Theory of spin excitations in Fe(110) multilayers

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We present theoretical studies of the nature of the spin excitations in ultrathin free standing Fe(110) films, and such films adsorbed on a semi-infinite W(110) substrate. The calculations are carried out within the framework of an itinerant electron theory, with a realistic underlying electronic structure. The energy bands of the film and substrate are described by a nine band empirical tight binding picture, to include the relevant *d* bands and overlapping *sp* complex. Ferromagnetism in the Fe film is driven by on site Coulomb interactions between the 3*d* electrons, treated in mean field theory while a description of the spin wave excitations is generated through use of the random phase approximation. Results are reported and discussed for Fe films three, four, and five layers in thickness. A principal conclusion which emerges from these studies is that the ''frozen magnon'' or adiabatic description of spin wave excitations proves inadequate in a qualitative sense, for systems such as those studied here. The spin waves are embedded in the Stoner continuum, with the consequence they are heavily damped by decay to Stoner excitations, save for the lowest lying mode at long wavelengths. A consequence is that throughout much of the surface Brillouin zone, the spectrum of spin excitations at each wave vector consists of a broad feature which shows dispersion, with no evidence of the sequence of standing spin wave modes predicted by a theory based on the adiabatic approximation. For the five layer film, even near the center of the surface Brillouin zone, while we find a weakly damped low lying acoustic spin wave, the first standing wave is quite broad, and the second standing wave structure is so broad it cannot be viewed as a well defined excitation. At large wave vectors, we find a single broad feature in the spectral density which shows dispersion, very similar to a spin wave, rather than a sequence of standing modes. The results are very similar in character to recent SPEELS data on the Co/Cu system. The physical reasons for the breakdown of the adiabatic method are discussed.

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I. INTRODUCTION

The dynamical response of spin systems in nanoscale magnetic structures is a topic of very active interest in the current literature. Much of the discussion places focus on the nature of collective excitations, the spin waves, whose wavelength is very long compared to the lattice constant and to other lengths which characterize the nanostructures as well. Such modes are excited in ferromagnetic resonance[1] and Brillouin light scattering studies.[2] It is fair to say at this point that the physics which controls the frequency of such modes is well understood. The systematics of the mode frequencies in ultrathin ferromagnetic films provides one with information on the unique forms of anisotropy in these structures,[1] and in multilayers one may obtain information on the strength, sign and character of interfilm exchange couplings. A topic under active exploration currently is the nature of damping mechanisms for spin motions which operate in the nanoenvironment. Recent discussions elucidate mechanisms unique to small scale magnetic systems.[3,4]

It is of fundamental importance to explore the physical nature of the spin wave excitations of short wavelength in ultrathin films fabricated from materials of current interest. These influence transport properties and mean free paths of hot electrons[5] through inelastic scattering processes, and also the understanding of spin motions on very small length scales will require knowledge of the nature of spin waves in these itinerant electron magnets with wavelength that is comparable with the lattice constant.

At the time of this writing, we have virtually no experimental information on the nature of short wavelength spin waves in ultrathin film structures. Interestingly, for the much studied bulk crystals of the 3*d* ferromagnetic metals it is also the case that experimental studies of spin waves well out into the Brillouin zone are very few in number.[6] For these materials, the excitation energy of short wavelength spin waves is sufficiently large that the kinetic energy of thermal neutrons is too low to create them in an inelastic scattering event. It is necessary to employ a spallation source for such studies; it is a challenge with such sources to achieve signal levels adequate to allow good angular resolution. Also for large scattering angles it is the case that the 3*d* form factor falls off rapidly with the consequence the excitation cross section decreases dramatically as wave vector increases. For the ultrathin films, one can conceive of employing a glancing incidence scattering geometry,[7] but again the excitation energies of short wavelength spin waves will be sufficiently high that thermal neutrons will not allow access to the modes, and the form factor issue persists.

In a series of papers, it has been argued that spin polarized electron energy loss spectroscopy (SPEELS) should offer the means of studying short wavelength spin waves on magnetic surfaces[8] and in ultrathin films.[9–13] In such an experiment, the electron kinetic energy will be in the range from a few

electron volts above the vacuum level, possibly up to a few tens of electron volts above the vacuum level. Thus, in contrast to thermal neutrons, the probe particle has kinetic energy sufficient to excite any spin wave found in the materials of interest. Surface sensitivity has its origin in the fact that in the energy range described, the inelastic electron mean free path is only two or three interatomic layer spacings. Confirmation that a given structure in a loss spectrum has its origin in spin wave losses can follow if a spin polarized incident beam is employed. One notes that excitation of a spin wave will decrease the angular momentum of the substrate by $\hbar$. Thus in a spin wave creation process, an electron with moment antiparallel to the substrate magnetization can suffer a spin flip and conserve angular momentum. Electrons with moment parallel to those in the substrate cannot create spin waves and conserve angular momentum at the same time, in the scattering process (assuming spin orbit coupling is weak). Thus a comparison between the loss spectrum with beam polarization first antiparallel and then parallel to the substrate magnetization will allow one to identify spin wave losses, and discriminate against spin independent scatterings such as those which produce the quasielastic peak. In response to a theoretical prediction that in Fe a clear spin wave signal should be detectable in this manner,[11] an experimental study reported the observation of a spin wave loss structure, in a SPEELS experiment on a four layer ultrathin Fe film deposited on W(110), with strength relative to the Stoner background in excellent accord with theory.[14] It was not possible to perform a detailed study of the spin wave dispersion relation with the apparatus used in this study. Experiments with improved spectrometers are underway presently, and spin wave signals have been detected with the new systems.[15,16] Indeed, for an eight layer Co film on Cu(100), spin wave dispersion measurements are in hand.[15]

In response to the considerations discussed in the previous paragraph, we have initiated calculations of the spin wave spectra of ultrathin film systems which will be candidates for study with the new generation of spectrometers. Earlier we have reported detailed studies of spin waves in the Fe monolayer adsorbed on the W(110) surface and for comparison purposes in a free standing Fe(110) film.[12,13] A striking result of these calculations is the prediction of a large in plane anisotropy of the exchange stiffness. Calculations of the anisotropy in the spin wave exchange stiffness tensor as a function of film thickness show it heals to the isotropic bulk form quite slowly.[13] In the present paper, we report calculations for Fe multilayers on W(110). Once again, we have examined free standing multilayers as well, to examine the influence of the substrate on the spin wave spectrum. We will present results for three, four, and five layer Fe(110) films. It is our aim that these studies will serve as a guide to the new generation of experiments. We discuss next aspects of the results presented here which provide insight into the underlying physics of short wavelength spin waves in ultrathin films.

We wish to elaborate on the last sentence of the previous paragraph, with the results presented below in mind. First, to make a general point, there is a fundamental difference between spin excitations in itinerant ferromagnets, and those in a lattice of localized magnetic moments commonly described by the Heisenberg model. In the latter case, the spin waves are rigorous elementary excitations out of the ground state, and as a consequence they have an infinite lifetime, at least at the absolute zero of temperature. Their lifetime remains quite small even at temperatures a fair fraction of the Curie temperature, since the damping provided by magnon-magnon interactions is modest until tolerably elevated temperature are attained.[17] The situation is very different for the spin wave excitations in itinerant systems. Such systems exhibit a spectrum of single particle excitations referred to as Stoner excitations, wherein a single electron may be excited from a majority spin state below the Fermi level to a minority spin state above. The angular momentum of the ferromagnetic ground state decreases by $\hbar$ by these spin flip excitations, very much as when a spin wave is excited. For a general wave vector $\vec{Q}$, the Stoner excitations form a continuum in frequency, ranging from a lower limit $\Omega_m(\vec{Q})$ to an upper limit $\Omega_M(\vec{Q})$; the values of these frequencies are controlled by the underlying band structure. The spin wave frequency $\Omega(\vec{Q})$ is, for all but the longest wave vectors, embedded in the Stoner continuum, i.e., $\Omega_m(\vec{Q}) < \Omega(\vec{Q}) < \Omega_M(\vec{Q})$. Under these conditions, even at the absolute zero of temperature, the spin wave in the itinerant material has a finite lifetime, since it is unstable with respect to decay into the particle-hole pairs of the Stoner continuum. This is, for spin waves, Landau damping similar to that which enters the theory of the collective plasmon excitations of metals. In simple single band models of infinitely extended ferromagnetic metals, the Landau damping of spin waves does not set in until a certain critical wave vector $\vec{Q}_c$ is reached as one moves out from the origin in any direction of the Brillouin zone.[18] Since the early theoretical studies of spin waves in bulk Fe and Ni reported by Cooke and his collaborators,[19] it has been clear that in real materials Landau damping of spin waves can be quite severe throughout much of the Brillouin zone, though the Goldstone theorem of many body theory insures it vanishes in the long wavelength limit, provided the underlying Hamiltonian is form invariant under spin rotations. This will be the case, for example, if spin orbit coupling is not present.

