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Ultrasonic attenuation in a quadrupolar solid

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A general expression for the high-temperature ultrasonic attenuation in a quadrupolar solid is derived. The four-point correlation function occurring in this expression is then factored into products of two-point functions so that the attenuation is proportional to the convolution of two of the usual two-point angular momentum correlation functions. Using first-principles results for these functions, which were previously obtained for solid H_2, the field-dependent or resonant part of the ultrasonic attenuation is then calculated as a function of ortho-H_2 concentration c. The result of this calculation indicates that the resonant attenuation for solid H_2 has a magnitude such that it could be observable by current techniques. A new kind of ultrasonic magnetic resonance is thus predicted.

I. INTRODUCTION

In this paper, a theory of the ultrasonic attenuation in solid H_2[1,2] and other quadrupolar solids[3] at temperatures large compared to the quadrupolar ordering temperature will be presented. In particular, the primary concern of the following discussion will be the calculation of the response of a quadrupolar solid to an externally applied, monochromatic ultrasonic wave. In addition, the possibility of a new kind of ultrasonic (or "acoustic") magnetic resonance[4,5] will be explored through an examination of the dependence of the attenuation on an external magnetic field.

In solid H_2 and other quadrupolar solids, the dominant orientationally dependent intermolecular forces originate from the electric quadrupole-quadrupole (EQQ) interaction.[1,2] In some sense then, this interaction plays a role in molecular solids similar to that which is played by exchange plus dipolar forces in a paramagnetic insulator. Since the dominant electron spin-phonon coupling mechanism in paramagnetic insulators is phonon modulation of the exchange plus dipolar interactions,[5-8] it is natural to assume, in analogy with that case, that the molecular-phonon interaction in a quadrupolar solid is described by phonon modulation of the EQQ interaction. Other authors have considered this kind of molecular-phonon interaction in solid H_2 in the context of the libron-phonon problem,[9,10] thermal conductivity,[11] renormalizations due to phonon effects,[1] and other problems.[1,2] However, thus far there has been no calculation of the ultrasonic attenuation due to this mechanism.

The facts that the EQQ interaction in quadrupolar solids has a relatively large magnitude and that current ultrasonic techniques[12] are capable of resolving acoustic attenuation effects of the order of 10^{-6} cm^{-1} are the primary motivations for the calculation of the ultrasonic attenuation in this system. Another motivation for this calculation is the fact that the dynamical angular momentum correlation functions which, as will be shown below, enter the theoretical expression for the attenuation in lowest order have recently been calculated at high temperatures in a *first-principles self-consistent* approximation[13,14] as a function of the o-H_2 concentration, c. Finally, as will also be shown below, the attenuation owing to this interaction can depend on the external magnetic field, which brings to mind the possibility of the observation of ultrasonic magnetic resonance in solid H_2 and other quadrupolar solids. Although the effects calculated in this paper have not yet been observed, it is hoped that this calculation will stimulate experimentalists in that direction.

The method used to obtain the ultrasonic attenuation is based upon a calculation of the phonon self-energy and is discussed in several places.[4-8,15-17] As will be shown below, the theoretical expression for the attenuation resulting from the use of this method is proportional to the Fourier transform of a fourpoint correlation function which is the time-ordered thermal average of four of the irreducible tensor angular momentum operators which are discussed elsewhere.[4,13,14,17-19] In analogy with the standard approximation for four-point correlation functions in magnetic-insulator problems,[5-8] it will be assumed that a good, lowest-order approximation for this function is that it can be factored into products of two-point correlation functions. Thus, with this approximation, the acoustic attenuation as a function of frequency becomes proportional to the convolution of two of the two-point angular momentum correlation functions which were determined for solid H_2 from first principles for various c in Refs. 13 and 14. By the use of the functions obtained in those references, a quantitative estimate of the acoustic attenuation as a function of frequency and orthoconcentration is then possible. Finally, the dependence of this attenuation on an external magnetic field is considered and an estimate of the resonant attenuation line shape and magnitude is made, with the result that this new kind of ultrasonic magnetic resonance ought to be observable for sufficiently large magnetic fields in the high-temperature phase of solid H_2 or other quadrupolar solids. This last prediction will hopefully stimulate some experimental activity in this area.

