Progress and Future Plans

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“Spin Polarized Probes of Magnetic Nanostructures”

D. L. Mills, Principal Investigator

Institute of Surface and Interface Science

University of California

Irvine, California 92697

Abstract:

This report summarizes progress to date in our theoretical research program, for the period from July 1, 2002 to November 1, 2003. In addition, our research priorities for the coming year are set forth. The reporting period has been a most exciting and significant one. For the past several years, one of our principal thrust areas has been development of the theory of spin dynamics in magnetic nanostructures with emphasis on the use of spin polarized electrons as probes of short wavelength spin dynamics in such entities. Our program stimulated the first experiment which detected large wave vector spin waves in ultrathin films in 1999 through spin polarized electron loss spectroscopy (SPEELS); the publication which announced this discovery was a joint publication between a group in Halle (Germany) with our theory effort. The continued collaboration has led to the design and implementation of the new SPEELS spectrometer and we now have in hand the first detailed measurements of spin wave dispersion in an ultrathin film. A second such spectrometer is now operational in the laboratory of Prof. H. Hopster, at UC Irvine. We are thus entering a most exciting new era in the spectroscopy of spin excitations in magnetic nanostructures. During the reporting period, we have completed very important new analyses which predict key aspects of the spectra which will be uncovered by these new instruments, and the calculations continue to be developed and to expand our understanding. In addition, we have initiated a new series of theoretical studies directed toward spin dynamics of single magnetic adatoms on metal surfaces, with STM based studies of this area n mind. In the near future, these studies will continue, and we will expand our effort into new areas of spin dynamics in magnetic nanostructures.

I. Introductory Remarks:

As noted in the abstract of this report, we have just entered a most exciting period in our program of research. In this introduction, we provide an overview of these developments.. Our efforts are directed toward a most important aspect of magnetically active nanostructures: the dynamical response characteristics of the spins within them. We note that while very considerable (and very successful) theoretical studies of small magnetic structures have been set forth in the past decade or two, virtually all of this has been directed toward what we may call ground state properties (spatial distribution of magnetic moments, orbital moments, anisotropy,.....) . So far as we know, our program is unique in its emphasis on microscopic understanding of spin dynamics in magnetic nanostructures. Our efforts have stimulated the design of new electron spectrometers directed toward the study of spin dynamics, and we now have very exciting new data in hand for the very first time. In essence, a new spectroscopy has been developed in two laboratories in direct response to our past proposals and calculations, and the new spectrometers are now operational. They will provide a major stimulus to our program. At the same time, a new thrust of our effort is directed toward studies of spin motions with very high spatial resolution, through use of a scanning tunneling microscope with spin polarized tunneling current emission. In this section, we provide an overview and historical setting for the new discoveries of the past year.

Spin excitations and related magnetic response characteristics of ultrathin ferromagnetic films have been studied experimentally for nearly two decades, it should be noted. However, the two methods used for these studies, Brilluoin light scattering (BLS) and ferromagnetic resonance spectroscopy (FMR) employ radiation whose wavelength is very long compared to, say, the underlying lattice constant or interatomic separations in the structures probed. It follows that these methods excite spin excitations or spin waves whose wavelength is macroscopic, orders of magnitude longer than the lattice constant. The resulting spin motions also have very low frequencies, in the microwave regime. From the theoretical perspective, such spin wave modes are accurately described through use of phenomenological Hamiltionians which contain only ground state parameters (anisotropy constants, magnetization, exchange stiffness, ...) Both FMR and BLS have by now provided us with fascinating and important information regarding the physical properties of ultrathin films and magnetic multilayers. However, it is the case that we still lack information on truly microscopic aspects of spin dynamics in such structures. As magnetic nanostructures become ever smaller in device applications, we will need to understand the nature of short wavelength spin motions in these systems, so the issues involved will prove important in the near future.

