Magnetism of an Fe monolayer on W(110)

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We discuss theoretical studies of magnetism in the Fe monolayer adsorbed on the W(110) surface. We first present density-functional studies of the ground state that provide us with basic magnetic parameters, under the assumption the ground state is ferromagnetic. We provide results for the spin and orbital magnetic moments, local density of states, the anisotropy, and measures of the exchange strength as provided by calculations of the energy difference between the ferromagnetic and antiferromagnetic states. We compare these results with those provided by the empirical tight-binding description we have used to describe spin waves in this system, to find good agreement between the two methods where results overlap. Inclusion of spin-orbit coupling within our tight-binding Hamiltonian produces anisotropy close in strength and character to that which emerges from the full density-functional analysis, for example. We then present new calculations of the nature of the spin waves in the monolayer. We point out that the large anisotropy that emerges from our analyses (similar to that found in earlier work), combined with our calculations of the effective exchange, provides a value for the Curie temperature of the film much larger than that found experimentally. An earlier experimental study has suggested that in this system, the effective exchange is much smaller than we find. We conclude that the magnetism in this much studied system is not well understood. We discuss reasons why this may be so.

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I. INTRODUCTION

The nature of the magnetism in monolayer systems is of fundamental interest. Of course, we have known for decades that the Mermin-Wagner system requires that there be no long-ranged order at any finite temperature in such a system, if its spin Hamiltonian is form invariant under spin rotations. This conclusion thus applies to any system modeled by the Heisenberg Hamiltonian, or to any system in the same universality class as the Heisenberg Hamiltonian. Included in the latter category is any Hamiltonian used to describe itinerant magnetism that contains only Coulomb interactions between the electrons, with spin-orbit effects omitted. Yet we know that both monolayers and ultrathin films can be ferromagnetic (FM) or exhibit other forms of long-ranged magnetic order up to substantial temperatures. The Curie temperature of the ferromagnetic monolayer which is the topic of this paper is 225 K, for example.[1] Symmetry breaking interactions such as anisotropy (and also magnetic dipole interactions between the magnetic moments) can result in long-ranged order with appreciable Curie temperatures in two dimensions, even if the strength of the symmetry breaking interactions is very weak compared to the strength of the exchange.[2,3]

The spin dynamics in such monolayers is thus of very great interest. Do the spin dynamics differ fundamentally from those in more conventional ordered magnets in three dimensions where exchange interactions alone are responsible for the long-ranged order? Partly for this reason earlier we have presented a detailed study of the spin-wave excitations out of the ground state of the much studied Fe monolayer on W(110), within the framework of a treatment which recognizes the itinerant character of the magnetism in this system, and which utilizes a realistic description of the electronic structure of the film/substrate combination.[4]

In this paper, we present a series of theoretical studies which revisit the nature of the ferromagnetism in this system. The reason for this is that, as we shall argue, the nature of its magnetism is not well understood. We wish to first present theoretical results that provide statements regarding the ferromagnetism of this system, within the framework of what we may call the standard picture. When this is done, we then note that the parameters that emerge from our studies provide a value for the Curie temperature of the monolayer which exceeds that found experimentally by a substantial amount. We thus have a considerable discrepancy between theory and experiment. As we shall see, it is also the case that a very different picture of this system has been set forth in the experimental literature. We are led to raise questions about this alternate scenario in the discussions below. We cannot resolve the issues we raise here in an unambiguous and satisfactory manner, with the information presently in hand. Our conclusion is that there are substantial open questions regarding the magnetism in this important, much studied system.

In Sec. II, we provide an overview of the issues. Then in Sec. III, we present the results of our two independent studies of the ground-state properties of the Fe monolayer. Section IV is devoted to our analyses of the spin dynamics, including calculations of the Curie temperature.

II. OVERVIEW

In Ref. 1, we find a lengthy discussion of the ferromagnetism of the Fe monolayer on W(110). It is argued here that the monolayer is well described as a Heisenberg ferromagnet, weakly perturbed by single-ion anisotropy whose

strength is roughly 0.1 meV/Fe atom. Measurements of the magnetization for temperatures very close to the Curie temperature show the critical exponent to be that of the two-dimensional (2D) Ising model. This conclusion is compatible with the theoretical prediction of Ref. 2 which showed that the two-dimensional Heisenberg model with single-ion anisotropy belongs to the universality class of the Ising model, even in the limit where the strength of the intersite exchange is very large compared to the single-ion anisotropy. Hence, the 2D Ising exponent is expected.

A qualitatively different picture of the magnetism of this system was set forth in Ref. 5. Scanning tunnel microscopy (STM) studies presented in this paper showed domain walls in the monolayer Fe/W(110) system to be astonishingly narrow, with widths in the range of 6 Å. It was argued that this means the anisotropy in the monolayer is very large indeed, in the range of 4 meV per Fe atom, while at the same time intersite exchange is far weaker than customarily encountered in transition metal magnets, on the order of 8 meV between nearest neighbors. The very large anisotropy suggested in Ref. 5 which is 40 times larger than that set forth in Ref. 1, combined with the very modest exchange, provided an account of the very narrow domain walls observed with the STM probe. The strength of the exchange was inferred from magnetization data fitted to the Ising model formula. Thus, the picture set forth in Ref. 5 differs qualitatively from that set forth in Ref. 1. The Fe monolayer is viewed here as an Ising system, as opposed to a Heisenberg system perturbed by weak anisotropy. We should remark that the Fe films studied in Ref. 5 were grown on slightly miscut substrates. In our discussions, we assume that the monolayer sections examined in this work are magnetically similar to monolayers grown on nominally flat substrates.

