Theory of Brillouin Light Scattering from Ferromagnetic Nanospheres

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We develop the theory of Brillouin light scattering (BLS) from spin wave modes in ferromagnetic nanospheres, within a framework that incorporates the spatial variation of the optical fields within the sphere. Our recent theory of exchange dipole spin wave modes of the sphere provides us with eigenvectors. When properly normalized, these eigenvectors allow calculation of the absolute cross section of various modes which contribute to BLS spectrum. We then present explicit calculation of the BLS spectrum associated with the first few dipole/exchange spin wave modes with emphasis on their relative intensity.

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I. INTRODUCTION

As lithographic techniques have rapidly developed in the last decade, it has become possible to assemble ordered magnetic elements on the nanometer scale. These have attracted interest as potential candidates in magnetic data-storage devices. Spin wave excitations control the dynamic response of the magnetization in the linear response regime, and the speed of real devices at least for small amplitude motions of the magnetization. This provides motivation for theoretical and experimental studies of spin waves in magnetic wires, dots and other nanoscale structures.

In our previous paper, we provided an analytical method to calculate eigenvalues and eigenvectors of dipole-exchange spin waves in ferromagnetic nanospheres, and we explored the properties of these modes. A question is how one may access these modes experimentally. We argue here that Brillouin light scattering spectroscopy (BLS) offers the means of probing the dipole-exchange spin waves of very small spheres. In this paper, we develop the theory of BLS from a single metallic nanosphere, and illustrate with explicit calculations that one may access the spectrum of dipole/exchange spin waves modes by this means.

As we know, there are two complementary experimental techniques to study spin waves in small structures such as nanoscale particles or spheres: ferromagnetic resonance (FMR) spectroscopy and Brillouin light scattering (BLS). FMR uses microwaves to excite spin waves, but the microwave wavelength is much larger than the radii of nanospheres. It is the case as well that their radius is far smaller than the skin depth of microwaves, which is typically a micron for materials of current interest. Thus, FMR produces a uniform exciting field inside nanospheres. This makes it impossible to excite the full spectrum of dipole-exchange modes of such an object. Only the uniform mode is excited in this circumstance. However, the laser photon used in BLS has a small skin depth at optical frequencies. This is typically 10-20 nm in the ferromagnetic metals of interest. This is comparable with the radius of nanoscale spheres, and the resulting non-uniform optical field can thus, in principle, excite a full spectrum of dipole-exchange modes. It is then of interest to develop an explicit description of the scattering cross sections associated with the various modes, and to obtain insight into the factors which control these cross sections. This is the purpose of the present paper. We shall see below from the calculations presented in the present paper that can indeed excite and study higher order dipole exchange modes through the BLS method. We shall conclude that BLS is a powerful tool which one can provide information about the nature of the spin wave spectrum of nanoscale spheres. This will be true of samples of less simple shape as well, of course. A quantitative theory of the BLS spectrum of more complex objects is quite non-trivial, unfortunately. While our calculations apply only to small spheres, our principal conclusion that the method can excite the dipole-exchange spin wave spectrum of nanoscale magnetic entities is quite general, in our view.

From the quantum point of view, the process of BLS in ferromagnets is viewed as an interaction between light photons and magnons. From the classical point of view, the thermal fluctuations of magnetization inside the sphere will change the dielectric tensor of the ferromagnetic material in a time dependent manner. The inelastic scattering event has its origin in the scattering of the laser light from these dynamic fluctuations in the dielectric constant. We use a semi-classic method here to address this issue.

In this paper, we present the theory of BLS from spin waves in ferromagnetic nanospheres, and we calculate the cross section of BLS from several low lying dipole exchange modes. We shall see that for small spheres, the cross section for exciting particular higher order dipole-exchange modes is appreciable, indeed larger than that of the uniform mode for sufficiently small spheres.

II. THE STRUCTURE OF THE THEORY

We consider a metallic ferromagnetic nanosphere with radius in the range of 10nm to 60nm, with vacuum outside. Since the linear dimensions of the sphere are large compared to the lattice constant, this allows us to use
 continuum theory to describe the spin motions in the sphere. The ferromagnetic nanosphere is magnetized uniformly along $z$-direction with saturation magnetization $M_s$. At small but finite temperatures, the thermally excited magnetization can be written by $\tilde{M}(\vec{r}, t) = M_s \hat{z} + m(\vec{r}, t)$, where $m(\vec{r}, t)$ is a small amplitude disturbance associated with thermal spin waves. As noted earlier, the eigenvectors which describe the dipole-exchange spin waves of the sphere can be obtained from our previous paper. These allow us to describe the spin wave contributions to $m(\vec{r}, t)$ provided that the eigenvectors are properly normalized. We remark that the prescription for normalizing such eigenvectors has been discussed only quite recently. In the regime where dipolar contributions to the excitation energy are appreciable, the prescription for normalizing the eigenvectors is not so obvious.