To acquire such microscopic information, one needs to employ a probe whose wavelength is comparable to lattice constants. In bulk materials, neutron scattering proves a powerful probe with this property. However, in the materials of current interest employed in magnetic nanoscale devices, the method is of little value. First, in the ferromagnetic transition metals, the short wavelength spin waves have excitation energies very large compared to the kinetic energy of thermal neutrons, so the probe particles have insufficient energy to excite such modes. One may employ high energy neutrons from spallation sources in principle, but then one encounters problems with the excitation cross section, since the 3d form factor falls off rapidly with wave vector transfer, thus reducing the excitation cross section dramatically in the large wave vector region of interest. For these reasons, we have remarkably little data in hand on even the short wavelength spin waves in the classical ferromagnetic metals Fe and Ni at this late stage of the game. Of course, when applied to ultrathin films, another complication with neutron probes is the lack of surface sensitivity.

Some years ago, we proposed that highly monoenergetic beams of electrons form, in principal, an ideal potential probe of shortly wavelength spin waves, through the spin polarized version of electron energy loss spectroscopy referred to as SPEELS. Electron loss spectroscopy (without spin polarized beams) has proved a powerful and versatile probe of vibrational quanta on crystal surfaces, with measurements that span the entire surface Brilluoin zone. The issue on our minds was to extend it to the study of spin excitations as well, and we set out to formulate the theory which would outline how this can be done and what might be observed and learned if the experiments can be carried out.

Indeed, in an earlier phase of the present DOE sponsored theory effort, which was in the form of a collaboration with Prof. S. Y. Tong, we developed and implemented a remarkably successful quantitative theory of the excitation of surface vibrational quanta in electron energy loss spectroscopy. Our direct collaboration with experimental groups led to the development of detailed models of surface lattice dynamics, and our theory allowed us to extract information on the nature of the polarization of individual vibrational modes, through comparison of theory with the energy and/or angle dependence of the excitation cross sections [1].

In the effort directed toward spin excitations studied by SPEELS, two areas were outlined that required theoretical attention. First, we had to extend our theory of the excitation of surface vibrational quanta to the excitation of spin waves. Second, in a lengthier and more challenging phase of the program, we needed to develop the quantitative theory of spin excitations in the itinerant materials of interest. In the arena of lattice dynamics, at the time we began our studies of electron probes of surface vibrations, there was a mature theory of bulk lattice vibrations, ranging from few parameter phenomenological models with clear physical content, to ab initio theories. In the case of spin excitations, the body of theory was very limited and it became clear early on we required calculations which address the itinerant character of the magnetism in the materials of interest. In the research completed in the period under review, our calculations have demonstrated that this is truly essential for the ultrathin films, and our key predictions are illustrated beautifully from the new data generated by the Kirschner group, at the Max Planck Institut in Halle, Germany.

It proved most difficult to excite interest from experimentalists in the study of spin waves via SPEELS. There are several reasons for this. Early in our program, we completed a quantitative calculation of the excitation cross section for creating spin waves in

SPEELS, to find it roughly three orders of magnitude smaller than the cross section for exciting surface vibrational quanta [2]. This combined with the desire to use spin polarized beams, which can discriminate between spin wave losses from a strong background of spin independent scattering necessarily present, presented the experimental community with a major challenge. There was an additional problem. In 1983, in a failed experiment, a leading figure in the field of electron loss spectroscopy was unable to observe spin wave losses on the Ni(100) surface [3]. This was known widely in the field, and others were discouraged to spend the time in effort to attempt such a difficult experiment. To succeed, we had to surmount a barrier, so to speak.

A major step forward was our quantitative theory of spin wave losses in SPEELS in Fe, where we calculated the intensity of the spin wave loss feature in a fully microscopic description of the excitation process, and normalized it to the previously observed bands of Stoner excitations in this material [4]. Our clear conclusion was that in Fe, the spin wave loss peak should be observable, if one can look in the low energy wing of the SPEELS loss spectrum. Our prediction stimulated Prof. J. Kirschner of the Max Planck Institute in Halle (Germany) to carry out a new experiment, and the result was his first observation of a loss feature from spin wave excitation in an ultrathin film of Fe grown on the W(110) surface. The paper which announced this discovery was a joint publication [5].