On physical grounds, one might expect very large anisotropy in this system by virtue of hybridization between the Fe 3*d* orbitals and the W 5*d* states, within which spin-orbit coupling is very strong. Recently Andersen and Hübner[6] have published a relativistic density-functional analysis of this system, to find values for the single Fe anisotropy in the range of 3 meV per Fe atom, quite close to the value suggested in Ref. 5. In view of the large discrepancy between the suggestions in Refs. 1 and 5 it seemed appropriate to perform new calculations of the anisotropy. The results reported below the present paper agree well with those reported in Ref. 6. In view of the very narrow domain walls and the two theoretical results just mentioned, in our view it is now well established that the anisotropy per Fe atom in this film is in the range of 3–4 meV per Fe atom.

The value of the effective exchange strength we calculate is far stronger than that suggested in Ref. 5. The exchange coupling we find is on the order of 40 meV between nearest neighbors. This is, it should be noted, comparable in strength to what one encounters generally in the bulk 3*d* transition metal ferromagnets.[7] We note that the nearest-neighbor exchange strength we calculate here is comparable to values presented earlier, for the uncapped Fe monolayer on Cu(100).[8] The point of these last remarks is that the values of the effective exchange produced by the theoretical calculations reported here are not particularly large or anomalous, compared with those found elsewhere in the literature.

There is thus a major discrepancy between our theoretical results for the exchange strength and the picture set forth in Ref. 5. One might inquire if there are strong temperature effects in the monolayer that reduce the apparent value of the exchange at the finite temperatures at which the experiments were carried out. The values we report here are appropriate to the ground state only. In such a two-dimensional system, one might inquire if temperature effects are very large as a consequence of large amplitude spin fluctuations found in reduced dimensionality. To explore this, we have explored the temperature dependence of the spin-wave frequencies, through use of a standard scheme, namely, the random-phase approximation (RPA) applied to an appropriate model Heisenberg Hamiltonian. This calculation provides us with the temperature variation of the effective exchange. We find the temperature effects to be modest. The amplitude of thermal spin fluctuations, very large in 2D magnets with weak anisotropy, is suppressed in this system by the strong anisotropy. By means of the RPA, we may also calculate the Curie temperature of our model film to find a value very much larger than the measured Curie temperature of 225 K, as remarked earlier. It is the case, as noted in the review of Frobrich and Kuntz,[9] that Curie temperatures in anisotropic 2D Heisenberg models generated through use of the RPA are in good accord with those found in Monte Carlo studies. Thus, we argue that our estimate of the Curie temperature is quite reasonable for the picture which emerges from our theoretical studies.

We then must inquire into the origin of the very large discrepancy between theory and experiment, in regard to the strength of the effective exchange for monolayer Fe on W(110). We offer two suggestions.

First, it has been reported recently that the W(110) surface, as commonly prepared, can contain carbon as a contaminant, at roughly the 2% level.[10] Evidently the carbon migrates to the surface from the bulk. The authors of Ref. 10 describe a means of preparing nominally carbon free surfaces. The W(110) surfaces so prepared, when probed by STM based spectroscopy, display electronic properties in excellent agreement with *ab initio* density-functional calculations[10] for the perfect surface. It is the case that when the Fe monolayer is grown on carbon free surfaces, the domain-wall width increases from 6 to 14 Å.[11] The domain-wall width, in simple models, scales as $(JH_A)^{1/2}$, with J the exchange strength, and H_A the spin-orbit-induced anisotropy. We suggest the value of the anisotropy, which is a single-ion property, may be insensitive to small amounts of carbon contamination. Indeed, the theory reported in Ref. 6 and in the present paper reproduce values for the anisotropy close to that put forth in the analysis of Ref. 5. If the increase in thickness of the domain walls in the Fe film on the clean surface is then due to a difference in the exchange strength, an increase in domain-wall thickness from 6 to 14 Å can be accounted for by an increase in the exchange strength by a factor of 5.44. This suggests that in the films prepared on a carbon free surface, the nearest-neighbor exchange would increase from 8 meV to 44 meV, a value very close to that produced by the theoretical studies reported here. If this suggestion is correct, the Curie temperature of the Fe monolayer on carbon free W(110) should then be very much larger than

225 K. A measurement of the Curie temperature of the Fe monolayer grown on carbon free W(110) would thus be of great interest.

There is a second issue raised by most interesting data and calculations reported in Ref. 12. This paper concerns the magnetic structure of the Mn monolayer on W(110). Early STM studies reported that this Mn monolayer is antiferromagnetic (AFM).[13] More precise STM measurements reported in Ref. 12 reveal that the Mn monolayer is not a simple antiferromagnet. These authors find a state that may be described as a nominal antiferromagnet, modulated by an incommensurate wave vector. It is pointed out in this paper that the lack of reflection symmetry through the plane of this adsorbed monolayer allows Dzyaloshinskii-Moriya (DM) exchange[14,15] to be "active" in the system. The authors argue that the presence of DM exchange can render the simple antiferromagnetic state to be unstable with respect to a modulated phase very similar in character to that seen in the new experiments. They then present calculations based on relativistic density-functional theory which demonstrate that indeed DM exchange is present, with strength sufficient to produce the modulated state. As in the case of the anisotropy, we may expect that the strength of the DM antisymmetric exchange (a spin-orbit-induced phenomenon) may be quite large for transition metal monolayers grown on W surfaces, by virtue of the hybridization between the Fe 3*d* and W 5*d* orbitals.

One may expect that DM exchange would be present in the Fe monolayer as well, with strength comparable to that found in Mn, since Mn and Fe are neighbors in the Periodic Table. If the exchange is as weak as suggested in Ref. 5, it is highly improbable that simple ferromagnetism can be stable in the presence of DM exchange comparable in strength to that reported for Mn in Ref. 12. Even for the much larger exchange strengths we calculate, it is possible that simple ferromagnetism may be unstable. Thus a second possibility is that the Fe monolayer on W(110) is not a simple ferromagnet as commonly assumed, but rather has a more exotic ground state with a net ferromagnetic moment. The spin-wave excitations could thus be very different in nature from those expected for a simple ferromagnet, so Curie temperatures calculated such as we do below may be inappropriate.