The fluctuation of magnetization causes a time-dependent change of the dielectric tensor of the material, which we write as $\delta\epsilon_{\mu\nu}(\vec{r}, t) = \sum_\lambda K_{\mu\nu\lambda}(\vec{r}, t)$. We retain terms only first order in the transverse magnetization, to generate a description of BLS by processes in which a single spin wave quantum is created or destroyed. When applied to static magnetization arrangements, the term just described controls Faraday rotation. An early theoretical analysis of the spin wave BLS spectra of 3d ferromagnetic films shows that a fully quantitative account of surface and bulk spin wave contributions follow by choosing $K_{\mu\nu\lambda} = K\varepsilon_{\mu\nu\lambda}$, where $\varepsilon_{\mu\nu\lambda}$ is the Levi Civita tensor, appropriate to bulk materials of cubic crystal structure. We adopt this picture here, so that we have

$$\delta\epsilon_{\mu\nu}(\vec{r}, t) = K \sum_\lambda \varepsilon_{\mu\nu\lambda} m_\lambda(\vec{r}, t).$$

As just discussed, thermal fluctuations produce time and space varying components to the magnetization, which in turn lead to fluctuations of the dielectric tensor. Now we turn to the description of the light scattering by electromagnetic radiation by dielectric tensor fluctuations in the sphere. We describe the electromagnetic fields in the vicinity of the sphere by Maxwell’s equations:

$$\nabla \times \vec{E}(\vec{r}, t) = -\frac{1}{c} \frac{\partial \vec{B}(\vec{r}, t)}{\partial t},$$  

$$\nabla \times \vec{B}(\vec{r}, t) = \frac{1}{c} \frac{\partial \vec{E}(\vec{r}, t)}{\partial t},$$

where within the sphere,

$$D_\mu(\vec{r}, t) = \epsilon E_\mu(\vec{r}, t) + \sum_\nu \delta\epsilon_{\mu\nu}(\vec{r}, t) E_\nu(\vec{r}, t),$$

with $\epsilon$ is the complex dielectric constant of the material from which the sphere is fabricated. Since $\delta\epsilon_{\mu\nu}(\vec{r}, t)$ varies slowly in time with respect to the laser field, from Eq. (4) we have,

$$\frac{\partial D_\mu(\vec{r}, t)}{\partial t} = -i\omega E_\mu(\vec{r}, t) - i\omega \sum_\nu \delta\epsilon_{\mu\nu}(\vec{r}, t) E_\nu(\vec{r}, t),$$

where $\omega$ is the frequency of the incident laser. Upon inserting this into Maxwell’s equations, we have

$$\nabla \times \vec{E}(\vec{r}, t) = i\omega \vec{B}(\vec{r}, t),$$

$$\nabla \times \vec{B}(\vec{r}, t) = -i\omega \vec{E}(\vec{r}, t) + \frac{4\pi}{c} \vec{J}^{eff}(\vec{r}, t).$$

We have defined $J^{eff}_\mu(\vec{r}, t) = \frac{1}{4\pi}\sum_\nu \delta\epsilon_{\mu\nu}(\vec{r}, t) E_\nu(\vec{r}, t)$. Upon using the relation between the dielectric tensor and the magnetization as in Eq. (1), we obtain the form of the effective current density, given by

$$J^{eff}_\mu(\vec{r}, t) = \frac{\omega K}{4\pi} \sum_{\nu,\lambda} \varepsilon_{\mu\nu\lambda} m_\lambda(\vec{r}, t) E_\nu(\vec{r}, t).$$

Since the thermal fluctuations are very small in amplitude, we will treat the effect of the term involving $\vec{J}^{eff}$ in Eq. (7) in the Born approximation. Thus, we may replace $E_\nu(\vec{r}, t)$ by $E_\nu^{(0)}(\vec{r}, t)$ in Eq. (5), with $E_\nu^{(0)}$ the amplitude of the incident field. Thus, we have

$$J^{eff}_\mu(\vec{r}, t) = \frac{\omega K}{4\pi} \sum_{\nu,\lambda} \varepsilon_{\mu\nu\lambda} m_\lambda(\vec{r}, t) E_\nu^{(0)}(\vec{r}, t).$$