For the purposes of this paper, "high temperature" will be taken to mean temperatures much greater than the quadrupolar-ordering temperature, where the infinite-temperature approximation for the dynamical angular momentum correlation functions is valid. Furthermore, this will be the only temperature regime considered in this paper. In the context of this paper, the term "resonant attenuation" will be taken to mean that contribution to the ultrasonic attenuation which depends upon an externally applied magnetic field. Also, in keeping with the terminology presently found in the literature, terms like "acoustic attenuation" and "acoustic magnetic resonance" will sometimes be used in place of the more correct terms "ultrasonic attenuation" and "ultrasonic magnetic resonance."

The remainder of this paper is organized as follows. Section II is devoted to a discussion of the physical model to be used and to a discussion of the notation that will be used throughout the paper. In Sec. III, the formalism for the calculation of the attenuation is briefly outlined and the approximations used in the calculation are discussed. Section IV contains the results of the calculation including predictions of the ultrasonic magnetic resonance line shape and the maximum resonant attenuation for a quadrupolar solid.

II. PHYSICAL MODEL

It is assumed that the Hamiltonian for the quadrupolar solid in the presence of phonons can be written as a sum of molecular, phonon, and molecular-phonon parts:

$$H = H_M + H_P + H_{MP} \ . \tag{1}$$

The molecular part of the Hamiltonian, H_M, will be assumed to be the pure EQQ interaction, which can be written[13,14]

$$H_M = \sum_{i \neq j} \sum_{m,m'} A_{2m}(i) J_{mm'}(i,j) A_{2m'}(j) \ , \tag{2a}$$

with (for solid H_2)

$$J_{mm'}(i,j) = \tfrac{1}{12}(70\pi)^{1/2}\Gamma\mu_J C_{mm'} \times (a/|\vec{r}_{ij}|)^5 [Y_4^{m+m'}(\Omega_{ij})]^* \ , \tag{2b}$$

where the $A_{2m}(i)$ are the irreducible tensor angular momentum operators for lattice site $\vec{r}_i$ which correspond to a rotational angular momentum $\vec{J}$ and which are discussed and defined in Refs. 17–19; $C_{mm'}$ is a shorthand notation for the Clebsch-Gordan coefficient $C(224;\, mm')$, Ω_{ij} specifies the direction of the intermolecular distance $\vec{r}_{ij}$ with respect to the c axis of the hcp crystal of solid H_2, $Y_4^{m+m'}$ is a spherical harmonic, a is the lattice constant, $\mu_J = \frac{1}{15}[2J(J+1)(J-1)(2J+3)]$, J is the angular momentum quantum number, and Γ is the EQQ coupling constant.

Following Refs. 4 and 5, it is assumed that since the effect of interest is the change in the phonon spectrum due to molecular-phonon interactions, it is sufficient to use the harmonic approximation for the phonons. In terms of phonon normal coordinates, this has the form[4,5,15]

$$H_P = \frac{1}{2}\sum_{\vec{q},\lambda}[\dot{Q}(\vec{q},\lambda,t)\dot{Q}(-\vec{q},\lambda,t) + \omega^2(\vec{q},\lambda)Q(\vec{q},\lambda,t)Q(-\vec{q},\lambda,t)] \ , \tag{3a}$$

where $\vec{q}$ is a wave vector in the first Brillouin zone, λ specifies the phonon branch, and $\omega(\vec{q},\lambda)$ is the harmonic frequency of the $(\vec{q},\lambda)$ phonon mode. The normal coordinate $Q(\vec{q},\lambda,t)$ and its canonical momentum $\dot{Q}(\vec{q},\lambda,t)$ satisfy the usual commutation relations:

$$[\dot{Q}(\vec{q},\lambda,t), Q(\vec{q}\,',\lambda',t)] = -i\hbar\delta(\lambda,\lambda')\delta(\vec{q}\,'+\vec{q}) \tag{3b}$$

and

$$[Q(\vec{q},\lambda,t), Q(\vec{q}\,',\lambda',t)] = [\dot{Q}(\vec{q},\lambda,t), \dot{Q}(\vec{q}\,',\lambda',t)] = 0 \ . \tag{3c}$$

