Theory of large-wave-vector spin waves in ultrathin ferromagnetic films: Sensitivity to electronic structure

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We present a series of theoretical studies of short wavelength spin waves in ultrathin ferromagnetic films, with attention to sensitivity of their dispersion relation to aspects of the electronic structure of the films. Our emphasis is on the influence of the magnitude of the intra-atomic Coulomb interaction U within the 3d shell. The calculations we report focus on eight layers of ferromagnetic Co adsorbed on the Cu(100) surface, a system whose spin wave dispersion relation has been studied experimentally for wave vectors throughout the surface Brillouin zone. We find the frequency of the short wavelength spin waves to be very sensitive to U. Appropriate values of this parameter produce a dispersion relation in very good accord with experiment, and a remarkably quantitative account of both the width and shape of the single feature in the spectral density found in the experiments. We have argued previously that in these systems, the adiabatic approximation (the "frozen magnon" approximation) breaks down qualitatively, with the consequence that our dynamical theory produces a single very broad feature in the spectral density at large wave vector, in contrast to the predictions of "frozen magnon" calculations, where a sequence of standing wave modes of infinite lifetime is predicted. In the present paper, we use adiabatic theory to calculate exchange constants for the ultrathin Co film adsorbed on Cu, and compare explicitly the predictions of "frozen magnon" theory with our dynamical calculations. The comparison provides insight into the origin of the broad features found in the dynamical calculations.

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I. INTRODUCTORY REMARKS

Knowledge of the collective excitations of a given class of solid materials lays the foundation from which their dynamical response to external probes may be understood, along with their thermodynamic behavior. For the last two decades, very considerable attention has been devoted to the study of ultrathin ferromagnetic films,[1] since the magnetism found in these materials has many unique features not realized in bulk matter, and also because multilayers formed from ultrathin ferromagnets have played a remarkable role in computer technology. For example, very small, highly sensitive read heads which exploit the giant magnetoresistance (GMR) found in magnetic multilayers have led to hard disks with storage densities two orders of magnitude higher than realized before the discovery of this phenomena. Other applications are under active study.

We know very little about the collective spin wave excitations of ultrathin ferromagnetic films, however. It is the case that the spin dynamics of these materials has been studied intensively by the technique of ferromagnetic resonance spectroscopy (FMR),[2] and also by Brillouin light scattering (BLS).[3] Both techniques excite the collective excitations associated with the magnetic degrees of freedom of the film, the spin waves. However, the wavelength of the modes probed by these methods is very long compared to a lattice constant. Thus, only a very tiny fraction of the surface Brillouin zone is explored, though surely we have learned a great deal from both techniques. It is a matter of fundamental interest to understand the physics of short wavelength spin wave excitations in this important class of materials; also as devices based on magnetic materials become very small, excitations of the spin system with large spatial gradients will necessarily be encountered. For the second reason, it is also a matter of practical importance to understand the nature of spin waves in the short wavelength regime.

We have been engaged in extensive theoretical studies of short wavelength spin waves in ultrathin magnetic films,[4–8], based on use of a realistic picture of the electronic structure of these itinerant ferromagnetic materials, and then through use of a dynamical theory of the spin waves (the random phase approximation) which does not resort to the commonly used adiabatic or "frozen magnon" approach. The method we have developed allows us to explore ultrathin films adsorbed on semi-infinite substrates, so the (sometimes considerable) influence of the substrate on the spin dynamics is included fully. Our calculations have predicted[4–7] that in the ultrathin films, at short wavelengths, the nature of the spectral densities, which describe the collective modes, differ qualitatively from the predictions of frozen magnon calculations. For a film of N layers, the collective modes are characterized by a wave vector parallel to the surface. Frozen magnon theories produce precisely N distinct spin wave modes for each value of this wave vector; each mode has infinite lifetime, so the

spectral densities associated with a given wave vector will consist of N sharp peaks, each one of zero width and thus described by a Dirac delta function of suitable weight. Our studies have shown this picture to be incorrect in a qualitative sense. At short wavelengths, we have predicted one should find a single very broad feature in the spectral density (sometimes with modest structure) whose maximum will display dispersion rather similar to that of a heavily damped spin wave. The origin of the damping is decay of the collective spin wave into spin triplet particle hole pairs, the Stoner excitations of the itinerant ferromagnet. This is a magnetic analog to the well-known Landau damping of plasmons in metals. We refer the reader to Ref. 7 for a complete discussion of an extensive series of calculations, for the case of Fe multilayers adsorbed on the W(110) surface.

Very recently, remarkable experimental studies have provided us with our first data on short wavelength spin excitations in ferromagnetic ultrathin films.[9] The method used was spin polarized electron energy loss spectroscopy (SPEELS). The technique allows unambiguous identification of the features in the loss spectrum through the polarization dependence of the excitation cross section.[10] The system studied was an eight-layer Co film, adsorbed on the Cu(100) surface. The spectra reported in Ref. 9 are fully compatible with the theoretical predictions discussed in the previous paragraph.