The problem is now that of computing the radiation fields generated by the effective current density within the sphere. Of interest to us are radiation fields in the far zone, as $r \to \infty$. We may proceed by following the approach outlined by Jackson. Far from the sphere, the radiation fields may be described as a superposition of fields of TM and TE character. In the far zone, the TM component of the radiation field has the magnetic field perpendicular to $\vec{r}$, the unit vector in the radial direction, while the electric field has both a radial and transverse component. The TE mode has the character of $\vec{E}$ and $\vec{B}$ interchanged. Given the radial component of $\vec{E}$ in the far zone, one can derive expressions for the transverse components of both $\vec{E}$ and $\vec{B}$ for the TM mode. Furthermore, one can derive equations satisfied by $\vec{r} \cdot \vec{E}$ everywhere:

$$[\nabla^2 + \frac{\omega^2}{c^2} \epsilon(\omega)](\vec{r} \cdot \vec{E}) = -\frac{4\pi \omega}{ic^2} \vec{r} \cdot \vec{J}^{eff}, (r < R),$$

$$[\nabla^2 + \frac{\omega^2}{c^2} ](\vec{r} \cdot \vec{E}) = 0, (r > R).$$

Similar statements can be made regarding the TE fields. One can derive equations satisfied by $\vec{r} \cdot \vec{B}$, and given $\vec{B}$ in the far zone, one can derive the remaining components of $\vec{B}$. The combination $\vec{r} \cdot \vec{B}$ satisfies:

$$[\nabla^2 + \frac{\omega^2}{c^2} \epsilon(\omega)](\vec{r} \cdot \vec{B}) = \frac{4\pi \omega}{ic} \vec{L} \cdot \vec{J}^{eff}, (r < R),$$

$$[\nabla^2 + \frac{\omega^2}{c^2} ](\vec{r} \cdot \vec{B}) = 0, (r > R),$$
where $\vec{L} = \frac{1}{i} \vec{r} \times \vec{\nabla}$ is the orbital angular-momentum operator of quantum mechanics.

We can calculate the radial components of the electromagnetic fields by using Green’s functions. By having solutions for the radial components of $\vec{B}$ and $\vec{E}$, we can generate a complete description of the TM and TE multipole modes in the far zone, where the fields have a radiative character. The analysis proceeds very much along the lines given in Jackson’s text, so we only provide the final results. The transverse field components in the far zone are of interest, since these control the radial flow of outgoing scattered energy. The expressions for these field components in the far zone may be written

\begin{align}
E_\theta^{(tot)} &= i \frac{e^{ik_0 r}}{r} \sum_{l,m} \frac{1}{(i)^l(l+1)} \left[ \Lambda^{(TM)}_{l,m} L_\varphi Y_{l,m}(\theta, \varphi) - \Lambda^{(TE)}_{l,m} L_\theta Y_{l,m}(\theta, \varphi) \right], \\
E_\varphi^{(tot)} &= -i \frac{e^{ik_0 r}}{r} \sum_{l,m} \frac{1}{(i)^l(l+1)} \left[ \Lambda^{(TM)}_{l,m} L_\theta Y_{l,m}(\theta, \varphi) + \Lambda^{(TE)}_{l,m} L_\varphi Y_{l,m}(\theta, \varphi) \right], \\
B_\theta^{(tot)} &= i \frac{e^{ik_0 r}}{r} \sum_{l,m} \frac{1}{(i)^l(l+1)} \left[ \Lambda^{(TM)}_{l,m} L_\theta Y_{l,m}(\theta, \varphi) - \Lambda^{(TE)}_{l,m} L_\varphi Y_{l,m}(\theta, \varphi) \right], \\
B_\varphi^{(tot)} &= i \frac{e^{ik_0 r}}{r} \sum_{l,m} \frac{1}{(i)^l(l+1)} \left[ \Lambda^{(TM)}_{l,m} L_\varphi Y_{l,m}(\theta, \varphi) + \Lambda^{(TE)}_{l,m} L_\theta Y_{l,m}(\theta, \varphi) \right],
\end{align}

where $k_0 = \omega/c$, and we note the angular momentum operators $L_\varphi = -i \frac{\partial}{\partial \theta} \frac{\partial}{\partial \varphi}$ and $L_\theta = -i \frac{\partial}{\partial \varphi} \frac{\partial}{\partial \theta}$. The quantities $\Gamma^{(TM)}_{l,m}$ and $\Gamma^{(TE)}_{l,m}$ can be viewed as the $(l, m)$ components of the effective current densities, with origin in the thermal fluctuations of the magnetization, as we have seen. The source $\Gamma^{(TM)}_{l,m}$ generates scattered radiation of TM character, while $\Gamma^{(TE)}_{l,m}$ generates scattered radiation of TE character. We have the forms

\begin{align}
\Gamma^{(TM)}_{l,m} &= \int \frac{d^3r}{R} j_i(kr) Y_{l,m}(\theta, \varphi) \cdot \vec{r} \times \vec{\nabla}, \\
\Gamma^{(TE)}_{l,m} &= \int d^3r j_i(kr) \vec{r} \cdot \vec{\nabla} Y_{l,m}^* \theta, \varphi,
\end{align}

where $k = k_0 \sqrt{\varepsilon}$ is the (complex) wave vector of electromagnetic radiation inside the sphere. The imaginary part of $k$ controls the penetration depth of the incident laser radiation.