There is, it should be noted, a recent theoretical study in which ferromagnetism in the Fe monolayer on W(110) is found to be unstable with respect to a modulated state even in the absence of spin-orbit coupling.[16] It is the case that the energy difference per atom between the modulated state and the ferromagnetic state is found to be very tiny indeed, on the order of 2 meV/Fe atom. We have found no evidence of such an instability in our calculations. If present, this would lead to a negative spin-wave exchange stiffness and we find, as in our earlier studies of this system,[4,17] the spin-wave stiffness to be positive always. The authors of Ref. 16 argue that the very strong single-ion anisotropy present in the monolayer will, in the end, stabilize ferromagnetism. It would be of great interest to see a fully relativistic study of the possibility of a modulated state in Fe which includes the DM interaction, as in Ref. 12.

Thus, we conclude from the results described below that there are fundamental issues yet to be understood in a much studied ultrathin ferromagnet, the Fe monolayer on W(110). We next describe the analyses which have led us to these conclusions.

III. STUDIES OF THE GROUND STATE

We begin with a description of our density-functional analysis of the Fe monolayer on W(110). In our first series of studies, we have performed first-principles calculations using the full potential linearized augmented plane-wave (FLAPW) method. This is, of course, an all-electron calculation. The generalized gradient approximation was adopted for the description of exchange correlation interactions among the electrons. A slab model that contained five W layers and two Fe layers, one on each side of the W film, was used to represent the system. The positions of all atoms in the structure were optimized through minimization of the ground-state energy. We used a plane-wave basis set with an energy cutoff of 13 Rydbergs, and spherical harmonics out to $l=8$ were employed in the expansion of the wave functions in the interstitial and muffin-tin regions, respectively. To sample the irreducible segment of the two-dimensional Brillouin zone, 10×14 grids were adopted.

When the calculations were completed, we found the Fe-W interlayer distance to be 3.79 atomic units (2.00 Å), a value rather close to that reported in Ref. 6. Our calculated spin and orbital moments, in units of Bohr magnetons, are 2.42 and 0.10, respectively. The presence of the Fe overlayer induces an appreciable magnetic moment in the first W layer under the Fe film, and this is antiparallel to the Fe moment. The spin moment in the first W layer is −0.10 Bohr magnetons, and the orbital moment is −0.02 in the same units. The size of the magnetic moments induced in the W layers decays rapidly as one moves into the bulk. The magnetic moments in the interior layers are all smaller than 0.01 Bohr magnetons. Our calculated spin moment in the Fe layer is close to that reported in Ref. 6. However, we are surprised and puzzled by the very tiny orbital moments in the Fe layer reported by these authors. The value they quote, 5×10^{-5} μ_B, seems very small indeed, and in our mind these tiny orbital moments are incompatible with the very substantial anisotropy they report.

To obtain a measure of the exchange coupling strength within the Fe overlayer, we have also studied the $c(2\times 2)$ antiferromagnetic state of the Fe. The total local magnetic moment of each Fe atom remains the same as in the ferromagnetic state, 2.52 Bohr magnetons, but the spin moment is reduced to 2.36 μ_B, while the orbital moment increases to 0.16. The ferromagnetic state is lower in energy than the antiferromagnetic state by 170 meV per Fe atom.

For the determination of the magnetocrystalline anisotropy (MCA) energy of our system, we have adopted the highly reliable torque approach. The spin-orbit coupling Hamiltonian for valence electrons was treated self consistently through second order. The method we employed is described in Ref. 18. The calculated energy differences $E[001]-E[110]$ and $E[\bar{1}10]-E[001]$ are 0.51 meV and −3.48 meV per Fe atom, respectively. Here the [110] axis is perpendicular to the Fe monolayer, the long axis in plane is

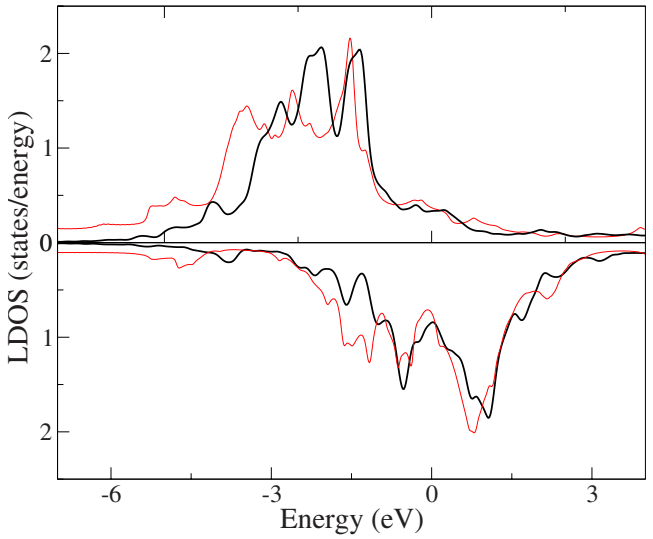

FIG. 1. (Color online) A comparison between the total local density of states in the Fe monolayer provided by the density-functional treatment (black curve) and by the empirical tight-binding description (red curve) of the Fe/W(110) system. The majority spin density of states is above the horizontal line, and the minority spin density of states is below. The zero of energy is chosen to be the Fermi energy.