One may then inquire how Landau damping in ultrathin ferromagnetic films compares to that found in the bulk material. Its influence in the films is expected to be more pronounced. The reason is that in the bulk, the decay process is such that all three Cartesian components of wave vector are conserved; the spin wave can decay only to Stoner excitations whose three-dimensional wave vector equals that of the spin wave. In an ultrathin film, breakdown of translational invariance normal to the surface means that wave vector components normal to the surface are not conserved. This breakdown of wave vector conservation opens decay channels not accessible to bulk spin waves. The severity of Landau damping of spin waves in the ultrathin film is illustrated by early calculations reported by Tang, Plihal, and Mills.[10] These authors calculated the spectral density of spin waves in free standing Fe(100) films, up to seven layers in thickness, for selected wave vectors in the surface Brillouin zone.

In a Heisenberg model description of such a seven layer film, for each two-dimensional wave vector parallel to the surfaces, one will predict seven standing spin waves, each with infinite lifetime and thus zero width in the spectral density. In the calculations reported in Ref. 10, one sees dramatic broadening of the standing waves as one moves up in frequency, at fixed wave vector. Indeed, the highest two or three standing wave modes are not even perceptible in the spectral density. The Landau damping is so very severe that they cannot be perceived as well defined elementary excitations in any sense.

This message is reinforced very strongly by the new and much more complete set of calculations reported in the present paper. We shall see, for the systems considered, that as one moves out into the surface Brillouin zone, the standing waves not destroyed fully by the Landau damping merge into one very broad structure which shows dispersion as a function of wave vector. In a SPEELS experiment, such a feature will be perceived as a single, very broad spin-wave-like structure. Indeed, very recent data taken on an eight layer Co film on Cu(100) shows just such a single, very broad structure well out into the surface Brillouin zone,[15] in striking accord with the results below which, of course, have been calculated for a different film/substrate combination. We have new studies of Co films on Cu(100) underway at present.

There is a most important conclusion one may draw from the remarks of the previous paragraph, and the results reported in Ref. 10 as well. The severe damping of the short wavelength spin wave modes we find is an illustration of the fact that the adiabatic approximation breaks down very badly in the case of ultrathin ferromagnetic films. The spectral composition of the spin fluctuations of a selected wave vector bear no *qualitative* resemblance to that provided by a Heisenberg like model, wherein spin wave dispersion relations are generated from a sequence of interatomic exchange interactions between spins in nearby unit cells. We note that in the current literature, one finds a number of papers wherein spin wave excitation spectra are calculated for itinerant electron ferromagnets through use of an adiabatic description of effective exchange interactions, calculated from density functional theory.[20] The results reported here establish that in the ultrathin films, such calculations are misleading in a *qualitative sense*. The issue is not one of quantitative detail associated with a particular approximate treatment of the ground state or excitation spectrum.

We hasten to add that calculations of effective exchange interactions in the static limit do indeed provide very useful information regarding ferromagnetic ultrathin films. Such information, combined with information on anisotropy and the dipolar interactions between the spins will lead to quantitative descriptions of (static) domain walls in such systems. If the anisotropy is sufficiently strong that the domain wall thickness is only a few lattice constants, use of a macroscopic parameter such as the exchange stiffness will not suffice. A microscopic description of the exchange between nearby spins is then required, and these are provided quite correctly by calculations which invoke the adiabatic approximation.

We conclude with two comments. First, ''frozen phonon'' calculations similar to the ''frozen magnon'' calculations just criticized clearly work very nicely for bulk metals and metals in ultrathin film form. One then must inquire about the difference between spin waves and phonons. First, the excitation energy of spin waves in the itinerant $3d$ magnets is very much higher than that of phonons, so the density of final particle-hole states is very much larger as a consequence. There are thus many more final states available for the spin wave to decay into. More important, however, is that the dimensionless coupling constant which controls the decay is, in the spin wave case of order unity. The coupling constant is roughly the product of the strength of the on site intra-atomic Coulomb interaction U between electrons in the $3d$ shell divided by the width W of the d band of the itinerant ferromagnet, and this must be of order unity or even larger for a magnetic ground state to obtain. The electron phonon interaction is very much weaker, when expressed in dimensionless form. Of course, the lifetime of the phonon is controlled by the square of the small coupling constant.

Second, our argument that the adiabatic approximation breaks down in ferromagnets for short wavelength spin waves is by no means new. Many years ago, it was demonstrated explicitly that the adiabatic approximation offers a valid means of calculating the spin wave exchange stiffness D which controls the dispersion relation of long wavelength spin waves in the ferromagnet.[21] It was demonstrated as well that the adiabatic approximation also breaks down if one wishes to move out to shorter wavelengths, beyond the regime of validity of the long wavelength form[21] $\hbar\omega(\vec{Q}) = Dq^2$. The calculations reported in this paper show the breakdown of the adiabatic approximation is particularly severe in the ultrathin films for the systems considered, to the point where it provides a description of the physics that is incorrect in a qualitative sense, if one wishes to apply the information so derived to a discussion of the short wavelength dynamics of the spins. In Sec. II, we comment on the description we use of the electronic structure of our system, and in Sec. III the results are presented, accompanied by a discussion of their implications.

II. THE ELECTRONIC STRUCTURE AND COMPUTATIONAL ASPECTS OF THE ANALYSIS

Our remarks on our means of describing the electronic structure of the systems of interest, and on our approach to the calculation of the spin wave spectral densities will be rather brief. The reason is that in previous publications[10,12] we have presented very detailed descriptions of these issues, with equations that provide the definition of the various quantities we utilize accompanied by the detailed structure of the equations that we solve to generate the spin wave spectra. We thus refer the reader interested in such matters to these earlier papers. Here, as noted above, we are employing the same methodology to extend the calculations of the spin dynamics to multilayers. Also, our description of the underlying electronic structure is the same as that utilized in our earlier studies of spin waves in the Fe monolayer on W(110).[12] We do comment briefly on our approach.

The underlying electronic structure is described through use of an empirical tight binding description of the d bands (the $3d$ bands of the Fe films and the $5d$ bands of the W substrate). The parameters employed here for this purpose are the same as employed before;[13] we simply now add Fe layers one by one. The W substrate is taken as semi-infinite in extent. Ferromagnetism in the Fe overlayer is driven by on site intra-atomic Coulomb interactions between electrons in the $3d$ shell, treated in mean field theory. In our past work, we have used two approaches to the modeling of the intra-atomic Coulomb interactions. In early work, a scheme used in atomic physics[22] was employed wherein a group theoretical analysis is used to express all intra-atomic Coulomb integrals in terms of an irreducible set of parameters adjusted to account for ground state properties of the bulk ferromagnet. For the case of d shells, three adjustable parameters are required. For Fe, their values are given in Ref. 10. Some years ago, Lowde and Windsor[23] performed computations within a much simpler scheme, where only a single parameter enters that can be fitted to the ground state magnetic moment of the bulk crystal. Their scheme is properly conserving, in the sense of many body theory, and inversion of the fundamental equation which emerges from the random phase approximation (RPA) we use to describe spin dynamics [Eq. (6) of Ref. 12] is far more efficient within the Lowde/Windsor scheme. In Ref. 12, we present a detailed comparison between the spin wave spectra calculated within the two schemes for the Fe monolayer on W(110) to find only small quantitative differences. We have thus adopted the Lowde/Windsor scheme for the calculations presented in the present paper. We note, by the way, the two schemes provide us with precisely the same value for the exchange stiffness,[12] provided the parameters are adjusted so the same magnetic moment is contained in each ground state.

