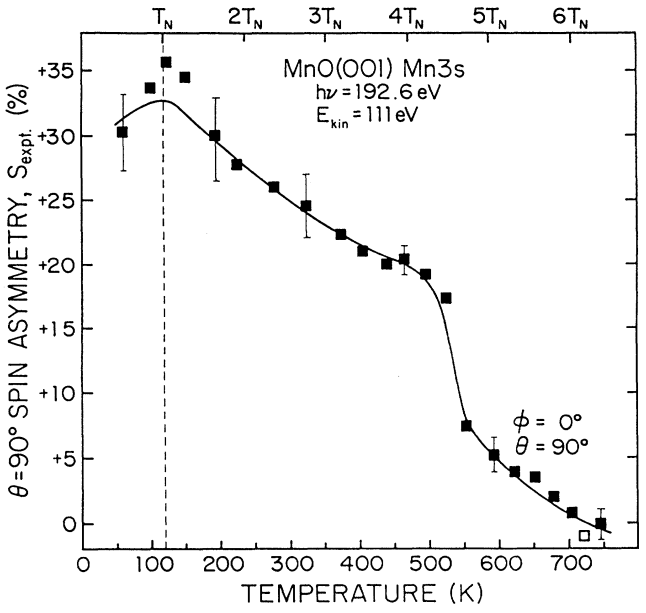

FIG. 1. Mn 3s experimental spin asymmetries S_{expt} with Mo $M\xi$ excitation are plotted as a function of temperature at a polar angle $\theta=90°$ (normal emission) and an azimuthal orientation of $\phi=0°$ ([010] azimuth). The temperature range covered is from $\simeq 50°$ below $T_{N,\mathrm{bulk}}$ to $\simeq 620°$ above it (from Ref. 8).

surfaces, beyond considering what configuration averages would be sensed by the experiment. Sinkovic and Fadley[5] considered only zero-temperature SPPD to simulate the spin-polarized photoelectron diffraction intensities from a small cluster. Friedman and co-workers[7] subsequently extended this theory to finite temperature, showing explicitly how the spin-polarized photoelectron diffraction intensity depends on averages of spin-spin correlation functions. They showed that the average of intensities at finite temperature is well approximated by the intensity calculated for a single configuration with all spins parallel (or antiparallel) to the emitter, but with effective scattering phase shifts involving averages over spin-spin correlation functions. They also showed that spin-spin correlation functions of the usual form and with the bulk transition temperature could not explain the steplike feature at the high temperature, even though the spin-spin correlation functions themselves showed abrupt changes near the bulk transition temperature. Finally, it has been shown that the overall change in S_{expt} across the high-temperature transition (e.g., 14% in MnO) is semiquantitatively predicted by spin-dependent diffraction calculations based only on results for zero temperature and a high-temperature paramagnetic limit.[8,9]

B. Other experiments on ferromagnetic systems

Two *ferromagnetic* surfaces for which the Curie temperature seems to be higher than the bulk Curie temperature have also been studied, using other experimental techniques.[11,12] Rau and Eichner[11(a)] first found evidence

for ferromagnetic order at Gd surfaces above the bulk Curie temperature, using deuteron electron capture spectroscopy. Weller *et al.*[11(b)] have used spin-polarized low-energy-electron diffraction and the magneto-optic Kerr effect to study epitaxial Gd(0001) films on W(110) and subsequently concluded that the surface transition temperature lies 22 K above the bulk Curie temperature of 293 K. This result has been confirmed by Tang *et al.*[11(c)] using spin-resolved electron spectroscopy, with a surface Curie temperature 60 ± 2 K above the bulk. In addition, Rau and Jin[12] have also previously studied the surface of Tb(0001) using electron capture, and have found a surface transition temperature that is 30 K above the bulk Curie temperature of 220 K.

C. Prior theoretical modeling of surface magnetic phase transition

Prior statistical mechanical simulations that predict surface transitions at significantly higher temperatures than the bulk have been performed mainly on simple-cubic ferromagnetic lattices with nearest-neighbor interactions only.[2,3] Binder and Hohenberg first studied the surface behavior of a semi-infinite simple-cubic Ising ferromagnet with nearest-neighbor interactions which could be modified in the surface layers with respect to those in the bulk. They did this both in mean-field theory and by means of high-temperature-series expansions,[2(a)] and found that, for sufficiently enhanced surface coupling, the surface ordered at a higher temperature than the bulk, and behaved like a two-dimensional (2D) Ising model above the bulk order temperature. The ranges over which the 2D behavior was seen were for the surface exchange integral $J_s > 1.25$ times the bulk exchange integral J_b in mean-field theory; and $J_s > 1.6J_b$ from high-temperature-series expansions. Binder and Landau later studied a simple-cubic Ising ferromagnet with a nearest-neighbor ferromagnetic exchange interaction J_b in the bulk and a nearest-neighbor antiferromagnetic exchange interaction J_s between surface spins, using Monte Carlo simulations.[2(b)] They investigated the ordering for a variety of values of $J_s/J_b < 0$ and for various temperatures and found that for $J_s < -2.01J_b$, the surface transition occurred at a higher temperature than the bulk transition. By studying the magnetization profile of the system, they also found that the magnetization under the surface layer was hardly affected by the antiferromagnetic ordering in the surface, and that the surface layer would be a very good approximation to a two-dimensional Ising antiferromagnet. Recently, Laudau and Binder[2(d)] reported more precise results of extensive Monte Carlo simulations of phase transitions and critical behavior at the surface of a simple-cubic Ising ferromagnetic model with nearest-neighbor exchange interactions only. By studying profiles of the magnetization and internal energy as a function of the distance from the surface, they extracted surface and bulk properties as a function of temperature and surface exchange interaction J_s. They found that the surface transition temperature $T_{C,\text{surf}}$ exceeded the bulk critical temperature $T_{C,\text{bulk}}$ when J_s/J_b was greater than 1.52. Schweika, Binder, and Landau[2(e)] studied surface-induced ordering in Ising models for frustrated face-centered-cubic alloys with antiferromagnetic nearest-neighbor coupling [$J(\text{NN}) < 0$] and ferromagnetic next-nearest-neighbor interactions [$J(\text{NNN}) > 0$]. Using Monte Carlo simulations with $J(\text{NNN})/J(\text{NN}) = -0.05$, they showed that the surface ordered at a high temperature than did the bulk. And they also noticed that if larger values of $J(\text{NNN})$ were used, the transition temperatures of the bulk increased faster than that of the free surface until eventually surface-induced ordering was either suppressed or was indistinguishable from the ordering of the bulk. Using the cluster variation method, Sanchez and Moran-Lopez[3] have also shown that a surface transition takes place at a higher temperature than the bulk when J_s/J_b is greater than 1.47 for (100) surfaces of simple-cubic lattices in the cubic approximation and greater than 2.25 and 1.778 for (100) surfaces and (111) surfaces of face-centered-cubic structures in the tetrahedron approximation, respectively.

As reviewed above, prior theoretical work deals primarily with the surface behavior of simple-cubic ferromagnetic Ising lattices with nearest-neighbor exchange interactions only, without considering the case of simple-cubic frustrated antiferromagnetic Ising lattices. In this paper, we will study the surface and near-surface behavior of both simple-cubic ferromagnets *and* antiferromagnets (both frustrated *and* nonfrustrated), in order to better understand the spin statistics behind such high-temperature transitions. Since SPPD, as well as electron capture by deuterons[11(a),12] and other surface-sensitive electron spectroscopies,[11(b),11(c)] are sensitive to spin-spin correlation functions on and near the surface, we will here also explore intralayer, interlayer, and overall spin-spin correlation functions on and near the surface in a simple Ising model with both nearest-neighbor (NN) *and* next-nearest-neighbor (NNN) interactions. We will also investigate the effects of varying degrees of enhancement of surface interactions on both surface and near-surface behavior, and the effects of varying relative strengths of next-nearest-neighbor interactions on the general behavior of near-surface spin-spin correlation functions.

In Sec. II, we give a brief description of the theoretical model and simulation method used. In Sec. III, our detailed results and analyses are presented. Here, we first study the ferromagnetic and antiferromagnetic Ising lattices with only a NN interaction, and then investigate frustrated antiferromagnetic Ising lattices with both NN and NNN interactions. Finally, in Sec. IV, we draw our conclusions and also make a few further remarks on the degree of applicability of such modeling, and promising directions for future work.

