

**Theory of collective spin-wave modes of interacting ferromagnetic spheres**

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We formulate the theory of the collective spin wave modes of arrays of spherical particles of ferromagnetic material, under the assumption that each sphere in the array is magnetized uniformly. In addition, the inter-sphere interactions have their origin in the magnetic fields generated by the precessing moments, appropriate to the case where there is no direct physical contact between the spheres. The formulation is a real space analysis, and thus can be applied in principle to disordered arrangements of spheres. While our formulation is quite general, and is directly applicable to the case where both exchange and dipolar interactions influence spin motions within an individual sphere, explicit calculations are presented for the case where exchange is absent. The numerical calculations we discuss explore the collective spin wave modes of square planar arrays of spheres, and consider the case where the spheres are magnetized both perpendicular and parallel to the plane.

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**I. INTRODUCTION**

Of course, magnetically ordered materials exhibit a spectrum of collective excitations known as spin waves. In various forms of bulk magnetic matter, the nature of the spin waves has been elucidated both in theory and in experiment for many years now. More recently, interest has centered on magnetic nanostructures, with attention to their response characteristics. In case of ultrathin films and magnetic superlattices or multilayers fabricated from ultrathin films, for some years now the spectrum of collective modes has been studied experimentally.<sup>1</sup> Both ferromagnetic resonance spectroscopy (FMR) and Brillouin light scattering (BLS)<sup>2</sup> provide access to these modes, which of course control the response characteristics of the structures, in the linear response regime. It is the case that in these systems, considerable theoretical effort has been devoted to the study of their collective spin waves as well. It is fair to say that at this point the physics of the collective excitations is well understood in principle, at least for modes characterized by spatial scales long compared to the underlying lattice constants of the media of interest.

Less clear by far is the nature of the collective spin wave excitations of textured magnetic media, where the basic underlying unit is not a film of infinite extent in the two directions parallel to the surface, but rather an entity of lower symmetry such as a thin circular disc, a nanowire, or a sphere. The latter case, that of the collective excitations of an array of small magnetic spheres is of particular interest, since magnetic recording media are in fact comprised of small roughly spherical objects packed closely together. We have been engaged in constructing the theory of the collective excitations of textured magnetic nanostructures. In a recent paper,<sup>3</sup> we have addressed the nature of the exchange/dipole spin wave spectrum of nanowires. The theory accounts nicely for doublets observed in FMR studies of nanowires of selected radii,<sup>4</sup> and BLS studies of size quantization effects on spin waves in small nanowires.<sup>5</sup> We have recently developed<sup>6</sup> the theory of the collective spin wave excitations of nanowire arrays, where the wires are not in direct physical contact, and thus magnetostatic coupling between these entities lead to collective spin wave modes.

In this paper, we present the theory of the collective spin wave modes of small magnetic spheres, once again for the case where the coupling between the spheres has its origin in magnetostatic fields generated by spin motions within the constituent spheres. Our formulation is very general in nature. For example, it is a real space formulation so it can be applied to small clusters of spheres, as well as to the periodic arrays we examine here in the numerical calculations presented below. It should be remarked, however, that the study of clusters which contain an appreciable number of spheres will require very large matrices to be handled numerically. Periodic arrays, in which the spin waves have well defined wave vector, may be studied efficiently. Our method is, in the formal sense, a multiple scattering method similar in nature to earlier work of Maystre *et al.*<sup>7</sup> in their explorations of the collective response of arrays of dielectric cylinders. In such approaches, one assumes that the response function of an isolated entity is known, and a self-consistent multiple scattering methodology frames the description of the collective modes of the array. In the magnetic case, through appropriate choice of the response function of the individual entity one can describe collective excitations of pure dipolar character, or if desired one can incorporate both exchange and dipolar interactions in the description of the response of an individual sphere. In our study of the collective excitations of nanowires, both dipolar interactions and exchange were included fully.

The extension of the basic formulation from arrays of cylinders to those of spheres requires a mathematical structure to be introduced. In the case of cylinders, an identity known as Graf's identity<sup>8</sup> is central to rendering the theory computationally accessible. One requires an equivalent for the spherical coordinate system used in three dimensions, for the description of spherical objects. We have recently developed the theory of the collective excitations of arrays of dielectric spheres<sup>9</sup> where we introduce a suitable identity similar in structure to the Graf identity which applies in cylindrical coordinate systems. This identity may be used as well in the present instance, to describe the collective spin wave modes of arrays of magnetic spheres, as we shall see. We also require, for the sphere, the function which describes