We were still left with a puzzling question. Why did Ibach fail to see spin waves on the Ni surface, in his 1983 experiment, whereas the loss feature stood out clearly in Kirschner's study of Fe? We found the answer to this puzzle. The explanation lies in the fact that in Ni, the ground state exchange splitting between minority and majority spin bands are some six times smaller than in Fe, with the consequence the Stoner continuum in the SPEELS spectrum washes out the spin wave loss peak, to leave one with a broad featureless spectrum in Ni in the large wave vector regime probed by Ibach. This left one final problem. Short wavelength spin waves have been probed by neutrons in Ni; why can they be seen by neutrons and not electrons? The answer is that the two probes explore different response functions. We calculate each, to find clear spin wave losses and almost no Stoner spectrum for neutrons, and only a broad featureless Stoner loss spectrum for the same wave vector transfer for electrons [6].

At this point, the theoretical issues are clear and with the 1999 Kirschner experimental data in hand, we knew that the signals were indeed there as predicted, and observable. Alas, the spectrometer in Halle did not have sufficient energy resolution to allow both wave vector and energy to be scanned and thus for actual dispersion curves to be measured. However, our predictions and the new data stimulated H. Ibach to design a new electron spectrometer explicitly oriented toward SPEELS studies of spin waves. This was built in Julich, moved to the Max Planck Institut in Halle, and has now produced the first data on spin wave dispersion in ultrathin films, measured out to the boundary of the surface Brilluoin zone [7]. It is the case as well that a new SPEELS spectrometer has been built at UC Irvine in the laboratory of Prof. Herbert Hopster. This is operational as well, and we shall see data emerge from it very shortly. As the new spectrometers were under construction, in our theory effort we have succeeded in developing programs which, within the framework of itinerant electron descriptions of the magnetism in ultrathin films deposited on semi infinite substrates, will perform fully quantitative calculations of the spin wave spectra. This has been a most exciting period; the next phase of the program will involve quantitative comparison between theory and real data. It has taken a long time to arrive at the position in which we find ourselves now, it should be remarked.

The comments in this opening section have focused entirely on our theoretical studies directed toward SPEELS studies of spin waves in ultrathin films. We have made very considerable progress in other arenas as well, as described in section II. However, at the moment, the major development is the appearance of SPEELS data, in response to the predictions and results which have emerged from our efforts of the past few years, so the focus of this overview section has been on this topic.

II. Research Completed During the Present Reporting Period:

This section of the report is devoted to specific research projects completed during the present reporting period.

A. Theory of Spin Wave Excitations In Multilayer Ultrathin Ferromagnetic Films; The Case of Fe on W(110):

We have completed a major piece of research during the present reporting period. We have now achieved the ability to calculate, within our full dynamical theory, the spin wave spectrum of multilayer ferromagnetic films adsorbed on metallic substrates [8]. The calculations use a fully realistic electronic structure for the itinerant system, with electronic states within the film fully hybridized with those in the substrate.

The electronic structure in this analysis is modeled within an empirical tight binding scheme, which includes nine bands, the five d bands of the film and the substrates, and the sp band structure which crosses and hybridizes with it. Film and substrate are linked through an appropriate choice of hopping integrals between the outer atomic layer of the substrate, and the innermost layer of the film. Ferromagnetism in the film is treated in the mean field approximation, driven by the intra atomic coulomb interactions within the 3d shell. All hopping integrals in our tight binding modeling of the electronic structure are chosen from fits to bulk electronic structure calculations. Are only adjustable parameters are the strengths of the intra atomic coulomb integrals within the 3d shell. These are adjusted to fit measured ground state properties of Fe. Most of our calculations are performed within a scheme introduced many years ago by Lowde and Windsor; there is only one parameter here chosen to reproduce the ground state moment, and the results for the spin dynamics with this simple picture agree nicely with those generated from a more sophisticate scheme we had used in earlier work. The scheme just described does not take proper account of the long ranged character of the Coulomb interaction. Ab initio studies show that the Coulmb interaction, when treated fully, leads to a ground state wherein electrical neutrality is maintained atomic plane by atomic plane, to an excellent degree of approximation. We thus adjust our single particle energy levels within the substrate/film combination to achieve charge neutrality on a plane by plane basis.

Once the ground state is set up as described, we use the random phase approximation to calculate the wave vector and frequency dependent susceptibility of the film in a mixed representation, where for a given frequency the response function is labeled by the wave vector parallel to the surface, and for the atomic planes. We have no adjustable parameters in our calculation of the spin dynamics, it should be remarked. It is the view of this writer that we regard it as a major accomplishment to carry through a full RPA description of the spin dynamics, for a ferromagnetic film adsorbed on the semi infinite substrate.