II. METHODS

A. Ising model with NN and NNN interactions

It is expected that magnetic order and critical behavior on a surface or near a surface will be different from that of the bulk for purely geometric reasons, namely the absence of the neighboring atoms above the surface, and the resultant lowering of the surface coordination number.

However, changes may also occur due to modified exchange interactions, and these changes may in turn be enhanced by surface relaxations or reconstructions of atomic positions (although we will not explicitly consider such geometry changes here). For example, Scholl *et al.*[13] have shown that the surface exchange integral J_s can be changed dramatically with respect to the bulk exchange integral J_b in different surface environments. They found that, by depositing a submonolayer of Fe on a clean Ni-Fe surface, J_s can be increased to as much as three times J_b.[13] A final surface effect is the reduction of the bandwidth and associated enhancement of local moments. This could be an important effect in itinerant models of surface magnetism.

We here adopt a three-dimensional simple-cubic Ising model in order to be able to account phenomenologically for changes in surface interaction parameters, as illustrated in Fig. 2. The Hamiltonian for this model is

$$H=-J_s(\mathrm{NN})\Sigma_{\mathrm{NN,surf}}S_iS_j-J_b(\mathrm{NN})\Sigma_{\mathrm{NN,bulk}}S_iS_j -J_s(\mathrm{NNN})\Sigma_{\mathrm{NNN,surf}}S_iS_j -J_b(\mathrm{NNN})\Sigma_{\mathrm{NNN,bulk}}S_iS_j\ , \quad (2)$$

where $S_{i,j}=\pm 1$; $J_s(\mathrm{NN})$ and $J_s(\mathrm{NNN})$ are the nearest-neighbor and next-nearest-neighbor exchange integrals, respectively, in the surface layer; and $J_b(\mathrm{NN})$ and $J_b(\mathrm{NNN})$ are the corresponding quantities in the "bulk" layers below the surface. The *interlayer* coupling between surface and bulk is assumed to be controlled by J_b (NN) and $J_b(\mathrm{NNN})$, although the interlayer interactions could in general vary near the surface as well. The NNN exchange interactions are included only when studying the frustrated antiferromagnetic systems.

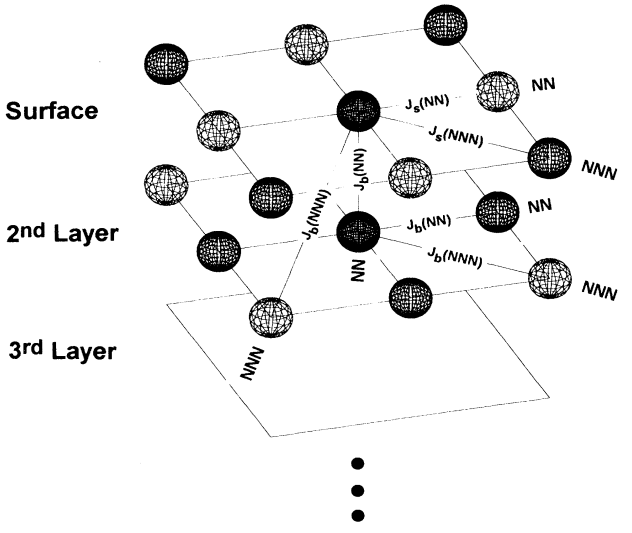

FIG. 2. Schematic illustration of the simple-cubic lattice considered here, with representative surface and bulk exchange interactions indicated.

B. Monte Carlo simulation

A standard single-flip Monte Carlo method[14,15] was used. Since SPPD and other probes of surface magnetic order are primarily sensitive to short-range spin-spin correlations,[9–12] the long-range effects in correlation length are not of primary interest here. Also, surface perturbations on underlying layers were found to be negligibly small after about the third layer, for the choices of parameter used here. Thus, most of the simulations were performed on a 10-layer lattice of spins with 40×40 spins in each layer instead of thicker, and much more time consuming, lattices. However, a few calculations were done on a $40\times 40\times 40$ lattice to be sure that the results reported here were not significantly influenced by finite-size effects. Periodic boundary conditions were imposed in the directions parallel to the surfaces, and free boundary conditions were used in the directions normal to the top surface and the bottom surface, which are equivalent to each other. Spin-spin correlation functions were calculated intralayer, interlayer, and over all spins in the lattice for the first five layers near the surface. The number of iterations per site used in our simulations was 3×10^3. A few calculations were done with 1.5×10^5 iterations per site on both ferromagnetic and antiferromagnetic systems to be certain that the systems studied here were adequately equilibrated. Our correlation function results were checked against high-temperature series expansions and exact results for a two-dimensional limit, and excellent agreement was found. Also, we checked the critical behaviors of a two-dimensional 40×40 square lattice and the $40\times 40\times 40$ simple-cubic lattice with nearest-neighbor exchange interactions under periodic boundary conditions in all directions, and found that the critical temperatures T_C are very close to $2.269J_b(\mathrm{NN})/k_B$ (Refs. 16 and 17) and $4.51J_b(\mathrm{NN})/k_B$,[17,18] which correspond to the well-known T_C values for two-dimensional and three-dimensional homogeneous cases, respectively. And k_B corresponds to Boltzmann constant in the paper.

III. RESULTS

A. Ferromagnetic and antiferromagnetic Ising lattices with NN interactions

In this section we present results for spin-spin correlation functions in the first five layers near a ferromagnetic surface with different relative surface:bulk interactions. The calculations were carried out on a simple-cubic Ising ferromagnetic system with nearest-neighbor interactions only.

In Fig. 3, we show the intralayer [Fig. 3(a)] and interlayer [Fig. 3(b)] spin-spin correlation functions between nearest neighbors calculated for the first five layers near the surface with $J_s(\mathrm{NN})=J_b(\mathrm{NN})=1.0$ (in units of k_BT). Since $J_s(\mathrm{NN})$ is equal to $J_b(\mathrm{NN})$, the only surface effect is reduced coordination number. Both of these figures show that the intralayer and interlayer spin-spin correlation functions fall off abruptly near the same bulk Curie temperature $T_{C,\mathrm{bulk}}[\approx 4.5J_b(\mathrm{NN})/k_B]$, and decrease smoothly at higher temperatures. Although these corre-

lation functions behave very much the same for all layers, the transition does occur at a slightly lower temperature in the surface layer, and then in the second layer, with the third-to-fifth layers being essentially identical and fully bulklike in their behavior. These small differences are expected from the lower surface coordination number and the resulting weaker surface order.

In Fig. 4, we now compare the spin-spin correlation functions for different relative strengths of surface:bulk $J_s(\mathrm{NN})/J_b(\mathrm{NN})=1.00$, 1.90, 2.75, 4.35, and 6.0. The corresponding spin-spin correlation functions were also calculated for a bulk system based upon a $40\times40\times10$ lattice with $J_b(\mathrm{NN})=1.0$ (denoted as "bulk" in the figure). The intralayer spin-spin correlation functions in the surface layer are shown in Fig. 4(a) and the interlayer spin-spin correlation functions between the surface and the second layer are shown in Fig. 4(b). Intralayer spin-spin correlation functions in the second layer appear in Fig. 4(c). The vertical dashed line indicates the bulk Curie temperature ($T_{C,\mathrm{bulk}}$). From Fig. 4(a), the surface critical temperature ($T_{C,\mathrm{surf}}$) corresponding to the abrupt steplike falloff of the intralayer spin-spin correlation functions is found to increase rapidly with an increase in the surface exchange interaction $J_s(\mathrm{NN})$. When $J_s(\mathrm{NN})/J_b(\mathrm{NN})=2.75$, $T_{C,\mathrm{surf}}[\approx 6.3J_b(\mathrm{NN})/k_B]$ is about 1.4 times $T_{C,\mathrm{bulk}}[\approx 4.5J_b(\mathrm{NN})/k_B]$; and when