$[\bar{1}10]$ and the short axis in plane is [001]. Clearly this places the easy axis in plane, and parallel to the long axis of the two-dimensional unit cell as found experimentally, while the hard axis is also in plane along [001]. The difference between the anisotropy energy between the short axis in plane ([001]) and that perpendicular to the plane ([110]) is rather small, so to first approximation we may think of the monolayer as having uniaxial anisotropy along the long axis, with strength in the 3.5 meV range.

We turn next to the description of the ground state of the Fe film provided by our empirical tight-binding scheme, which we use later to obtain a description of the spin-wave excitations of the system. Our studies of the spin dynamics will be presented in Sec. IV. We begin by remarking that a detailed discussion of our general approach and also aspects of the numerical calculations may be found in Ref. 4. The present calculations employ tight-binding parameters generated by a fit to the electronic structure analysis of Fe by Wood.[19] It should be remarked that through use of our approach, the substrate is taken to be fully semi-infinite, as opposed to the five W layers utilized in the density-functional analysis just described.

When our self-consistent description of the Fe monolayer ground state is completed, we find that the spin moment is 2.47 Bohr magnetons, very close to the density-functional value of 2.42. In Fig. 1, we provide a comparison between the local density of states provided by the two schemes. The black curve is that extracted from the density-functional analysis of the ground state, and the red curve is that which results from the empirical tight-binding scheme. While there are differences in details here and there, our view is that the two calculations yield very similar results.

Within the empirical tight-binding scheme, we have also calculated the (adiabatic) exchange integrals between various neighbors. The convention we use is that the energy of the spin system is written in the form $-\Sigma_{ij}J_{ij}\hat{e}_i\cdot\hat{e}_j$. We summarize our calculations of the exchange integrals in Table I.

TABLE I. A tabulation of our calculated (adiabatic) exchange integrals. The units are meV, and the distance from the spin at the origin, and the spin in a (110) plane to which it is coupled is expressed in the form $(a_0/2)[n_x\hat{x}+n_y\hat{y}+n_z\hat{z}]$, with a_0 being the lattice constant as defined for a bulk bcc structure.

Neighbor	J_{ij} (meV)
[1,1,1]	+42.5
[0,0,2]	+3.72
[2,2,0]	+0.46
[1,1,3]	−0.86
[2,2,2]	−0.51
[0,0,4]	+0.84
[3,3,1]	−0.21
[2,2,4]	−1.06
[3,3,3]	−0.24
[1,1,5]	−0.15

From the exchange integrals listed in Table I, one might be tempted to calculate the energy difference between the fully aligned FM state and the $c(2\times2)$ AFM structure examined in the density-functional calculation. If we do the calculation of the FM/AFM energy difference, we find the energy difference per Fe atom to be 328 meV, nearly a factor of 2 larger than the energy difference generated by the all-electron FLAPW analysis.

It is the case, however, that the exchange integrals in Table I are calculated from perturbation theory, assuming neighboring spins are tipped relative to each other by an infinitesimal amount. In contrast to this, the rotation of neighboring spins by 180° to form the AFM state will produce a substantial change in the electronic structure of the system. Note, for instance, there are significant differences in the spin and orbital magnetic moments between the two states within the FLAPW calculation. While the comparison of the energy difference between the FM and the AFM state on the basis of the exchange integrals tabulated in Table I allows us to conclude that the "magnetic stiffness" of the system generated by the two very different calculations is similar in magnitude, a precise comparison is not possible since we cannot calculate absolute energies meaningfully in our tight-binding scheme. Of interest in the present context are density-functional calculations presented by Sandratskii, Sasioglu and Bruno[20] in their study of the Fe monolayer on W(100). These authors calculated the energy difference between the FM and the AFM state within density-functional theory, and then within the same scheme they calculated adiabatic exchange integrals such as those displayed in Table I. When they used the exchange integrals to estimate the energy difference between the FM and the AFM state of their monolayer, they found a difference virtually identical to ours: Use of the exchange integrals produced an energy difference very close to a factor of 2 larger than that found from the full calculation.

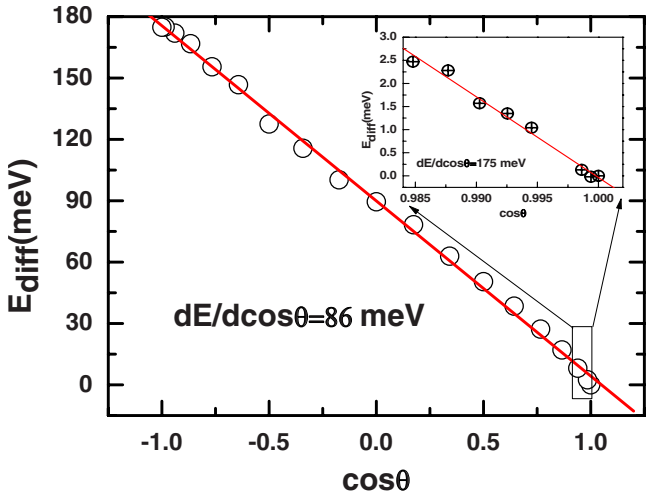

FIG. 2. (Color online) Calculations of the energy change produced by canting spins on one sublattice relative to those on the second sublattice, as one rotates spins from full ferromagnetic alignment to the $c(2\times2)$ antiferromagnetic state. The figure shows the change in energy per unit cell of the AFM lattice. This contains two Fe atoms. The results are generated through use of the VASP code, as discussed in the text. The red curves are best fits to a cos θ form in the angular regions covered by the plots.

