Electromagnetic response of an array of particles: Normal-mode theory

R. Rojas* and F. Claro
Facultad de Física, Pontificia Universidad Católica de Chile, Casilla Postal 6177, Santiago 22, Chile
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A normal-mode theory for the electromagnetic excitations in an array of polarizable particles is presented. The response of the system to an external field is completely described by these modes, which are uncoupled and react independently. They are characterized by purely geometry-dependent strengths and depolarization factors. The two-sphere case is discussed in detail, and it is shown that, at small separations, excitations of pole character higher than dipole are the leading terms in the absorption spectrum.

I. INTRODUCTION

The optical properties of systems composed by small metallic particles have been the subject of continuous attention in the last ten years.[1-7] From a theoretical point of view, an explanation of the anomalous far-infrared absorption is lacking, since experiments show much greater values than the predictions of classical theories.[8-10] Several models have been proposed to solve this problem. Some of them are models for the intrinsic response of a small particle and consider in detail the quantization of the electronic levels due to the finite and small size of a particle.[11-13] More recent models consider the contribution of the different cluster types present in experimental samples.[14,15] This problem also has motivated the development of new approaches for the electromagnetic wave propagation in heterogeneous mediums, including multipolar effects induced by the excitation field.[16-20]

In the far-infrared limit, an electrostatic approach to the problem of small particles in an electric field is sufficient; in such limit the wavelength of the external field is much greater than the particle diameter and typical distances between particles. This greatly simplifies a theory of the optical absorption in such systems. In this limit, Fuchs obtained the single-particle susceptibility in terms of the particle surface normal modes, each appearing as an independent contribution to a sum. Cast in this form the theory enables a simple estimation of each normal mode contribution to the optical absorption.[21]

In a work by Claro,[18] the collective surface resonant modes induced by the Coulomb interaction in an N-particle system are considered. This theory includes in an effective manner all the interactions between particles and can be taken as a starting point to obtain optical properties in systems with interacting particles. In Sec. II of the present work we reformulate the above theory in terms of normal mode depolarization factors and strengths. In this way we get an expansion for the absorption coefficient in terms of collective normal modes; in this sense we make an extension of the single-particle theory of Fuchs to an array of arbitrary geometry and number of particles. The surface collective modes are obtained considering the coupling between particles and with the external field using a spherical harmonics expansion of the potential and charge densities. As an illustration we apply in Sec. III the formalism to the simple case of two spheres. Numerical results are obtained for the collective normal modes of this system. The main finding is that although at large separations the dipole resonance dominates, as the particles approach each other higher pole resonances gain strength and eventually overcome the excitation of dipolar character.

II. N-PARTICLE FORMALISM

We consider an array of N uncharged, polarizable, spherical particles composed of a homogeneous isotropic material of dielectric constant $\epsilon=1+4\pi\chi$, where χ is the material susceptibility. We study the response to an applied electric field associated with an electromagnetic wave of wavelength much greater than the dimensions of our particles and their separation. The particles are assumed to be sufficiently small so that we may ignore the magnetic excitations.

The applied electric field and the electric charge distribution of other particles induces in the ith particle a charge distribution of multipolar moments q_{lmi} given by[22]

$$q_{lmi}=-\frac{2l+1}{4\pi}\alpha_{lmi}V_{lm}(i)\ , \tag{1}$$

where $V_{lm}(i)$ is the coefficient of order (l,m) in the expansion of the local potential about the center of the ith particle, and α_{lmi} defines the corresponding polarizability of the particle. For a sphere of radius a_i, the multipolar polarizabilities are[23]

$$\alpha_{lmi}=\frac{l(\epsilon-1)}{l(\epsilon+1)+1}a_i^{2l+1}\ . \tag{2}$$

This expression can be written in terms of the material susceptibility and the depolarization factors n_l^0 of an isolated sphere,

$$\alpha_{lmi}=4\pi n_l^0 a_i^{2l+1}\frac{1}{\chi^{-1}+4\pi n_l^0}\ , \tag{3}$$

where $n_l^0=l/(2l+1)$. If $l=1$ in the above relations, we recognize the dipolar depolarization factor of the sphere: $n_1^0=\frac{1}{3}$, and the value $\epsilon^*=-2$ corresponding to the dipolar resonance of an isolated sphere.