The molecular-phonon interactions to be considered in this paper are taken to be linear in the strains or displacements from equilibrium and quadratic in the tensor angular momentum operators $A_{2m}(i)$. Interactions of higher order in the strains or the angular momentum operators should be smaller.[5] Since the EQQ interaction, Eq. (2), depends on the distance between the molecules, lattice vibrations modulate this coupling. This is mathematically represented by expanding the potential $J_{mm'}(i,j)$ in a power series in the displacements from equilibrium. If one keeps only terms which are linear in the displacements $\vec{U}(i)$, the molecular-phonon interaction has the form

$$H_{MP} = \sum_{i \neq j}\sum_{mm'} A_{2m}(i)A_{2m'}(j)\vec{K}_{mm'}(i,j)\cdot[\vec{U}(i) - \vec{U}(j)], \tag{4a}$$

where

$$\vec{K}_{mm'}(i,j) = \nabla_{ij}J_{mm'}(i,j) \ . \tag{4b}$$

Here ∇_{ij} means the gradient with respect to $\vec{r}_{ij}$. The lattice displacement $\vec{U}$ can be expanded in phonon normal coordinates as[4,5,15]

$$\vec{U}(i) = (MN)^{-1/2}\sum_{\vec{q},\lambda}\hat{e}(\vec{q},\lambda)\,e^{i\vec{q}\cdot\vec{r}_i}Q(\vec{q},\lambda) \ , \tag{4c}$$

where $\hat{e}(\vec{q},\lambda)$ is the unit polarization vector of the

$(\vec{q}, \lambda)$ phonon mode, M is the molecular mass, and N is the number of molecules. Substitution of Eq. (4c) into Eq. (4a) yields

$$H_{MP} = (MN)^{-1/2} \sum_{i \neq j} \sum_{m,m'} \sum_{\vec{q},\lambda} A_{2m}(i) A_{2m'}(j) \times \vec{K}_{mm'}(i,j) \cdot \hat{e}(\vec{q},\lambda) Q(\vec{q},\lambda)(e^{i\vec{q}\cdot\vec{r}_i} - e^{i\vec{q}\cdot\vec{r}_j}) \ . \tag{4d}$$

Equation (4d) is the form of the molecular-phonon interaction that will be considered in this paper.

III. CALCULATION OF THE ACOUSTIC ATTENUATION

The method that will be used to calculate the acoustic attenuation due to the molecular-phonon process H_{MP} described by Eq. (4d), is the same as that used in Refs. 4–7, so it will only be briefly discussed here. The acoustical properties of the system are conveniently discussed by the use of the phonon Green's function.[15]

$$D(\vec{q}, \lambda, t-t') = (i/\hbar) \langle (Q(\vec{q},\lambda,t) Q(-\vec{q},\lambda,t'))_+ \rangle \ , \tag{5}$$

where the angular brackets $\langle \cdots \rangle$ denote an average in the canonical ensemble and the circular brackets $(\cdots)_+$ denote the Wick time-ordering operation. $D(t)$ is Fourier transformed via the usual prescription[15]:

$$D(\omega_\nu) = \int_0^{-i\hbar\beta} dt\, e^{i\omega_\nu t} D(t) \ , \tag{6}$$

where $\omega_\nu = \pi\nu / i\hbar\beta$, $\beta = 1/kT$, and ν is an even integer. By the use of Eqs. (3a), (3c), (4d), and the Heisenberg equation of motion

$$i\hbar \frac{\partial x}{\partial t} = [x, H_P + H_{MP}] \ , \tag{7}$$

an exact equation of motion can be obtained for the phonon Green's function. This equation is

$$\left(\frac{\partial^2}{\partial t^2} + \omega^2(\vec{q},\lambda)\right) D(\vec{q},\lambda,t-t') = \delta(t-t') - (MN)^{-1/2} \sum_{i,j} (e^{-i\vec{q}\cdot\vec{r}_i} - e^{-i\vec{q}\cdot\vec{r}_j}) \times \langle (F(-\vec{q},\lambda,i,j,t) Q(-\vec{q},\lambda,t'))_+ \rangle \ , \tag{8a}$$

where

$$F(\vec{q},\lambda,i,j,t) = \sum_{m,m'} A_{2m}(i,t) A_{2m'}(j,t) \vec{K}_{mm'}(i,j) \cdot \hat{e}(\vec{q},\lambda) . \tag{8b}$$