It would be of great interest if we could explore the energy of noncollinear spin arrangements in the Fe film through use of the all-electron FLAPW scheme, so we could extract exchange couplings which may be directly compared with those generated by our tight-binding scheme, and displayed in Table I. This would allow us to verify that the factor of 2 difference discussed above persists in the full FLAPW calculation. However, to do so would require very lengthy, massive computations. As a consequence, we employed a pseudopotential method described by the Vienna *ab initio* simulation package (VASP)[21,22] for the determination of $E(\theta)$, the energy of the system as a function of the canting angle θ as spins on one sublattice are rotated continuously from ferromagnetic to antiferromagnetic alignment, to form in the end the $c(2\times2)$ AFM structure. We used the projector augmented wave (PAW) potentials for the description of core states and an energy cutoff of 350 eV for the plane-wave basis expansion. To sample the Brillouin zone, $6\times8\times1$ Monkhorst-Pack $\vec{k}$ points were employed. The film was modeled by a monolayer of Fe adhered on a five layer W(110) surface. It should be noted that the VASP calculations give quite different magnetic moments if we use the same structure which emerges from the full FLAPW calculations, so we moved the Fe atoms outward by 0.4 Å to get a comparable magnetic moment of 2.5 Bohr magnetons.

Our calculations of $E(\theta)$ within the scheme just described are illustrated in Fig. 2. What is shown is the change in energy per unit cell of the AFM state. This contains two Fe atoms. Thus, the energy change per Fe atom is close to half of that provided by the more accurate all-electron calculation. The inset presents a calculation which focuses on the regime of small canting angles, below 10°. If we compare the slope $dE/d[\cos\theta]$ deduced from the small-angle regime with this same quantity obtained by a best fit of the complete curve to a cos θ form, we find the former to be very close to a factor of 2 larger than the latter, in agreement with the study of the Fe/W(100) system reported in Ref. 20, and also in agreement with the comparison with the energy difference calculated with the exchange integrals in Table I and the full FLAPW calculation.

Thus, while the VASP code does not produce magnetic properties of Fe/W(110) as accurately as those which emerge from the all-electron FLAPW analysis, these calculations reinforce the notion that adiabatic exchange strengths extracted from small-angle tipping of spins relative to each other out of the ferromagnetic ground state may be expected to be larger than those obtained from comparison of the energy difference between the FM and AFM state by a factor close to 2. Thus, we argue that the exchange strengths displayed in Table I, which are generated from the tight-binding scheme, are compatible with the FM/AFM energy difference that emerges from the FLAPW analysis.

We have incorporated spin-orbit coupling into our tight-binding description of the ground state, as mentioned earlier. We include spin-orbit effects within the d states only, for both the Fe monolayer and of course for the W substrate, within which spin-orbit effects are very strong. We add to our basic Hamiltonian on site spin-orbit coupling by taking the matrix elements of the form $\xi\vec{s}\cdot\vec{l}$ within d orbitals located on each lattice site of the monolayer/substrate system. The parameters ξ_{Fe} and ξ_{W} were chosen to be $\xi_{\mathrm{Fe}}=5.5$ mRy, and $\xi_{\mathrm{W}}=30$ mRy. These values are compatible with those found in the literature.[23,24] After the spin-orbit anisotropy is introduced into our Hamiltonian, we then carry through a mean-field description of the ground state. We start out with the magnetization aligned along the long axis in plane, and then rotate it through a sequence of small angles, and calculate the energy change associated with each step through use of the spin torque method. We may add up the energy changes so calculated to generate a description of the anisotropy energy as a function of the direction of the magnetization, as we rotate first from the long axis, to the short axis in plane, and then from the short axis to the perpendicular to the film. We shall present a detailed discussion of how we proceed elsewhere.[25] In Fig. 3, we show the angular dependence of the anisotropy energy as this path is swept out.

We see from Fig. 3 that the description of the anisotropy given by our tight-binding scheme is very similar to that which emerges from the full FLAPW calculation. The energy difference $E[001]-E[110]$ agrees remarkably well with the FLAPW result. It is the case that the energy difference $E[\bar{1}10]-E[001]$ is -1.63 meV as opposed to -3.48 meV, which is the number that emerges from the full FLAPW calculation. We regard the agreement between these two calculations as very adequate, when one considers the sensitivity of anisotropy calculations to the description of electronic structure. Both calculations select the long axis in plane as the easy axis, in agreement with experiment, and the short axis in plane is the hard axis.

From the discussion above, we conclude that our tight-binding approach provides a description of the ground state of the Fe monolayer on W(110) compatible with that produced by the highly reliable FLAPW scheme. If we accept

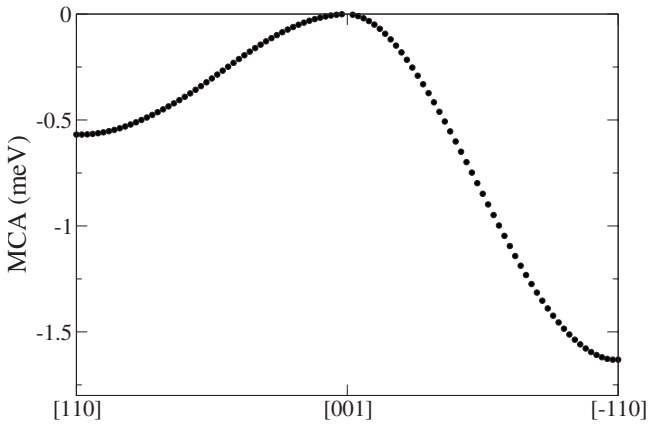

FIG. 3. The MCA as a function of direction of the magnetization, as it is swept from the long axis in plane ([$\bar{1}$10]) to the short axis in plane ([001]) and then to the perpendicular to the plane ([110]). The results have been generated with the tight-binding method, as discussed in the text.