The electric potential at the position of the ith particle is given by the sum of the applied external field V^{ext} and the contribution due to the other particles,

$$V_{lm}(i)=V_{lm}^{\text{ext}}(i)+\sum_{j\,(\neq i)} V_{lm}^{j}(i)\ , \tag{4}$$

where $V_{lm}^{j}(i)$ is the field from the jth particle. Writing this potential in terms of the $q_{l'm'j}$ moments of the j-particle charge distribution and combining expressions (4), (3), and (1) one obtains[18,24]

$$q_{lmi}=-la_i^{2l+1}(\chi^{-1}+4\pi n_l^0)^{-1}\times\left[V_{lm}^{\text{ext}}(i)+\sum_{l',m',j}(-1)^{l'}A_{lmi}^{l'm'j}q_{l'm'j}\right]\ , \tag{5}$$

where

$$A_{lmi}^{l'm'j}=\begin{cases}0 \ \text{ if } i=j\ ,\\ (-1)^{m'}\dfrac{[Y_{l+l'}^{(m-m')}(\theta_{ij},\phi_{ij})]^*}{R_{ij}^{l+l'+1}}\left[\dfrac{(4\pi)^3(l+l'+m-m')!(l+l'-m+m')!}{(2l+1)(2l'+1)(2l+2l'+1)(l+m)!(l-m)!(l'+m')!(l'-m')!}\right]^{1/2}\ , \ \text{if } i\neq j\ .\end{cases} \tag{6}$$

Equation (5) is a system of linear equations that couples the excited multipoles in the particles composing our sample. It is convenient to rewrite it in the more compact form

$$\sum_{\mu'}(\chi^{-1}\delta_{\mu\mu'}+R_\mu^{\mu'})\bar{q}_{\mu'}=G_\mu\ , \tag{7}$$

where $\bar{q}_\mu=a_i^l q_\mu$ with μ representing the triplet of indices (l,m,i), $\delta_{\mu\mu'}$ is a product of three Kronecker deltas, and

$$G_\mu=-la_i^{l+1}V_{lm}^{\text{ext}}(i)\ , \tag{8}$$

$$R_\mu^{\mu'}=4\pi n_l^0\delta_{\mu\mu'}+(-1)^{l'}la_i^{l+1}a_j^{l'}A_\mu^{\mu'}\ . \tag{9}$$

Written in matrix form, Eq. (7) becomes

$$(\chi^{-1}\underline{I}+\underline{R})\mathbf{Q}=\mathbf{G}\ , \tag{10}$$

where $\mathbf{Q}$ and $\mathbf{G}$ are column vectors whose elements are $\bar{q}_\mu$ and G_μ, respectively, $\underline{R}$ is a matrix of elements $R_\mu^{\mu'}$ and $\underline{I}$ is the identity matrix with elements $\delta_{\mu\mu'}$. Note the important feature that $\underline{R}$ depends only on the geometry of the array. Assuming $\underline{U}$ is a matrix that diagonalizes $\underline{R}$, multiplying Eq. (10) on the left by $\underline{U}^{-1}$ we obtain the relation:

$$(\chi^{-1}\underline{I}+\underline{R}')\mathbf{Q}'=\mathbf{G}'\ , \tag{11}$$

where

$$\underline{R}'=\underline{U}^{-1}\underline{R}\,\underline{U},\quad \mathbf{Q}'=\underline{U}^{-1}\mathbf{Q},\quad \mathbf{G}'=\underline{U}^{-1}\mathbf{G}\ . \tag{12}$$