In response to this new data, we initiated calculations of the spin wave spectrum for ultrathin Co films on the Cu(100) surface.[8] While the calculated spectra show large widths at large wave vectors quite close to those seen experimentally, in fact at and near the Brillouin zone boundary the spin waves produced by theory were considerably stiffer than found experimentally. At the boundary of the surface Brillouin zone in the [110] direction, the data shows the spectral density peaks near 250 meV, whereas the calculated spin wave frequency was a bit above 400 meV.

This discrepancy has led us to explore the sensitivity of the calculated spin wave spectra to features in the electronic structure, in our description of the Co film. The electron energy bands in the film and the substrate are described within the empirical tight binding scheme, with parameters taken from fits to *ab initio* calculations of the bulk electronic structure. We refer the reader to Table 3 and Fig. 6 of Ref. 4, where for ultrathin Fe(100) films, comparison is made between the method used here and *ab initio* density functional calculations of the moment distribution and spatial variations in the local density of states. Use of bulk tight binding parameters provides good accounts of these properties for the Fe(100) films and we assume the scheme is adequate also for the Co(100) films of interest here. We require in addition a description of the Coulomb interaction within the 3d shell of the Co atom, in the ferromagnetic state. Our recent calculations employ a one-parameter scheme introduced many years ago by Lowde and Windsor.[11] We have compared[5] the predictions of the Lowde-Windsor approach with a more sophisticated scheme used in an early study,[4] to find the two in very good quantitative agreement. The one parameter we employ, U, is the Coulomb interaction in the 3d shell per Bohr magneton. Himpsel has compiled data on the intra d shell Coulomb interactions found in 3d transition metal ions in various solid state environments, and has argued that a value close 1 eV/Bohr magneton for U emerges from photoemission data as a universal parameter.[12] The calculations we reported earlier, where the short wavelength spin waves were found to be much stiffer than the data at short wavelengths, utilized this value for U.

Upon examining Fig. 2 of Himpsel's paper, where his survey is summarized, one notes that for bulk hcp Co, the value of U lies below the universal curve by some 20% or so. This suggests that for Co one should employ a value of this parameter slightly smaller than the universal value. This thought stimulated us to carry out a new series of calculations of spin waves in the Co film, with attention to the sensitivity of the short wavelength modes to the value of U. We find, in fact, that the short wavelength spin waves are surprisingly sensitive to U, far more so than the spin magnetic moment of the film. By simply reducing U from 1 eV to 0.85 eV, we are able to generate a dispersion curve quite close to that found experimentally, while as argued below the spin moment in the center of the film is reduced only a little below our earlier value. We have "tuned" other aspects of our description of the electronic structure of the film as described below, but these changes do not have an effect as dramatic as that of the intra atomic Coulomb interaction.

Thus, we conclude that data such as that presented in Ref. 9 presents a challenge to the theory of ferromagnetism in ultrathin films of transition metal elements. We find the short wavelength, dynamic response of the spin system is remarkably sensitive to the description of the electronic structure. We shall also see below that the same is true of the exchange stiffness D, which controls the dispersion relation of spin waves in the long wavelength limit, in an exchange only theory with dipolar couplings neglected. Thus, the study of short wavelength spin dynamics in the ultrathin films emerges as a remarkably sensitive test of theories of ferromagnetism in the ultrathin film limit. We remark also that a virtue of our empirical tight binding scheme is that we do have the flexibility to test sensitivity of the calculated results to features of the electronic structure, if desired. This would not be possible in a full *ab initio* calculation. Of course, our scheme also allows us to carry through complete calculations of the spin dynamics in rather thick films, with the electronic structure of the assumed semi-infinite substrate taken into account fully. As we have seen earlier[5–7], the influence of the substrate can be very substantial, most particularly for the very thin films with less than four or five layers. It is our understanding that at present, within the framework of time dependent density functional theory, a study of spin dynamics of multilayer adsorbed films remains very challenging from the computational perspective. In view of our conclusions regarding the sensitivity of the spin wave spectra to electronic structure, it would be most intriguing indeed to see complete parameter free calculations for the Co/Cu(100) system, and for other systems that will be the attention of subsequent experimental studies.