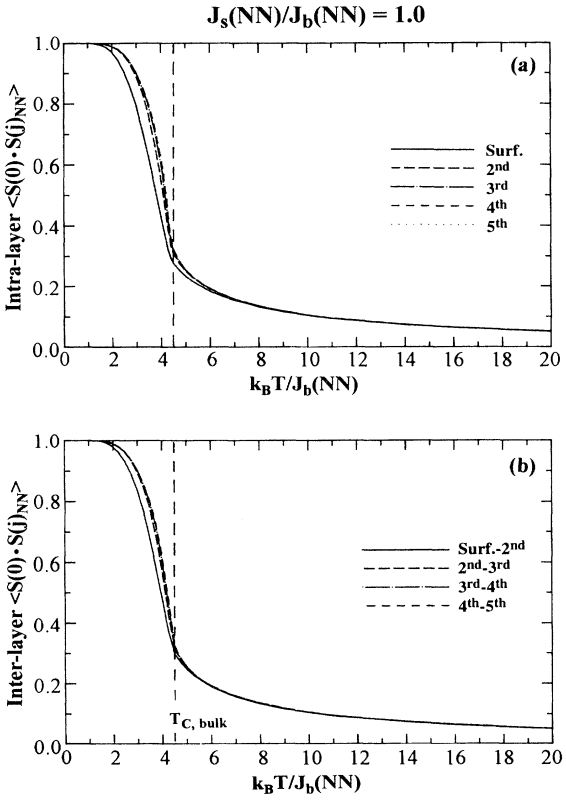

FIG. 3. Temperature dependence of near-surface intralayer and interlayer spin-spin correlation functions between nearest neighbors (NN) for a ferromagnetic system with $J_s(\mathrm{NN})/J_b(\mathrm{NN})=1.0$. (a) Intralayer spin-spin correlation functions from the surface to the fifth layer. (b) Interlayer spin-spin correlation functions between different pairs of adjacent layers near the surface.

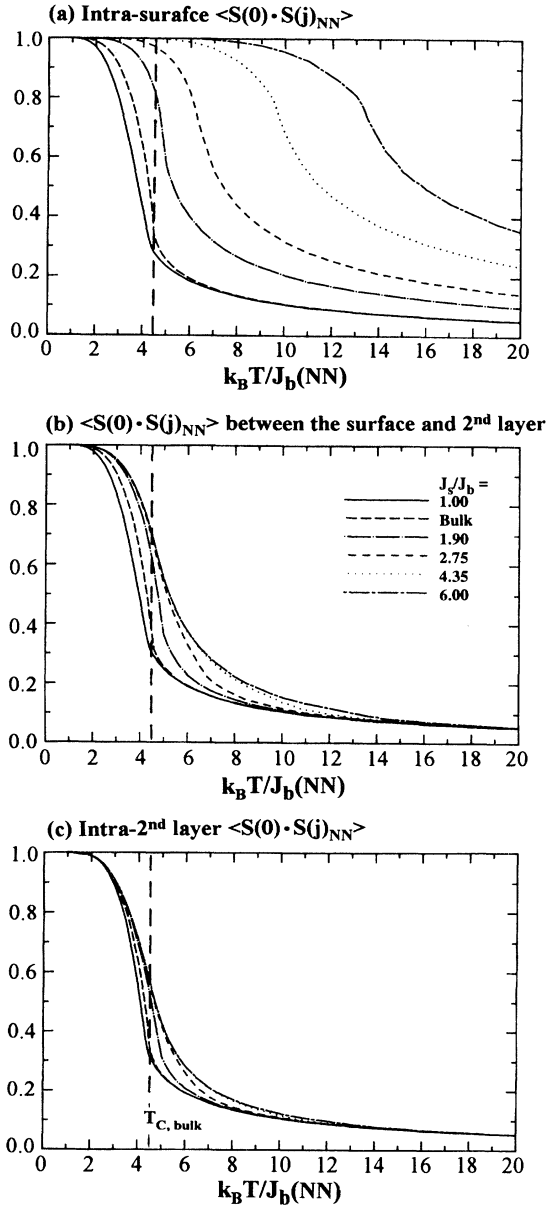

FIG. 4. Temperature dependence of intralayer and interlayer spin-spin correlation functions between nearest neighbors (NN) for different relative strengths of surface exchange: $J_s(\mathrm{NN})/J_b(\mathrm{NN})=1.0$, 1.90, 2.75, 4.35, and 6.0, and for a reference bulk case. (a) Intralayer spin-spin correlation functions for the surface layer and for the bulk case; (b) as (a), but interlayer spin-spin correlation functions between the surface and the second layer; (c) as (a), but in the second layer.

J_s(NN) is increased to 6.0, $T_{C,\text{surf}}[\approx 13.4 J_b(\text{NN})/k_B]$ increases to about three times $T_{C,\text{bulk}}$. However, Fig. 4(b) shows that the spin-spin correlations between the surface and the second layer are affected very little by these changes in surface exchange interactions. Finally, the intralayer spin-spin correlation curves for the second layer in Fig. 4(c) are much closer to each other for these five different values of $J_s(\text{NN})/J_b(\text{NN})$, and do not differ very much from the curve for the bulk case: thus, changing J_s(NN) does not significantly affect the spin arrangements in the second layer.

The two-dimensional critical temperature $T_{C,2D}$ for a simple square ferromagnetic lattice is equal to $2.269 \cdot J_s(\text{NN})/k_B$, where J_s(NN) here corresponds to the exchange integral in the two-dimensional case.[16,17] This relationship leads directly to $T_{C,2D}=6.24 J_b(\text{NN})/k_B$ for $J_s(\text{NN})/J_b(\text{NN})=2.75$, and $13.61 J_b(\text{NN})/k_B$ for $J_s(\text{NN})/J_b(\text{NN})=6.0$. Our calculated values of $T_{C,\text{surf}}$ for both of these cases [about $6.3 J_b(\text{NN})/k_B$ for $J_s(\text{NN})/J_b(\text{NN})=2.75$ and $13.4 J_b(\text{NN})/k_B$ for $J_s(\text{NN})/J_b(\text{NN})=6.0$] are very close to these $T_{C,2D}$ values.

As a more detailed look at the approach to this limiting behavior, Fig. 5 shows $T_{C,\text{surf}}/T_{C,\text{bulk}}$ from our calculations and $T_{C,2D}/T_{C,\text{bulk}}$ from prior analytical treatments as a function of $J_s(\text{NN})/J_b(\text{NN})$.[16–18] We find that, when $J_s(\text{NN})/J_b(\text{NN})$ is greater than about 3.0, $T_{C,\text{surf}}/T_{C,\text{bulk}}$ increases almost linearly with $J_s(\text{NN})/J_b(\text{NN})$, and is essentially identical to $T_{C,2D}/T_{C,\text{bulk}}$. Thus, for $J_s(\text{NN})/J_b(\text{NN})$ greater than 3.0, the exchange interactions between the surface and the second layer are expected to have negligible effects on the spin arrangements in the surface layer, and, as a result, the behavior at the surface should be rather close to that of a purely two-dimensional Ising ferromagnetic lattice.

We next consider in more detail the case of a strongly enhanced surface interaction with $J_s(\text{NN})/J_b(\text{NN})=6.0$, and show the intralayer spin-spin correlation functions in each layer [Fig. 6(a)], the interlayer spin-spin correlation functions between pairs of adjacent layers [Fig. 6(b)], and the overall spin-spin correlation function [Fig. 6(c)]. In Fig. 6(c), each layer has been averaged with equal weight, although photoemission and other electron capture or

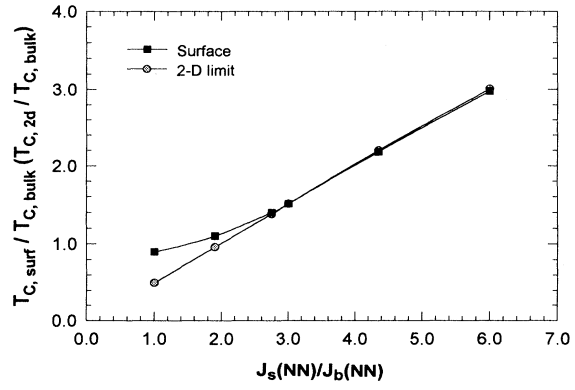

FIG. 5. The ratio of surface critical temperature $T_{C,\text{surf}}$ (or $T_{N,\text{surf}}$) and two-dimensional critical temperature $T_{C,2D}$ to bulk critical temperature $T_{C,\text{bulk}}$ (or $T_{N,\text{bulk}}$) as a function of $J_s(\text{NN})/J_b(\text{NN})$.