Because $\underline{R}'$ is diagonal, we can solve immediately Eq. (11). The solution is

$$\bar{q}_\mu=\sum_{\mu'}\frac{C_\mu^{\mu'}}{\chi^{-1}+4\pi n_{\mu'}}\ , \tag{13}$$

where the depolarization factors n_μ are defined through $(R')_\mu^{\mu'}=4\pi n_\mu\delta_{\mu\mu'}$, and the amplitudes

$$C_\mu^{\mu'}=\sum_{\mu''}U_\mu^{\mu'}(\underline{U}^{-1})_{\mu'}^{\mu''}G_{\mu''} \tag{14}$$

satisfy the sum rules

$$\sum_{\mu'}C_\mu^{\mu'}=G_\mu\ . \tag{15}$$

We have thus succeeded in expressing the multipolar moments as a sum over the N-particle-system normal modes, with each normal mode characterized by a depolarization factor $n_{\mu'}$ and a set of amplitudes $c_\mu^{\mu'}$. The minima of the denominators in Eq. (13) determine the values of ϵ corresponding to the normal mode resonances. In the absence of damping the resonances are poles of Eq. (13) and occur at the sequence of values $\epsilon_\mu^*=1-1/n_\mu$. Our Eq. (13) is an extension to all multipolar couplings of the dipolar theory of Fuchs.[21] Its equivalence with the multipolar theory of Ref. 18 is proven in the Appendix.

The sum rule (15) suggests that for $G_\mu\neq 0$ one may define weight factors $C_\mu^{\mu'}/G_\mu=\bar{C}_\mu^{\mu'}$ that add up to 1. The weighted average depolarization factor

$$\langle n_\mu\rangle=\sum_{\mu'}\bar{C}_\mu^{\mu'}n_{\mu'}=n_l^0+\frac{a_i^{l+1}}{4\pi G_\mu}\sum_{\mu'}(-1)^{l'}a_{i'}^{l'}A_\mu^{\mu'}G_{\mu'}\ , \tag{16}$$

does not depend on the fields when only one component is nonzero. For instance, in a uniform external field an $l=1$ field is present only and (16) reduces to

$$\langle n_{1mi}\rangle=\frac{1}{3}\left[1+a_i^2(1-3\delta_{m0})\times\sum_{i'}\frac{P_2(\cos\theta_{ii'})}{R_{ii'}^3}a_{i'}(1-\delta_{ii'})\right]\ . \tag{17}$$

One may easily verify then the additional sum rule

$$\sum_{m=-1}^{1} \langle n_{1mi} \rangle = 1 \tag{18}$$

valid for an arbitrary configuration.

III. CASE OF A PAIR

The simplest case we can study that includes multipolar couplings corresponds to two equal spheres in a uniform electric field. This problem has been treated in the past by Goyette and Navon[25] using bispherical coordinates and by Claro[18,26] in a multipolar expansion. The pair is known to exhibit all qualitative features of symmetric arrays and because of its low dimensionality one can get good numerical results with a moderate computational effort.[18] In the case of disordered systems the structure in the spectrum will disappear but certain features such as the proximity broadening effect discussed in Ref. 27 is likely to remain.[28] We shall thus apply our results to a pair as the simplest representative system of more complex arrays.

If we consider two equal spheres of radius a, one of them at the origin and the other at the point $z=D$ on the z axis, then the coefficients $A_\mu^{\mu'}$ in Eq. (6) satisfy

$$A_\mu^{\mu'} = A_{lmi}^{l'mi'} \delta_{mm'} , \tag{19}$$

a symmetry condition that suppresses couplings in the m index and permits to study separately Eqs. (7) with different m indices. If we also separate optically active modes (OA modes) from optically inactive modes (OI modes), we can use the relation

$$q_{lm2} = (-1)^{l+\Delta} q_{lm1} , \tag{20}$$

where $\Delta=1$ for OA modes and $\Delta=0$ for OI modes. With these simplifications one can do explicitly the sum over the i' index in Eq. (7), obtaining

$$\sum_{l'} (\chi^{-1}\delta_{ll'} + \tilde{R}_l^{\,l'}) \bar{q}_{l'} = G_l , \tag{7'}$$

where

$$\tilde{R}_l^{\,l'} = 4\pi n_l^0 \delta_{ll'} + (-1)^\Delta l a^{l+l'+1} A_{lm1}^{l'm2} , \tag{9'}$$

$$A_{lm1}^{l'm2} = \begin{cases} 4\pi \begin{pmatrix} l+l' \\ l \end{pmatrix} \dfrac{1}{[(2l+1)(2l'+1)]^{1/2}} \cdot \dfrac{1}{D^{l+l'+1}} & \text{if } m=0 , \\[2ex] -\left[\dfrac{l \cdot l'}{(l+1)(l'+1)} \right]^{1/2} A_{l01}^{l'02} & \text{if } m=\pm 1 . \end{cases} \tag{6'}$$