We address a second issue in this paper. As remarked above, and discussed in detail elsewhere, the adiabatic or frozen magnon approach provides a qualitatively misleading picture of the spin wave spectrum at short wavelengths, in these itinerant ultrathin films. Indeed, even near the center of

the surface Brillouin zone, our calculations show the standing wave modes to be very heavily damped with the higher modes not even evident in the spectral functions. It is only the low lying acoustic spin wave mode that has a very long lifetime, by virtue of the Goldstone theorem[13] which insures its lifetime becomes infinite in the limit of zero wave vector. However, the authors of Ref. 9 used a Heisenberg model to describe the dispersion relation deduced from the broad feature revealed by their data, and extracted parameters from the data that are surely reasonable. It is thus of interest to make a comparison between the predictions of a Heisenberg-type model, with effective exchange interactions calculated in adiabatic theory, with the results of our full dynamical theory for the same underlying electronic structure. We shall see that while the adiabatic theory, suitably interpreted, can serve as a crude guide to the excitation spectrum of the ultrathin film, its predictions cannot be used for quantitative purposes, unfortunately. In Sec. II, we describe our new dynamical calculations, and in Sec. III we present a comparison between the full dynamical calculations and spin fluctuation spectra generated by adiabatic theory.

II. SENSITIVITY OF SHORT WAVELENGTH SPIN WAVES TO ELECTRONIC STRUCTURE: Co on Cu(100)

In this section, we present our studies of the sensitivity of spin waves in ultrathin Co films on Cu(100) to the features of electronic structure discussed in Sec. I. In the interest of brevity, we confine our attention to the case where the Co film is eight layers in thickness, so the theory may be compared with the SPEELS data reported in Ref. 9. In our previous publication,[8] we have studied the dependence of various aspects of the spin waves spectrum to the thickness of the Co film. Our conclusion there was that for films with thickness greater than three or four layers, the dispersion relations, linewidths and exchange stiffness vary only modestly with film thickness. Since we have every reason to believe that similar conclusions will be found within our new series of calculations, we confine our attention here to the specific case of the eight-layer film.

As remarked above, the electronic structure of both the ultrathin film and the Cu substrate are described within an empirical tight binding scheme, with nine orbitals per site. Our description includes the five 3*d* bands, and the 3*p*/4*s* complex that overlaps and hybridizes with the 3*d* bands. Our tight binding parameters are extracted by fitting *ab initio* electronic structure calculations.[14] We use mean field theory to describe the ferromagnetism in the ultrathin film, driven by intra-atomic Coulomb interactions described by the parameter *U* introduced by Lowde and Windsor.[11] For reasons described in Sec. I, we varied *U* in the range of 0.8 eV to 1.0 eV in the series of calculations we report below.

One issue is the question of orbital occupancy in various layers of the film. In a full *ab initio* calculation the long ranged part of the Coulomb interaction is present in the Hartree term, so the electrons automatically arrange themselves within the orbitals in the various layers so the system is very close to electrical neutrality on a layer by layer basis. In our approach, we retain only the intra-atomic Coulomb interaction, and the long ranged piece is absent when we generate our description of the ground state. In earlier calculations[4–8] we have adjusted the orbital energies on a layer-by-layer basis to impose local electrical neutrality. When this is done for the case of Co on Cu(100), the *d* shell within the surface layer is filled to the extent that the moment in the surface layer is actually smaller than that in center of the film. We can see this from the last row of entries in Table I, where for the four outermost layers of the film the spin only moments are tabulated for the case where we require strict electrical neutrality on a layer-by-layer basis. We see that the surface moment is indeed smaller than that in the center of the film for this case. In fact, *ab initio* calculations[15] for the (100) surface of fcc Co show that the complex of nine orbitals in the surface have 8.66 electrons rather than 9.0, so the surface layer carries a slight positive charge. When we build this feature into our calculation, we find an enhanced surface moment as one sees from the remaining entries in Table I. It is the case that in the center of the film, our calculated spin only moment is smaller than that given in Ref. 15 by 15% for the case where $U=0.85$ eV, which will emerge as our preferred value for this parameter. It is the case that for ultrathin films of Co on Cu(100), one finds very lovely measurements of the absolute magnetization as a function of film thickness, for films that cover the range of interest to us.[16] Ney and co-workers measure the absolute magnetization as a function of film thickness for Co films deposited on the Cu(100) surface with outer surface free, and also for such Co films on which a Cu(100) overlayer is added. They are able to draw inferences on the magnitude of the enhancement of the surface moment and that for the Co layer next to the interface with Cu from the thickness dependence of the total magnetization for the two types of sample. Through extrapolation of the data to infinite thickness, they determine the moment in the center of the film to be 1.74 Bohr magnetons. We regard this value as quite reliable, though in our minds there are questions[17] about the quantitative significance of the surface and interface moments, which emerge from the analysis in Ref. 16. When the moment in the film center reported in Ref. 16 is compared to the entries in our Table I, it must be recognized that we have tabulated the spin only moment, whereas the authors of Ref. 16 measure the total moment, spin and orbital combined. Because of lattice mismatch, in

TABLE I. For the four outermost layers of the Co film, we list the spin only magnetic moments in our various calculations, in Bohr magnetons. The layer S is the surface layer of the film, S-1 is the first interior layer, and so on.