We comment briefly on aspects on the numerical calculations of the spin wave spectra. One requires the one electron Green's functions for electrons in the Fe film, to synthesize the irreducible particle hole propagator which serves as the kernel in the RPA integral equation. Of course, the one electron states in the Fe film hybridize with those in the underlying W substrate, taken as semi-infinite. These one electron Green's functions are calculated as follows. First, the one electron Green's function of the (noninteracting) electrons in the W substrate is generated by numerical methods which are by now well established and efficient.[24] Then, as described earlier,[12] once this function is in hand the appropriate one electron Dyson equation may be used to introduce the ultrathin film into the electronic structure, by switching on the hopping integrals between the substrate and the film. We consider the Fermi energy of the film and the W substrate to coincide, of course, and adjust the center of the d orbital complex in each plane to insure charge neutrality in each of the Fe planes. Within the Lowde Windsor scheme, in the ground state there is a local exchange splitting common to all d orbitals on a given plane, so for the ground state calculation it is equivalent to splitting the local d orbitals by the amount $I(n_\uparrow - n_\downarrow)$ with n_σ the number of d electrons of spin σ on the site of interest. Following Himpsel's suggestion,[25] we choose the parameter $I=1$ eV. All integrations over the two dimensional Brillouin zone have been performed with 1008 special points in its irreducible part, and we have carried out careful checks of convergence before adopting this grid. We turn to the presentation of our results in the next section, along with comments on their implications.

III. RESULTS AND DISCUSSION

We arrange the geometry of the (110) surface so that if one atom is at the origin of the xy plane, its nearest neighbors are located at $(a_0/2)(\pm\sqrt{2}\hat{x}\pm\hat{y})$. Then in the ΓX direction, the boundary of the surface Brillouin zone is located at $(2\pi/a_0)(1/\sqrt{2})$, whereas in the ΓY direction, it is located at $(2\pi/a_0)(3/4)$. In the discussion of the results, we shall use reduced wave vectors, where the wave vector is in units of $2\pi/a_0$.

To obtain the results displayed below, we have calculated the wave vector and frequency dependent transverse susceptibility in a representation appropriate to ultrathin film structures. This function, denoted by $\chi_{+,-}(\vec{Q}_\parallel,\Omega;l,l')$ is defined and introduced in our earlier publications.[10,12] Its physical meaning is as follows. First, suppose we have a ferromagnetic film with magnetization directed parallel to the $\hat{z}$ direction, and apply an externally generated, circularly polarized time dependent magnetic field of the form $\vec{h}(\vec{r},t)=h(l)[\hat{x}+i\hat{y}]\exp(i\vec{Q}_\parallel\cdot\vec{r}_\parallel - i\Omega t)$. Here $\vec{Q}_\parallel$ is a two-dimensional wave vector in the xy plane parallel to the film surfaces, while the index l labels the planes within the film. Thus we have an applied transverse field of frequency Ω which exhibits a wavelike variation in the direction parallel to the film surfaces, and has arbitrary spatial variation in the direction perpendicular to the film surfaces. If $\langle S_+(\vec{r}_\parallel,l;t)\rangle$ is the transverse magnetic moment induced in atomic plane l by such a field, then

$$\langle S_+(\vec{r}_\parallel,l;t)\rangle=\left\{\sum_{l'}\chi_{+,-}(\vec{Q}_\parallel,l,l')h(l')\right\}\times\exp[i\vec{Q}_\parallel\cdot\vec{r}_\parallel-i\Omega t]. \quad (1)$$

Suppose we wish to model the film as a Heisenberg ferromagnet, with a localized spin $\vec{S}(\vec{l})$ located on each lattice site. Then the spin wave eigenmodes are plane waves, characterized by the wave vector $\vec{Q}_\parallel$ parallel to the film surfaces. For each value of $\vec{Q}_\parallel$ in the surface Brillouin zone, one has precisely N spin wave normal modes each of infinite lifetime, with N the number of layers in the film. A rather general description of the character and frequency distribution of these modes, along with thermodynamic properties of such a film was given a number of years ago.[26] We may introduce a subscript α to label the modes associated with a particular wave vector. If $e_\alpha(\vec{Q}_\parallel,l)$ is the eigenvector associated with the mode $(\vec{Q}_\parallel,\alpha)$ and $\Omega_\alpha(\vec{Q}_\parallel)$ is its frequency, then one may show that

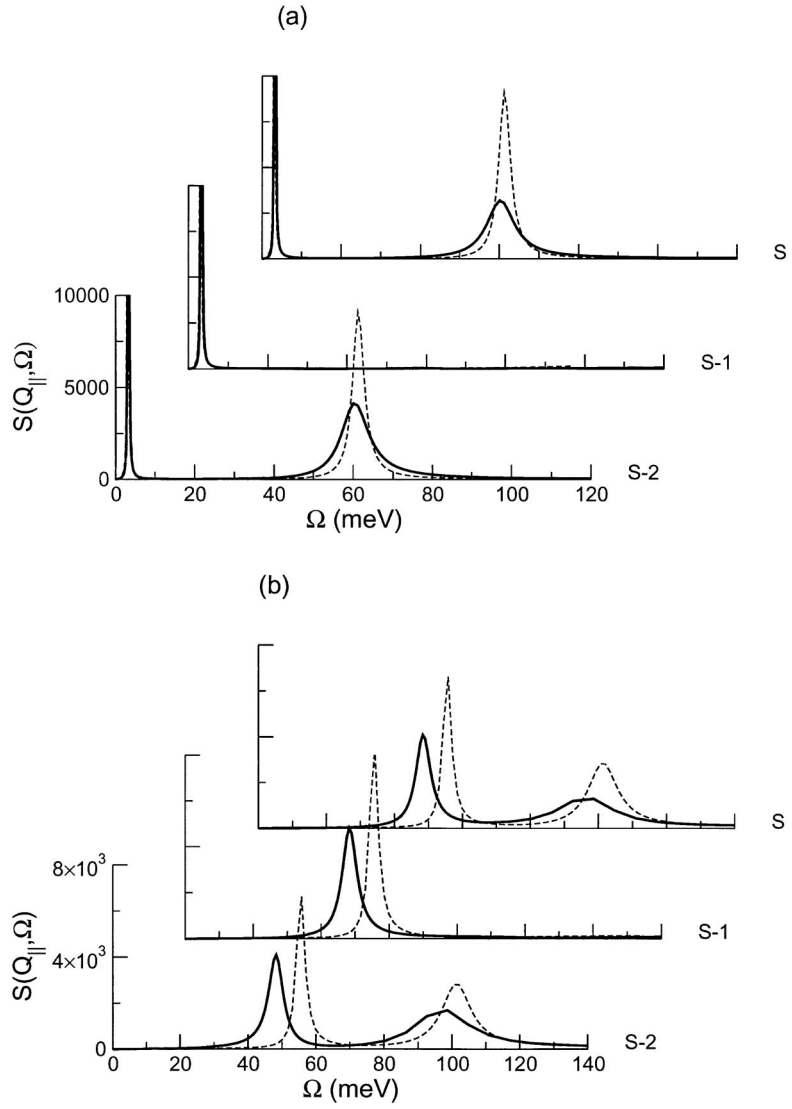

FIG. 1. For two values of the reduced wave vector Q_x along the ΓX in the surface Brillouin zone, (a) $Q_x=0.05$ and (b) $Q_x=0.2$ we show the energy variation of the spectral density function defined in Eq. (2) for the three layer film. The solid curves refer to the film adsorbed on the W(110) surface and the dotted curves are for the free standing film. The curves labeled S are the outer or surface layer of the three layer film, S-1 is the middle layer, and S-2 the innermost layer in direct contact with the substrate for the adsorbed film.

$$S(\vec{Q}_\parallel,\Omega;l)=\frac{1}{\pi}\{\mathrm{Im}\,\chi_{+,-}(\vec{Q}_\parallel,\Omega-i\eta;l,l)\}$$

$$=\sum_{\alpha=1}^{N}|e_\alpha(l)|^2\delta[\Omega-\Omega_\alpha(\vec{Q}_\parallel)]. \qquad (2)$$

Thus, a plot of the spectral density function defined in Eq. (2) for a film modeled as a Heisenberg ferromagnet will show, for each value of $\vec{Q}_\parallel$ a total of N peaks, one for each of the spin wave normal modes of the wave vector. The integrated strength of each peak provides one with a measure of the square of the amplitude of the mode on the plane in question.