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the linear response of an individual sphere to a microwave field of arbitrary spatial variation. This is not so simple, unfortunately, since the fact that the sphere is magnetized lowers its symmetry from that of an object invariant under arbitrary spatial rotations about its center, to one invariant only to rotations about the axis along which the magnetization is directed. Below we show that if we are willing to ignore exchange, and describe the response of the sphere within the magnetostatic approximation, then the appropriate response function may be constructed. For the case where the sphere is so small that exchange influences its response, the appropriate response function of the isolated sphere is not yet in hand, though we have this topic under study at the time of this writing. Thus, in the present paper, in our numerical studies, we confine our attention to the pure magnetostatic problem, wherein both interactions between spheres and the intrasphere response may be described by magnetostatic theory. Thus, our considerations apply to ferromagnetic spheres whose radius exceeds the exchange length  $l_C = (D/4\pi M_s)^{1/2}$ . If we have in mind arrays of particles made from the 3d transition metal ferromagnets, the theory presented here is applicable to spheres whose diameter is larger than a few tens of nanometers. We hasten to emphasize that our formulation of the coupling between the spheres is quite general and is fully applicable to small spheres wherein the response of the individual spheres is influenced by exchange as well as dipolar fields generated by spin motions. Thus, when developed, the response function of a single dipole/exchange coupled sphere may be inserted directly into the formalism described here, to give us a full theory valid for particles whose diameters are comparable to or smaller than the exchange length. This question is under active study at present, as noted above. (Recently, we have completed a computationally tractable formulation of the response function of an isolated sphere, with exchange included.)

This paper is organized as follows. In Sec. II we present the formalism that we have developed, and in Sec. III we describe results of calculations we have carried out, to provide an illustration of the collective mode spectrum of particular cases.

## II. THE THEORETICAL FORMULATION

We consider an array of ferromagnetic spheres arranged in some manner in space, each uniformly magnetized. In principle, each sphere need not be identical, but in the interests of simplicity we assume this to be so in what follows. The magnetizations of the spheres are parallel to each other, and parallel to the axis we call the  $\hat{z}$  axis. Imagine the magnetizations of the spheres in the array are excited, possibly by an external applied magnetic field. As the magnetization of a given sphere engages in its precessional motion at some particular frequency  $\Omega$ , it generates a spatially nonuniform dipolar field which couples to the precessing magnetization of the other spheres in the array, with the result that the array exhibits a spectrum of collective spin wave modes.

To obtain a description of these collective modes, we proceed with a logic similar to that employed in a different physical context by the authors of Ref. 7. We locate the

origin of the coordinate system at the center of one selected sphere, and examine its response to the combination of the assumed externally generated driving field and the sum of the dipolar fields generated by the motions of the magnetization in the other spheres of the array. In the vicinity of the origin, the total driving field seen by the selected sphere has the form

$$\vec{h}^{(\text{tot})}(\vec{r}, t) = \vec{h}^{(\text{tot})}(\vec{r}) \exp(-i\Omega t) = -\vec{\nabla} \Phi_M^{(\text{tot})}(\vec{r}) \exp(-i\Omega t). \quad (1)$$

Since all fields are described in the magnetostatic approximation, the fields near the origin may be expressed in terms of the gradient of the magnetic scalar potential  $\Phi_M^{(\text{tot})}(\vec{r})$ . In the vicinity of the origin, outside the other spheres and outside the sources which generate the external driving field, we may cast the magnetic scalar potential in the form

$$\Phi_M^{(\text{tot})}(\vec{r}) = \sum_{l=1}^{\infty} \sum_{m=-l}^l C_{lm}^{(\text{tot})} r^l P_l^m(\cos \theta) \exp(im\varphi), \quad (2)$$

where  $(r, \theta, \varphi)$  are the usual spherical coordinates and  $P_l^m(\cos \theta)$  is the associated Legendre function of the first kind. We find it more convenient for our purposes to work with these objects, rather than the closely related spherical harmonics.

The analysis can be broken down into two distinct steps. The first is to describe the response of the sphere at the origin to driving fields generated by the magnetic scalar potential in Eq. (2), and the second step is to express the coefficients  $C_{lm}^{(\text{tot})}$  in terms of appropriate amplitudes which describe the motions of the magnetization of the other spheres in the array, and also that of the external driving field. This will generate a set of equations which, upon setting the amplitude of the driving field to zero, will lead to an array of equations whose homogeneous form allows us to study the collective modes of the array, and whose inhomogeneous form leads to a description of its microwave response whose nature, of course, is controlled by the collective mode spectrum. We turn to each step in the analysis next.