U(eV)	S	S-1	S-2	S-3
Ab initio[a]	1.85	1.69	1.73	1.71
0.80	1.59	1.45	1.33	1.25
0.85	1.66	1.53	1.50	1.59
0.90	1.70	1.56	1.53	1.53
1.00	1.73	1.59	1.57	1.57
1.00[b]	1.39	1.62	1.59	1.60

[a]From Ref. 14.

[b]For the electrically neutral surface.

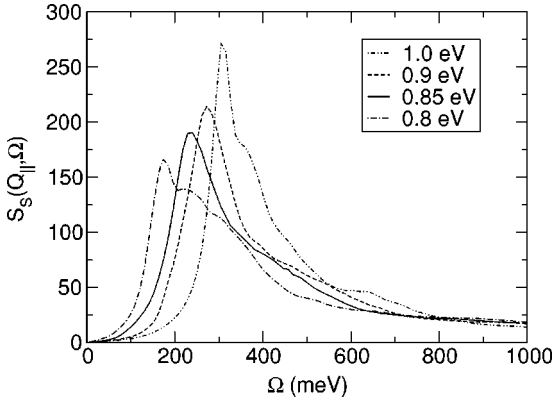

FIG. 1. We show the spin wave spectral density for spin fluctuations in the outermost layer S of the eight layer Co film on Cu(100), for the four values of the intra atomic Coulomb parameter U shown in the inset. The wave vector used in the calculations is directed along the [110] direction in the surface Brillouin zone, and has magnitude $Q=0.5$ in units of $2\pi/a_0$. The spectral density function we plot in this figure is defined in Ref. 19, and discussed in detail in Ref. 7.

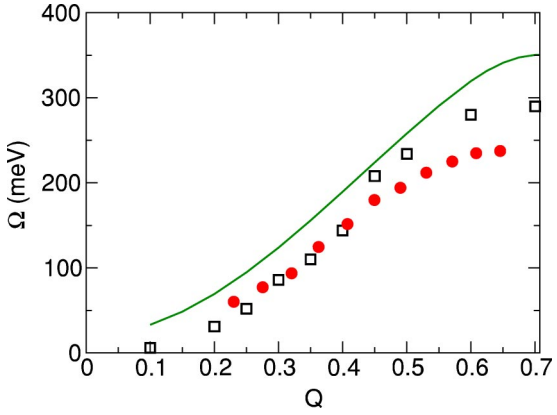

FIG. 2. The effective dispersion relation for the spin wave feature in the spectral density, from theory for the case $U=0.85$ eV (open squares) and the data reported in Ref. 9 (solid circles). The reduced wave vector Q is in units of $2\pi/a_0$, and is directed along the [110] direction of the surface Brillouin zone. The solid line is a calculation based on the adiabatic or frozen magnon approach. The calculations are for eight layers of Co on Cu(100).

fact the films are actually slightly tetragonal. This will enhance the orbital moment over that appropriate to a perfect fcc film. We understand that it is reasonable to suppose the orbital angular momentum lies in the range of 0.1–0.2 Bohr magnetons[18] for such films. This suggests our value for the spin only moment in the film center is quite reasonable, and close to what one expects for these films.

As we decrease the value of U from 1 eV used in the calculations we reported earlier to 0.85 eV, the moment in the center of the film decreases by only a bit less than 5%. However, the short wavelength spin wave frequencies decrease dramatically, as we see from Fig. 1. The figure shows the frequency spectrum of the spin fluctuations[19] in the outer layer of the Co film, for a reduced wave vector of 0.5 in the [110] direction of the surface Brillouin zone of the film. Recall that in this direction, which is that probed in the experiments reported in Ref. 9, the surface Brillouin zone boundary lies at $(1/\sqrt{2})(2\pi/a_0)$. As we lower the value of U from 1.0 eV to 0.8 eV, the peak in the spectral density decreases in frequency by almost a factor of 2! This figure contains the principal conclusion of the present paper: the frequencies of short wavelength spin waves such as those reported by Vollmer and co-workers[9] are remarkably sensitive to details of the electronic structure of the film, most particularly to the Coulomb interaction within the 3*d* shell of the magnetic ion. Thus, such data presents a remarkable challenge to the theory of spin dynamics in these materials. As remarked in Sec. I, it would be of very great interest indeed to see full *ab initio* calculations of the spin wave spectrum to see if, with no flexibility in the input parameters, the SPEELS data can be reproduced.