If the adiabatic approach discussed in Sec. I is used to calculate effective exchange couplings between nearby spins, and a spin wave spectrum constructed from the information so obtained, then the spectral density constructed from the spin wave eigenvectors will have precisely the form given in Eq. (2). One has modeled the film as a Heisenberg-like ferromagnet, not recognizing its itinerant character through the presence of (low lying) Stoner excitations. As we discussed in section I, a major conclusion of this paper, and of earlier descriptions of spin waves in itinerant ferromagnets, this viewpoint is inadequate in a qualitative sense.[10–13,18,21]

We illustrate this first by our studies of the spin wave spectral density defined in Eq. (2) for the three layer Fe film on W(110) and also for the free standing three layer film. In Fig. 1(a), we show the spectral density function calculated for each of the three layers of the film for a reduced wave vector $Q_x=0.05$ along the ΓX direction. The results for the adsorbed film are shown as solid lines, whereas the dashed curves are for a free standing film. The curves labeled S here and elsewhere in the paper are the outer or surface layer of the film, $S-1$ the first interior layer, and so on. The low lying mode is an acoustic spin wave, in which the amplitude on each plane is closely equal throughout the film. We see this mode is a very long lived eigenmode, as judged by the narrow width of the features in the spectral density. Indeed, the width of the acoustic mode displayed in this figure may be partly of numerical origin. One may prove that within the computational scheme we are using, which is a ''conserving approximation'' in the language of many body theory, the

width of the acoustic mode must vanish identically in the limit of vanishing wave vector $\vec{Q}_{\parallel}$. That this is so for the film is proved explicitly in the Appendix of Ref. 10. We also see the first standing spin wave excitation feature of the film, close in energy to 60 meV. For the free standing film, the eigenvector of this mode has odd parity, and thus its amplitude vanishes for the central layer of the film. Adsorption of the film on the substrate leads to a breakdown of reflection symmetry in the film, so we see the amplitude near the substrate is somewhat larger for this mode than in the outer layer of the film, though its amplitude remains very small in the central layer.

Notice the substantial width of the standing wave feature in Fig. 1(a). This is the Landau damping discussed in Sec. I, and also in the present section. While the first standing wave mode is rather heavily damped, clearly in qualitative terms we may view it as a well defined elementary excitation. Note that the lifetime of the mode is considerably longer for the free standing film, when we compare with the adsorbed film. Clearly the precessing spins in the adsorbed film may decay also to Stoner excitations in the substrate, whose *d*-like density of states near the Fermi level is substantial. Hybridization between the one electron wave functions in the film and substrate allow spin motions in the Fe film to communicate to the particle/hole manifold of the substrate.

In a Heisenberg-like picture, we should expect to see a third standing wave mode for the three layer film. This mode is broadened so severely that we do not see it as a well defined feature in the spectral density, for either the free standing or the adsorbed film. We recall that in earlier studies of free standing Fe(100) films, it is also the case that the higher frequency standing spin wave resonances were washed out by Landau damping. We thus have here our first illustration of the inadequacy of the adiabatic approach to calculation of spin wave spectra of ultrathin films, since such calculations would generate a third mode of infinite lifetime, as illustrated in Eq. (2).

In Fig. 1(b), we show results for $Q_x = 0.2$. We see now that acoustic spin wave has moved up in frequency as expected, as does the first standing wave mode. The Landau damping of each of these modes has increased substantially. We note also that there is appreciable softening of the acoustic mode for the adsorbed film, when its frequency is compared with that of the free standing film. It is interesting to inquire about the effective exchange stiffness of the adsorbed film. If we assume that the dispersion relation remains quadratic out to $Q_x = 0.2$, then from the acoustic mode frequency in Fig. 1(b) one estimates an exchange stiffness of 270 meV A^2, close to that of bulk Fe and very considerably smaller than that provided for the earlier results for the adsorbed Fe monolayer.[12] It is the case that the value just quoted is also in very good accord with our earlier direct calculations, via the adiabatic approach, of the exchange stiffness as a function of layer thickness.[13] (As demonstrated earlier for bulk magnets[21] and for ultrathin films,[12] the adiabatic approach suffices for the calculation of the exchange stiffness.) From the results above, it would appear that the effective exchange stiffness normal to the film surfaces is quite small. We may make an estimate of this from the results in Fig. 2(b) if we assume a dispersion relation of the form $\Omega(\vec{Q}) = D_{xx}Q_x^2 + D_{\perp}Q_{\perp}^2$. Use of the frequency of the acoustic mode for which $Q_{\perp} = 0$ yields the estimate D_{xx} stated above, and $D_{\perp}$ can be estimated from the difference in frequency between the acoustic mode and the first standing spin wave mode in Fig. 1(b), which we assume describes standing wave with wavelength $4(a_0/\sqrt{2})$. We find that $D_{\perp} = 102$ meV A^2 from this criterion.

In Fig. 2(a), we present results for $Q_x = 0.4$, which is a bit more than halfway from the center of the surface Brillouin zone to the boundary. We see now that for the adsorbed film, the standing wave spin wave and the acoustic mode are so broad that they essentially form a single feature in the spectral density with a long tail that extends out to rather high excitation energy. It remains the case for this thin film that the Landau damping for the adsorbed film is more pronounced than for the free standing case. By the time we reach $Q_x = 0.6$, quite close to the boundary of the surface Brillouin zone, we see a single asymmetric structure in the spectral density. For the free standing film, interestingly we do see in these calculations a signature of all three modes in the spectrum of the outer two layers. What has happened here is that the first two lowest modes have now moved up high enough in frequency that they become nearly as heavily damped as the highest mode, but for the free standing film one can still barely resolve the modes as individual entities. Of course, as a given mode becomes more heavily damped, the peak height of the corresponding feature in the spectral density is necessarily reduced as one appreciates upon comparing the peak heights in Fig. 2(b) with those in Fig. 1(a). Actually it is the case that the reduction in peak height is considerably greater than one would expect from the increase in damping; the integrated area of the spin wave peaks decrease, by virtue of the transfer of oscillator strength from the spin wave to the Stoner continuum. We will elaborate on this point and its implications in Sec. IV.

We turn next to our results for the four layer film. In Figs. 3 and 4, we show the spectral densities for the same four wave vectors discussed in the case of the three layer film. Qualitatively the message in these results is quite similar to that for the three layer film. We can estimate the value of the effective exchange constant perpendicular to the surface from the frequency of the first standing spin wave mode, assuming its wavelength is $6(a_0/\sqrt{2})$ appropriate to a zero slope boundary condition at both surfaces. We find a value very close to 120 meV A^2, distinctly larger than that for the three layer film. Of course, in the bulk material, cubic symmetry dictates that the exchange stiffness is isotropic. Thus, for very thick films, the perpendicular exchange stiffness should approach the much larger D_{xx} which, again for the four layer film is somewhat less than that of the three layer film (in accord with the calculations of Ref. 13) but still close to the value for bulk Fe. Our earlier studies of the in plane anisotropy of the exchange stiffness shows that the isotropic bulk limit is approached rather slowly, and we believe this to be true of the perpendicular effective exchange stiffness as well.