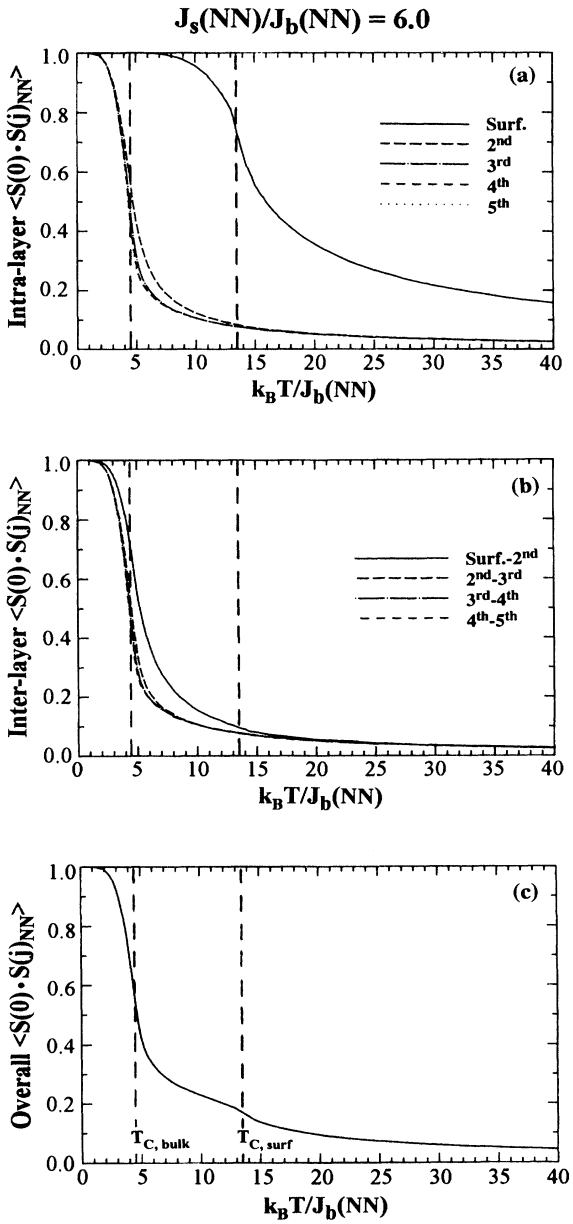

FIG. 6. Temperature dependence of intralayer and adjacent interlayer spin-spin correlation functions between nearest neighbors for a ferromagnetic system with $J_s(\text{NN})/J_b(\text{NN})=6.0$. (a) Intralayer spin-spin correlation functions from the surface to the fifth layer; (b) interlayer spin-spin correlation functions between different neighboring layers near the surface; (c) the overall spin-spin correlation function for the first five layers near the surface, averaged with equal weight per layer.

emission experiments tend to weigh the near-surface layers much more strongly.

The spin-spin correlation curve corresponding to the surface layer in Fig. 6(a) has a significantly higher transition temperature of $T_{C,\text{surf}}/T_{C,\text{bulk}} \approx 3.0$, as compared to the curves for the other layers, which have almost the same transition temperature as the bulk. All of the interlayer spin-spin correlation curves [Fig. 6(b)] show similar features and drop off suddenly at about the bulk Curie temperature $T_{C,\text{bulk}}[\approx 4.5 J_b(\text{NN})/k_B]$. The overall spin-spin correlation curve in Fig. 6(c) has a steplike falloff at about $T_{C,\text{bulk}}$, then decreases smoothly with increasing temperature until at $T_{C,\text{surf}}$, where another steplike falloff appears. For temperature above $T_{C,\text{surf}}$, the spin-spin correlation function decreases slowly to zero. Overall, there are thus only two distinct transitions in these spin-spin correlation curves: one very close to the bulk critical temperature $T_{C,\text{bulk}}$ and the other at a much higher temperature $T_{C,\text{surf}}$ due to surface-enhanced interactions. There is no evidence of any intermediate steplike features or significant broadening due to intermediate transition temperatures for subsurface layers.

By flipping spins on one sublattice, one can easily see that the Hamiltonians of ferromagnetic and antiferromagnetic systems are equivalent to each other in the simple-cubic Ising model with NN interactions only. Thus, the above results for a ferromagnetic Ising lattice would be the same as those for an antiferromagnetic Ising lattice, except that the spin-spin correlation functions between neighbors would have opposite sign, and $T_{C,\text{bulk}}$ and $T_{C,\text{surf}}$ would be replaced by $T_{N,\text{bulk}}$ (bulk Néel temperature) and $T_{N,\text{surf}}$ (surface Néel temperature), respectively.

Considering these results as also representative of an antiferromagnetic surface without frustration, we note that the curve in Fig. 6(c) is at least qualitatively similar to the experimental SPPD data for MnO in Fig. 1: in both cases, there are two distinct transitions, one at around $T_{N,\text{bulk}}$ and the other at what can now be termed $T_{N,\text{surf}}$, which can be much higher than $T_{N,\text{bulk}}$ if $J_s(\text{NN})/J_b(\text{NN})$ is large enough. However, our calculated spin-spin correlation functions show no evidence of a high-temperature transition that is limited to short-range magnetic order (SRMO), and in fact none is expected within this model. Thus, it is possible that the high-temperature transitions seen in SPPD are surface-specific transitions, with the high surface sensitivity of the experiment leading to a greater emphasis of the surface transition.

B. Frustrated antiferromagnetic Ising lattices with NN and NNN interactions

We now consider a more realistic model of the near-surface behavior in an antiferromagnetic lattice such as $KMnF_3$ and MnO, in which spin frustration is included via next-nearest-neighbor interactions, as already introduced in Eq. (2). In estimating the relative strengths of the NN and NNN interactions, we make use of prior experimental data for some similar systems. For example, studies of the spin-wave dispersion curves of antiferromagnetic $KMnF_3$ (Refs. 19 and 20) and $RbMnF_3$ (Refs. 20 and 21) at 4.2 K by means of neutron inelastic scattering indicate that the next-nearest-neighbor exchange interaction $J(\text{NNN})$ is much smaller than the nearest-neighbor exchange interaction $J(\text{NN})$. For $KMnF_3$, $J(\text{NN})/k_B = -3.8 \pm 0.04$ K and $J(\text{NNN})/k_B = 0.11 \pm 0.02$ K; for $RbMnF_3$, $J(\text{NN})/k_B = -3.4 \pm 0.3$ K and $J(\text{NNN})/k_B = 0.0 \pm 0.2$ K. Thus, to begin this discussion, the exchange interaction between next-nearest neighbors $J(\text{NNN})$ has been arbitrarily, but plausibly, set to 0.1 times the corresponding value for nearest neighbors: $J(\text{NNN}) = 0.1 \cdot J(\text{NN})$; and the signs of both have been reversed, as compared with the ferromagnetic case of the last section. Later in this paper we investigate various $J(\text{NNN})/J(\text{NN})$ values for completeness.

In Fig. 7, the spin-spin correlation functions for different surface exchange strengths $J_s(\text{NN})/J_b(\text{NN}) = 1.00$, 1.90, 2.75, 4.35, and 6.00 are compared with the corresponding functions calculated for a frustrated bulk system with $J_b(\text{NN}) = -1.0$ and $J_b(\text{NNN}) = 0.1 J_b(\text{NN})$. The intralayer spin-spin correlation functions in the surface layer are shown in Fig. 7(a) and the interlayer spin-spin correlation functions between the surface and the second layer are shown in Fig. 7(b). Figure 7(c) shows the spin-spin correlation functions in the second layer. This figure is thus the antiferromagnetic analogue of Fig. 4, and both the intralayer and interlayer spin-spin correlation functions are very similar to those in Fig. 4, except for a trivial change in sign. For these choices of parameters, as in the case of ferromagnetic systems, there is no indication of transition temperatures significantly different from $T_{N,\text{bulk}}$ in the second to fifth layers. Again in parallel with the ferromagnetic case, the temperature $T_{N,\text{surf}}$ for the steplike falloff of the surface intralayer spin-spin correlation functions increases rapidly with an increase in exchange interactions [Fig. 7(a)]. When $J_s(\text{NN})/J_b(\text{NN}) = 2.75$, $T_{N,\text{surf}}/T_{N,\text{bulk}}$ is about 1.8, but when $J_s(\text{NN})/J_b(\text{NN})$ is increased to 6.0, $T_{N,\text{surf}}/T_{N,\text{bulk}}$ is increased to about 3.9. The interlayer spin-spin correlations between the surface and the second layer and the intralayer spin-spin correlations in the second layer [Figs. 7(b) and 7(c)] are affected very little by the change in surface exchange interactions. This again indicates that the surface layer can exhibit magnetic behavior very different from that of the bulk, and rather close to the corresponding two-dimensional case.