Through a series of spectral density calculation such as those displayed in Fig. 1, we may generate an effective spin wave dispersion curve by plotting the position of the maximum in the spin wave spectral density in the outermost layer as a function of wave vector. The dispersion curve so constructed is compared with the data of Vollmer and coworkers in Fig. 2, for the case of $U=0.85$ eV. In the figure the open squares show the trajectory of the peaks in the spectral density obtained from our dynamical theory. The solid line is the result of a "frozen magnon" calculation and will be discussed later. The frequencies which emerge from the full calculation are still slightly too stiff at the zone boundary, but we regard the agreement as very good, given the sensitivity of the frequencies to Coulomb interaction displayed in Fig. 1. It is the case that our approach is not a full *ab initio* calculation, and we see little point in tuning parameters further to improve the fit.

In Fig. 3, we show a comparison between the theoretical line shape, and the data. In Fig. 3(b), we have artificially shifted the experimental data upward in frequency, so the

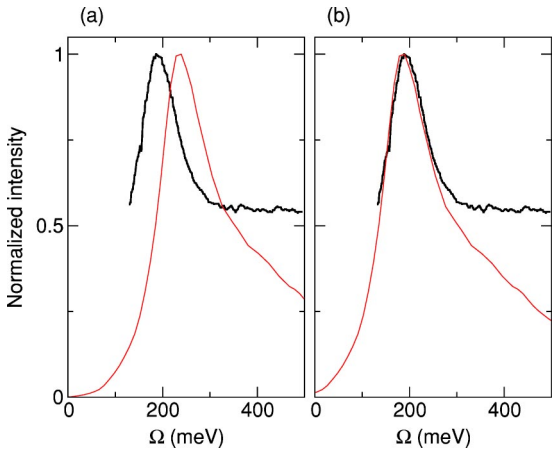

FIG. 3. (a). For the wave vector employed in Fig. 1, we show a comparison between the calculated and measured spin wave loss features. Quite clearly, the experimental trace is the curve which has small amplitude noise present. (b) Here we make the comparison between theory and experiment again, but we have shifted the data up in energy artificially so the linewidth and line shapes may be compared.

peak in the experimental SPEELS loss spectrum coincides with that in the theoretical spectrum. We see that both the width and line shape in the theory are in remarkable accord with the data. At higher energies, in the data one sees the onset of the Stoner spectrum, whereas this feature is absent in the dynamic transverse susceptibility generated by our solution of the RPA equations. As discussed at length in Ref. 7, the response function actually probed in the SPEELS experiment is not the transverse susceptibility but rather a different response function χ_{SPEELS} described and calculated in earlier studies.[20] While the dynamic susceptibility provides one with a good description of the spin wave portion of the spectrum, to obtain a proper description of the Stoner regime one needs the full response function. We refer the reader to Ref. 7 for a discussion of this issue, including a description of the underlying physics associated with the two response functions. It is our hope to generate calculations of χ_{SPEELS} for ultrathin films in the future; this requires a nontrivial extension of the calculations reported here.

We turn our attention next to a comparison between the full dynamical calculations reported here and in our earlier papers with spin wave spectra calculated within the framework of the frozen magnon, or adiabatic approach.

III. COMPARISON BETWEEN THE ADIABATIC DESCRIPTION OF THE SPIN WAVE SPECTRUM AND DYNAMICAL THEORY

As stated earlier, this section is devoted to a comparison between results calculated within the framework of the full dynamical theory described in the previous section, and the adiabatic or "frozen magnon" approach used commonly to generate spin wave spectra of itinerant ferromagnets.[21] While we have argued earlier that the latter approach is misleading for the ultrathin films, by virtue of its neglect of the very strong Landau damping evident in the results presented in Sec. II and in our earlier work, it is the case that an explicit comparison between the two approaches will prove useful, particularly if both analyses are carried out for the same underlying description of the electronic structure of the system.

In the adiabatic approach, one calculates exchange interactions $\mathbf{J}_{\boldsymbol{l},\boldsymbol{l}'}$ between the magnetic moment in unit cell $\boldsymbol{l}$ with that in unit cell $\mathbf{l}'$. One imagines rotating the magnetic moment in cell $\boldsymbol{l}$ away from the perfectly aligned ferromagnetic state, and then one may calculate the torque experienced by the moment in unit cell $\boldsymbol{l}'$ and deduce $\mathbf{J}_{\boldsymbol{l},\boldsymbol{l}'}$ from this torque. For the case of ultrathin films, a formal expression from which one may calculate such exchange constants is found in an earlier publication of ours.[5] With the exchange constants so calculated in hand, one may then describe the spin system within the framework of the Heisenberg model, with spins of unit length in each unit cell. The effective Hamiltonian is then

$$H = -\sum_{\mathbf{l},\mathbf{l}'} \mathbf{J}_{\mathbf{l},\mathbf{l}'} \hat{e}(\boldsymbol{l}) \cdot \hat{e}(\boldsymbol{l}), \tag{1}$$