In Eq. (7') the m index and the particle index are implicit, with the identification $\bar{q}_l = \bar{q}_{lm1}$ and $G_l = G_{lm1} = G_{lm2}$. In a uniform external field (long-wavelength limit) only the $m=0,\pm 1$ cases are relevant and these are the only ones included in the above expressions. If the field is along the line joining the particle centers (parallel excitation) only the $m=0$ modes are excited, whereas for a field perpendicular to this line the degenerate $m=\pm 1$ modes are excited. By using Eq. (17), the following very simple expressions for the average parallel and perpendicular depolarization factors in a uniform electric field are obtained for the axial and transverse fields, respectively:

$$\begin{aligned} \langle n^{\parallel} \rangle &= \langle n_{10i} \rangle = \frac{1}{3} \left[1 - \frac{1}{4\sigma^3} \right] , \\ \langle n^{\perp} \rangle &= \langle n_{1,\pm 1,i} \rangle = \frac{1}{3} \left[1 + \frac{1}{8\sigma^3} \right] , \end{aligned} \tag{17'}$$

where $\sigma = D/2a$. These are exact expressions that show that at large separation there is no difference between axial and transverse excitation so both averages approach the same value $\frac{1}{3}$. For nearer spheres the excitation direction is important, the values for touching spheres being $\langle n^{\parallel} \rangle = \frac{1}{4}$ and $\langle n^{\perp} \rangle = \frac{3}{8}$. We may easily check that the sum rule (18) is obeyed for all values of σ.

In the numerical computations we need to limit the number of equations in the infinite set given by Eq. (7'). Because the coupling coefficients $A_{lm1}^{l'm2}$ decrease with increasing indices l or l', we can get the so-called 2^L-polar approximation by suppressing all couplings with l or l' greater than L. Then Eq. (7') gives a set of L simultaneous linear equations that can be solved to obtain depolarization factors and strengths. The L value required in a

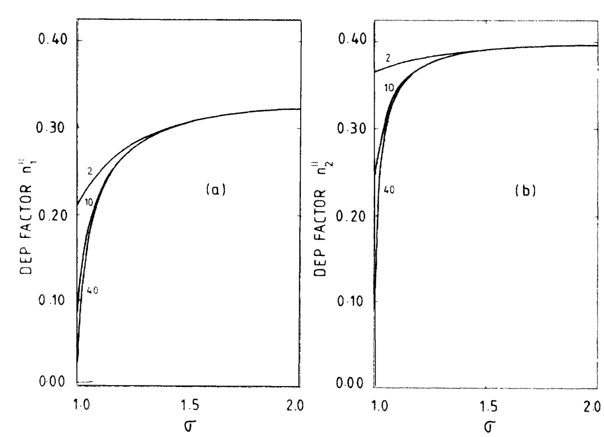

FIG. 1. Dipolar (a) and quadrupolar (b) depolarization factors for two equal spheres a distance $\sigma=D/2a$ apart excited by a field along the line joining their centers. Integers label the order (L) of the approximation used.