Following Refs. 4–7 the second term on the right-hand side of Eq. (8a) can be expanded in powers of H_{MP}. Since H_{MP} is linear in the normal coordinate $Q(\vec{q},\lambda,t)$, only odd powers of that interaction will contribute to the expansion. When such an expansion is made, Eq. (8a) can be written

$$\left(\frac{\partial^2}{\partial t^2} + \omega^2(\vec{q},\lambda)\right) D(\vec{q},\lambda,t-t') = \delta(t-t') + \int_0^{-i\hbar\beta} d\bar{t}\, \Pi(\vec{q},\lambda,t-\bar{t}) D(\vec{q},\lambda,\bar{t}-t') \ , \tag{9a}$$

where, to lowest order in H_{MP},

$$\Pi(\vec{q},\lambda,t-\bar{t}) = \frac{i}{MN} \sum_{iji'j'} (e^{-i\vec{q}\cdot\vec{r}_{i'}} - e^{-i\vec{q}\cdot\vec{r}_{j'}}) \times (e^{-i\vec{q}\cdot\vec{r}_{i'}} - e^{-i\vec{q}\cdot\vec{r}_{j'}}) \langle (F(-\vec{q},\lambda,i,j,t) \times F(\vec{q},\lambda,i',j',\bar{t}))_+ \rangle \ . \tag{9b}$$

Upon Fourier transforming and rearranging, Eq. (9a) can be rewritten

$$D(\vec{q},\lambda,\omega_\nu) = [-\omega_\nu^2 + \omega^2(\vec{q},\lambda) - \Pi(\vec{q},\lambda,\omega_\nu)]^{-1} \ , \tag{9c}$$

where $\Pi(\vec{q},\lambda,\omega_\nu)$ is now, by definition,[15] the phonon self-energy.

From Eqs. (8b) and (9b), it can be seen that the phonon self-energy $\Pi(\vec{q},\lambda,\omega_\nu)$ is, to lowest order in H_{MP}, proportional to the Fourier transform of the four-point correlation function

$$L_{mm'\bar{m}\bar{m}'}(i,j,i',j',t,\bar{t}) = \langle (A_{2m}(i,t) A_{2m'}(j,t) A_{2\bar{m}}(i',\bar{t}) A_{2\bar{m}'}(j',\bar{t}))_+ \rangle \ . \tag{10a}$$

In order to proceed further, this four-point function must therefore be evaluated. To calculate this function from a strictly first-principles approach would certainly be a formidable task, although progress has recently been made on the calculation of four-point correlation functions in paramagnetic systems.[20] Because of the complexity of this problem, such a first-principles calculation of L will not be attempted here. Instead, following the procedure that other authors[5–8] have used in the paramagnet case, this function will be approximated here by the sum of all possible products of two-point correlation functions containing different times:

$$L_{mm'\bar{m}\bar{m}'}(i,j,i',j',t,\bar{t}) \simeq \langle (A_{2m}(i,t) A_{2\bar{m}}(i',\bar{t}))_+ \rangle \langle (A_{2m'}(j,t) A_{2\bar{m}'}(j',\bar{t}))_+ \rangle + \langle (A_{2m}(i,t) A_{2\bar{m}'}(j',\bar{t}))_+ \rangle \langle (A_{2m'}(j,t) A_{2\bar{m}}(i',\bar{t}))_+ \rangle \ . \tag{10b}$$

Correlations that are independent of time do not contribute to the phonon self-energy at finite frequencies and are neglected. Following other authors,[5–8] this decomposition is expected to be most valid at temperatures large compared to the quadrupolar-ordering temperature. Also, preliminary calculations have shown that this approximation is the first term in an expansion of L in a power series in H_M and the two-point correlation functions.[21]