where $\hat{e}(\boldsymbol{l})$ is a unit vector. Quite clearly, if one makes *static* deformations in the spin system, Eq. (1) provides an adequate description of the energy change, so long as the angle between adjacent spins is small (if this is not the case, then one must be concerned about the role of biquadratic exchange, and other higher order couplings). The question we address here is whether the Hamiltonian in Eq. (1) can be used to describe *dynamic* motions of the spin system. It has been established for many years that if spin waves with wavelength long compared to a lattice constant are of interest, then the dispersion relation may be described through use of the exchange stiffness constant D, and adiabatic theory correctly generates this parameter.[22] At issue is the relevance of the adiabatic approach for calculation of the properties of short wavelength spin waves, such as those probed in the SPEELS experiment in Ref. 9.

For the ultrathin film, the spin wave normal modes of a Heisenberg system have been discussed many years ago.[23,24] The small amplitude spin waves may be described by the appropriate linearization of the equations of motion for the operator (in ourcurrent notation) $\hat{e}_+(\boldsymbol{l})$. For the film one seeks solutions of the form $\hat{e}_+(\hat{l}) = \hat{n}(l_\perp;\alpha)\exp[i\boldsymbol{Q}_\parallel \cdot \boldsymbol{l}_\parallel - i\Omega_\alpha(\boldsymbol{Q}_\parallel)t]$, where the index α ranges from 1 to N, with N the number of layers in the film. The eigenmodes may be either surface modes, localized to one or the other surface in the short wavelength limit, or standing wave bulk modes. In terms of the layer amplitudes $\hat{n}(l_\perp;\alpha)$, the spectral density functions such as those displayed in Fig. 1 are given by, in the Heisenberg picture,

$$S_{l_\perp}(\boldsymbol{Q}_\parallel,\Omega) = \sum_{\alpha=1}^{N} |\hat{n}(l_\perp,\alpha)|^2 \delta(\Omega - \Omega_\alpha(\boldsymbol{Q}_\parallel)). \tag{2}$$

As discussed above, within the framework of the adiabatic or frozen magnon approach, for any wave vector, we have N modes each with infinite lifetime.

We have calculated the intersite exchange couplings which enter Eq. (1) for our eight layer Co film on Cu(100), through second nearest neighbors utilizing the expression given in Ref. 5. We remark that it appears to be the case, if one examines the array of exchange constants calculated for bulk fcc Co, [25] that reliable dispersion curves should be generated with exchange constants through second neighbors.[25] Our results for the film are tabulated in Table II. With these exchange constants in hand, we may calculate the dispersion curves and eigenvectors for the eight normal modes associated with each wave vector $\boldsymbol{Q}_\parallel$.

In Fig. 4, for a reduced wave vector of 0.6 in the [110] direction of the surface Brillouin zone (recall that in these units, the zone boundary is located at the reduced wave vector of $1/\sqrt{2}$), we plot the layer dependent spectral densities $S_{l\perp}(\boldsymbol{Q}_\parallel,\Omega)$ generated by the full dynamical calculations, and compare to those generated by the Heisenberg model. Of course, we have artificially broadened the sharp features present in the Heisenberg model a bit. Generally speaking, as we have argued earlier,[7] the broad structures evident in the dynamical calculations can be viewed as having their origin in an array of Heisenberg-type eigenmodes, each broadened severely by the strong Landau damping present in the ultrathin film. In the end, in the spectral density we have a single, broad structure that shows dispersion and which has the appearance of a single, short-lived excitation. The lowest

TABLE II. Exchange coupling constants for the eight layer film, in meV. The nearest neighbor interplane interactions in column S-N describe coupling between moments in plane S-N and S-N-1, while the next nearest neighbor interaction in column S-N describes coupling between moments in plane S-N and plane S-N-2.

Layer No.	S	S-1	S-2	S-3	S-4	S-5	S-6	S-7
nn, in plane	19.32	10.82	10.63	10.18	10.22	10.90	9.28	14.08
nnn, in plane	0.15	1.86	2.27	2.09	2.07	2.29	1.98	1.91
nn, interplane	16.20	9.95	11.01	10.22	10.84	9.90	10.63	
nnn, interplane	2.14	2.34	2.21	2.25	2.64	1.54		

eigenmode which contributes to the spectral density in the outer layer of the film is a surface spin wave mode whose amplitude decays as one moves into the film interior. The authors of Ref. 9 argued that such a surface mode dominated the loss spectra they measured, it should be noted. Similarly, the lowest mode evident in the spectral density of the innermost layer of the Co film is also a surface spin wave whose frequency lies lower than the mode in the outer layer.