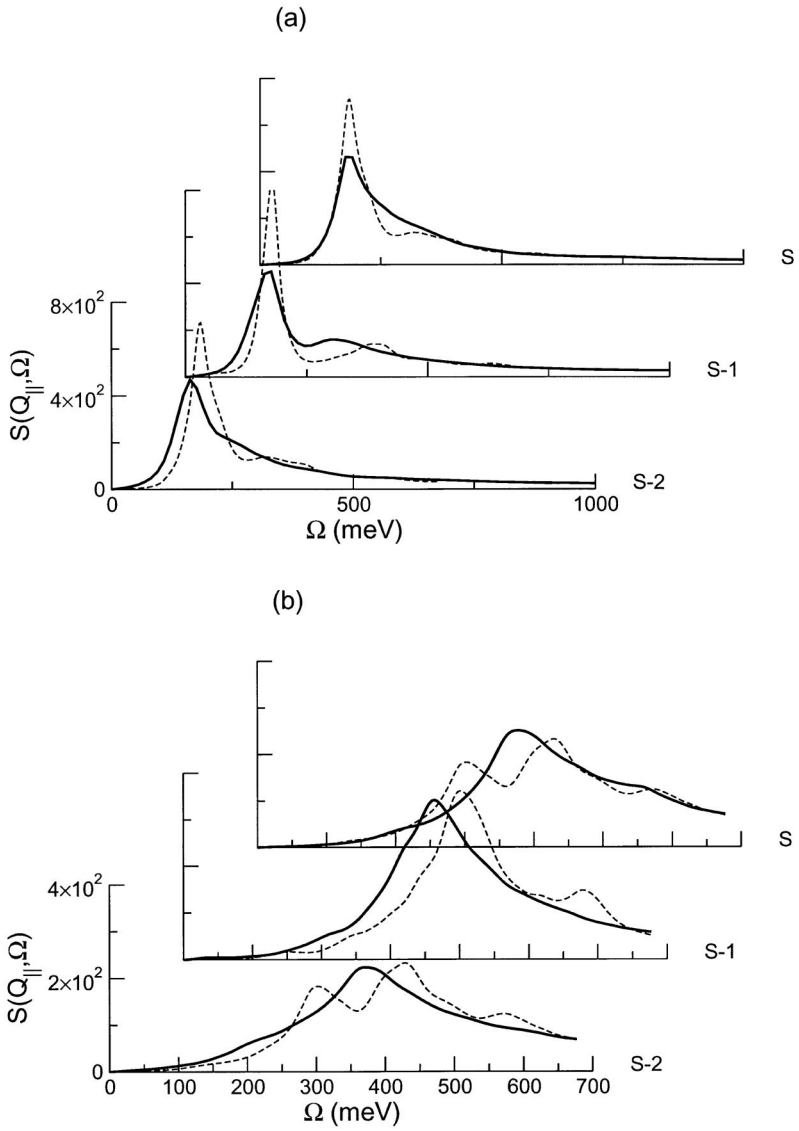

FIG. 2. The same as Fig. 1, except now we give results for (a) $Q_x=0.4$ and (b) $Q_x=0.6$.

It is interesting to compare the maximum peak heights in various layers for the acoustic mode in the free standing film, and in the adsorbed film, for $Q_x=0.2$. Quite clearly, in the free standing film the amplitude of the mode is constant across the film, as expected for a uniform mode in a film with no spin pinning at the surfaces. However, when the film is adsorbed, the amplitude of the mode falls off distinctly, as one moves from the film surface into the interior. This suggests that the mode is in fact a surface spin wave in this case. In Heisenberg model descriptions of spin waves near surfaces, under commonly encountered conditions, the lowest lying eigenmode in the band of spin wave states characterized by the wave vector $\vec{Q}_\parallel$ parallel to the surface is a surface spin wave.[26,27] When the wavelength of the mode parallel to the surface is long compared to a lattice constant, the amplitude of the surface wave falls off rather slowly as one moves into the crystal. In the case of the well known Rayleigh surface acoustic wave, the penetration depth in the long wavelength limit scales as $|\vec{Q}_\parallel|^{-1}$, whereas in the case of surface spin waves on the Heisenberg ferromagnet, the penetration depth is larger by a factor of $1/|\vec{Q}_\parallel|a_0$. It would be most intriguing to follow the eigenvector out to larger values of the wave vector, where it should become more localized to the surface but as one sees from Fig. 4(a) the modes are Landau damped sufficiently at larger wave vectors that one cannot isolate the lowest lying spin wave mode as a clear, distinct excitation. We see in Figs. 4(a) and 4(b) that well out in the zone, the various modes coalesce into one rather broad feature with a high frequency tail which extends out to higher energy. Notice that in the outer layer, for the case $Q_x=0.6$ [Fig. 4(b)] that in the outer surface, there is very little difference between the structures in the spectral density for the free standing and the adsorbed film. By this point, short wavelength spin motions in the outer layer are not affected by the substrate. Interestingly, the centroid of the feature we see in the innermost layer has lower frequency for the adsorbed film, and it is broader as well, so the spins at the boundary with the tungsten substrate are indeed affected by the particle hole manifold below them.

Results for the five layer film are summarized in Figs. 5–8. In Figs. 5(a) and 5(b) we give results for $Q_x=0.05$ and

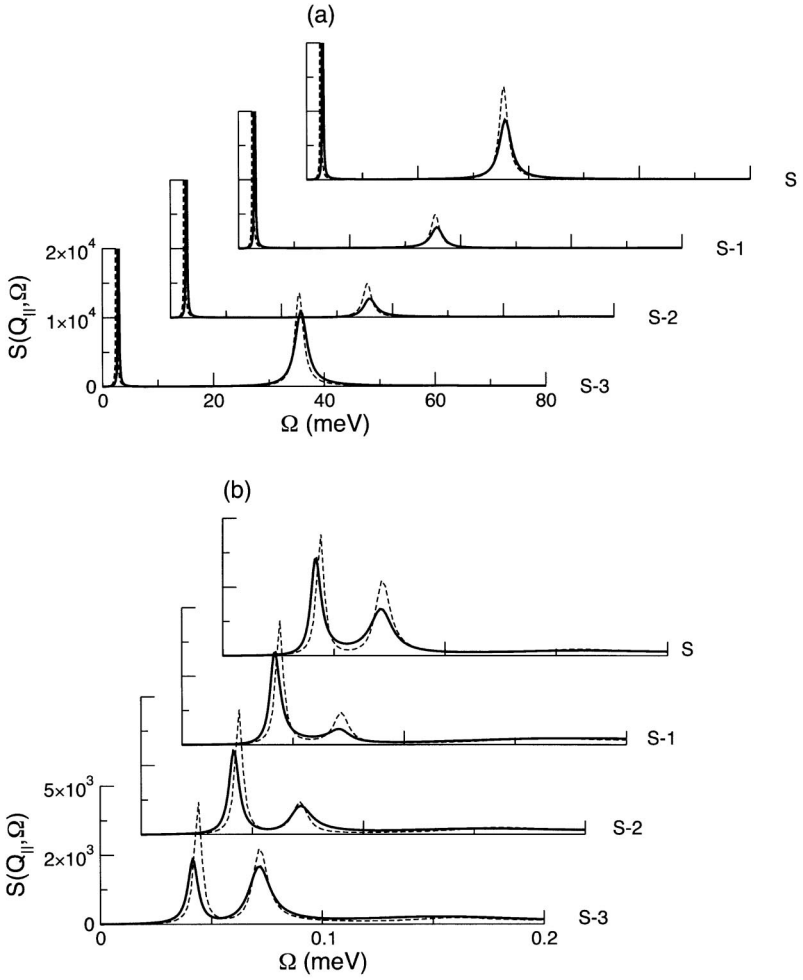

FIG. 3. The same as Fig. 1, but now we have results for the four layer film for (a) $Q_x=0.05$ and (b) $Q_x=0.2$.