In addition to providing quantitative predictions for the nature of the spin wave excitations in this classic system, our results clearly establish a number of qualitative points. For instance, there is what may be described as a small industry of theorists with backgrounds in density functional theory, who are calculating effective exchange integrals between magnetic moments in the cells of itinerant ferromagnets, including the ultrathin films, using the adiabatic approach wherein spins are tipped statically, resulting torques calculated, and then effective exchange couplings are deduced from the torques. Given a complete set of such exchange integrals, spin wave spectra may be generated through resort to an effective Heisenberg Hamiltonian. When applied to the ultrathin film of N layers, such an approach would generate N standing wave spin wave branches for each value of the wave vector parallel to the surface. Each such spin wave would have infinite lifetime. Thus, if one were to make a plot of the spectral density associated with each wave vector parallel to the surface, one would see a sequence of N Dirac delta functions, one for each spin wave mode of the structure.

We find results that differ qualitatively from the picture just mentioned; we have established that, in fact, in the ultrathin films we have examined, the adiabatic approximation breaks down severely. Throughout much of the surface Brilluoin zone, instead of a sequence of standing spin wave modes as described in the last paragraph, we see a single, very broad structure which exhibits dispersion rather like a single spin wave mode. Near the center of the surface Brillouin zone, there is a standing spin wave mode whose width is small, but we see only two or three standing spin waves at higher energy rather than the N which would emerge from a Heisenberg like Hamiltionian. Except for the acoustic mode, the remaining modes are very heavily damped. The new results from Halle are fully compatible with our picture, it should be remarked, since their spectra indeed show a single, broad asymmetric peak very similar in nature to that which emerges from our calculations.

It was our intent to perform calculations on the system to be explored first by the Halle group. Alas, they had difficulty resolving signals from multilayer Fe films, and found instead that multilayer Co films on Cu(100) worked much more nicely. Hence, during the next research year, our attention will be devoted to the Co/Cu system.

B. Many Body Effects and Spin Dependent Tunneling in Spin Valve Structures:

There is great interest in spin dependent tunneling in spin valve structures. One has a three layer system, with two ferromagnetic films separated by an insulating barrier. The magnetization of one layer is pinned, possibly via the exchange bias provided by an antiferromagnetic substrate upon which the structure is grown. The tunneling current then exhibits a strong dependence on the orientation of the magnetization in the topmost layer, relative to that of the pinned layer underneath.

On qualitative grounds, one understands the origin of the spin dependent tunneling quite easily. When the magnetizations of the two ferromagnetic films are parallel, majority spin electrons emitted from one film tunnel into majority spin states in the second, and similarly for minority spins. Upon reversing the magnetization of the upper film, electrons emitted from a majority spin band tunnel into a minority spin band, and conversely. Thus, the tunneling asymmetry can result straightforwardly from simple density of states effects.

However, when one begins to examine details, simple theory fails to describe the observations. At relatively high biases, the tunneling magnetoresistance falls off with voltage substantially, in disagreement with elementary theory based on electrons tunneling through a simple barrier. In addition, there is a substantial asymmetry with the sign of the applied voltage which does not emerge from the simple models. Various proposals have been put forward to explain these effects; the most prominent of which invokes inelastic tunneling mediated by spins very near the barrier [9]. (The principal investigator has serious reservations about aspects of the calculation set forth in ref. [9], it should be stated.)

We have proposed an intrinsic effect of many body origin which may explain both effects described in the previous paragraph. This follows from the earlier work completed in our DOE supported program, on the quantitative theory of the spin dependence of the hot electron mean free path in the 3d ferromagnetic metals [10]. There we found that excited electrons in these materials couple very strongly to spin waves, when their energy is within one or two electron volts of the Fermi energy—this is precisely the range where the tunneling magnetoresistance anomalies occur. We argue that in strong ferromagnets, the excited electrons should not be regarded as simple band electrons, but rather they are renormalized by their interaction with spin waves. The renormalization becomes more pronounced with increasing excitation energy, and influences the tunneling characteristics as the bias increases. Furthermore, these effects can lead to substantial asymmetries in the dependence of the tunneling magnetoresistance on the sign of the voltage, if as is usually the case the two ferromagnets incorporated into the junction are fabricated from different materials.