If we look at the frequency of the surface mode in the outer layer in Fig. 4, its frequency is distinctly higher than the peak in the spectral density which emerges from the full dynamical calculation, so the Heisenberg model is not so accurate from a quantitative point of view. We view the origin of this frequency shift in the following terms; what follows is an intuitive picture, it should be remarked. As we move from the adiabatic approach to a full dynamical theory, we may view the Landau damping as having its origin in the imaginary part of the self-energy which enters the spin wave propagator. If we call this self-energy function $\Pi(\boldsymbol{Q}_{\parallel},\Omega)$, then the adiabatic theory incorporates only $\Pi(\boldsymbol{Q}_{\parallel},0)$ where as the full dynamical theory adds in $\Delta\Pi(\boldsymbol{Q}_{\parallel},\Omega)=\Pi(\boldsymbol{Q}_{\parallel},\Omega)-\Pi(\boldsymbol{Q}_{\parallel},0)$. The strong damping has its origin in $\mathrm{Im}(\Delta\Pi(\boldsymbol{Q}_{\parallel},\Omega))$. But the Kramers Kronig relation requires that $\mathrm{Re}(\Delta\Pi(\boldsymbol{Q}_{\parallel},\Omega))$ be nonzero as well. This introduces frequency shifts in the full dynamical theory absent from the adiabatic theory. In our earlier study of spin waves of the Fe monolayer of W(110),[5] we found this frequency shift to be not so large, and this seems to be the case for the surface mode localized on the innermost layer of the Co film considered presently. In the presence of damping as strong as we see in the dynamical calculations, the shift can be appreciable.

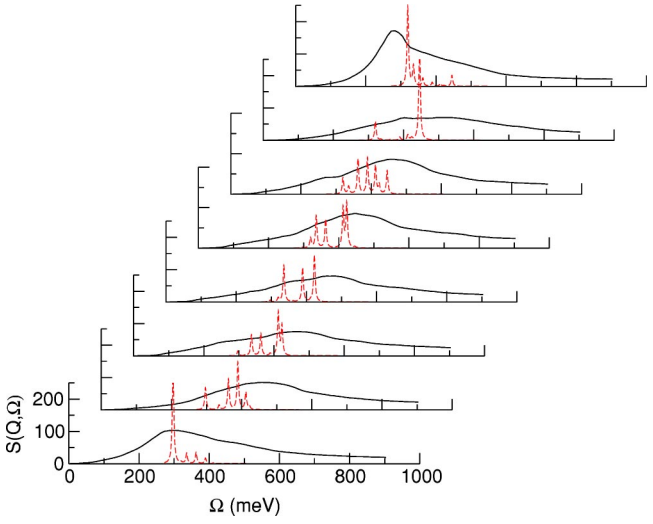

FIG. 4. For the reduced wave vector of 0.6 along the [110] direction of the surface Brillouin zone, we compare the spectral density functions generated by the full dynamical calculation (smooth curves) with those generated from adiabatic theory, using the exchange constants given in Table II. We show each layer in the eight layer film. The outermost layer is at the top of the figure, and the innermost layer is at the bottom.

In Fig. 5, we show a comparison between the spectral densities at smaller values of the reduced wave vector along the [110] direction, $Q=0.3$. The adiabatic results do not provide us with much insight into the results which emerge from the full calculations. Another issue arises. This is that as the reduced wave vector Q becomes smaller, the eigenvector of the surface mode on the outer layer of the film penetrates throughout the film, and the same is true of the surface mode localized on the inner layer, which we see from Fig. 4 occurs at a somewhat lower frequency than the mode localized on the outer layer. We thus see in Fig. 5 that on the outermost layer, the lowest frequency structure comes from the tail of the surface mode localized on the inner layer, though it is the case that the surface mode localized on the outer layer has a much stronger influence on the spectral density. If one

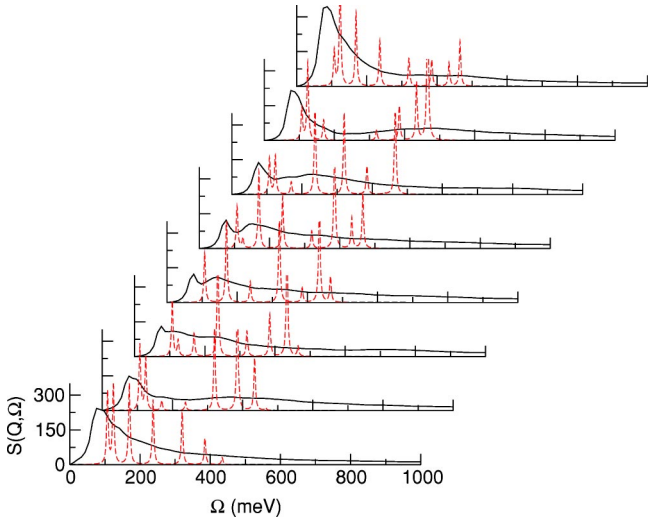

FIG. 5. The same as Fig. 4, but now the reduced wave vector is $Q=0.3$.

wishes to plot a single effective dispersion curve identified as the dominant structure throughout the entire surface Brillouin zone, it is not so clear to us which mode to choose.