In Eq.(12), the factors $\Lambda^{(TM)}_i$ and $\Lambda^{(TE)}_i$ have the following physical interpretation. The effective currents just described create scattered radiation inside the nanosphere; this radiation must be transmitted through the surface of the sphere to the outside. These quantities are the transmission coefficients for TM and TE waves of partial wave character $l$, respectively. We need to discuss properties of these transmission coefficients to appreciate the reason why the numerical results in section III display the systematics we shall see. We begin by quoting the general expressions for $\Lambda^{(TM)}_i$ and $\Lambda^{(TE)}_i$, and then we pause to explore their behavior in the limit of interest to us.

We express the factors as

\begin{align}
\Lambda^{(TM)}_i &= \frac{4\pi k_0 k^2}{\varepsilon} \left( \frac{j_i(kR)h^{(1)}_{l-1}(kR) - j_{l-1}(kR)h^{(1)}_l(kR)}{\epsilon H_l(k_0 R)j_i(kR) - h^{(1)}_l(k_0 R)G_i(kR)} \right), \\
\Lambda^{(TE)}_i &= \frac{4\pi k^2}{\varepsilon} \left( \frac{j_i(kR)h^{(1)}_{l-1}(kR) - j_{l-1}(kR)h^{(1)}_l(kR)}{H_l(k_0 R)j_i(kR) - h^{(1)}_l(k_0 R)G_i(kR)} \right).
\end{align}

We have introduced $G_l(x) = x j_{l-1}(x) - l j_l(x)$ and $H_l(x) = x h^{(1)}_{l-1}(x) - l h^{(1)}_l(x)$, where $j_l(x)$ is the spher-
ical Bessel function of the first kind, and \( h_1^{(1)}(x) \) is the spherical Hankel function of the first kind. The wave vector \( k \) in Eq.(14) is again the (complex) wave vector of electromagnetic radiation within the material from which the nanosphere is fabricated. Thus, the magnitude of the product \( kR \) is roughly the radius of the nanosphere divided by the optical skin depth \( \delta \). As noted in section I, this ratio will be in the range of unity, for samples of interest. The quantity \( k_0 \) is the free space wave vector of electromagnetic radiation of frequency \( \omega \) and for spheres whose diameter is near the smaller end of the the range of 10-60 nm we shall have \( k_0 R \ll 1 \). Thus, of interest is the relative magnitudes of \( \Lambda_l^{(TM)} \) and \( \Lambda_l^{(TE)} \) when \( k_0 R \ll 1 \).

Upon using small argument forms for the relevant Hankel functions, one finds in this limit

\[
\Lambda_l^{(TM)} \approx \frac{(k_0 R)^{l+2}}{(2l-1)!!} \frac{4\pi k^2 R}{c} \left\{ \frac{j_l(kR)h_1^{(1)}(kR) - j_{l-1}(kR)h_1^{(1)}(kR)}{lj_j(kR) + G_l(kR)/\epsilon} \right\}, \quad \text{(15a)}
\]

and

\[
\Lambda_l^{(TE)} \approx \frac{(k_0 R)^{l+1}}{(2l-1)!!} \frac{4\pi k^2 R}{c} \left\{ \frac{j_l(kR)h_-^{(1)}(kR) - j_{l-1}(kR)h_-^{(1)}(kR)}{lj_j(kR) + G_l(kR)} \right\}. \quad \text{(15b)}
\]

In Eq.(15a) and Eq.(15b), the factors in curly brackets are close in magnitude to each other. We thus see that in the limit that the wavelength of the laser radiation is large compared to the radius of the sphere, the intensity of TE radiation with angular momentum \( l \) (its strength is controlled by \( \Lambda_l^{(TE)} \)) will be stronger than the TM radiation with angular momentum \( l \) (its strength is controlled by \( \Lambda_l^{(TM)} \)) by the large factor of \( (k_0 R)^{-2} \). This follows since \( \Gamma_{l,m}^{(TM)} \) and \( \Gamma_{l,m}^{(TE)} \) are roughly the same order of magnitude. This observation will be very important when we discuss the relative scattering intensities of the various dipole-exchange spin wave modes below, in the limit the radius of the sphere is small.