### A. Response of an individual sphere to an inhomogeneous driving field

Suppose we consider a single sphere of radius  $R$  with center at the origin, driven by an externally applied magnetic field whose vector potential is given by  $\Phi_{l,m}^{(\text{ext})} r^l P_l^m(\cos \theta) \exp(im\varphi)$ . Our interest will center on the description of the total magnetic field outside the sphere, including the contribution generated by the motion of its magnetization. Quite generally this may be derived from a magnetic potential we write as

$$\Phi_M^{(0)}(\vec{r}) = \Phi_{l,m}^{(\text{ext})} \left\{ r^l P_l^m(\cos \theta) + \sum_{l'} s_{l,l'}^m(\Omega) \frac{R^{l+l'+1}}{r^{l'+1}} P_{l'}^m(\cos \theta) \right\} \exp(im\varphi). \quad (3)$$

The form in Eq. (3) recognizes that our problem has symme-

try lower than spherical symmetry, since the presence of the spontaneous magnetization lowers the symmetry of the spherical object so that the only rotational symmetry which remains is that about the  $\hat{z}$  axis. Thus, in language borrowed from quantum mechanics, the azimuthal quantum number  $m$  remains a good quantum number, but this is not true for the quantum number  $l$ . Hence, in general, the response function  $s_{l',l}^m(\Omega)$  introduced in Eq. (3) will not be diagonal in  $l$ . As noted in Sec. I, we shall examine the response of the sphere at the origin in the magnetostatic limit, with exchange ignored. In this special limit, we show below that the response function is diagonal in the index  $l$ , but it is useful to keep the discussion general for the moment.

To construct the response function of the sphere in the magnetostatic approximation, we may utilize treatments which appeared many years ago. In a classic paper, Walker<sup>10</sup> analyzed the magnetostatic modes of elliptical samples. Of course, the sphere is a special limit of the more general geometry considered by him. More relevant to the present analysis is the paper by Fletcher and Bell.<sup>11</sup> These authors consider the special case of the sphere in detail, providing analytic formulae for the characteristic equations from which frequencies of the various normal modes can be determined. They also describe the response of the sphere to an external microwave field, so in fact from their paper one can construct the response function defined in Eq. (3). We provide a brief sketch of the analysis, since this allows us to introduce the various quantities which we require.

Outside the sphere, the total magnetic potential obeys Laplace's equation, whereas inside the sphere in the magnetostatic limit it obeys an anisotropic form of Laplace's equation commonly referred to as the Walker equation. This can be written as

$$(1 + \kappa) \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) \Phi_M^{(0)} + \frac{\partial^2}{\partial z^2} \Phi_M^{(0)} = 0. \quad (4)$$

If  $\Omega$  is the frequency of the spin motion in the sphere and  $M_s$  is its magnetization, we introduce the dimensionless measure of frequency  $\omega = \Omega / 4\pi\gamma M_s$ , where  $\gamma$  is the gyromagnetic ratio. Then  $\kappa = \omega_H / (\omega_H^2 - \omega^2)$  and we shall encounter  $\nu = \omega / (\omega_H^2 - \omega^2)$ . If  $H_0$  is the dc field which is applied parallel to the magnetization, and  $H_i = H_0 - 4\pi M_s / 3$  is the internal field, then  $\omega_H = H_i / 4\pi M_s$ . Thus, in what follows, frequency and magnetic fields are expressed as multiples of  $4\pi M_s$ .

The solutions of Laplace's equation are well known and elementary, and it is possible to generate families of solutions to the Walker equation by expressing these in Cartesian coordinates, then scaling the  $z$  coordinate appropriately. However, the problem of matching solutions at the boundary of the sphere then leads to a rather complex set of equations. Walker noted closed form solutions can be obtained by resorting to bispherical coordinates within the interior. In the special case of the sphere itself, Fletcher and Bell find solutions that are quite simple in structure. In what follows, we shall be interested in reduced frequencies  $\omega > \omega_H$  where  $\kappa < 0$ . The transformation to bispherical coordinates  $(\xi, \eta, \varphi)$  then takes the form

$$x = R(-\kappa)^{1/2}(1 - \xi^2)^{1/2} \sin \eta \cos \varphi, \quad (5a)$$

$$y = R(-\kappa)^{1/2}(1 - \xi^2)^{1/2} \sin \eta \sin \varphi, \quad (5b)$$

and

$$z = R(\kappa/[1 + \kappa])^{1/2} \xi \cos \eta. \quad (5c)$$

On the sphere of radius  $R$  the coordinate  $\xi$  assumes the constant value  $\xi_0 = ([1 + \kappa]/\kappa)^{1/2}$ , while  $\eta$  coincides with the polar angle  $\theta$  of spherical coordinates. In the bispherical coordinate system, the Walker equation admits separable solutions of the form  $P_l^m(\xi)P_l^m(\cos \eta)\exp(im\varphi)$ .