If one uses the approximation, Eq. (10b) for L, keeps only the angular momentum conserving two-point correlation functions so that

$$\langle (A_{2m}(i,t)\,A_{2\bar{m}}(i',\bar{t}))_+\rangle = \delta_{\bar{m},-m}\langle (A_{2m}(i,t)\,A_{2,-m}(i',\bar{t}))_+\rangle$$
$$\equiv \delta_{\bar{m},-m}(-1)^m\,G_{2m}(i,i',t-\bar{t}). \tag{11}$$

substitutes Eqs. (10b) and (11) into Eq. (9b) and Fourier transforms that equation, the result, after some manipulation, is

$$\Pi(\vec{q},\lambda,\omega_\nu) = \frac{2}{MN\beta}\sum_{mm'}\sum_{\vec{q}'}\sum_{\nu'}\left|\hat{e}(\vec{q},\lambda)\right.$$
$$\left.\circ\,[\vec{K}_{mm'}(\vec{q}') + \vec{K}_{mm'}(\vec{q}-\vec{q}')]\right|^2$$
$$\times\,\mathcal{G}_{2m}(\vec{q}',\omega'_\nu)\,\mathcal{G}_{2m'}(\vec{q}-\vec{q},\omega_\nu-\omega_{\nu'})\ , \tag{12}$$

where $\mathcal{G}_{2m}(\vec{q},\omega_\nu)$ is the Fourier transform of the function defined in Eq. (11) and $\vec{K}_{mm'}(\vec{q})$ is the Fourier transform of $\vec{K}_{mm'}(i,j)$. The functions $\mathcal{G}_{2m}(\vec{q},\omega_\nu)$ are related to the correlation functions discussed in Refs. 13 and 14 by the fluctuation-dissipation theorem.[22]

In order to compute the acoustic attenuation from $\Pi(\vec{q},\lambda,\omega_\nu)$, one uses the prescription[15]

$$\alpha(\vec{q},\lambda,\omega) = \mathrm{Im}[\Pi(\vec{q},\lambda,\omega+i\epsilon)/2V_\lambda\omega(\vec{q})]\ , \tag{13}$$

where ϵ is a positive infinitesimal, ω is a real frequency, and V_λ is the velocity of sound for the $(\vec{q},\lambda)$ phonon mode.

By subsitution of Eq. (12) into Eq. (13), a general formula for the acoustic attenuation can be obtained. If an external magnetic field, $\vec{H}_0$, is present, however, as will be the case for the acoustic magnetic resonance problem discussed in Sec. IV, it is necessary to make one further step before proceeding with the calculation. The crystal of primary interest in the present paper is solid H_2, which has a hcp structure in the high-temperature phase.[1,2] In the presence of a magnetic field, the convenient coordinate system for discussing the angular momentum operators A_{2m} is one which has its axis coincide with the direction of $\vec{H}_0$. On the other hand, the convenient coordinate system for discussing the phonons is one in which the z axis is along the c axis of the hcp crystal. In general, $\vec{H}_0$ will make an angle (θ,ϕ) with respect to the c axis. The transformation that takes the angular momentum operators from one coordinate system to another is a generalization for the hcp crystal of that used in Refs. 4 and 5 and will be discussed in Appendix A.

After making such a coordinate transformation on Eq. (12), assuming that the $\vec{q}$ dependence of the angular momentum correlation functions in that equation can be neglected as in Refs. 13 and 14, using the fluctuation-dissipation theorem[22] to relate the correlation functions in Eq. (12) to those calculated in Refs. 13 and 14, using the prescription given by Eq. (13), and taking the high-temperature limit the general formula for the high-temperature acoustic attenuation has the form

$$\alpha(\vec{q},\lambda,\omega) = \frac{\beta\omega}{M\omega(\vec{q},\lambda)V_\lambda}\sum_{mm'} S_{mm'}(\vec{q},\lambda)\,I_{mm'}(\omega)\ , \tag{14a}$$

where

$$S_{mm'}(\vec{q},\lambda) = \frac{1}{N}\sum_{\vec{q}'}\left|\hat{e}(\vec{q},\lambda)\cdot[\vec{M}_{mm'}(\vec{q}') + \vec{M}_{mm'}(\vec{q}-\vec{q}')]\right|^2 \tag{14b}$$

and

$$I_{mm'}(\omega) = \int_{-\infty}^{\infty}\frac{d\omega'}{\pi}\,g_{2m}(\omega')\,g_{2m'}(\omega-\omega') \tag{14c}$$

or equivalently

$$I_{mm'}(\omega) = \int_0^\infty dt\,\cos\omega t\,G_{2m}(t)\,G_{2m'}(t)\ . \tag{14d}$$