We have one final step. In the definition of the effective current density \( \vec{j}^{eff}(\vec{r},t) \) given in Eq.(10), we require an expression for the incident laser field \( \vec{E}^{(0)}(\vec{r}) \) inside the nanosphere. If we consider a plane wave incident on a conducting sphere of arbitrary radius, the expression for this field is cumbersome. Here we exploit the fact that our sphere has radius \( R \) small compared to the wavelength of the laser light in vacuum. In this limit, to excellent approximation, we may assume that the sphere is placed in the spatially uniform field \( \vec{E}^{(0)} = \vec{z} \cdot \vec{E}^{(0)} \exp(-i\omega t) \). In the region just outside the sphere, we are far into the near zone, to use language appropriate to radiation theory, so the spatial form in the field outside the sphere may be taken to be that given by the equations of electrostatics. However, the radius of the sphere is not small compared to the optical skin depth, so we must solve for the electric field inside through use of the full electromagnetic theory. We surely have \( \nabla \cdot \vec{E} = 0 \) inside the sphere, so the electric field obeys the Helmholtz equation

\[
\nabla^2 \vec{E}(\vec{r}) + k^2 \vec{E}(\vec{r}) = 0 \quad \text{(16)}
\]

everywhere inside the sphere, where as above \( k^2 = (\omega/c)^2 \epsilon \) with \( \epsilon \) the optical frequency dielectric constant.

Outside the sphere, in response to the incident field we have the classical electrostatic field of a polarized sphere. In the near zone and this has components

\[
E_r^g = \frac{2a}{r^3} \cos \theta; \quad E_\theta^g = \frac{a}{r^3} \sin \theta, \quad \text{(17a)}
\]

whereas inside the sphere the fields have the form

\[
E_r^i = b \frac{2j_1(kr)}{r} \cos \theta; \quad E_\theta^i = \frac{b}{r} \frac{d}{dr} (r j_1(kr)) \sin \theta. \quad \text{(17b)}
\]

Application of the boundary conditions is straightforward, and yields

\[
b = 3 \frac{E^{(0)}R}{2c j_1(kR) + \frac{d}{dr}(rj_1(kR)) R}. \quad \text{(18)}
\]

The expressions for optical fields inside a conducting nanosphere under conditions similar to those considered here will prove useful for a variety of applications. Thus we pause by arranging the results into a more useful format. We define

\[
\tilde{f} = \frac{R}{r (\epsilon - 1) j_1(kR) + \sin(kR)} (j_1(kr) + \sin(kr)) \quad \text{(19a)}
\]
and \( \Delta f = \frac{R}{r} \left( \frac{\epsilon + 2}{\epsilon + 2} - \frac{\epsilon - 1}{\sin(kR)} \right) (3j_1(kr) - \sin(kr)) \). (19b)

Of course, inside the sphere, the field is invariant under rotation around the \( z \) axis. The two non zero components of the field in cylindrical coordinates are given by

\[
E_z(r) = \frac{3E^{(0)}(r)}{\epsilon + 2} (f' + \Delta f(r) \cos(2\theta)) 
\]

and \( E_\theta(r) = \frac{3E^{(0)}(r)}{\epsilon + 2} \Delta f(r) \sin(2\theta) \). (20b)

In the limit that the skin depth is large compared to the radius of the sphere, the function \( f(r) \) approaches unity everywhere, and \( \Delta f(r) \) approaches zero. We are then left with the elementary expression for the fields inside a dielectric sphere exposed to a spatially uniform electric field. When the skin depth is comparable to or smaller than the radius of the sphere, the use of Eqs. (20) allows us to account for the spatially non uniform character of the laser field inside the sphere. It should be remarked that in the numerical calculations we report below, the magnetization of the sphere is always taken parallel to the \( z \) axis, and we will explore the Brillouin cross section when the laser field is not necessarily parallel to the \( z \) axis. Of course, in this case one may apply a simple coordinate rotation to the field components in Eqs. (20) to generate expressions for the exciting field inside the nanosphere.

We now have all the ingredients in place to obtain an expression for the Brillouin scattering cross section. One evaluates the Poynting vector in the far zone, integrates it over solid angle, and divides the result by the energy per unit time which illuminates the sphere. One finds the following for the total Brillouin scattering cross section:

\[
\sigma_{BLS} = \frac{1}{|E^{(0)}|^2} \sum_{l,m} \frac{1}{l(l+1)} (|A_{l,m}^{(TM)}|^2 + |A_{l,m}^{(TE)}|^2) \Gamma_{l,m}^{(TM)} \Gamma_{l,m}^{(TE)}.
\]

(21)