When analyzing the response of the sphere to an external potential of the form  $r^l P_l^m(\cos \theta) \exp(im\varphi)$  one must match the magnetostatic potential in the interior of the sphere to that outside. One boundary condition is that the magnetic potential be continuous; this insures continuity of tangential components of the magnetic field  $\vec{h}$  derived from the gradient of the potential. Quite clearly, from the remarks in the previous paragraph, a solution inside the sphere proportional to  $P_l^m(\xi)P_l^m(\cos \eta) \exp(im\varphi)$  may be matched to the form  $\{r^l + s_l^m(R^{2l+1}/r^{l+1})\} P_l^m(\cos \theta) \exp(im\varphi)$  applicable outside, suggesting that in Eq. (3)  $s_{l',l}^m = \delta_{l',l} s_l^m$ , i.e., it is diagonal in the index  $l$ . In addition, the radial component of the magnetic induction  $\vec{b}$  must be conserved as well. After considerable algebra, one may show that one may conserve radial components of the magnetic induction as well with this special form. We shall omit details, and just quote the final form for the response function. We find

$$s_{l',l}^m = \delta_{l',l} s_l^m = \frac{(l - m\nu)P_l^m(\xi_0) - \xi_0 P_l^m(\xi_0)'}{([l + 1] + m\nu)P_l^m(\xi_0) + \xi_0 P_l^m(\xi_0)'} \delta_{l',l}. \quad (6)$$

In Eq. (6), the symbol  $P_l^m(\xi_0)'$  denotes the derivative of the function with respect to its argument. Notice that in the frequency range  $\omega_H < \omega < [\omega_H(\omega_H + 1)]^{1/2}$ , the quantity  $\xi_0$  is real and positive, whereas when  $\omega > [\omega_H(\omega_H + 1)]^{1/2}$  it is pure imaginary and is written as  $\xi_0 = -i|\xi_0|$ .

As remarked above, with the response function of the single sphere in hand, we may now turn our attention to the response of the array of spheres. Before we address the array of spheres, we should point out that we have yet to address a complication not present in the earlier discussion of the dielectric spheres.<sup>9</sup> We will describe an array of ferromagnetic spheres, each magnetized uniformly, with magnetizations of all spheres parallel. The array is then placed in an external magnetic field, as in the discussion just presented. For the isolated sphere just described, then quite clearly the internal dc magnetic field is spatially uniform, and assumes the value  $H_i = H_0 - 4\pi M_s / 3$  introduced above. Now when the sphere at the origin is surrounded by magnetized spheres in its near vicinity, the internal field will differ from this value, by virtue of the static dipole fields produced by the neighboring spheres. This dipole field from neighbors is in fact spatially non-uniform. We will argue below that it suffices as a first approximation to retain only the spatially uniform portion of the dipole field from the neighbors. This will then lead us to employ the single sphere response function just derived, but the internal field  $H_i$  is to be replaced by  $H_0 - 4\pi M_s(1 + \Gamma)/3$  where  $\Gamma$  is a correction to the internal field felt by the

sphere at the origin, from the dc field generated by its neighbors. We shall give explicit forms for the correction below. It should be noted that in discussion of granular magnetic materials such as employed in recording media, the same approximation is widely used,<sup>12</sup> and provides excellent quantitative accounts of such materials.

### B. Response of an array of spheres to an external driving field; the collective spin wave modes of the spherical array

To begin, we need to set up a coordinate system. As in the previous section, we shall focus attention on a single sphere of radius  $R$  whose center is located at the origin of the coordinate system, and we shall use the standard spherical coordinates  $(r, \theta, \varphi)$  to designate points in the vicinity of this sphere. A vector from the origin of the coordinate system to the center of sphere  $j$  is  $\vec{R}_0(j)$  and its direction is specified by the polar and azimuthal angles  $\theta_0(j)$  and  $\varphi_0(j)$ . A vector from the center of sphere  $j$  to a point of interest is  $\vec{r}(j)$  and its direction is specified by the polar and azimuthal angles  $\theta(j)$  and  $\varphi(j)$ . It will be convenient to introduce the two functions  $R_{l,m}(\vec{r}) = r^l P_l^m(\cos \theta) \exp(im\varphi)$  and  $I_{l,m}(\vec{r}) = r^{-(l+1)} P_l^m(\cos \theta) \exp(im\varphi)$  which are regular and irregular at the origin respectively, and vice versa at infinity.

We imagine the array of spheres to be driven by an externally applied magnetic field, also described within the magnetostatic approximation. It is thus generated through use of an external magnetic potential whose sources lie outside the array of spheres under consideration. Thus, in the near vicinity of the origin, we may write

$$\Phi_M^{(\text{appl})} = \sum_{l,m} \Phi_{l,m}^{(\text{appl})} R_{l,m}(\vec{r}). \quad (7)$$

For the moment, the coefficients  $\Phi_{l,m}^{(\text{appl})}$  need not be specified in detail.