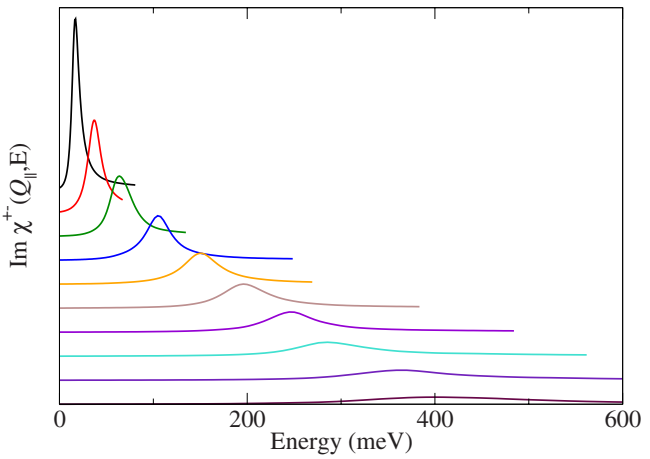

FIG. 4. (Color online) Spectral density plots, for the case where the wave vector is directed along the ΓL direction in the surface Brillouin zone. The wave vector is then directed parallel to the short axis in real space. The wave vector associated with the topmost curve is 0.3 Å^{-1}, and it increases from top to bottom in steps of 0.1 Å^{-1}.

the notion, as found in Ref. 20 and reinforced by our VASP calculations, that use of adiabatically calculated exchange integrals such as those in Table I overestimate the FM/AFM energy differences by close to a factor of 2, then we may argue that the values for the exchange integrals which emerge from our tight-binding analysis give an appropriate description of the "magnetic stiffness" of the monolayer. These values of the exchange set the energy scale for the spin-wave excitations of the monolayer, and they are also central to our calculation of the Curie temperature. As remarked in Sec. II, it is our view that they are also compatible with the thickness of the domain wall in the monolayer, as measured on carbon free W(110) surfaces. We turn next to our description of the spin-wave excitations in the monolayer, and the Curie temperature.

IV. SPIN DYNAMICS, MAGNETIZATION CURVES AND THE CURIE TEMPERATURE

As we have done in earlier publications,[4,17,26] we have studied the nature of the spin waves in the adsorbed Fe monolayer through application of the RPA applied to the ground-state configuration described in Sec. III, as generated by the empirical tight-binding approach. Detailed discussions of our method and approach are given in Ref. 4. The scheme generates the wave vector and frequency dependent susceptibility $\chi_{+,-}(\vec{Q}_{\parallel},\Omega;l,l')$ and from this we form the spectral density functions $\rho(\vec{Q}_{\parallel},\Omega;l)=\frac{1}{\pi}\text{Im}\,\chi_{+,-}(\vec{Q}_{\parallel},\Omega;l,l)$. The spectral density functions, calculated for a selected wave vector $\vec{Q}_{\parallel}$, provide us with the frequency spectrum of spin fluctuations in layer l of the sample. Of course, for the Fe monolayer, we encounter only $l=1$. The spin-wave excitations appear as peaks in the spectral density, and the trajectory of the peaks as a function of $\vec{Q}_{\parallel}$ provides us with an effective dispersion relation for the spin waves. For multilayer films, as we have emphasized in earlier publications,[26] use of adiabatic exchange integrals such as those tabulated in Table I provides a qualitatively misleading picture of the spin-wave spectrum, by virtue of a severe breakdown of the adiabatic approximation. However, as we have seen earlier,[4] for a monolayer with its single spin-wave branch, the spin-wave dispersion relation calculated from the exchange integrals is close to that generated through the full dynamical theory. In Fig. 4, we show a series of spectral density plots for a series of wave vectors directed along the ΓL direction (wave vector parallel to the short axis in real space).

We see, as discussed in our earlier publications, the damping produced by decay of the spin wave into the spin triplet particle-hole pairs (Stoner excitations) of the film substrate combination, as discussed in our earlier publications.[4,26] At the largest wave vectors shown, the damping becomes very severe indeed, to the point where one may question whether the spin wave exists as a well-defined collective excitation. We remark that the Brillouin zone boundary in this direction is very close to 1.5 Å^{-1} as we shall see in Fig. 5, so that in the outer regions of the surface zone, the damping of the spin waves is very severe indeed.

In Fig. 5, we show spin-wave dispersion curves calculated in two ways. The dashed lines are frequencies calculated through use of the exchange integrals tabulated in Table I with Heisenberg spins of unit length attached to each lattice site. (See the remarks in Sec. II on our convention for defining the exchange integrals). Of course, the Heisenberg model is quite misleading here if taken literally, because within this picture the spin waves have infinite lifetime (zero damping) throughout the entire surface Brillouin zone, in dramatic contrast to the results of the full calculations as displayed in Fig. 4. The circles and squares are frequencies extracted from the maxima in spectral density functions generated as displayed in Fig. 4. As in our earlier work[4,26] use of adiabatically calculated exchange integrals leads to short wavelength spin waves softer than those which emerge from the complete dynamical theory in which the full spectral density functions are generated. That this is so is evident from the differences

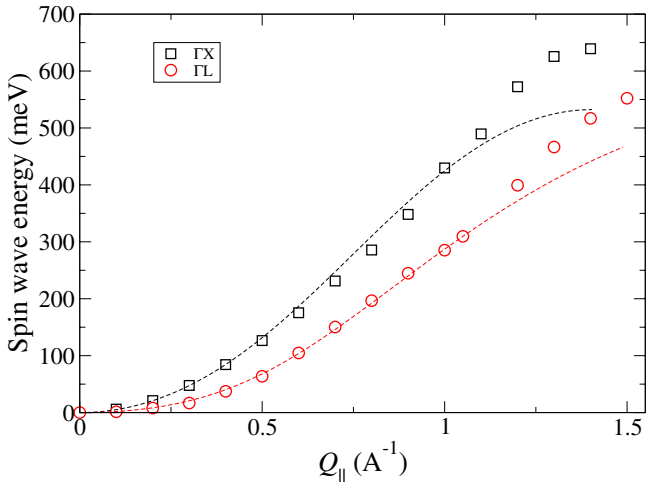

FIG. 5. (Color online) Spin-wave dispersion relations along the two principal directions in the surface Brillouin zone, calculated as described in the text.