In Fig. 8, we show $T_{N,\text{surf}}/T_{N,\text{bulk}}$ as a function of $J_s(\text{NN})/J_b(\text{NN})$ (plotted circles). For comparison, the corresponding curve of $T_{C,\text{surf}}/T_{C,\text{bulk}}$ from the ferromagnetic case in the prior section (Fig. 5) is also shown (plotted squares). We find that $T_{N,\text{surf}}/T_{N,\text{bulk}}$, like $T_{C,\text{surf}}/T_{C,\text{bulk}}$ for nonfrustrated systems, increases almost linearly with $J_s(\text{NN})/J_b(\text{NN})$ when $J_s(\text{NN})/J_b(\text{NN})$ is greater than about 3.0. As in ferromagnetic systems, this indicates that the magnetic behavior at the surface is close to that of the purely two-dimensional case. However, $T_{N,\text{surf}}/T_{N,\text{bulk}}$ increases more rapidly than $T_{C,\text{surf}}/T_{C,\text{bulk}}$ with $J_s(\text{NN})/J_b(\text{NN})$. This is because frustration lowers the critical temperature in the bulk more than it does in the surface layer, in this case. That

is, there is a higher degree of frustration in the bulk than in the surface for a fixed $J(\mathrm{NNN})/J(\mathrm{NN})$, since the ratio of NNN to NN coordination numbers $[N(\mathrm{NNN})/N(\mathrm{NN})]$ is higher in the bulk: $N_{\mathrm{bulk}}(\mathrm{NNN})/N_{\mathrm{bulk}}(\mathrm{NN})=12/6$ and $N_{\mathrm{surf}}(\mathrm{NNN})/N_{\mathrm{surf}}(\mathrm{NN})=8/5$.

In Fig. 9, we show for this antiferromagnetic case the intralayer spin-spin correlation functions between nearest neighbors [Fig. 9(a)] and next-nearest neighbors [Fig. 9(b)] for $J_s(\mathrm{NN})/J_b(\mathrm{NN})=6.0$ and again $J(\mathrm{NNN})=0.1\cdot J(\mathrm{NN})$. Figure 9(c) shows the overall value of spin-spin correlation functions between nearest neighbors

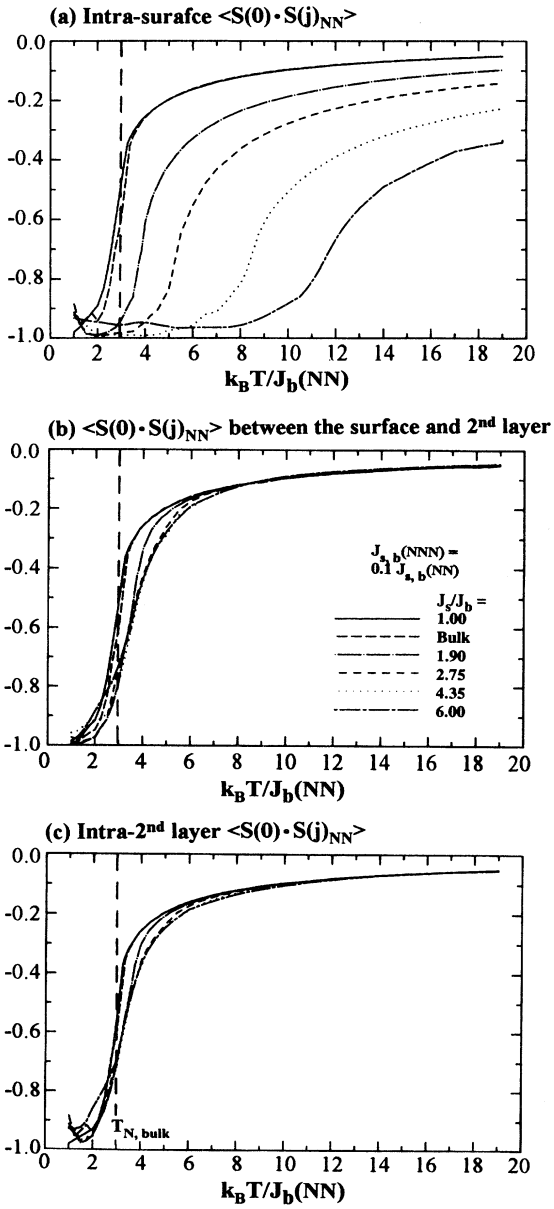

FIG. 7. As for Fig. 4, but for a frustrated antiferromagnetic system, with the next-nearest-neighbor interaction added as $J(\mathrm{NNN})=0.1\cdot J(\mathrm{NN})$.

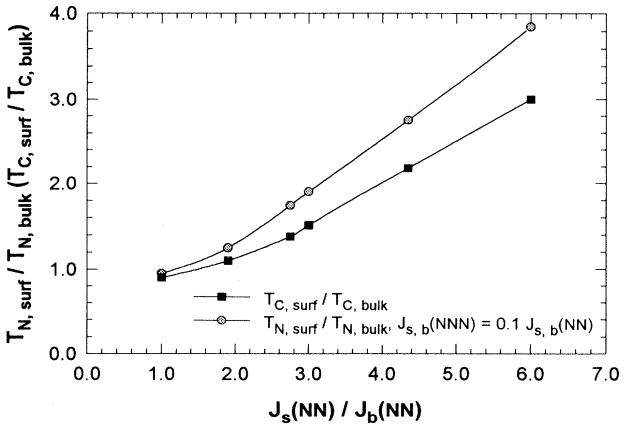

FIG. 8. The ratio of surface critical temperature $T_{N,\mathrm{surf}}$ to bulk critical temperature $T_{N,\mathrm{bulk}}$ as a function of $J_s(\mathrm{NN})/J_b(\mathrm{NN})$ for a frustrated antiferromagnetic system with $J(\mathrm{NNN})=0.1\cdot J(\mathrm{NN})$. For comparison, the corresponding ferromagnetic curve from Fig. 5 is also presented here.

averaged for the first five layers near the surface, with each layer again equally weighted [this is similar to Fig. 6(c) for the ferromagnetic case, but with next-nearest neighbors now included]. The spin-spin correlation functions show similar features for both the NN and NNN cases. The intralayer spin-spin correlation curves at the surfaces have a significantly higher transition temperature compared with the curves for the other layers, with the latter having almost the same transition temperature as the bulk for both the NN and NNN cases. The overall spin-spin correlation curve in Fig. 9(c) has a steplike feature at about $T_{N,\mathrm{bulk}}$, then increases smoothly with increasing temperature until about $T_{N,\mathrm{surf}}$, where another steplike feature appears. At temperatures higher than $T_{N,\mathrm{surf}}$, the spin-spin correlation function increases slowly to zero. Thus, in frustrated antiferromagnetic systems, the overall spin-spin correlation function still shows the same trends as the SPPD experimental data in Fig. 1, and these results give no indication of a higher-temperature transition that is unique to SRMO. This further suggests that the high-temperature transitions observed in both $KMnF_3$ and MnO could be due to surface-specific phenomena.

We have also studied the behavior of spin-spin correlation functions on and near the surfaces of antiferromagnetic systems for various $J(\mathrm{NNN})/J(\mathrm{NN})$ values ranging from -0.25 to 0.35 and with $J_s(\mathrm{NN})/J_b(\mathrm{NN})=3.0$ and 6.0, respectively. In general, we find that frustration does not have major effects on the general behavior of the spin-spin correlation functions. For example, the magnetic behavior at the surface is rather unique and affected very little by the other layers, like the results we have shown before for the $J(\mathrm{NNN})/J(\mathrm{NN})=0$ and 0.1 cases. The various spin-spin correlation functions exhibit distinct steplike features at only two temperatures, one of which is due to the bulk transition and the other to the

surface transition.