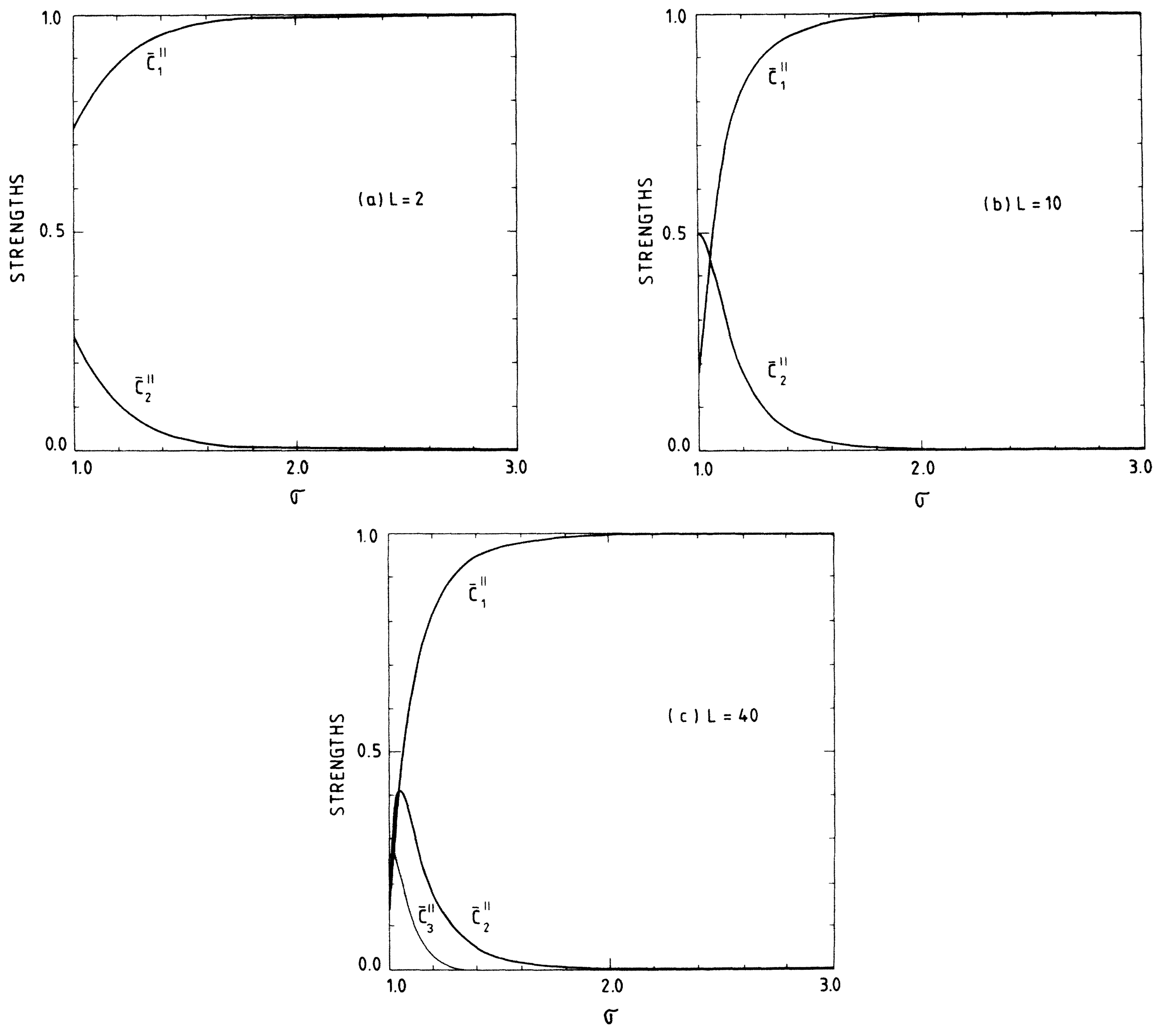

FIG. 2. Dipolar and quadrupolar mode strengths for two spheres excited by a field along the line joining their centers, in the 2^L polar approximations: (a) $L=2$, (b) $L=10$, (c) $L=40$.