The magnetic field associated with the external potential sets the magnetizations of all the spheres in the array in motion, with the consequence that other spheres in the array generate magnetostatic fields which combine with that of the external field to drive the magnetization of the sphere at the origin. The spatial part of the time dependent magnetic potential in the vicinity of the origin which drives the magnetization of the sphere there may then be written in the form

$$\Phi_M^{(\text{ext})} = \sum_{l,m} \left\{ \Phi_{l,m}^{(\text{appl})} R_{l,m}(\vec{r}) + \sum_{j \neq 0} B_{l,m}(j) I_{l,m}[\vec{r}(j)] \right\}, \quad (8)$$

where  $\vec{r}(j) = \vec{r} - \vec{R}_0(j)$ . The aim of this section is to derive a set of self-consistent equations for the coefficients  $B_{l,m}(j)$ . Central to our ability to do so is the identity<sup>13,14</sup>

$$\begin{aligned} I_{l,m}[\vec{r}(j)] &= I_{l,m}[\vec{r} - \vec{R}_0(j)] \\ &= (-1)^l \sum_{l'=0}^{\infty} \sum_{m'=-l'+m}^{l'+m} (-1)^{m'-m} \binom{l+l'-m'}{l-m} \\ &\quad \times R_{l',m-m'}(\vec{r}) I_{l+l',m'}[\vec{R}_0(j)]. \end{aligned} \quad (9)$$

The identity is valid when  $r < R_0(j)$ . In Eq. (9), the quantity

$\binom{n}{m} = n! / m!(n-m)!$  is the binomial coefficient. The statement in Eq. (9) allows us to cast the external potential in Eq. (8) in the form

$$\Phi_M^{(\text{ext})}(\vec{r}) = \sum_{l=0}^{\infty} \sum_{m=-l}^{m=l} \Phi_{l,m}^{(\text{ext})} R_{l,m}(\vec{r}), \quad (10)$$

where

$$\begin{aligned} \Phi_{l,m}^{(\text{ext})} &= \Phi_{l,m}^{(\text{appl})} + \sum_{j \neq 0} \sum_{l'=0}^{\infty} \sum_{m'=-l'}^{l'} B_{l',m'}(j) (-1)^{l'+m} \\ &\quad \times \binom{l+m+l'-m'}{l'-m'} \times I_{l+l',m'-m}[\vec{R}_0(j)]. \end{aligned} \quad (11)$$

From the discussion in the previous section, the magnetic potential outside the sphere at the origin due to the precession of its magnetization has the form

$$\Phi_M^{(0)} = \sum_{l'=0}^{\infty} \sum_{m'=-l'}^{l'} B_{l',m'}(0) I_{l',m'}(\vec{r}), \quad (12)$$

where, in this instance, identifying  $\Phi_{l,m}^{(\text{ext})}$  in Eq. (11) with the quantity  $\Phi_{l,m}^{(\text{ext})}$  of the previous subsection

$$B_{l',m'}(0) = \sum_{l=0}^{\infty} \Phi_{l,m}^{(\text{ext})} s_{l,l'}^{m'}(\Omega) R^{l'+l+1}. \quad (13)$$

The statement in Eq. (13) combined with Eq. (11) provides us with a self-consistent set of equations for the amplitudes  $\{B_{l,m}(j)\}$ . These are the formal results on which the calculations reported below are based. One may study the microwave response of the array of spheres by solving the inhomogeneous equations generated by this array, once a form for the external driving potential is known or chosen. Alternatively, one may see the frequencies of the collective spin wave modes by finding the frequencies which allow a nontrivial solution of the homogeneous equations formed by setting the external potential to zero. If one considers a periodic array of spheres, as we do in the next section, then the amplitudes may be assumed to have the Bloch form  $B_{l,m}(j) = B_{l,m}(0) \exp[i\vec{k} \cdot \vec{R}_0(j)]$ , where the wave vector  $\vec{k}$  lies in the appropriate Brillouin zone.

In the concluding remarks of the previous subsection, it was noted that when the response function  $s_{l',l}^m(\Omega)$  is generated, we must take due account of the influence of the dc magnetic field generated by the magnetized spheres which surround the sphere at the origin. It is a straightforward exercise, after making use of the identity in Eq. (9), to generate an expression for the dc magnetic potential from which this field may be generated. The expansion is in the form of a series of the functions  $R_{l,m}(\vec{r}) = r^l P_l^m(\cos \theta) \exp(im\varphi)$ . In general, as remarked above, this field is spatially nonuniform, and thus a full and complete inclusion of its influence is nontrivial. However, for the periodic arrays we consider below, the leading term in the series describes a spatially uniform magnetic field which, as mentioned at the end of the previous subsection may be incorporated into the analysis by a suitable redefinition of the internal field experienced by the

sphere at the origin. We can proceed, in principle, by incorporating this uniform component of the field then calculating the spectrum of collective modes which obtains as a first approximation. If further refinement is desired, one can treat the spatially nonuniform terms through an appropriate perturbation theory. Since the angular average of the spatially nonuniform fields over solid angle is zero, it follows they contribute first only in the second order of perturbation theory. We confine our attention here to the mode spectrum calculated in first approximation, wherein the dc field produced by the neighboring spheres is approximated as spatially uniform. We remark, as noted earlier, the same approximation is utilized widely in the literature on granular magnetic media.<sup>12</sup>