In Fig. 10, we summarize this behavior by showing $T_{N,\text{surf}}$, $T_{N,\text{bulk}}$, and $T_{N,\text{surf}}/T_{N,\text{bulk}}$ as a function of $J(\text{NNN})/J(\text{NN})$ for the two cases $J_s(\text{NN})/J_b(\text{NN})=3.0$ and 6.0. Figure 10(a) shows that $T_{N,\text{surf}}$ decreases monotonically as $J(\text{NNN})/J(\text{NN})$ increases from -0.25 to 0.35, as expected due to the concomitant increase in frustration. However, $T_{N,\text{bulk}}$ behaves differently as a function of $J(\text{NNN})/J(\text{NN})$: it decreases monotonically as $J(\text{NNN})/J(\text{NN})$ increases from -0.25 to 0.25, and reaches its minimum [about $1.1J_b(\text{NN})/k_B$] at $J(\text{NNN})/J(\text{NN})=0.25$. Then, it increases monotonically to about $1.7J_b(\text{NN})/k_B$ as $J(\text{NNN})/J(\text{NN})$ increases from 0.25 to 0.35. This phenomenon is caused by the switching of the bulk antiferromagnetic spin configurations of simple-cubic frustrated antiferromagnetic lattices at $J(\text{NNN})/J(\text{NN})=0.25$.

For example, in Fig. 11 we show two possible ground-state spin configurations on simple-cubic antiferromagnetic lattices with NN and NNN interactions. The Hamiltonian in the bulk is, for this model

$$H=-J_b(\text{NN})\Sigma_{\text{NN,bulk}}S_iS_j-J_b(\text{NNN})\Sigma_{\text{NNN,bulk}}S_iS_j \ . \tag{3}$$

For the spin configuration in Fig. 11(a), the ground-state energy is

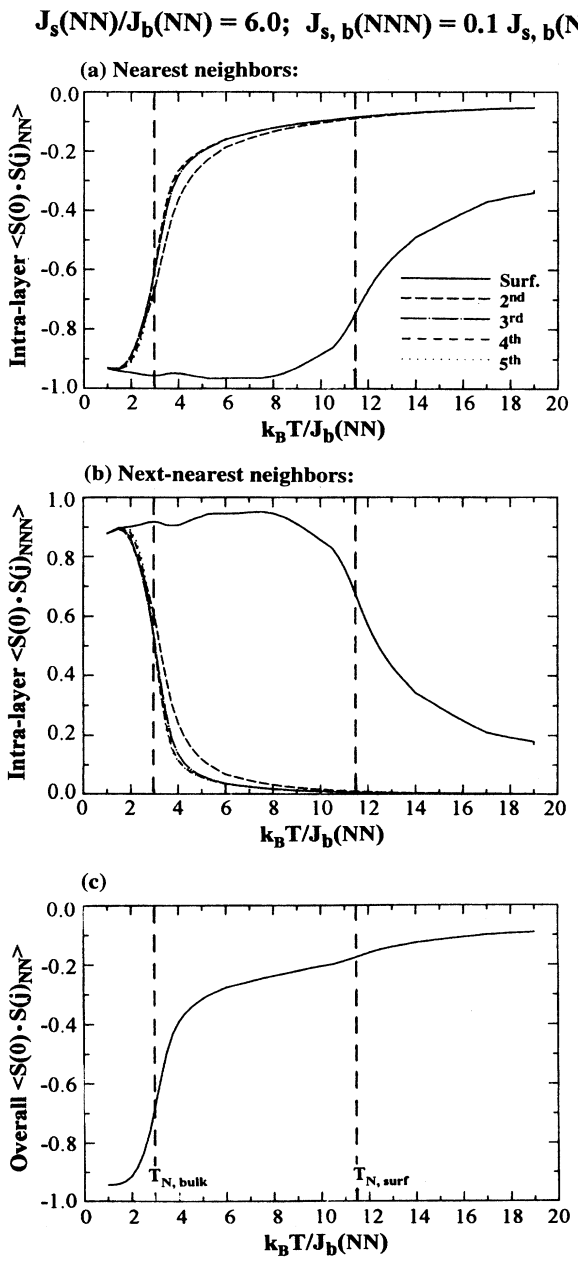

FIG. 9. (a) and (b) Temperature dependence of intralayer spin-spin correlation functions from the surface to the fifth layer for a frustrated antiferromagnetic system with $J_s(\text{NN})/J_b(\text{NN})=6.0$ and $J(\text{NNN})=0.1\cdot J(\text{NN})$. (a) Between nearest neighbors; (b) between next-nearest neighbors; (c) the overall spin-spin correlation function for the first five layers near the surface averaged with equal weight per layer.

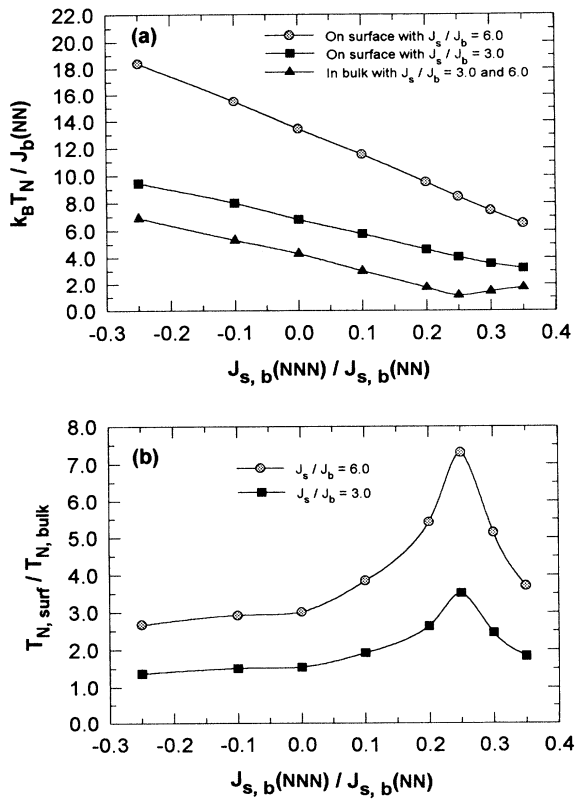

FIG. 10. (a) Surface critical temperature $T_{N,\text{surf}}$ and bulk critical temperature $T_{N,\text{bulk}}$ as a function of $J(\text{NNN})/J(\text{NN})$ for antiferromagnetic systems when $J_s(\text{NN})/J_b(\text{NN})=3.0$ and 6.0, respectively; (b) $T_{N,\text{surf}}/T_{N,\text{bulk}}$ as a function of $J(\text{NNN})/J(\text{NN})$ when $J_s(\text{NN})/J_b(\text{NN})=3.0$ and 6.0, respectively [derived from (a)].

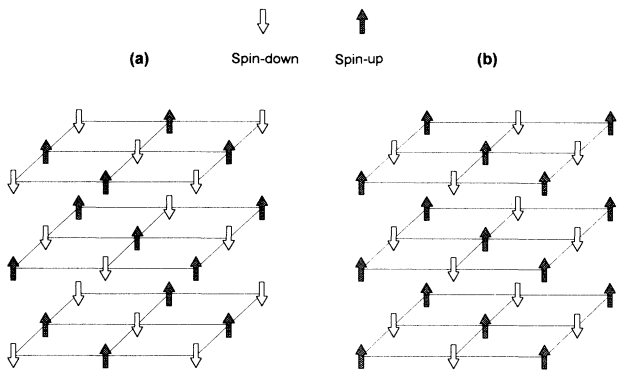

FIG. 11. Two possible antiferromagnetic spin configurations on simple-cubic antiferromagnetic lattices with NN and NNN interactions.

$$U_a = J_b(\mathrm{NN})\{6 - 12J_b(\mathrm{NNN})/J_b(\mathrm{NN})\} \tag{3a}$$

and for the spin configuration in Fig. 11(b) the ground-state energy is

$$U_b = J_b(\mathrm{NN})\{2 + 4J_b(\mathrm{NNN})/J_b(\mathrm{NN})\} \ . \tag{3b}$$