We have developed a model description of the effects described in the previous paragraph, and carried out a series of model calculations of the tunneling resistance of spin valves, to find the effect is indeed substantial [11]. While our model description is too primitive to provide quantitative accounts of tunneling characteristics of real spin valve structures, clearly the results establish the importance of this intrinsic mechanism. It would be of very considerable interest to extend this analysis to more realistic models, but the real spin valve structures are fabricated from complex, poorly characterized materials so in our judgment the point is made clearly by our simple model, and this suffices for the moment.

C. Theory of Spin Dynamics of Adatoms on Metal Surfaces:

As discussed in section I, we now have in place a new spectroscopy of spin excitations in ultrathin ferromagnetic films (and, we assume on other kinds of magnetic surfaces eventually) by means of the SPEELS technique. One may inquire about whether other methods can be developed which allow high spatial resolution; of course the SPEELS measurement averages over a large surface area.

We have been intrigued by recent developments in STM based spectroscopies. It is now possible with STMs to perform a spectroscopy on the vibrational motions of isolated, selected adatoms or molecules adsorbed on surfaces, and to probe their electronic structure as well. It would be intriguing if one could probe the spin dynamics of single adatoms or very small nanostructures through use of this technique. In the Wiesendenger group in Hamburg, spin polarized currents have been generated through STM tips, and used to image magnetic structures on surfaces with atomic scale spatial resolution. In our view, it should be possible to perform the analog of the SPEELS measurements discussed in section I not with an external electron beam, but rather with the spin polarized current from a scanning tunneling microscope. We have initiated new theoretical studies with this possibility in mind, and during the last reporting period we completed our first theoretical study in this area. Before continuing on, we should remark that we hope to have experiments underway directed to this possibility at UC Irvine, in the very near future. The group of Prof.Wilson Ho now has operational a remarkable new STM from which spin polarized currents have been generated. It is the first apparatus anywhere which can make measurements in a magnetic field, with field sweeps as the measurement is underway. The Hamburg group can only make measurements in fixed external field, since in their apparatus a change in external magnetic field alters the tip/sample separation in an uncontrolled manner. At UC Irvine, the Ho group has under construction a new low temperature, high field STM apparatus which should be operational within the next year. This apparatus will be perfectly suited to address the issues under discussion here.

An important first question to address is the nature of spin dynamics of magnetic adatoms and small units, when they are found on the metal surfaces used typically in STM studies. With this issue in mind, we have completed quantitative calculations of the dynamic susceptibility of Co and Mn atoms on the Cu(100) surface, within the framework of the empirical tight binding scheme employed in our studies of the spin wave excitations of ultrathin films.

These calculations explore a most interesting and fundamental theoretical issue. We have used a Hamiltonian which contains only the one electron hopping integrals of empirical tight binding theory in combination with intraatomic coulomb interactions between electrons, suuplemented by a Zeeman interaction of spins with an external fields. Such a Hamiltionian is invariant under spin rotations, and communtes with the operator which describes the total z component magnetic moment of the system. One may then show that if the system is driven by a time dependent transverse magnetic field, the *total moment* of the system exhibits a resonance response with no g shift, and with zero width. Thus, in an ESR experiment, for our model system, there would be no g shift and the linewidth would be zero. In real materials, there is a g shift and linewidth, of course, but these have their origin in terms which violate spin rotation invariance. Spin orbit coupling is an example. However, in the STM experiment we envision, the core of the moment will be probed by the instrument, rather than the total moment. Thus, even in the absence of spin orbit coupling, a linewidth will be present in the response function relevant to the measurement, with origin in coupling of the magnetic moment core to Stoner excitations in the subtrate (Landau damping again, in essence).