between the frequencies as described by the circles and squares in Fig. 5, in comparison with the dashed lines. The two sets of calculations agree well for values of Q less than 1 Å^{-1}, but there are significant differences at larger wave vectors. We note that the spin-wave frequencies displayed in Fig. 4 are somewhat softer than those which appear in our earlier study of the Fe monolayer on W(110). The origin of this lies in the different parameterization we use here. The tight-binding parameters employed presently, and also in our studies of Fe monolayers, do a more satisfactory job of reproducing spin waves in bulk Fe than those utilized in Ref. 4, so we have turned to these when we addressed multilayer Fe films and we use them here. Finally, for the present parameterization scheme, we have earlier calculated[17] the exchange stiffness parameters in the two principal directions to find $D(\Gamma X)=505$ meV.Å^2 and $D(\Gamma L)=205$ meV.Å^2.

We next inquire into the finite temperature properties of the monolayer that result from the parameters which have emerged from our studies of the ground state, and of the spin excitations out of the ground state. For this purpose, we have resorted to the random phase approximation (known also as Tyablikov decoupling) applied to a suitably constructed Heisenberg Hamiltonian which describes a lattice of localized spins. We remark that a detailed description of this scheme has been given by Tahir-Kheli and ter Haar[27] many years ago, and more recently it has been applied to diverse studies of ultrathin films, as modeled in the Heisenberg localized spin framework.[9] As remarked above, this scheme in two dimensions generates Curie temperatures in good agreement with those calculated with Monte Carlo schemes.[9]

Before we present our calculations of the magnetization, we wish to comment on the direct calculation of the Curie temperature. It is evident that the calculations of the Curie temperature of ultrathin films presented in Ref. 8 employ an inappropriate expression for this quantity. The result of this is a serious quantitative error in the results tabulated in this paper. An explicit expression for the Curie temperature of the Heisenberg ferromagnet, as calculated in the RPA, is given by the authors of Ref. 27. If this is expressed in terms of the zero-temperature form of the spin-wave dispersion relation, and written in notation appropriate to the 2D system of present interest, for a lattice of spins of length S we have

$$\frac{1}{k_B T_C} = \frac{3}{S+1}\frac{1}{N_{\parallel}}\sum_{\vec{k}_{\parallel}}\frac{1}{E(\vec{k}_{\parallel})}, \tag{1}$$

where $N_{\parallel}$ is the number of spins in the basic quantization area, and of course the sum over wave vector ranges over the first Brillouin zone. In Ref. 8, the Curie temperature is calculated from the expression

$$\frac{1}{k_B T_C} = \frac{6}{M}\frac{\mu_B}{N_{\parallel}}\sum_{\vec{k}_{\parallel}}\frac{1}{E(\vec{k}_{\parallel})}. \tag{2}$$

If we write for the magnetization per atom $M=2S_{\mathrm{eff}}\mu_B$, we see that the Curie temperature calculated through use of Eq. (2) must be multiplied by the factor $(S_{\mathrm{eff}}+1)/S_{\mathrm{eff}}$ to provide the proper Curie temperature as generated through use of the RPA. Evidently the authors of Ref. 8 have used the large spin limit of the formula, which is an inappropriate limit. For the Fe monolayer on Cu(100), the moment is found to be 2.8 μ_B, so it may be argued that S_{eff} is 1.4, and the Curie temperatures displayed in Eq. (2) should be multiplied by a factor of 1.71. It should be noted that some years ago, Wang, Prange and Korenman[28] have noted and discussed the role of the factor $(S+1)/S$. Within the framework of a Heisenberg model, the Curie temperature was explored as a function of S within a picture where both single site anisotropy and dipolar coupling between the spins were incorporated.[29]

This means that the Curie temperature for the Fe monolayer on Cu(100), as deduced from the parameters in Ref. 8, should be 882 K, as opposed to the 515 K tabulated in Table II of that paper. This very high value of the Curie temperature would seem to be a consequence of the very large value taken for the anisotropy in Ref. 8. The anisotropy assumed by these authors results in a zero wave-vector gap in their model dispersion relation (their parameter Δ) of 1.9 meV. If we interpret this in the language we have used above, this would be an anisotropy per Fe atom of 0.95 meV.[30] While such a value is unphysically large for the Fe films on Cu of interest to these authors, both the strength of the exchange and the anisotropy employed in Ref. 8 are quite comparable to the parameters we have used in the calculations we now present.

Strictly speaking the Heisenberg model applies to integer and half integer spins. In our calculations of the temperature dependent magnetization, we have performed two sets of calculations for each parameter set, one with a moment of 2 μ_B ($S=1$) and one for a moment of 3 μ_B ($S=3/2$). In Fig. 6, we show two sets of calculations of the magnetization as a function of temperature. The left-hand panel employs the exchange interactions tabulated in Table I, and the right-hand panel shows calculations which utilize the very weak exchange set forth in Ref. 5. A comment is in order regarding how one maps the exchange integrals given in Table I onto the Heisenberg model of a lattice of spins S. Recall, as mentioned above, that the numbers in Table I assume the exchange contribution to the energy has the form $-\Sigma_{ij}J_{ij}\hat{e}_i\cdot\hat{e}_j$, with $\hat{e}_i$ being a unit vector attached to each site. In the