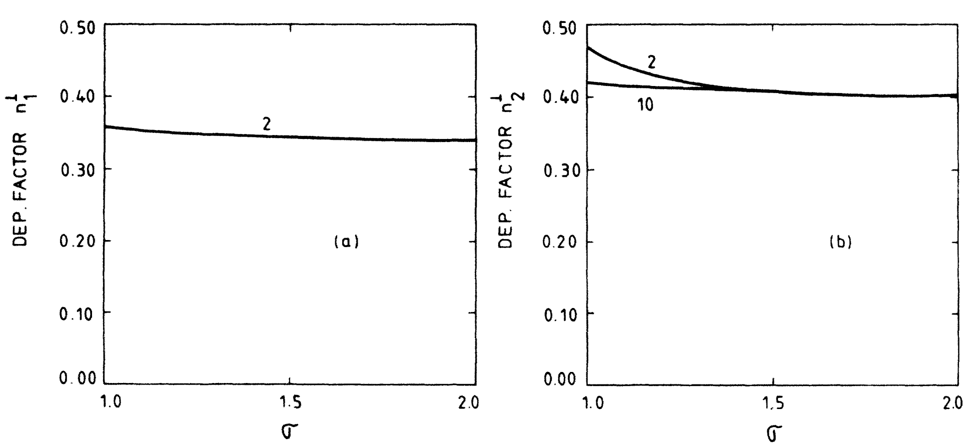

FIG. 3. Same as Fig. 1 with a field perpendicular to the line joining the sphere centers. (a) The approximation of order $L=10$ and $L=40$ coincide with the curve shown. (b) The approximation of order $L=40$ gives the same results as that with $L=10$.

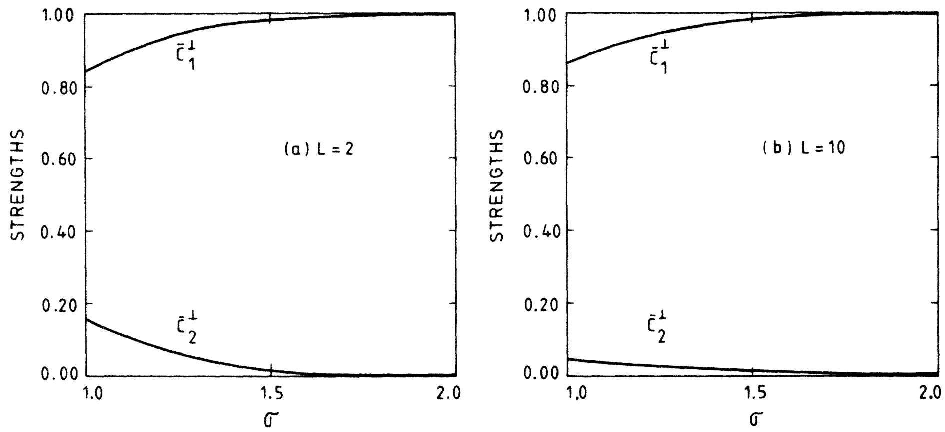

FIG. 4. Same as Fig. 3, with a field perpendicular to the line joining the sphere centers. Here the approximation of order $L=40$ gives the same results as that with $L=10$.

given computation is a decreasing function of the normalized distance σ between spheres.[27] In our computations we have used up to $L=160$.

Figure 1 contains the dipolar (a) and quadrupolar (b) depolarization factors $n_1^{\parallel}$ and $n_2^{\parallel}$ as a function of σ, in the 2^2-, 2^{10}-, and 2^{40}-polar approximations. We note that these factors have the expected limit behavior for large σ, their value approaching the first two terms in the sequence $n_l^0 = l/(2l+1)$ (where $l=1,2,3,\ldots$) of depolarization factors for an isolated sphere. In decreasing σ, $n_l^{\parallel}$ decreases faster for larger L. This indicates that a value of σ close to 1 requires a large value of L to obtain a valid result. Convergence of $n_l^{\parallel}$ is faster, however, the smaller the value of l.

The dipolar strengths $\bar{C}_l^{\parallel}$ corresponding to the depolarization factors $n_l^{\parallel}$ are shown in Fig. 2. Figure 2(a) is for $L=2$, an approximation in which the dipolar mode dominates at all values of σ. Figs. 2(b) and 2(c) show the strengths of the first two modes in the approximations $L=10$ and 40, respectively. There we see that for near-touching spheres ($\sigma=1$) the dipolar mode ceases to be the mode of largest strength. For $L=10$, the mode of greatest strength is the quadrupolar, while for $L=40$ the octupolar mode is stronger. This means that particles that are very close will absorb predominantly by excitation of modes of pole order higher than the dipole.