For Co and Mn on Cu(100), we have calculated the response function, with attention to both the linewidth, and apparent g shift. Both are quite substantial, since they have their origin in the intra atomic coulomb interaction, rather than the very much weaker spin orbit interaction. Furthermore, the magnitude of both is affected sensitively by the electronic structure of the adsorbate. The Co ion has its minority spin virtual level virtually overlapping with the Fermi level, with the consequence its motions are quite heavily damped. In contrast, for Mn, the Fermi level falls roughly midway between the majority and minority spin virtual levels, with the consequence the damping is much weaker.

These calculations establish basic principles which can guide the experimentalist in the choice of adsorbate/substrate combinations. Also, we now have programs in hand which can explore diverse systems, if desired. This is the initial study in what we envision to be a sequence of studies of the spin dynamics of very small magnetic entities on metallic surfaces. We envision a rich spectroscopy will emerge from STM probes of magnetic excitations.

D. The Collective Excitations of Nanoscale Dielectric Sphere Arrays:

During the past decade, much of the focus of the present program has been on the nature of spin excitations in ultrathin ferromagnetic films and structures which incorporate them, and the means of probing such enetities, with SPEELS as an example. In the renewal proposal, the program set forth had a broader perspective, with a new emphasis on other classes of nanoscale entities. The analysis of the spin dynamics of single ad atoms described in the previous subsection is an example of the new thrust we envision. The same is true of our theory of the enhancement of dynamic dipole moments of molecules and other adsorbates under an STM tip, by virtue of their excitation of localized plasmons of the tip/substrate combination [13], [14]. In the present reporting period, we have addressed another issue: the collective plasmon excitations of arrays of interacting metallic spheres, modeled as dielectrics with an appropriate frequency dependent dielectric constant [15].

The motivation for the analysis here came from very interesting experiments in the laboratory of Prof. J. C. Hemminger, at UC Irvine..In his laboratory, graphite surfaces are grown with very straight, parallel steps. Nanosized Au spheres are deposited on the surface, and it is found they migrate to the steps, to form linear arrays of spheres nearly touching each other. A question arises regarding the nature of the collective excitations of such arrays, their response to electric fields associated with radiation incident on the surface, and ultimately on their influence on the reflectivity of the surface. At issue is whether one may use such self organized arrays to tailor or design a surface with desired optical response characteristics, with absorption bands located in appropriate spectral regions. Of course, the presence of such linear arrays will render the reflectivity anisotropic as well.

In the literature on photonic band gap structures, one finds multiple scattering formalisms which allow one to analyze electromagnetic propagation in periodic arrays of dielectric spheres. The formalism is quite formidable in nature; it was developed for structures whose length scales are comparable to the wavelength of the radiation incident on them, so the full Maxwell equations with retardation included are required in these studies. The nanosphere arrays of interest to us involve spheres whose diamter is perhaps 100 times smaller than the wavelength of the radiation, and the spheres nearly touch. Under such circumstances, retardation effects are quite negligible, and we can use a quasi-static approach. Within such a framework, we have developed a versatile, real space formulation of the problem that is readily and easily programmed for numerical calculations. We should remark that it will be possible to extend the formalism to arrays of magnetic spheres, to address their microwave response. With our formalism, we have carried out studies of the collective exciations of linear arrays of spheres, along with their response to a spatially uniform electric field of appropriate frequency. The results are quite fascinating in our view, and suggest that one can indeed tailor the reflectivity of surfaces by using linear arrays of spheres of appropriate diameter and spacing. We wish to actually calculate the reflectivity of such a model surface in the future, as the reader will see from our discussion of future plans, presented below.

III. Research Plans for the Coming Grant Year, November 1, 2003-October 31, 2004:

In this section, we describe the specific research projects we wish to pursue in the upcoming grant year. Much of the motivation and background for these investigations has been covered in the discussions given in section I and section II. Thus, the descriptions below are rather brief, with reference to the more elaborate comments above.

A. Quantitative Calculations of the Spin Wave Spectra of Ultrathin Films: The Case of Co on Cu[100]:

As we have seen in section I, very exciting new SPEELS data is now available which provides us our first information on the spin wave dispersion in an ultrathin (eight layer) film of ferromagnetic FCC Co on Cu(100). While we have carried out extensive studies of spin waves in ultrathin ferromagnetic films in the reporting period covered by this report, the system we chose (which we thought would be that explored first experimentally!) was the classic system Fe on W[110]. The highest priority for our research program in the coming year is to carry out similar calculations for Co on Cu[100]. To do this, we have to adapt our programs to the different symmetry, notice. We will be most eager to see the results, since this should offer us the first opportunity to test the quantitative accuracy of our approach.