0.2, respectively. We may estimate the exchange stiffness D_{xx} along ΓX by using the frequency of the low lying acoustic mode in Fig. 5(b) as we did for the three layer film. We find the considerably smaller value of 210 meV A^2. This is also consistent with the calculations reported in Ref. 13, where it is found by direct calculation of D_{xx} that it approaches the value appropriate to bulk Fe in a nonmonotonic manner. For the monolayer, as found also in our earlier study of the adsorbed monolayer, D_{xx} assumes a value substantially larger than that of bulk Fe. It decreases with increasing film thickness to pass through a minimum of roughly 200 meV A^2 in the five layer thickness region before approaching the bulk value. From the difference in excitation energy between the acoustic and first standing spin wave mode in Fig. 5(a) we can estimate the exchange stiffness perpendicular to the surfaces, assuming the standing wave mode has a wavelength of $8(a_0/\sqrt{2})$. This yields the estimate $D_{\perp}=140$ meV A^2, again somewhat larger than that for the four layer film. Evidently the perpendicular exchange stiffness approaches the bulk value rather slowly as a function of film thickness, as we have seen earlier for the two in plane components of exchange stiffness.[13] We remind the reader that earlier studies of free standing Fe(100) films up to seven layers in thickness also produced estimates of the perpendicular exchange stiffness smaller than the bulk by nearly a factor of 2, using interpretations of calculated spectral densities similar to those discussed here.[10] In Fig. 5(b), we can now see clearly that for the adsorbed film, the amplitude of the lowest spin wave mode is not uniform throughout the film, but falls off distinctly as one moves into the film interior. If this were a surface spin wave, then the amplitude should fall off exponentially as one moves into the interior. In Fig. 6, we plot the logarithm of the peak height versus layer number, and we indeed find that the amplitude is very close indeed to an exponential. This strongly suggests that we do have here a surface magnon, with amplitude localizing on the outer layer with increasing wave vector. As one moves to larger wave vectors, the amplitude should become progressively more localized, if this mode behaves similar to the surface spin waves discussed for Heisenberg magnet surfaces and films.[27] Alas, it is difficult to address this question, since as we move to shorter wavelengths, the mode becomes sufficiently broad that its spectral feature merges with the higher lying excitations, as noted above.

In Figs. 7(a) and in 7(b), we show the spectral densities on the ΓX direction, for the choices $Q_x=0.4$ and $Q_x=0.6$, respectively. Again we see a very broad structure formed by the merger of the various Landau damped standing wave modes. Upon following the peak position in, say, the outer layer with increasing wave vector, one finds a feature which disperses in a manner very similar to that of a single spin wave mode. Thus, in an experiment, this feature could be

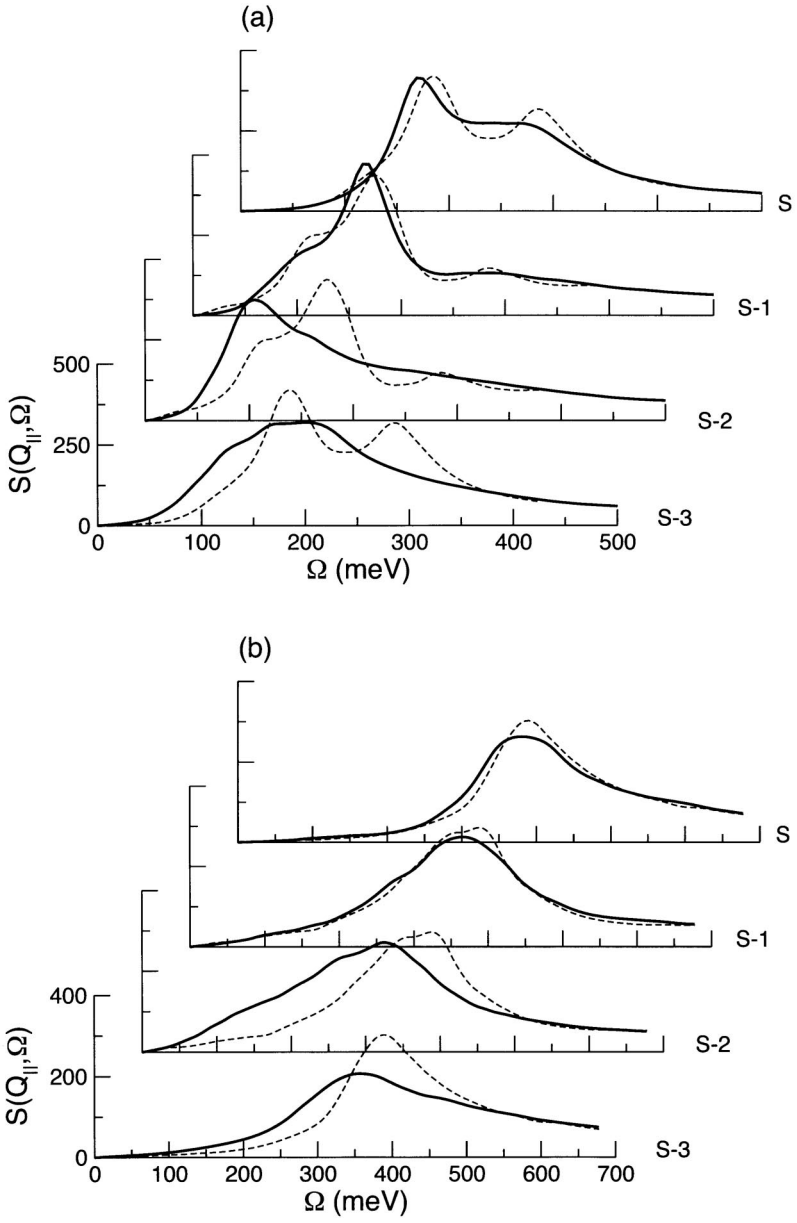

FIG. 4. The same as Fig. 3, but the results are for the four layer film with (a) $Q_x = 0.4$ and (b) $Q_x = 0.6$.

interpreted very reasonably as produced by one mode that has suffered very substantial lifetime broadening. These calculations are very similar to the data reported in Ref. 15, and in fact the interpretation offered by these authors is that just mentioned.

In Figs. 8(a) and 8(b), for the five layer film we show examples of spectral density calculations along the ΓY direction. In Fig. 8(a) we show calculations for $Q_y = 0.2$ and in Fig. 8(b) we show results for $Q_y = 0.6$. It is evident upon comparing the frequency of the acoustic spin wave in Fig. 8(a) with that in Fig. 5(a) that the exchange stiffness D_{yy} is substantially smaller than D_{xx}. We find here that D_{yy} is 130 meV A^2, again close to our earlier calculations using the adiabatic approach.[13] By comparing Fig. 8(b) with Fig. 7(b), we see the softness in the spin system response along ΓY persists out to short wavelengths. In Fig. 8(b), in the outermost layer the peak in the spectral density is in the vicinity of 250 meV, while in Fig. 7(b) it is just below 400 meV. In the next section, we present a summary of our principal conclusions, along with comments on the implications of the results just discussed.

IV. COMMENTS AND DISCUSSION

We have presented detailed studies of the nature of the short wavelength spin wave excitations of ultrathin Fe films adsorbed on a semi-infinite W(110) substrate, within a framework which recognizes the itinerant character of the magnetic film, and in which the adiabatic or ''frozen magnon'' approximation is not invoked. A principal conclusion is that qualitative features of the spin wave spectrum are influenced strongly by nonadiabatic aspects of the electronic response of the film/substrate combination. This is true even near the center of the surface Brillouin zone, where we have seen the presence of a sharp, long lived acoustic spin wave mode, but the higher lying standing spin wave resonances of the ultra-