We return to Fig. 2, where we have superimposed onto the data and the results of the dynamical calculations of the dispersion curve of the surface mode localized on the outer layer. It is the case that this mode is considerably "stiffer" than that found experimentally. As Q approaches zero, this mode evolved into the first standing wave mode of the film, whereas the lower frequency surface mode localized on the inner layer at large Q evolves into the uniform mode.

If we follow the procedure of Ref. 9, and fit the results of the full dynamical calculation in Fig. 2 to the form $8JS(1-\cos(qa_0))$, with a_0 the nearest neighbor distance, and q the wave vector along the [110] direction of the surface Brillouin zone, we then find a value of JS quite close to the value 15 meV discussed by these authors. This yields an exchange stiffness in the range of 390 meV A^2, quite close to that reported in an early neutron study of a bulk fcc $Co_{0.92}Fe_{0.08}$ alloy.[26] However, a direct calculation of the spin wave exchange stiffness produces the smaller value 238.7 meV A^2. Evidently the data at the lowest wave vectors explored in Ref. 9 does not quite take it into wave vectors small enough for the long wavelength quadratic dispersion law Dq^2 to be applicable. We note that the calculated exchange stiffness is also sensitive to the value of U. When $U=1$ eV, the exchange stiffness D assumes the much larger value of 489 meV A^2,[27] and we find a roughly linear variation of D with U in the range $0.8\ \text{eV} < U < 1.0$ eV. We should note that Brillouin light scattering studies of much thicker Co films grown on Cu(100), with thickness in the 200 A range, produce the value 466 meV A^2.[28] It would be of great interest to see direct measurements of the exchange stiffness on true ultrathin films of Co on Cu(100).

IV. FINAL REMARKS

We have presented a series of calculations which illustrate that the frequency spectrum of short wavelength spin waves in ultrathin films of Co on Cu(100) are very sensitive indeed to the Coulomb interaction within the 3d shell of the Co atoms. With a value of this Coulomb interaction only 15% smaller than Himpsel's suggested "universal value" for transition metal ions in the solid-state environment, we generate a dispersion curve in good agreement with the SPEELS data presented in Ref. 9. Agreement between theory and experiment in regard to the very large linewidths measured experimentally is excellent, as we see in Fig. 2. The value for the intra d shell Coulomb interaction we use here, 0.85 eV, is in fact fully compatible with that deduced by Himpsel for bulk hcp Co, and thus seems quite reasonable.

As remarked above, it would be of very great interest indeed to see fully parameter free time dependent density functional calculations carried out for this system. Our analysis suggests that the dynamic response of the material in the short wavelength regime will provide a much more demanding test of theory than ground state properties, such as the magnetic moment. It is our hope that the present discussion will stimulate interest in such calculations.

We have also presented a direct comparison of the predictions of the adiabatic or frozen magnon theories of spin waves, with the results of our dynamical calculations. In our view, the adiabatic theories provide us with only limited insight into the spin wave spectrum of such itinerant ultrathin films. While such adiabatic calculations work splendidly for the generation of phonon spectra for almost all solid materials, they fail badly with regard to the short wavelength spin excitations of itinerant ferromagnets, as we see in this paper. Of course, phonons also are damped by decay to particle-hole pairs. But the electron phonon matrix element that controls this decay is small compared to unity when expressed in dimensionless form, with the consequence that the damping is weak. In contrast to this, for spin waves in itinerant ferromagnets, the dimensionless coupling constant is of order unity and the damping is strong. In the bulk, for very long wavelengths the Goldstone theorem[22] ensures that the damping rate is small, with the consequence that spin waves are long-lived at small wave vectors. The theorem has been extended to untrathin films,[4] where it assures us that at small wave vectors the low-lying acoustic spin wave has a long lifetime. As we and others have proved,[5,22] a consequence is that the exchange stiffness may be calculated rigorously within adiabatic or "frozen magnon" theories. However, such calculations cannot be extended to large wave vectors. We refer the reader to our earlier publications for a more complete discussion of this point.[4–8]

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[18] We are grateful to Professor Ruqian Wu for a discussion of this issue.
[19] What is plotted in Fig. 1 is the spectral density function $S_l(\mathbf{Q}_\parallel,\Omega)=\mathrm{Im}\{\chi_{+,-}(\mathbf{Q},\Omega;l,l)\}/\pi$ introduced in Ref. 4 and explored in or more recent papers. A detailed discussion of this function, and its relationship to the SPEELS data reported in Ref. 9 has been given by us in Ref. 7.
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