Here, $\vec{M}_{mm'}(\vec{q})$ is related to $\vec{K}_{mm'}(\vec{q})$ via the coordinate transformation discussed above, the $g_{2m'}(\omega)$ are the infinite-temperature spectral functions calculated in Ref. 13 and $G_{2m'}(t)$ are the infinite-temperature correlation functions calculated in Ref. 14. These spectral and correlation functions are therefore known from first principles for the case of solid H_2 for various orthohydrogen concentrations c.

IV. RESULTS IN THE ULTRASONIC MAGNETIC RESONANCE CASE

The formula given by Eqs. (14a)–(14d) is the result for the general acoustic attenuation in a quadrupolar solid. In this section this formula will be evaluated numerically for the special case of the resonant attenuation in ultrasonic magnetic resonance in solid H_2. As was stated earlier, the term "resonant" attenuation is taken to mean that portion of the attenuation which depends upon the external magnetic field $\vec{H}_0$. Although the correlation functions computed in Refs. 13 and 14 were obtained in the absence of a magnetic field, one can easily modify these solutions to include the effect of such a field.[4,16,17] It is shown in Appendix B that the functions calculated in Refs. 13 and 14 can be used directly in the evaluation of the attenuation in the presence of a magnetic field if one shifts the origin of the function $I_{mm'}(\omega)$ from $\omega = 0$ to $\omega = (m+m')\omega_0$, where $\omega_0 = \gamma|\vec{H}_0|$, with γ the gyromagnetic ratio. In other words, if one makes the replacement $\omega \rightarrow \omega - (m+m')\omega_0$ in $I_{mm'}(\omega)$, the field dependence of $\alpha(\vec{q},\lambda,\omega)$ is taken into account. Furthermore, the main interest is now the evaluation of α at frequencies in the neighborhood of $\omega = (m+m')\omega_0$.

Since only acoustic phonons are of interest here, the approximation $\omega(\vec{q},\lambda) = V_\lambda|\vec{q}|$ may be made in Eq. (14a) and it is thus also appropriate to take the small $|\vec{q}|$ limit in that equation. Also, since only resonant terms are of interest, $\omega = \omega(\vec{q},\lambda)$

[thus $|\vec{q}| = \omega(\vec{q}, \lambda)/V_\lambda = \omega/V_\lambda = (m+m')\omega_0/V_\lambda$]. When these facts are taken into account, when only the resonant terms in that equation are kept, and when the replacement $\omega \to (m+m')\omega_0$ is made in $I_{mm'}(\omega)$ as discussed above and in Appendix B, the result is

$$\alpha(\vec{q}, \lambda, \omega) = \frac{\omega^2}{MV_\lambda^3 kT} \sum_{m,m'}{}' \hat{e}(\vec{q}, \lambda) \cdot \overleftrightarrow{F}_{mm'}(\vec{q}, \lambda) \cdot \hat{e}(\vec{q}, \lambda) I_{mm'}[\omega - (m+m')\omega_0] , \qquad (15a)$$

where

$$\overleftrightarrow{F}_{mm'}(\vec{q}, \lambda) = \sum_{nn'\bar{n}\bar{n}'} \Gamma_{nm}\Gamma_{n'm'}\Gamma_{\bar{n}'-m'}\Gamma_{\bar{n}'m'} \times \sum_{\vec{r}}{}' (\hat{q} \cdot \vec{r})^2 \nabla_{\vec{r}} J_{nn'}(\vec{r}) \nabla_{\vec{r}} J_{\bar{n}\bar{n}'}(\vec{r}) . \qquad (15b)$$

Here, the prime on the mm' sum means only resonant terms are kept, $\hat{q}$ is a unit vector in the direction of $\vec{q}$, the prime on the $\vec{r}$ sum means that it goes only over occupied sites, and Γ_{nm} is the coordinate transformation matrix discussed in Appendix B.