To calculate the correction to the internal field from the surrounding magnetized spheres, one proceeds as follows. First, if one considers a uniformly magnetized sphere, then outside the sphere it is well known that the static field is that of a point dipole of strength  $4\pi M_s R^3/3$ . If this sphere is placed at an arbitrary point in space, the magnetostatic potential near the origin may be calculated from the formalism given above, noting the only nonzero coefficient  $B_{lm}(j)$  in Eq. (8) is that with  $l=1, m=0$ . Then when one uses Eq. (9) to express the resulting potential in terms of coordinates reckoned relative to the origin, and averages the resulting field over the sphere at the origin, the only term that survives is the term proportional to  $R_{10}(\vec{r})$ . The field averaged over the sphere at the origin has the same value as the field from an array of point dipoles (it is, again, rigorous to treat the spheres which generate the field as point dipoles) evaluated at the center of the sphere at the origin, i.e., at the origin of the coordinate system. We have carried out numerical studies of the collective modes for two cases, one where a square two-dimensional lattice of spins are magnetized parallel to the plane, and one where they are magnetized perpendicular to the plane. The relevant local field can be expressed in terms of two-dimensional (2D) dipole sums, which can be converted to rapidly converging series using methods set forth many years ago.<sup>15</sup> For the 2D square lattice magnetized in plane, the total internal field is given by  $H_0 - (4\pi M_s/3)(1 - [R/D]^3\lambda)$ , and for the case where the 2D lattice is magnetized perpendicular to the plane we have for the internal field  $H_0 - (4\pi M_s/3)(1 + 2[R/D]^3\lambda)$ , where one has for the parameter  $\lambda$  the sum  $\lambda = (4\pi^2/9)[1 + 24\sum_{n=1}^{\infty}\sum_{l=1}^{\infty}n^2K_2(2\pi nl)] = 4.517$ , and  $D$  is the distance between spheres. We turn next to numerical calculations based on the formalism just derived.

### III. STUDIES OF THE COLLECTIVE MODES OF AN ARRAY OF FERROMAGNETIC SPHERES AND THEIR MICROWAVE RESPONSE

In the magnetostatic description of the response of individual spheres employed here, one finds the isolated sphere admits a large number of standing spin wave modes in the frequency regime above the frequency  $\gamma H_i$ , where  $H_i = H_0 - 4\pi M_s/3$ . A careful and remarkably complete discussion of this mode spectrum can be found in the paper by Fletcher and Bell.<sup>11</sup> Most of these modes may be described as high

order multipole modes, characterized by rather large values of the indices  $l, m$  in the discussion of Sec. II A.

We have carried out numerical studies of the spectrum of collective waves of two-dimensional square lattice of ferromagnetic spheres for two cases. In the first, the spheres are all magnetized perpendicular to the plane, and in the second the spheres are magnetized parallel to the plane. The collective modes then have Bloch character, with dispersion relations characterized by the two dimensional wave vector  $\vec{k}_{\parallel}$  which lies within the appropriate two-dimensional Brillouin zone. For any choice of this wave vector, one finds a large number of collective mode branches. Each reverts to a dispersionless, Einstein-oscillator-like branch whose frequency equals that of the isolated sphere modes described by Fletcher and Bell,<sup>11</sup> in the limit that the lattice constant of the square lattice becomes very large. The question we have explored in our studies is the evolution of this collective mode spectrum as the lattice constant becomes comparable to the diameter of the spheres themselves. We have explored sphere separations  $D$  down to  $2.2R$ , with  $R$  the radius of the individual spheres. For smaller lattice constants, a rather large basis set is required to avoid convergence problems. In the results presented below, the maximum value of  $l$  included in our basis set is  $l=2$ . We have checked convergence to find the results for the low lying modes nicely converged with this choice.

Nearly all of the modes examined show very little dispersion, even for lattice constants as small as  $2.2R$ . The reason is that these are derived from high order multipole modes of the isolated sphere, which generate dynamic dipole fields which fall off rapidly outside a given sphere, and which also are very weak in magnitude, since the pattern of dynamical magnetization in the spheres contains nearby regions where magnetostatic potential differs in sign. An exception is the uniform mode of the sphere which, in the language of Sec. II A is characterized by the quantum numbers  $l=m=1$ . This mode disperses markedly, for the lattice constants we have examined. It crosses and hybridizes with higher order multipole modes which have relatively flat collective mode branches. Thus, qualitatively speaking, the collective mode spectrum of the lattice of spheres consists of what one might describe as a forest of dispersionless modes formed from high order multipole modes of the individual spheres, crossed by a dispersive branch with origin in the uniform mode of the isolated spheres. This dispersive branch crosses and hybridizes with the flat multipole branches it encounters. In the cases we have explored in our studies, we find hybridization with a single branch.