The expression in Eq. (21), written out in full, is a quadratic form in the transverse magnetization components, with terms proportional to the combination \( m_\mu(r', t)m_\nu(r', t) \) integrated over the volume of the sphere; one has an integral over both \( r' \) and \( r'' \). One takes a thermal average over this quadratic form. The resulting expression provides one with the Brillouin cross section, integrated over all solid angle, and integrated also over all spin wave modes which contribute to the Brillouin signal. One can break the correlation functions \( <m_\mu(r', t)m_\nu(r', t)> \) down into sums over each of the spin wave eigenmodes of the sphere, and thus extract from Eq. (21) the Brillouin cross section integrated over solid angle for each individual mode. When we refer to the Brillouin cross section of an individual mode, we include both the Stokes and the anti Stokes component. The algebra associated with the decomposition just described is complex, and since the resulting forms are of little general interest, we omit the details here. We refer the reader to the discussion given in ref. [6] for a description of how this is done. It is the case, as noted in this paper, that in the regime where dipolar contributions to the spin wave excitation energy are of comparable magnitude, the algorithm for normalizing the eigenvectors is not so obvious, as noted above. The means of doing so are derived in ref. [6].

We turn next to our numerical studies of the Brillouin scattering intensities of the various normal modes.

III. RESULTS AND DISCUSSION

We first begin with some general comments. It is the case that the nature of the spin wave modes in ferromagnetic spheres and related sample shapes is a classic topic in magnetism. Motivation for the early theoretical studies was provided by the first generation of FMR experiments, which were largely carried out on spherical samples of yttrium iron garnet (YIG). These samples had linear dimensions that were macroscopic. As a consequence, the long wavelength spin wave modes excited by the microwave fields used in FMR could be described by magnetostatic theory with exchange ignored. In a remarkable paper that is now classic, Walker presented an analytic theory of the magnetostatic spin wave modes of uniformly magnetized ferromagnetic ellipsoids. The special case of the sphere was discussed by Fletcher and Bell, who provide a very large and interesting array of useful formulae for describing the eigenfrequencies and eigenvectors as well. As the radius of the sphere is made small, to the point where the radius lies in the few nanometer range, exchange enters importantly in the description of the spin wave eigenmodes. This is particularly the case for the materials of current interest, which
are the 3d ferromagnets or alloys fabricated from these elements. By virtue of the itinerant character of their ferromagnetism, along with their high Curie temperatures, the exchange stiffness $D$ is very large. The first description of the exchange/dipole spin wave modes of a ferromagnetic sphere has been developed only recently by Arias and the present authors; this paper extends the analysis of ref.[9] and ref.[10] to include exchange as well as dipolar interactions between the spins. A complete discussion of the response of such a sphere to spatially inhomogeneous microwave frequency magnetic fields is given as well.

As discussed in ref.[1], a measure of the importance of exchange in the description of the spin wave modes of a ferromagnetic sphere is the dimensionless parameter $r_{ex} = \frac{D}{4\pi M_s (\frac{\pi}{R})^2}$, where $R$ is the radius of the sphere. Consultation of Fig.4 of ref.[1] shows that the contribution of exchange to the excitation energy of spin waves in spheres is substantial even for values of $r_{ex}$ as small as 0.2. The calculations presented below are for Fe nanospheres, for which $4\pi M_s = 2 \times 10^6$ gauss, and $D = 2.5 \times 10^{-8}$ gauss cm$^2$. For these parameters, and for an Fe sphere with radius of 10 nm, we have $r_{ex} \approx 1$. We call the reader’s attention to the Appendix of ref.[1] where simple analytic expressions are provided for the excitation energies of spin waves in the limit of strong exchange, with the first correction from dipolar interactions included. As noted in ref.[1], this expression accounts nicely for the excitation energies calculated from the full theory for a rather wide range of $r_{ex}$.

In this paper, we refer to the dipole exchange spin wave modes by the quantum number scheme used in ref.[1], which is adapted from ref.[9] and ref.[10]. First, suppose we consider the magnetostatic limit. Then for each eigenmode of the ferromagnetic sphere, the magnetic dipole field generated outside the sphere by the spin motion has an angular dependence controlled by a single spherical harmonic $Y_{l,m}(\theta, \phi)$. Inside the sphere, the magnetic potential which describes these fields is not simple, if wishes to express it in terms of spherical harmonics. It is the case that each mode can be labeled by the two quantum numbers $l, m$ associated with the spherical harmonic just mentioned, and a radial quantum number $n$ which may be viewed as a radial quantum number, to make an analogy with quantum mechanics. Hence, in the magnetostatic limit, each mode is labeled by the three numbers $(l, m, n)$. The uniform mode, which is the mode seen in FMR when the sphere is exposed to a spatially uniform microwave field, is the $(1,1,0)$ mode in this notation. When exchange is added, it is no longer true that the magnetic potential outside the sphere is proportional to a single spherical harmonic. However, as the limit that the exchange stiffness $D$ is allowed to approach zero, each mode of the exchange/dipole spectrum smoothly and continuously reduces to a particular magnetostatic mode which can be labeled by the scheme just described. Thus, we label each dipole/exchange mode of the sphere by the same three quantum numbers employed for the mode in the limit $D \to 0$. We recall ref.[1] that with no spin pinning at the surface of the sphere (the case considered here), the FMR mode is unaffected by exchange, and remains a uniform mode even with exchange present. We now turn to our results.