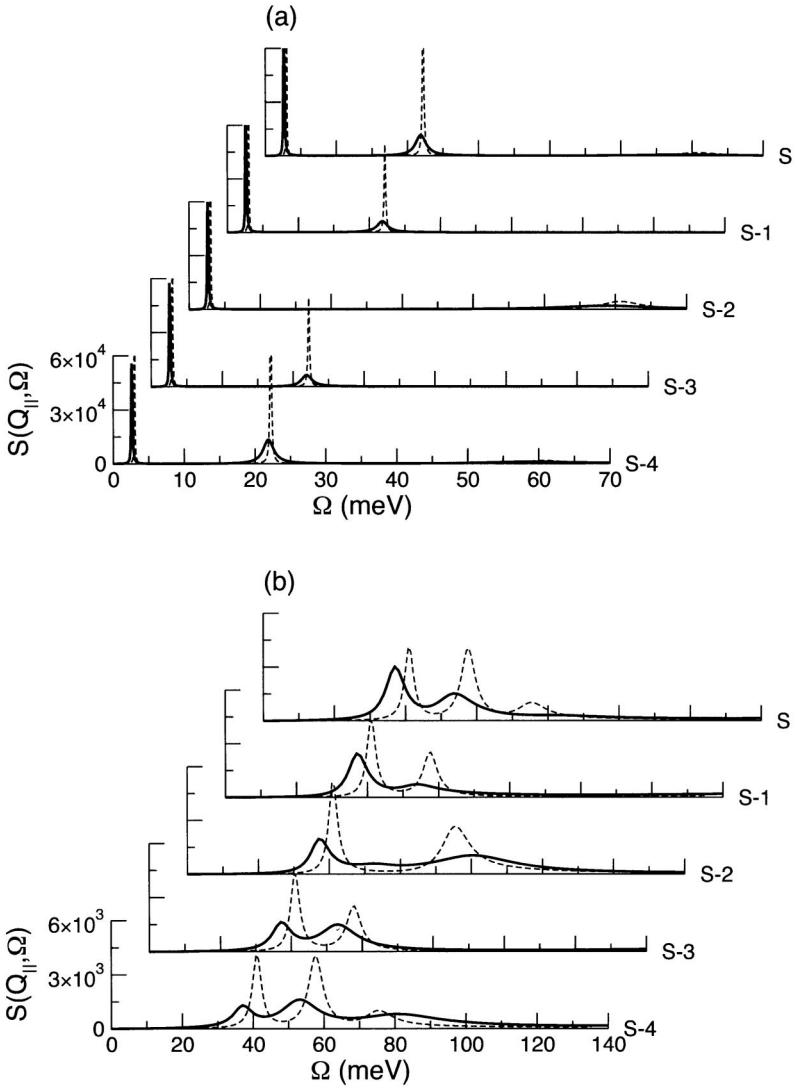

FIG. 5. The same as Fig. 1, except now the results are for the five layer film with (a) $Q_x = 0.05$ and (b) $Q_x = 0.2$.

thin film are damped so strongly by decay to Stoner excitations that for the examples discussed, only the (lifetime broadened) first standing wave appears as a well defined excitation in the spectral density. At short wavelengths, in the spectral density we see only a single rather broad feature which exhibits dispersion as a function of wave vector $\vec{Q}_{\|}$, but the lifetime broadening is sufficiently severe that individual standing mode resonances are not resolved.

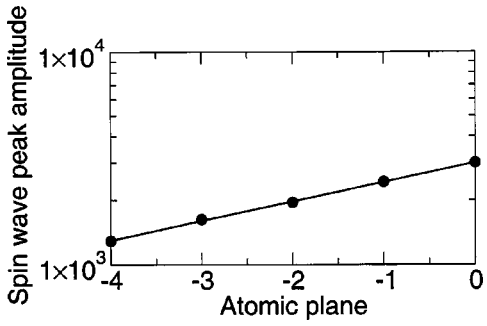

FIG. 6. We plot the logarithm of the amplitude of the lowest lying spin feature in the spectral density of Fig. 5(b) as a function of layer number, to demonstrate the amplitude of the mode falls off exponentially with layer number, measured from the surface layer of the film.

We should remind the reader about the relationship of the response function we have calculated, the wave vector and frequency dependent transverse susceptibility $\chi_{+,-}$, to the SPEELS experiments which motivated the calculations reported here. As discussed in the opening remarks of Sec. III, this function describes the response of the film to an applied transverse magnetic field of the time and spatial variation described just before Eq. (1). Some years ago, this function also entered into an early version of the theory of SPEELS.[28] Clearly, to speak in classical language, when a beam electron excites a spin wave, the magnetic moments in the substrate necessarily rotate a bit from the direction of the substrate magnetic moment. In the theory just cited, it was assumed that the moment rotated as a rigid entity unchanged in shape, an approximation which worked very well indeed for modeling the electron phonon matrix element in theories of excitation of surface vibrational quanta in electron loss spectroscopy.[29] It was demonstrated in Ref. 28 that if the moment rotates rigidly in the course of the excitation pro-

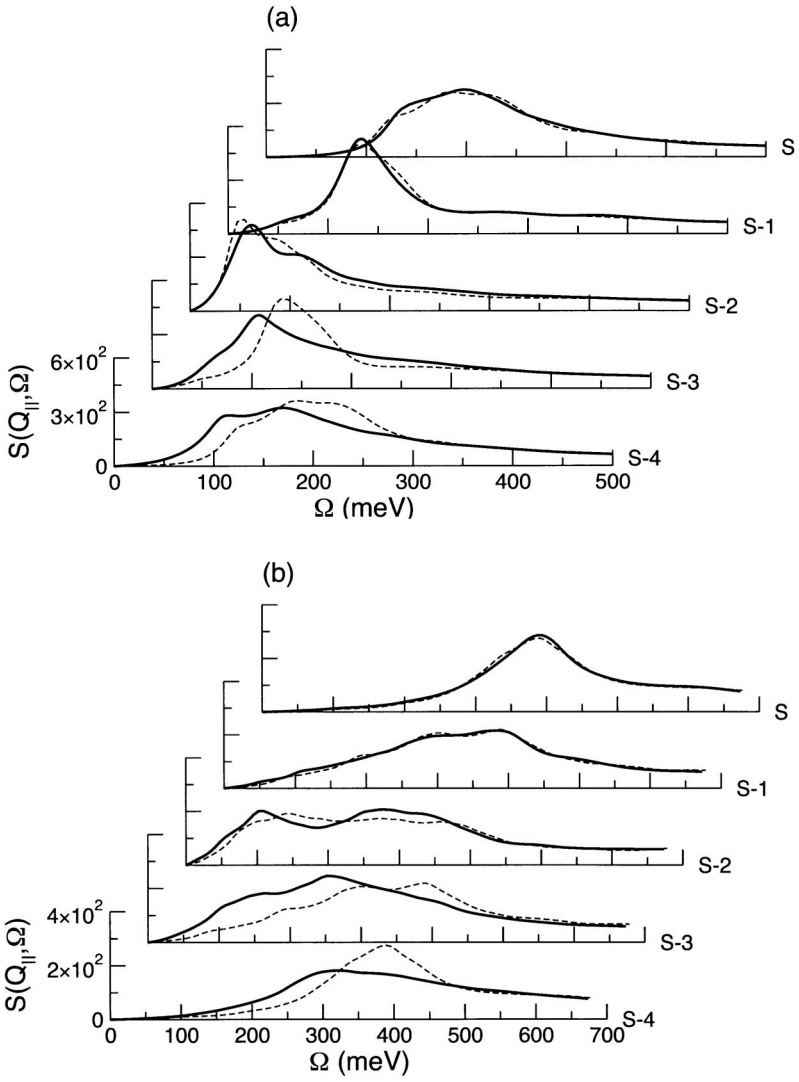

FIG. 7. The same as Fig. 5, but now we have (a) $Q_x = 0.4$ and (b) $Q_x = 0.6$.

cess, then the electron magnon matrix element may be expressed in terms of phase shifts for scattering from a ground state muffin tin potential. Also, the SPEELS spectrum can be expressed in terms of the response function $\chi_{+,-}$ discussed in Sec. III.

However, while our calculations of $\chi_{+,-}$ for both bulk Fe and free standing ultrathin Fe films displayed the spin wave structures very nicely, as we see above, in fact there is virtually no oscillator strength in the higher energy Stoner excitations,[10] which for Fe occur in the vicinity of 2 eV, the average exchange splitting in the *d* band complex. This is very problematical for the theory of SPEELS, since in early classic experiments, the Stoner excitations appear prominently in the loss spectra.[30] From a theoretical perspective, the origin of this difficulty is as follows. First, in the bulk material, a theorem of many body theory based on the spin rotation invariance of the underlying Hamiltonian requires all of the oscillator strength of $\chi_{+,-}$ must reside in the spin wave mode only, in the limit of zero wave vector. Our calculations then demonstrated that even far out into the Brillouin zone of both the bulk material and the film, the Stoner excitations are very weak indeed in this response function. The present studies confirm this conclusion once again.