In order to study spheres at σ very close to 1, we have increased L to the value 160 obtaining converged results for $\sigma=1.001$. For an axial external field ($m=0$) we get only two fully converged strengths and depolarization factors while for a transverse field ($m=1$) all significant strengths are converged. We show these numerical results in Table I where the relative importance of the dipolar mode is evident; while it is clearly the predominant mode for a transverse field, it is not for an axial field because at least the quadrupolar mode has a larger strength than the dipolar one. The numerical results for a field perpendicular to the line joining the particle centers are shown in Fig. 3 (depolarization factors) and Fig. 4 (strengths). The results with $L=10$ and 40 are the same which indicates a fast convergence even at very small σ. Figures 5(a), 5(b), and 5(c) are plots of the strength versus depolarization factor corresponding to diverse situations. Comparison of the axial and transverse cases shows that while in the former the strength is broadly distributed among the first few multipolar excitations, in the latter the strength is concentrated in the dipolar mode. In all cases the sum rules (17′) are obeyed since they remain exact independently of the value of L.

TABLE I. Converged strengths and depolarization factors in the approximation $L=160$ and $\sigma=1.001$.

	$m=0$		$m=1$	
l	$C_l^{\parallel}$	$n_l^{\parallel}$	$C_l^{\perp}$	$n_l^{\perp}$
1	0.0274	0.0319	0.8674	0.357
2	0.0406	0.0725	0.0403	0.419

IV. CONCLUSIONS

We have developed a formalism for the problem of N spherical particles in an external electric field that separates the response of the system in independent terms each corresponding to an individual normal mode. This decomposition is advantageous in discussing absorption by clusters since the strength with which each mode contributes to the spectrum is a function of geometry only and therefore universal for a given array. Thus, for an array of metallic spheres, for instance, if the depolarization factors and strengths are known, the mode $l'm'$ participates in the absorption through a term of the form

$$\frac{1}{4\pi}\frac{\Omega^2 C_{1m}^{l'm'}}{(\Omega^2 - n_{l'm'})^2 + \Gamma^2\Omega^2},$$

where a Drude model dielectric function has been assumed, with $\Omega=\omega/\omega_p$ and $\Gamma=1/\omega_p\tau$. Note that all terms behave as ω^2 at very low frequencies in agreement

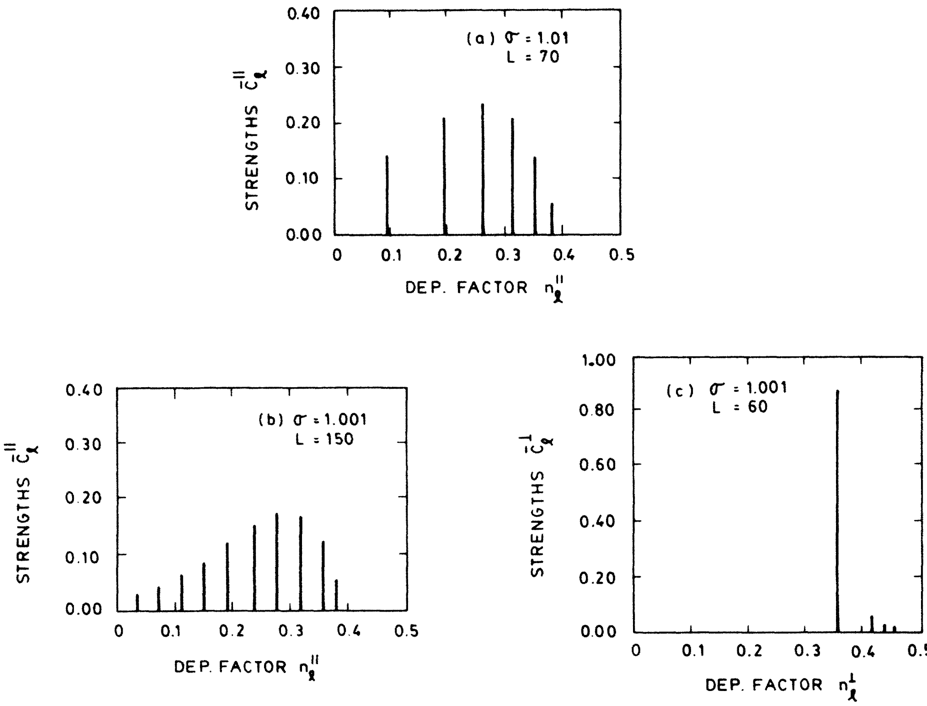

FIG. 5. Mode strengths as a function of depolarization factors for two equal spheres excited by a field along the line joining their centers (a),(b) and perpendicular to this line (c). Data shown in (a) and (c) are fully converged but in (b) only the strengths corresponding to the two smallest depolarization factors are fully converged.