We shall illustrate the point just made for the case where the spheres are magnetized perpendicular to the plane, a geometry of interest in the case of granular media for perpendicular recording. For this case, it should be remarked, the spin wave collective mode spectrum breaks down into modes of two different symmetry classes. This follows by noting in Eq. (11) the second term involves the associated Legendre function  $P_{l'+1}^{m'-m}$  for the case where the angle  $\theta = \pi/2$ . The function  $P_L^M[\cos(\pi/2)] = P_L^M(0)$  vanishes whenever the sum  $L+M$  is odd. It follows that if  $l$  and  $m$  are both even, the coefficients  $B_{l,m}(0)$  are coupled only to coefficients for which

both indices are either even, or both are odd integers. Similarly, if the indices in  $B_{l,m}(0)$  are both odd, then also the equation couples this amplitude only to coefficients whose indices are both even or both odd. We refer to the modes so described as *ee-oo* modes. If one of the two indices is even ( $l$  or  $m$ ) and the other is odd ( $m$  or  $l$ ), then the coefficient couples only to coefficients in which one index is even, or the other odd. We refer to such modes as *eo-oe* modes. Upon noting the identity which applies to the spherical harmonics  $Y_{l,m}(\theta, \varphi) = (-1)^{l+m} Y_{l,m}(\pi - \theta, \varphi)$  one sees that the scalar potential associated with the *ee-oo* modes is even under reflection in the  $xy$  plane, whereas that associated with the *eo-oe* modes is odd. If one excites the spheres with an externally applied microwave field parallel to the  $xy$  plane, it is the *ee-oo* modes that will be excited.

We note that in our earlier discussion of the collective modes of planar arrays of dielectric spheres,<sup>9</sup> we found for the same reason that the collective modes can be decomposed into the two symmetry classes described. In the case of the array of magnetic spheres, this decomposition obtains only for the case where the magnetization is perpendicular to the plane of the spheres. The magnetization is an axial vector, left unchanged by reflection in a plane perpendicular to itself. However, if the magnetization is in plane, or canted with respect to the normal to the plane, then reflection symmetry in the  $xy$  plane is no longer a “good symmetry” since the component of magnetization parallel to the plane changes sign under this reflection. In our mathematics, the breakdown of this symmetry is expressed by the requirement that the  $z$  axis be chosen parallel to the magnetizations of the spheres in the array. Thus, if the magnetization is canted with respect to the normal to the plane, it is no longer the case that all of the Associated Legendre functions on the right-hand side of Eq. (11) are evaluated for  $\theta = \pi/2$ .

In Fig. 1, for the case  $D/R=3.0$ , we show the spectrum of collective modes of the square array of ferromagnetic spheres, in the frequency regime where one finds the dispersive branch associated with the uniform mode of the widely separated spheres. The wave vector is directed along the [11] direction in plane. At this separation, we see considerable dispersion, and we note that the mode hybridizes with an Einstein-like branch associated with a higher order multipole mode. We show also the next higher branch, for which the dispersion is very modest at this separation. As we have seen, in computer simulations of arrays of small particles (in the calculation of hysteresis loops, for example) the small spheres often are approximated as structureless point dipoles. The hybridization phenomenon displayed in Fig. 1 is a reflection of the fact that from the dynamic point of view, the finite sphere is not equivalent to a simple point dipole with a single resonant frequency, but has internal structure with higher order multipole modes which may hybridize and mix with the collective branch formed from the uniform mode, as illustrated in Fig. 1. In Fig. 2, we show the collective modes in the same region of the spectrum, for the case where the spheres are brought closer together, to the point where  $D/R=2.2$ . Two things are evident in this case. First, the whole spectrum has been downshifted in frequency, by virtue of the dipole fields set up in the rather dense lattice. These are the fields incorporated in the correction factor  $\Gamma = \lambda(R/D)^3$

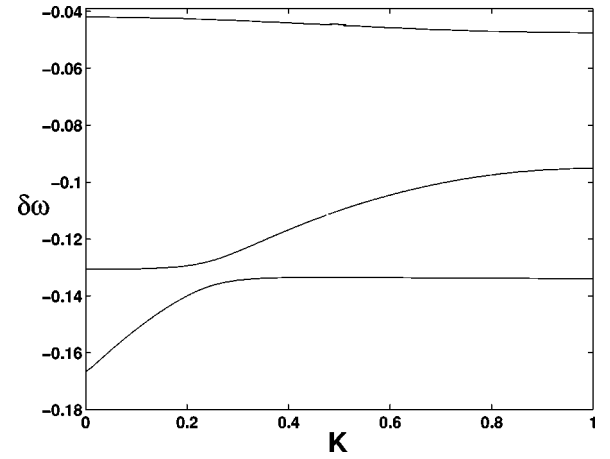


FIG. 1. For the two-dimensional lattice of ferromagnetic spheres magnetized perpendicular to the plane, we show the wave vector dependence of the dispersive branch of the *ee-oo* mode spectrum discussed in Sec. III and nearby branches, for the case  $D/R=3.0$ , with  $R$  the radius of an individual sphere in the lattice, and  $d$  the separation between the centers of the spheres. The wave vector is directed along the [11] direction of the lattice, and is expressed in units of  $\pi\sqrt{2}/D$ . Frequency is in units of  $(\Omega - H_0)/4\pi M_s$  with  $H_0$  the externally applied dc field.