B. Spin Dynamics of Small Magnetic Structures on Metal Surfaces:

As we saw in section IIC, we have completed studies of the spin dynamics of isolated magnetic ions adsorbed on Cu[100]. The motivation for these calculations is the possibility of probing such spin excitations, through inelastic STM based tunneling spectroscopy. It will be a real challenge to see a single ion, such as that we have studied, however, since the precession frequency is in the microwave region. This frequency is far below the resolution of any conceivable STM system, including the very low temperature, high magnetic field systems currently under construction in various laboratories. It is the case that one can conceive of methods for probing the spin motions, For instance the ion can be driven by an externally applied microwave field, and there will be modulation of the tunneling current at the precession frequency of the driven moment. One may then perform in indirect electron spin resonance experiment by sweeping the applied DC magnetic field through resonance. This is a possible, but challenging experiment.

It is much more likely that one can probe, say, a pair of magnetic ions on neighboring sites, or probe a small structure which will have spin excitations in the form of internal degrees of freedom. These can be expected to have reasonably high excitation energies controlled by, in effect, nearest neighbor exchange interactions. If such entities are placed on a metal surface, as would be expected for an STM measurement, then once again the issue of Landau damping and its influence will arise. We plan calculations of the excitation spectra of pairs of magnetic ions on neighboring sites, and other structures. For instance, at UC Irvine, the in laboratory of Prof. Wilson Ho, linear structures are created artificially on the NiAl[110] surface. So far, these have been fabricated only from noble metal atoms, but it is our understanding that one dimensional magnetic chains of atoms can be fabricated as well. Studies of the magnetism of such entities will be fascinating, and we shall explore their spin excitation spectra. It should be remarked that as we address such systems, we shall be in active interaction with Prof. Ruqian Wu, who has ab initio calculations underway of the anisotropy realized in such novel structures.

C. The Dipole/Exchange Spin Wave Modes of Ferromagnetic Spheres; Macroscopic Theory:

In the previous section, we discussed calculations of the spin wave excitation spectra of very small few atom magnetic entities, within the framework of microscopic theory, wherein a realistic electronic structure forms the basis for the analysis. When the object becomes sufficiently large that it contains many moment bearing ions, such an approach breaks down. In addition, new physics becomes important. For example, the low lying excitations commonly encounterd in FMR and BLS experiments have excitation energies influenced importantly by spatially non-uniform dipolar fields excited by the spin motions.

As the entity becomes large, the low lying excitations may be described accurately within the framework of macroscopic theory. In the next research period, we shall study the theory of exchange/dipole spin wave modes of spherical ferromagnetic particles. With exchange ignored, and only Zeeman and dipolar energies included in the analysis, the problem was solved in very elegant papers which appeared in the late fifties. The most notable work was that of Walker, and the magnetostatic modes of the ferromagnetic sphere are referred to often as Walker modes. His analysis, carried out for the more general case of a sample of elliptical shape, is a most formidable piece of mathematical physics. Later authors addressed the specific case of the sphere, which can be handled somewhat more straightforwardly from the mathematical perspective. These classic theoretical papers were directed toward studies of macroscopic samples of YIG and related materials. Now we are in an era where nanosized particles are the focus of attention in the magnetic data storage industry, and it has become necessary to comtemplate incorporating exchange in the treatment. An understanding of the character of the exchange/dipole modes of the ferromagnetic sphere is imperative if the spin dynamics of the very small magnetic particles now used in data storage is to brought under control.

In our formulation, we shall describe the influence of exchange through introduction of the appropriate exchange stiffness parameter D, and the dipolar fields will be generated through use of a magnetostatic potential, as in earlier theories. The problem is formidable; while the model particle is spherical, and we know very well in theoretical physics how to address spherical geometries, the presence of the spontaneous magnetization breaks the symmetry, thus rendering the problem most challenging/ We remark that while we did not discuss this in section II, during the reporting period we devoted considerable effort to the problem. We have completed a very elegant and lovely analysis which, in the end, leads us to a hierarchy of coupled radial equations which, when solved, will lead to the mode structure. It is a formidable task to carry through a complete formal solution. We plan to continue with this analysis during the coming research period.