Evidently the assumption that the moment rotates rigidly as it fluctuates in direction in response to spin wave excitation must be corrected, for the itinerant ferromagnets; once again we have a difference between the description of phonon physics and that of spin excitations in these materials. To correct this, one must employ a microscopically generated matrix element to describe the electronic rearrangement within the *d* shell, as the spin wave is excited. In essence, the moment distorts in shape as well as direction, in response to the excitation process. When this feature is incorporated into the theory, one encounters a new response function χ_{SPEELS} (Ref. 11) whose structure is such that even at zero wave vector, the Stoner excitations enter fully, in addition to the spin wave structures. Calculations of loss spectra using χ_{SPEELS} appear very realistic. From the comparison between theory and experiment given in Ref. 14, one sees that this response function provides a fully quantitative account of the relative intensity of the spin wave loss, and the background of Stoner excitations in the first experimental detection of a spin wave signal.

The calculation of χ_{SPEELS} requires information of the

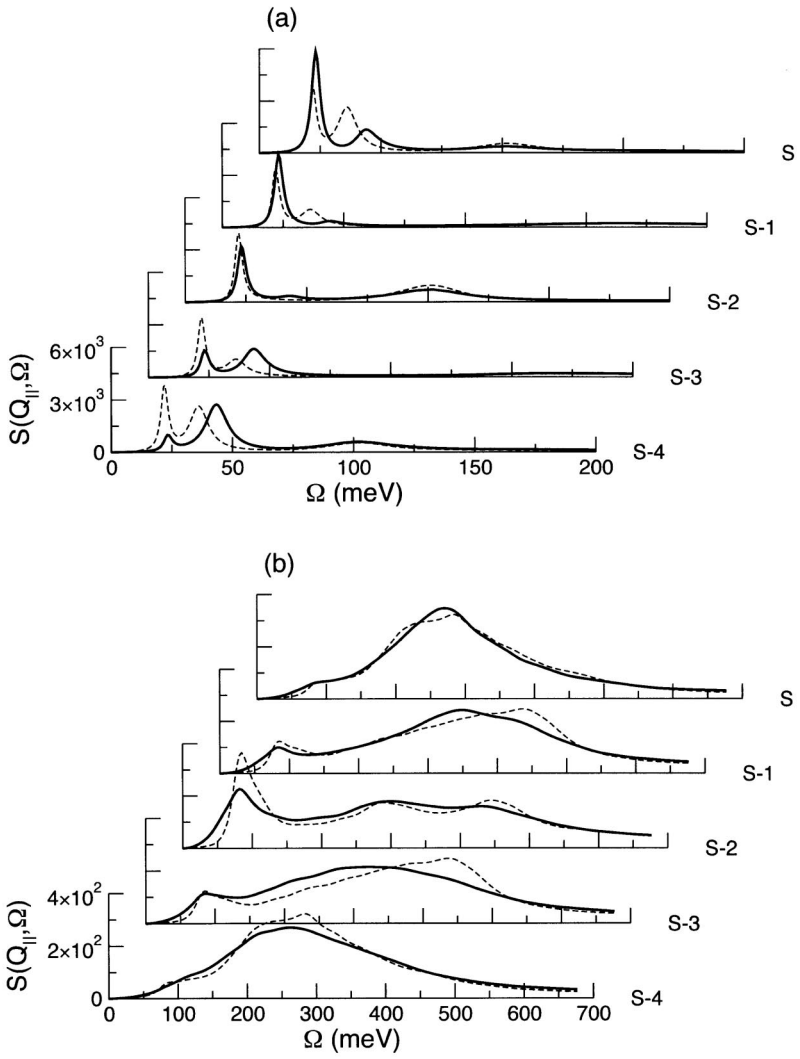

FIG. 8. For the five layer film, we show calculations of the spectral density for two values of the wave vector along the direction. We have (a) $Q_y = 0.2$ and (b) $Qy = 0.6$.

spatial form of the $3d$ wave functions in the Fe film. In Refs. 11 and 14 the calculations reported were done for bulk Fe. It is a nontrivial extension of the scheme used in these references to move from the bulk material to the ultrathin film, and in fact the cost in computational complexity to generate χ_{SPEELS} for the systems studied here would be substantial. Since it is the case the much more accessible response function $\chi_{+,-}$ contains full information on the character of the spin wave excitations in the ultrathin films, we have been content to confine our attention to it. It would be of very great interest indeed to compute full SPEELS spectra, utilizing a multiple scattering theory of the electron/sample interaction, in combination with a full description of χ_{SPEELS} so theory can provide an account a full account of the experiments, including the energy and angle variation of the excitation cross section and the loss spectrum from the spin wave regime up to and including the Stoner spectrum. Such a program has been carried out with substantial success, for the case of electron energy loss by surface vibrational quanta, it should be remarked.[29,31] The full multiple scattering description of SPEELS is in place and it has been used to calculate the absolute cross section for excitation of spin waves within the rigid rotating moment picture.[28] Evidently the results are in good accord with recent measurements.[32] What is required for the next step is the development of a description of the exchange matrix element in a form that allows computation of χ_{SPEELS}. This can then be combined with the multiple scattering development in Ref. 28 to provide the complete theory . Experiments such as those reported in Ref. 15 provide motivation for such a development.

It is perhaps appropriate to discuss the relationship between the calculations reported here, and the very beautiful experimental data presented in Ref. 15, which reports the first measurements of spin wave dispersion in ultrathin films via the SPEELS technique. The measurements extend out to the boundary of the surface Brillouin zone, so these are experimental studies of short wavelength spin excitations in these materials, in the spin wave frequency region. Alas, a direct comparison between our calculations and the data cannot be made, since the experiments probe an eight layer Co film deposited on Cu(100). However, we may make comments on general trends. First, it was not possible to access the vicinity of the zone center with sufficient resolution to access the standing spin wave features we have discussed here, since the resolution realized in the experiment is in the range of 40 meV. Under these conditions, one will observe

the acoustic spin wave, and the first standing mode will skew the feature observed by contributing to the high energy tail in the acoustic spin wave feature. It is the case that farther out in the surface Brillouin zone, only a single, very broad feature is observed which displays spin-wave-like dispersion. This is very similar to the trends found in our calculations. Surely the data provides no evidence for the sequence of standing spin wave modes predicted by adiabatic calculations of the spin wave spectra of ultrathin films, very much in accord with our conclusions. It should be remarked that we have calculations underway that will explore Co films on Cu(100), so we shall provide more detailed comparisons between theory and experiment in subsequent work.

We conclude by describing a connection between the studies reported here, and recent discussions of mechanisms which control the ferromagnetic resonance (FMR) linewidth in ultrathin ferromagnetic films.[4] In recent years, it has become evident that in ultrathin ferromagnetic films, and in multilayers which contain ultrathin ferromagnetic films, mechanisms not operative in bulk materials influence FMR linewidths substantially. One such mechanism is the following. When spins are excited in an ultrathin ferromagnet film in contact with a metallic substrate, or surrounded by metal films, a contribution to the linewidth has its origin in transfer of angular momentum from the precessing spins in the ferromagnet, to conduction electrons outside the ferromagnet.[4] We have seen in the calculations reported here that our spin wave linewidths in the ultrathin film are larger for the adsorbed film than for the free standing film. The origin of this difference is in transfer of angular momentum from the ultrathin ferromagnetic film, to the underlying substrate, very much as envisioned in the references just cited. In these theoretical studies of FMR linewidth, very simple models of the film and the surrounding media are used to make estimates of the role of this effect. The models are so schematic that such estimates are of semiquantitative validity only, clearly. Embedded in the calculations reported here is a description of precisely this effect, for a model whose underlying electronic structure is fully realistic. It would thus be of great interest to explore, within the framework used here, the transfer of angular momentum from the ultrathin film to the substrate when spin waves are excited in the ultrathin films. This should enable us to provide estimates of the magnitude of this effect, for realistic models of idealized systems, whose interfaces are perfect on the atomic scale. We hope to address this issue in future studies.

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