Comparing the two ground-state energies and noting that $J_b(\mathrm{NN})$ is negative for antiferromagnetic lattices, we find that $U_a < U_b$ when $J_b(\mathrm{NNN})/J_b(\mathrm{NN}) < 0.25$, $U_a = U_b$ when $J_b(\mathrm{NNN})/J_b(\mathrm{NN}) = 0.25$, and $U_a > U_b$ when $J_b(\mathrm{NNN})/J_b(\mathrm{NN}) > 0.25$. Thus, when $J_b(\mathrm{NNN})/J_b(\mathrm{NN}) < 0.25$, the lattices studied here prefer the antiferromagnetic spin configuration shown in Fig. 11(a). This spin configuration will switch to the one shown in Fig. 11(b) when $J_b(\mathrm{NNN})/J_b(\mathrm{NN}) > 0.25$, because in this case the spin configuration in Fig. 11(b) has lower internal energy than in Fig. 11(a). From Eqs. (3a) and (3b), we can also see that U_a increases as $J_b(\mathrm{NNN})/J_b(\mathrm{NN})$ increases, while U_b decreases as $J_b(\mathrm{NNN})/J_b(\mathrm{NN})$ increases. The internal energy reaches its maximum at $J_b(\mathrm{NNN})/J_b(\mathrm{NN}) = 0.25$, where $U_a = U_b$. Since the Néel temperature is related to the net statistically averaged interactions of neighboring spins on a given spin, it is expected that the bulk Néel temperature decreases when $J_b(\mathrm{NNN})/J_b(\mathrm{NN})$ increases from -0.25 to 0.25, reaches a minimum at $J_b(\mathrm{NNN})/J_b(\mathrm{NN}) = 0.25$, then increases with increasing $J_b(\mathrm{NNN})/J_b(\mathrm{NN})$ from 0.25 to 0.35, as shown in Fig. 10(a). The antiferromagnetic spin configuration switching phenomenon has also been observed by Landau and Binder,[22] and by Selke and Fisher[23] for simple *square* antiferromagnetic lattices at $J(\mathrm{NNN})/J(\mathrm{NN}) = 0.5$. These two spin configurations differ in their ordering wave vector κ. The magnetic structure factor of the spin configuration in Fig. 11(a) would exhibit a peak at $\kappa = (\pi, \pi, \pi)$, while in Fig. 11(b) it would have a peak at $\kappa = (\pi, \pi, 0)$.

In Fig. 10(b), we also show $T_{N,\mathrm{surf}}/T_{N,\mathrm{bulk}}$ as a function of $J(\mathrm{NNN})/J(\mathrm{NN})$: This quantity increases monotonically as $J(\mathrm{NNN})/J(\mathrm{NN})$ increases from -0.25 to 0.25 and reaches a sharp maximum [about 3.6 for $J_s(\mathrm{NN})/J_b(\mathrm{NN}) = 3.0$, and 7.3 for $J_s(\mathrm{NN})/J_b(\mathrm{NN}) = 6.0$] when $J(\mathrm{NNN})/J(\mathrm{NN})$ is about 0.25. Then, $T_{N,\mathrm{surf}}/T_{N,\mathrm{bulk}}$ decreases to about 1.9 for $J_s(\mathrm{NN})/J_b(\mathrm{NN}) = 3.0$ and 3.8 for $J_s(\mathrm{NN})/J_b(\mathrm{NN}) = 6.0$, as $J(\mathrm{NNN})/J(\mathrm{NN})$ increases from 0.25 to 0.35. Also, over the full range of $J(\mathrm{NNN})/J(\mathrm{NN})$ values, $T_{N,\mathrm{surf}}/T_{N,\mathrm{bulk}}$ at $J_s(\mathrm{NN})/J_b(\mathrm{NN}) = 6.0$ is about twice that at $J_s(\mathrm{NN})/J_b(\mathrm{NN}) = 3.0$, indicating that the surface magnetic order depends primarily on $J_s(\mathrm{NN})$, and further showing that the surface order behaves almost independently of the bulk.

Finally, consider the behavior of the Curie-Weiss constant at the surface (Θ_{surf}) with respect to its behavior in the bulk (Θ_{bulk}), for the antiferromagnetic lattices studied above. Based on mean-field theory, the Curie-Weiss constant should represent a sum over all NN and NNN interactions.[24] Also, prior SPPD results have suggested that there might be a connection between Θ_{bulk} and the high-temperature transition observed experimentally.[7-9] We have therefore calculated $\Theta_{\mathrm{surf}}/\Theta_{\mathrm{bulk}}$ as a function of $J_s(\mathrm{NN})/J_b(\mathrm{NN})$ and $J(\mathrm{NNN})/J(\mathrm{NN})$. From the expressions below,

$$\frac{\Theta_{\mathrm{surf}}}{\Theta_{\mathrm{bulk}}} = \frac{4[J_s(\mathrm{NN})/J_b(\mathrm{NN})][1 + J(\mathrm{NNN})/J(\mathrm{NN})] + \{1 + 4[J(\mathrm{NNN})/J(\mathrm{NN})]\}}{6\{1 + 2[J(\mathrm{NNN})/J(\mathrm{NN})]\}} \ , \tag{4a}$$

$$\frac{\Theta_{\mathrm{surf}}}{\Theta_{\mathrm{bulk}}} = \frac{1}{3}\left[\frac{J_s(\mathrm{NN})}{J_b(\mathrm{NN})} + 1\right] + \frac{2[J_s(\mathrm{NN})/J_b(\mathrm{NN})] - 1}{6\{1 + 2[J(\mathrm{NNN})/J(\mathrm{NN})]\}} \ , \tag{4b}$$

and these results are shown for some representative choices of parameters in Figs. 12(a) and 12(b), respectively. In Fig. 12(a), $\Theta_{\mathrm{surf}}/\Theta_{\mathrm{bulk}}$ shows the expected linear increase with an increase in $J_s(\mathrm{NN})/J_b(\mathrm{NN})$. For the case of $J(\mathrm{NNN})/J(\mathrm{NN}) = 0.1$, $\Theta_{\mathrm{surf}}/\Theta_{\mathrm{bulk}}$ is about 1.9 when $J_s(\mathrm{NN})/J_b(\mathrm{NN}) = 2.75$ and about 3.9 when $J_s(\mathrm{NN})/J_b(\mathrm{NN}) = 6.0$. These values are close to the corresponding $T_{N,\mathrm{surf}}/T_{N,\mathrm{bulk}}$ values from Fig. 8 [about 1.8 for $J_s(\mathrm{NN})/J_b(\mathrm{NN}) = 2.75$ and about 3.9 for $J_s(\mathrm{NN})/J_b(\mathrm{NN}) = 6.0$]. However, in Fig. 12(b), we show that $\Theta_{\mathrm{surf}}/\Theta_{\mathrm{bulk}}$ decreases monotonically with an increase in $J(\mathrm{NNN})/J(\mathrm{NN})$. This is expected due to the fact that,

as J(NNN)/J(NN) increases, the sum over NN and NNN interactions increases faster in the bulk than in the surface layer—because the ratio of coordination numbers of NNN to NN is higher in the bulk than in the surface [i.e., Θ_{bulk} increases faster than Θ_{surf} as J(NNN)/J(NN) increases]. This is different from the behavior of $T_{N,\text{surf}}/T_{N,\text{bulk}}$ as a function of J(NNN)/J(NN) shown in Fig. 10, and such a difference is understandable because the Néel temperature T_N is related to the net statistically averaged interactions of neighboring spins on a given spin, while the Curie-Weiss constant Θ is just the simple sum of all the neighboring interactions.

IV. CONCLUSIONS

We have studied spin-spin correlation functions near both ferromagnetic and antiferromagnetic (frustrated and unfrustrated) surfaces, as derived from Monte Carlo simulations on simple-cubic Ising lattices. The relative strengths of surface exchange interactions were modified in an attempt to model prior experiments in which magnetic transition temperatures higher than those of the bulk have been observed, with particular emphasis on spin-polarized photoelectron diffraction studies of antiferromagnets.

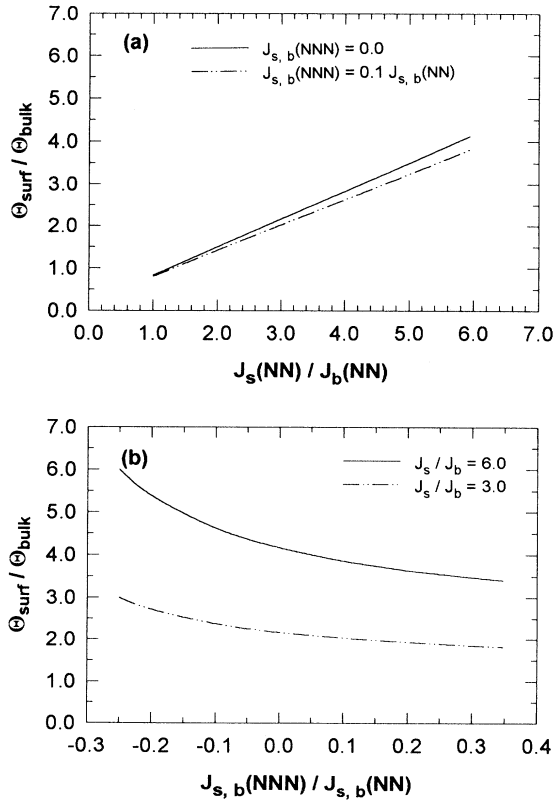

FIG. 12. (a) The ratio of surface Curie-Weiss constant Θ_{surf} to bulk Curie-Weiss constant Θ_{bulk} as a function of J_s(NN)/J_b(NN) when J(NNN)/J(NN)=0.0 and 0.1, respectively. (b) The same ratio, but as a function of J(NNN)/J(NN) with J_s(NN)/J_b(NN)=3.0 and 6.0, respectively.