D. Theory of STM Based Spectroscopy of Spin Excitations:

In section IIC and section IIIB, we discussed completed and proposed studies of spin excitations of few atom magnetic entities adsorbed on metal surfaces, with STM based inelastic tunneling spectroscopy as a possible means of accessing such spin motions. In essence, we envision an STM based spectroscopy which might be viewed as a SPEELS experiment with atomic scale spatial resolution.Vibrational spectroscopy has now been performed with atomic or molecular scale spatial resolution, i. e. it is now possible to place an STM tip directly over a selected adsorbate, and measure its vibrational normal modes as it sits on a metal surface. Prof. M. Persson has formulated a theory of the intensity of the various vibrational normal modes seen in this spectroscopy, and this theory has been most impressive in its quantitative success. In essence, he calculated the spatial variation of the electron-phonon matrix element which controls the STM based vibrational excitation probability, with impressive success indeed.

We wish to formulate a similar theoretical description of STM based excitation of spin excitations in small structures, and carry through explicit quantitative calculations of the excitation probability to see if, in fact, the excitation cross sections are large enough to observe. The recent successes of the SPEELS experiments are very encouraging to us. If one can see spin excitations with an external electron beam as the exciting source, one would think that one should be able to see them as well with the electron emitted from an STM tip as the exciting source. Indeed, the basic excitation mechanism will be of an exchange character, and we also know from beam based polarized electron experiments that the excitation cross section increases with decreasing energy. Hence, the use of electrons with energy in the range of 1 eV or so above the Fermi energy should be advantageous.

We have established a collaboration with Prof. Persson, on this project. He recently spent a sabbatical year at UC Irvine, and during this period we made substantial progress with the general formulation of the problem. He will return to us for a stay of six weeks near the beginning of the next calendar year, with the purpose of discussing the next step. This will thus be pursued actively in the next grant period.

E. The Reflectivity of Dielectric Surfaces Decorated with Metallic Nanospheres:

In section IID, we discussed our completed project which explores the collective excitations of arrays of nanospheres, along with their interaction with spatially uniform electric fields. Of considerable interest would be the calculation of the reflectivity of surfaces such as those in the Hemminger laboratory, which involve one dimensional line of metallic spheres deposited along steps on a graphite surface. Since the steps are monatomic in nature, we can model the system as a perfectly flat surface on which parallel, one dimensional strings of spheres are depositied.

We will explore the extension of our analysis to the actual calculation of the reflectivity of the surface. At the time of this writing, we do not have a formal approach within which such an analysis can be framed. We envision dividing the region into three segments. We can have the vacuum which begins just above the layer of spheres. Here we have the incident and reflected light. The second region will contain the spheres, and within this region, the quasi static description will be applied to the fields, with the combination of the fields from the incident and the reflected light the external driving fields. Then in the substrate, which is the third region, we shall have the transmitted wave.

References for Sections I-III:

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IV. Manuscripts Completed During the Reporting Period:

“Many Body Effects on the Tunneling Magnetoresistance of Spin Valves”, Jisang Hong, R. Wu and D. L. Mills, Phys. Rev. B**66**, 100406 (2002).

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“Local Spin Dynamics of Magnetic Moments on Metal Surfaces”, R. B. Muniz and D. L. Mills, Phys. Rev. B (in press).

“Theory of Spin Excitations in Fe(110) Multilayers”, A. T.Costa, R. B. Muniz and D. L. Mills, Phys. Rev. B (in press).

V. Personnel Supported During the Present Reporting Period:

In addition to one month’s summer salary for each of the two summers which fall in the reporting period, the grant provided salary support to a visiting Postdoctoral Researcher, Dr. Rodrigo Arias. In addition, salary support in the form of a Graduate Student Research Assistantship was provided to Ping Chu in the summer of 2003. Ms. Chu will continue to be actively involved with the program during the 2003//2004 academic year, though present plans call for her support to come from the Department of Physics and Astronomy.

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