We have carried out calculations of the BLS cross sections for the $(110)$ mode, the $(2m0)$ modes and the $(3m0)$ modes, and also the $(410)$ mode. Since there is little interest in the absolute BLS cross section for a single, isolated sphere, we shall present the result in the form of cross section ratios of the form $\sigma_{lm0}/\sigma_{110}$. Our principle conclusions are summarized in Fig.1 and Fig.2, for the case where the incident photons have the energy of 2.5 eV. In Fig.1, for the case where the electric field vector would
of the incident radiation is aligned with the magnetization of the sphere, we show two of the cross section ratios as a function of the radius of the nanosphere. We have taken material parameters appropriate to Fe, but clearly the trends are robust, and not affected by details of the material parameters. We see that as the radius of the sphere is reduced, the scattering power of the (210) mode increases dramatically relative to the FMR mode, while that of the (310) mode remains roughly similar to that of the FMR mode.

What is responsible for the behavior of the cross sections in Fig.1 is the operation of what we may call a quasi selection rule. First, for the FMR mode, one may show that all of the coefficients \( \Gamma_{l,m}^{(TE)} \) vanish identically, and the only non zero contribution to the cross section comes from \( \Gamma_{l,m}^{(TM)} \). Thus, we can say that in the limit when \( k_0 R \ll 1 \), the dominant contribution to the BLS cross section for the FMR mode (that from \( \Gamma_{11}^{(TE)} \)) is “silent”, and the FMR mode scatters only by virtue of the contribution \( \Gamma_{11}^{(TM)} \), which as we see from Eqs.(15) leads to a contribution to the cross section smaller than that from the leading term by the factor of \( (k_0 R)^2 \). For a nanosphere with a radius of 10 nm, we have \( k_0 R \approx 0.13 \), whereas when we reach 50 nm, \( k_0 R \) is sufficiently large that the quasi selection rule is not operative. For the (210) mode, \( \Gamma_{11}^{(TE)} \) is substantial for the smaller spheres. The quasi selection again suppresses the cross section of the (310) mode in the limit of small radii.

In Table I, we tabulate the cross section ratio for several modes, and for sphere radii of 10 nm, and also 50 nm. We see that for the larger sphere, the (110) has the largest cross section, but the (310) mode is nearly as strong. As the sphere is reduced in size the quasi selection rule dominates, and the \( l = 2 \) modes dominate the spectrum, to have very large cross sections save for the \( m = 2 \) case, where the cross section is very small; we insert a zero into the Table for this reason.

The angular dependence of the cross section ratios is also of interest. In Fig.2, we show the cross section ratios as a function of the angle between the incident laser field and the magnetization for the two modes which dominate the spectrum for small spheres. We see that in the limit \( k_0 R \ll 1 \) the large cross section for the (210) mode remains large for all angles, whereas the scattering from the (200) mode is suppressed as electric field of the incident laser is rotated from parallel to the magnetization, to the equatorial plane of the sphere. Experimental observation of this angular dependence would be of great interest, as a test of the picture put forward in this paper.

Insight into the origin of the quasi selection rule which controls the results above in the limit of small radii can be obtained from the limiting forms displayed in Eqs.(15) and in the structure of the quantities \( \Gamma_{l,m}^{(TE)} \) and \( \Gamma_{l,m}^{(TM)} \) defined in Eqs.(13). For the form of the coupling between the light and the spin system described earlier, and for the case where the incident electric field is aligned with the magnetization of the sphere, one has the explicit forms:

\[
\Gamma_{l,m}^{(TE)} = A \int_V d^3r j_l(kr) \bar{f}(\vec{r}) [\frac{\partial Y_{lm}(\theta, \varphi)}{\partial \theta} m_\rho(\vec{r}) + im \cot(\theta) Y_{lm}(\theta, \varphi) m_\varphi(\vec{r})] \tag{22a}
\]

and

\[
\Gamma_{l,m}^{(TM)} = iA \int_V d^3r \frac{r}{R} j_l(kr) \bar{f}(\vec{r}) Y_{lm}(\theta, \varphi) m_\varphi(\theta) \sin(\theta) m_\varphi(\vec{r}). \tag{22b}
\]

In these expressions, the integration is over the volume of the nanosphere, we have defined \( A = [3\omega K E^{(0)} / 4\pi(\epsilon + 2)] \), and the components of the transverse magnetization associated with a given spin wave mode are expressed in cylindrical coordinates.