discussed above. Also, we see increased dispersion, and a considerably larger hybridization gap. We remark that we have calculated the microwave response of our lattice of spheres by subjecting them to a long wavelength driving field, simulated by choosing  $\Phi^{(\text{appl})}(\vec{r})$  to have the form  $\exp(-Qz)\exp(i\vec{Q}\cdot\vec{r})$ . If the wave vector  $\vec{Q}$  is chosen near the center of the two-dimensional Brillouin zone, then we can simulate a field whose scale of spatial variation is long compared to a lattice constant. By calculating the energy absorbed by such a lattice as a function of frequency, we find the only mode that has substantial oscillator strength is the lowest frequency zero wave vector mode in Figs. 1 and 2. Thus, a ferromagnetic resonance study of such a material would show only a single mode spectrum, in the limit the spheres have radius very small compared to the microwave wavelength. A similar statement will apply to Brillouin light

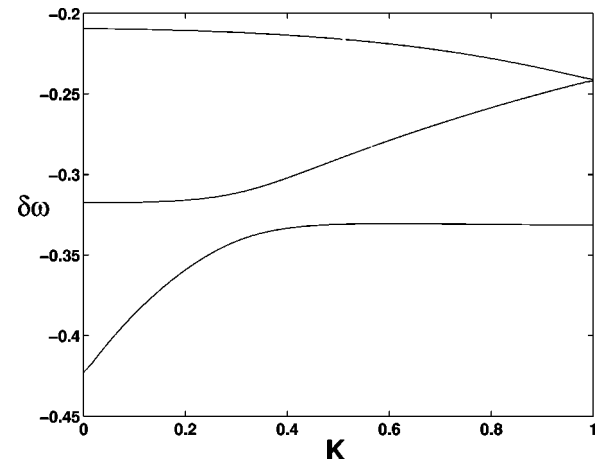


FIG. 2. The same as Fig. 1, but now  $D/R=2.2$ .

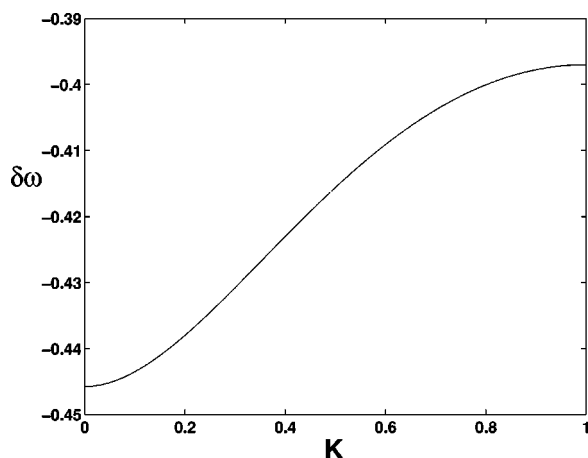


FIG. 3. For the case  $D/R=2.2$ , and for the [11] direction in the two-dimensional Brillouin zone, we show an example of the dispersion relation of an  $eo$ - $oe$  mode. Note the modest amount of dispersion present, when compared to the example in Fig. 2.

scattering studies. Finally, in Fig. 3, in the spectral region where the strong, dispersive branch is found, we show a dispersion curve for the  $eo$ - $oe$  mode in this region, for  $D/R=2.2$ . Clearly, when this is compared with the calculations presented in Fig. 2, we see very little dispersion indeed.

As remarked above, we have also carried out a series of calculations for the case where the spheres are magnetized in plane, to find results rather similar in character to those shown above. In this case, there is no simple symmetry decomposition one can make for the collective modes, so all branches appear in a single calculation.

#### IV. CONCLUDING REMARKS

We have developed the formalism through which one may analyze the collective spin wave modes of arrays of ferromagnetic spheres where interactions between the precessing magnetizations in the spheres are controlled by the dynamic dipole fields generated by spin motions in the array. The formalism is quite general, in that one can apply it to clusters or small collections of spheres, as well as the two-dimensional periodic lattice we have chosen to study in our numerical analyses.

One requires the response function for the individual spheres in the array as defined in Eq. (3), in order to carry out explicit calculations. For the case where the internal response of the sphere can be described by magnetostatic theory, we have generated an explicit expression for this response function, given in Eq. (6). The numerical calculations presented in Sec. III employ this form. With the current interest in magnetic nanostructures in mind, it would be highly desirable to have in hand an extension of the expression in Eq. (6) to the case where exchange as well as dipole interactions influence the response of the single sphere. We remark that we have devoted very considerable effort to the task of generating such a form, and the challenge of doing so is formidable, at least for the general case where dipole and exchange effects are comparable in magnitude.

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