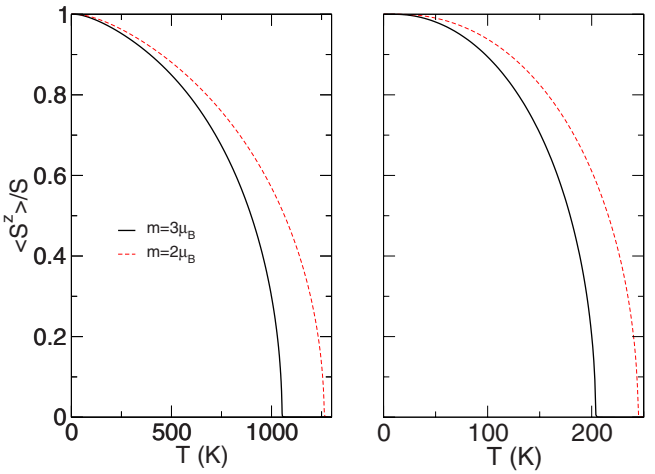

FIG. 6. (Color online) The magnetization as a function of temperature, as calculated from a Heisenberg model which utilizes two different parameter sets. The solid curves are calculations for the case where the spin $S=3/2$, and the dotted curve is for $S=1$. The left-hand panel employs uniaxial anisotropy of 3.5 meV, and exchange parameters taken from Table I. The right-hand panel presents calculations which use the very modest exchange strengths proposed in Ref. 5, along with uniaxial anisotropy of 3.5 meV.

Heisenberg model, of course we have $-\Sigma_{ij}\tilde{J}_{ij}\vec{S}_i\cdot\vec{S}_j$, so $\tilde{J}_{ij}=J_{ij}/S^2$. Hence, for $S=3/2$, the interspin exchange $\tilde{J}$ is smaller than for the case where $S=1$. This leads to a somewhat smaller Curie temperature for $S=3/2$ than realized for $S=1$, in contrast to the case where the strength of the exchange is held fixed as spin S is increased.

We see from the left-hand panel that use of exchange strengths and anisotropies compatible with the values which emerge from our studies of the ground state produce a Curie temperature several times larger than found experimentally (225 K). One may inquire if perhaps the anisotropy strength used in these calculations is too large. We note that for the 2D Heisenberg model with weak uniaxial anisotropy, the Curie temperature has only a weak logarithmic dependence on the strength of the anisotropy.[2,3] If, for instance, we lower the anisotropy to 1 meV, in the range given by our tight-binding analysis, the Curie temperatures we calculate are only 200 K or so lower than those illustrated in the left-hand panel of Fig. 6. We see no simple way for the measured Curie temperature to be reconciled with the parameters which emerge from our analyses of the ground-state properties of the Fe monolayer. The disagreement between theory and experiment is so striking that small adjustments will not resolve the discrepancy. If we use an anisotropy of 0.1 meV per Fe atom, as proposed in Ref. 1, we still find Curie temperatures of 570 K ($S=3/2$) and 680 K ($S=1$).

It is, of course, very striking indeed that the very weak exchange strengths suggested by the narrow domain walls observed in Ref. 5 yield a Curie temperature very close to that found experimentally. This suggests to us that the monolayer explored in this experiment is very different in its magnetic properties from the "ideal theorist's" monolayer studied in Sec. III. Recall that in Sec. II, we suggested carbon contamination as one possible issue which may affect the average exchange strength seriously. It is also possible that the ground state is not a simple ferromagnet, but a more complex state with a net ferromagnetic moment.

V. FINAL REMARKS AND DISCUSSION

We have presented theoretical studies of the ground-state properties of the much studied Fe monolayer on the W(110) surface. We are then led to parameters which describe the effective adiabatic exchange interactions between nearby magnetic moments and the spin-orbit-induced anisotropy. The latter is in good accord with the value suggested in Ref. 5 and also with the previous theoretical result of Anderson and Hübner.[6] The effective exchange strength we find, as judged from that between nearest-neighbor sites, is very much larger than that proposed in Ref. 5, though our results are comparable in magnitude to exchange found in the various forms of $3d$ transition metal magnetism. The combination of the exchange and the single-ion anisotropy leads to theoretically predicted Curie temperatures that are very high indeed, several times the observed value for this system. Interestingly, the parameters proposed in Ref. 5 led to quite reasonable values for the Curie temperature.

Our conclusion is then that the Fe monolayers commonly studied are quite different in character from the "theorist's ideal" monolayer explored in the studies reported in the present paper. We have discussed reasons why this may be so in Sec. II. It is our view that there is compelling evidence that the magnetic properties of the monolayer Fe/W(110) system are sensitive to small amounts of carbon contamination. The fact that monolayers grown on carbon free W(110) surfaces have domain walls 1.4 nm in thickness,[11] as opposed to the 0.6 nm reported in Ref. 5, is most interesting in this regard. This suggests that the exchange strength is sensitive to carbon contamination, and in fact the parameters which emerge from our studies are compatible with the 1.4 nm wall thickness. It would be of very great interest indeed to see measurements of the Curie temperature of such carbon free structures. Of course, another intriguing possibility is that the ground state of the Fe monolayer is not a simple ferromagnet as assumed here, but is more complex in character, as we see from the example of the Mn monolayer on W(110).[13]

We have yet to comment on very interesting studies of the spin-wave spectrum of the Fe bilayer on W(110), by the method of spin polarized electron-energy-loss spectroscopy (SPEELS).[31] Not mentioned in this paper is the fact that the exchange stiffness inferred from the data is in very good accord with our previously published value[17] when the data is extrapolated to zero temperature. However, at large wave vectors, the spin-wave frequencies which we calculate are larger than those found experimentally by roughly a factor of 2. We have discussed these issues elsewhere.[32] In our view, it will be necessary to acquire a more complete understanding of the magnetism in the Fe monolayer before the nature of the large wave-vector spin waves in the bilayer may be understood.

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