with experiments, since depolarization factors are nonzero for finite separation of the particles. In addition, we have found that in the two-particle cluster the lowest pole-order excitations such as the dipole and even quadrupole, may become weaker than higher multipolar excitations, a result that stresses the fact that the dipole approximation is poor for very close particles.

It is worthwhile to emphasize that this work confirms earlier results on the limitations of the dipolar approximation. According to those, the dipolar approximation fails when the particles are closer than about three-particle radii from each other. Beyond this separation, multipolar excitations amount to an effect of less than 1% in the relevant quantities.[25,26] As we show here, this result depends purely on geometry and not on the dielectric properties of the particles.

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APPENDIX

The formal equivalence between our expression (13) and Eq. (11) of Ref. 18 that exhibits an alternate decomposition of the couplings between particles may be easily proven. This last equation can be written in the form

$$q_\mu=\frac{J_\mu}{1-Q^\mu_\mu}\,, \tag{A1}$$

where

$$J_\mu=\sum_{\mu'}\frac{\alpha_{\mu'}G_{\mu'}}{4\pi n^0_{l'}a_{i'}^{l'+1}}\cdot[Q^{\mu'}_\mu+\delta_{\mu\mu'}(1-Q^{\mu'}_\mu)]\,, \tag{A2}$$

and

$$Q^{\mu'}_\mu=1+\frac{\det(\underline{B}-\underline{I})}{\operatorname{cof}(B^\mu_\mu-1)}\,, \tag{A3}$$

with

$$B^{\mu'}_\mu=(-1)^{l'+1}\frac{2l+1}{4\pi}\alpha_\mu A^{\mu'}_\mu\,. \tag{A4}$$

Because $\det(\underline{B}-\lambda\underline{I})$ is a polinomial in λ, it can be written in the form

$$\det(\underline{B}-\lambda\underline{I})=(\lambda-\lambda_1)(\lambda-\lambda_2)\cdots(\lambda-\lambda_t)\,, \tag{A5}$$

where $\{\lambda_1,\lambda_2,\ldots,\lambda_t\}$ is the set of eigenvalues of matrix $\underline{B}$ [i.e., the solutions to the algebraic equation $\det(\underline{B}-\lambda\underline{I})=0$]. By making a partial fraction expansion we obtain

$$[\det(\underline{B}-\lambda\underline{I})]^{-1}=\sum_{s=1}^{t}\frac{T_s}{1-\lambda_s}\,, \tag{A6}$$

where

$$T_s = \left[\frac{d}{d\lambda} [\det(\underline{B} - \lambda \underline{I})] \right]^{-1}_{\lambda=\lambda_s} , \qquad \text{(A7)}$$

Finally, introducing Eqs. (A3), (A5), and (A7) in Eq. (A1) we obtain

$$q_\mu = \widetilde{J}_\mu \sum_{s=1}^{t} \frac{T_s}{1-\lambda_s} , \qquad \text{(A8)}$$

where

$$\widetilde{J}_\mu = -J_\mu \operatorname{cof}(B^\mu_\mu - 1) . \qquad \text{(A9)}$$

With the identifications

$$T_s = \frac{\chi a_i^l}{\widetilde{J}_\mu} \sum_{\mu''} U^s_\mu (\underline{U}^{-1})^\mu_s G_{\mu''} , \qquad \text{(A10)}$$

$$\lambda_s = -4\pi \chi n_s , \qquad \text{(A11)}$$

Eq. (A8) is just Eq. (13).

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*Permanent address: Departamento de Física, Universidad Federico Santa María Casilla Postal 110-V, Valparaíso, Chile.

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