From the structure of the eigenvectors given in ref.[1], one may see that each of the dipole/exchange spin wave modes of the sphere has well defined parity, even or odd, under reflection through the equatorial plane of the sphere. The parity of the mode may be written as \((-1)^{l+m}\). Now the spherical harmonic which appears in Eq.(22a) along with the factor of \( \sin(\theta) \) also have parity \((-1)^{l+m}\) under this reflection, whereas the parity of the integrand in Eq.(22a) is \((-1)^{l+m+1}\). Then we also note that for a given mode with label \((l_0, m_0, n)\) the angular integration over \( \varphi \) renders all the quantities in Eqs.(22) zero except for the contributions for which \( m = m_0 \).

Thus, we can see from the previous paragraph that the term which provides the leading contribution to the cross section, \( \Gamma_{11}^{(TE)} \) vanishes for the (110) mode, which is the mode excited in FMR. It is the case that for this mode, the only non zero contribution to these coefficients is \( \Gamma_{11}^{(TM)} \). The FMR mode is thus forbidden to scatter in lowest order, in the limit \( k_0 R \ll 1 \). When we turn to the \( l = 2 \) modes, for the (200) mode \( \Gamma_{10}^{(TE)} \) is non zero and substantial, and for the (210) mode the same is true of \( \Gamma_{11}^{(TE)} \). For the (220) mode, the azimuthal integrations render all the leading terms, \( \{\Gamma_{1,m}^{(TE)}\} \), zero and we obtain our first substantial contribution from \( \Gamma_{22}^{(TM)} \).
The cross section for this mode is thus very small, in the limit \( k_0R \ll 1 \). For the (310) mode, \( \Gamma_{11}^{(TE)} \) is forbidden by reflection symmetry through the equatorial plane, and the first non zero contribution is from \( \Gamma_{11}^{(TM)} \). The cross section for this mode is thus comparable to that for the (110) mode, the FMR mode. We can continue on to understand the small cross sections for the (320) and the (330) mode. By the time we reach the (410) mode, the angular momentum content of the spin wave is sufficiently high that the scattering cross section is small.

From the calculations presented in this section, we can see that BLS spectroscopy can allow one to access selected higher order dipole exchange modes, whereas ferromagnetic resonance spectroscopy probes only the single FMR mode in the limit of small sphere radii.

### IV. CONCLUDING REMARKS

From the calculations presented here, we can see that BLS spectroscopy of the spin wave modes of small nanospheres can provide one with access to certain higher order dipole/exchange modes. In the limit of small sphere radii, in the sense described above, we see that the \( l = 2 \) modes are predicted to dominate the spectrum.

The analysis presented here explores the BLS cross section for the various spin wave modes, for a single, isolated nanosphere. Of course, any experimental study will necessarily explore an array of such objects. Indeed, Steinmuller et al. have reported BLS studies of Fe nanospheres deposited on a GaAs surface\[^{11}\]. It is difficult to comment in detail on their results, since the film consisted of clusters of Fe spheres arranged in a disordered fashion on the surface. We can comment on one issue, however. These authors observe Stokes/anti Stokes asymmetries in their spectra, and argue this provides evidence that the spin excitations they study should be regarded as collective excitations of the array of spheres, which interact through dipolar coupling. We recall that Stokes/anti Stokes asymmetries in BLS spectra taken from the surfaces of ferromagnetic crystals and in films were reported many years ago, and theory based on the physical picture used here provides a fully quantitative account of the data\[^{12,13,14}\]. A question raised with these experiments is whether the Stokes/anti Stokes asymmetry is truly evidence for collective mode behavior, or whether in fact such asymmetries are found for the scattering of light from spin waves in a single, isolated sphere such as that studied here. In section III, we presented only total cross sections, integrated over both the Stokes and anti Stokes data\[^{12,13,14}\].

It would be highly desirable to see BLS studies from ordered lattices of ferromagnetic nanospheres, as opposed to the disordered films studies in ref.\[^{11}\]. Then, of course, one would observe collective spin wave modes of the lattice of spheres where the intersphere interaction is dipolar in nature, if one assumes that neighboring spheres do not touch. We remark that we have developed a theoretical description of such collective modes, for the case where both exchange and intrasphere dipolar interactions are comparable in strength\[^{15}\]. In our paper, one finds explicit calculations which describe the nature of such modes. It will be a straightforward matter to adapt the description of the BLS spectrum of an isolated sphere set forth in the present paper to the description of scattering from the collective modes. Experimental data on ordered arrays of spheres will provide motivation for this extension.

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