For sufficient enhancement of the surface interactions, the overall spin-spin correlation function near the surface showed behavior that is qualitatively similar to what has been observed in experiment. Two distinct steplike features occurred for this function: one is around the bulk critical temperature $T_{N,\text{bulk}}$ (or for ferromagnet $T_{C,\text{bulk}}$), and the other is around the surface critical temperature $T_{N,\text{surf}}$ (or for ferromagnets $T_{C,\text{surf}}$). If the surface-to-bulk ratio of nearest-neighbor exchange interactions J_s(NN)/J_b(NN) is high enough, $T_{N,\text{surf}}$ (or $T_{C,\text{surf}}$) can be well above $T_{N,\text{bulk}}$ (or $T_{C,\text{bulk}}$), as observed experimentally. For J_s(NN)/J_b(NN) greater than about 3.0, the magnetic behavior at the surface is found to be essentially two-dimensional and very little affected by the other layers. The abrupt steplike falloff of spin-spin correlation functions well above the bulk transition temperature $T_{N,\text{bulk}}$ (or Curie temperature $T_{C,\text{bulk}}$) is due to the enhanced exchange interactions in the surface layer, and gives rise to a surface-specific transition. All the other spin-spin correlation functions (except that in the surface layer) exhibit similar behavior, and an abrupt falloff around the bulk transition temperature $T_{N,\text{bulk}}$ (or $T_{C,\text{bulk}}$).

The addition of frustration through antiferromagnetic next-nearest-neighbor interaction also does not have major effects on the general behavior of the spin-spin correlation functions. Thus, this model does not predict a distinct high-temperature transition in *short-range* magnetic order (SRMO), although this has been proposed as one possible explanation for the prior SPPD results.[4–6,8,9] On the basis of these results and the fact that the experimental spin asymmetry in SPPD is directly linked to spin-spin correlation functions, we conclude that the falloff of the spin asymmetry well above the bulk critical temperature could be due to a surface-specific magnetic order transition. However, it will require more experimental data and a more quantitative knowledge of the actual bulk and surface coupling parameters for real systems to make this a definitive conclusion. The results presented here should also be useful in assessing the phenomenology of other surface-specific magnetic transitions.

Finally, we note that, for real materials, a more appropriate model would be the Heisenberg Hamiltonian, unless some form of surface relaxation induces a high magnetic anisotropy. Pure Heisenberg systems cannot have a separate finite-temperature surface transition, owing to the Mermin-Wagner theorem.[25] However, the correlation length of the two-dimensional Heisenberg model increases very rapidly as T is lowered. Thus it may be that surface spin order at moderate distances will appear at rather higher temperatures than the bulk temperature ($T_{C,\text{bulk}}$ for ferromagnets and $T_{N,\text{bulk}}$ for antiferromagnets), in a manner that is qualitatively similar to what we have observed here. The much greater computational complexity of the Heisenberg model has prevented our exploring it here, but this would certainly be of interest for future work.

ACKNOWLEDGMENTS

This work was supported in part by the Department of Energy, Basic Energy Sciences, Materials Sciences Division, under Contract No. DE-AC03-76SF00098, and the Office of Naval Research under Contract Nos. N00014-90-5-1457 and N00014-94-1-0162. One of us (S.T.) is supported by Pacific Northwest Laboratory which is a multiprogram national laboratory operated for Department of Energy by Battele Memorial Institute under Contract No. DE-AC06-76RLO 1830.

[1]L. M. Falicov *et al.*, J. Mater. Res. **5**, 1299 (1990); H. C. Siegmann, Condens. Matter **4**, 8395 (1992); H. C. Siegmann, Rev. Solid State Sci. **4**, 817 (1990).

[2](a) K. Binder and P. C. Hohenberg, Phys. Rev. B **6**, 3461 (1972); **9**, 2194 (1974); (b) K. Binder and D. P. Landau, Surf. Sci. **151**, 409 (1985); (c) K. Binder, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and J. Lebowitz (Academic, London, 1983), Vol. 8; H. W. Diehl, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and J. Lebowitz (Academic, London, 1986), Vol. 10; (d) D. P. Landau and K. Binder, Phys. Rev. B **41**, 4633 (1990); (e) W. Schweika, K. Binder, and D. P. Landau, Phys. Rev. Lett. **65**, 3321 (1990).

[3]J. M. Sanchez and J. L. Moran-Lopez, in *Magnetic Properties of Low Dimensional Systems,* edited by L. M. Falicov and J. L. Moran-Lopez, Springer Proceedings in Physics Vol. 14 (Springer-Verlag, Berlin, 1986); Phys. Rev. Lett. **58**, 1120 (1987); J. L. Moran-Lopez and J. M. Sanchez, Phys. Rev. B **39**, 9746 (1989).

[4]B. Sinkovic, B. Hermsmeier, and C. S. Fadley, Phys. Rev. Lett. **55**, 1227 (1985).

[5]B. Sinkovic and C. S. Fadley, Phys. Rev. B **31**, 4665 (1985).

[6]B. Hermsmeier, J. Osterwalder, D. J. Friedman, and C. S. Fadley, Phys. Rev. Lett. **62**, 478 (1989).

[7]D. J. Friedman, B. Sinkovic, and C. S. Fadley, Phys. Scr. **41**, 909 (1990).

[8]B. Hermsmeier, J. Osterwalder, D. J. Friedman, B. Sinkovic, T. Tran, and C. S. Fadley, Phys. Rev. B **42**, 11 895 (1990).

[9]B. Sinkovic, D. J. Friedman, and C. S. Fadley, J. Magn. Magn. Mater. **92**, 301 (1991).

[10]C. S. Fadley, in *Synchrotron Radiation Research: Advances in Surfaces and Interface Science,* edited by Robert Z. Bachrach (Plenum, New York, 1992), Vol. 1.

[11](a) C. Rau and S. Eichner, Phys. Rev. B **34**, 6347 (1986); (b) D. Weller, S. F. Alvarado, W. Gudat, K. Schroder, and M. Campagna, Phys. Rev. Lett. **54**, 1555 (1985); (c) H. Tang, D. Weller, T. G. Walker, J. C. Scott, C. Chappert, H. Hopster, A. W. Pang, D. S. Dessau, and D. P. Pappas, *ibid.* **71**, 444 (1993).

[12]C. Rau and C. Jin, Phys. Lett. A **138**, 334 (1989).

[13]D. Scholl, M. Donath, D. Mauri, E. Kay, J. Mathon, R. B. Muniz, and H. C. Siegmann, Phys. Rev. B **43**, 13 309 (1991).

[14]*Monte Carlo Methods in Statistical Physics,* edited by K. Binder (Springer, Berlin, 1979).

[15]*Applications of the Monte Carlo Method in Statistical Physics,* edited by K. Binder (Springer, Berlin, 1984).

[16]L. Onsager, Phys. Rev. **65**, 117 (1944).

[17]C. Domb, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and M. S. Green (Academic, London, 1974), Vol. 3, p. 425.

[18]H. B. Tarko and M. E. Fisher, Phys. Rev. B **11**, 1217 (1975).

[19]S. J. Pickart, M. F. Collins, and C. G. Windsor, J. Appl. Phys. **37**, 1054 (1966).

[20]E. Schleitzer-Steinkopf, *Gmelin Handbook of Inorganic Chemistry: Manganese* (Verlag Chemie, Weinheim, 1973), Vol. C1.

[21]C. G. Windsor and R. W. H. Stevenson, Proc. Phys. Soc. **87**, 501 (1966).

[22]D. P. Landau and K. Binder, Phys. Rev. B **31**, 5946 (1985).

[23]W. Selke and M. W. Fisher, Z. Phys. B **40**, 71 (1980).

[24]J. E. Smart, *Effective Field Theories of Magnetism* (Saunders, New York, 1966).

[25]N. D. Mermin and H. Wagner, Phys. Rev. Lett. **17**, 1133 (1966); **17**, 1307 (1966).