The angular dependences for the various phonon modes and resonances which would be obtained from Eqs. (15a) and (15b) are in general very complicated for the hcp lattice of solid H_2. Also, the difficulties of obtaining a single crystal of solid H_2,[23] which is necessary for such phonon modes to have meaning, and the difficulties of growing the crystal so that one can orient the ultrasonic transducer in a specific plane thus allowing a particular phonon mode to be excited, probably preclude an experimental-theoretical comparison of angular dependences anyway. However, progress has been made with regard to the latter point in rare-gas solids.[24] For these reasons, the explicit angular dependences for the various phonon modes and resonances will not be computed. In a polycrystalline sample of solid H_2, one would see an attenuation which is proportional to an angular average of the Γ_{nm} matrices in Eq. (15b). This average is a number of the order unity and will thus be approximated by one in the following discussion, where only an order of magnitude estimate of the maximum resonant attenuation will be considered. Finally, for purposes of this order of magnitude estimate, Eqs. (15a) and (15b) will be specialized to the case of externally applied longitudinal phonons [$\vec{q} \parallel \hat{e}(\vec{q}, \lambda)$] in an ideal sample where the transducer can be oriented in a plane normal to the c axis of the hcp crystal (thus $\hat{c} \parallel \hat{e} \parallel \vec{q} \parallel \hat{z}$). Both approximations discussed in this paragraph will affect the *magnitude* of the maximum resonant attenuation by numbers of the order-of-unity, but will *not* affect the resonant attenuation *line shape*.

Only one more assumption is necessary before order-of-magnitude estimates of the resonant attenuation for solid H_2 may be made from Eqs. (15a) and (15b) and that is an assumption regarding the dependence of those equations on the o-H_2 concentration c. This c dependence will come from two sources; from the dependence of the function $I_{mm'}(\omega)$ on c and from the sum over occupied lattice sites in Eq. (15b). The c dependence of $I_{mm'}(\omega)$ is contingent, through Eqs. (14c) and (14d) on the dependence of the functions $g_{2m}(\omega)$ and $G_{2m}(t)$ on c. Since these functions have been calculated in Refs. 13 and 14 as functions of c, this dependence will automatically be contained in the results of the numerical evaluation of the integrals of Eqs. (14c) and (14d) using the solutions found in those references. For the c dependence of the $\sum_{\vec{r}}'$ occurring in Eq. (15b) the ansatz

$$\sum_{\vec{r}}{}' = h(c) \sum_{\vec{r}} \qquad (16)$$

will be made, where the sum on the right-hand side is to be taken *as if* all of the lattice sites were occupied and $h(c)$ is a function that has the following limiting forms:

$$\lim_{c\to 1} h(c) \to c \qquad (17a)$$

and

$$\lim_{c\to 0} h(c) \to c^{22/3} B , \qquad (17b)$$

where B is a constant which will be taken to be unity here.

Equation (17a) holds universally for large c and has been discussed and justified in Refs. 13 and 25. On the other hand, Eq. (17b) holds *only* for the sum over the particular functions in Eq. (15b) and needs further justification. A detailed discussion of these limiting cases will be deferred to Appendix C. Although, of course, at intermediate values of c, $h(c)$ must realistically interpolate smoothly between Eqs. (17a) and (17b), in the following discussion it will be assumed that Eq. (17a) holds for $0.2 \le c \le 1.0$ and that Eq. (17b) holds for all $c < 0.2$. This arbitrary point at which $h(c)$ changes was chosen to approximately coincide with the point at which the c dependence of other measurable quantities, notably the nuclear spin-lattice relaxation time T_1, changes.[13,14,26,27] Because this choice is somewhat arbitrary, the computed attenuation discussed below will be quoted in terms of $h(c)$ as well as in terms of a definite numerical value. The relations, Eqs. (17a) and (17b), along with the known c dependence of $I_{mm'}(\omega)$, obtained from Refs. 13 and 14 [$I_{mm'}(\omega) \sim c^{-1/2}$ for large c and $I_{mm'}(\omega) \sim c^{-5/3}$ for small c] indicate that the resonant attenuation has the following limiting c dependences:

$$\alpha \sim c^{1/2}, \quad c \to 1 \qquad (18a)$$

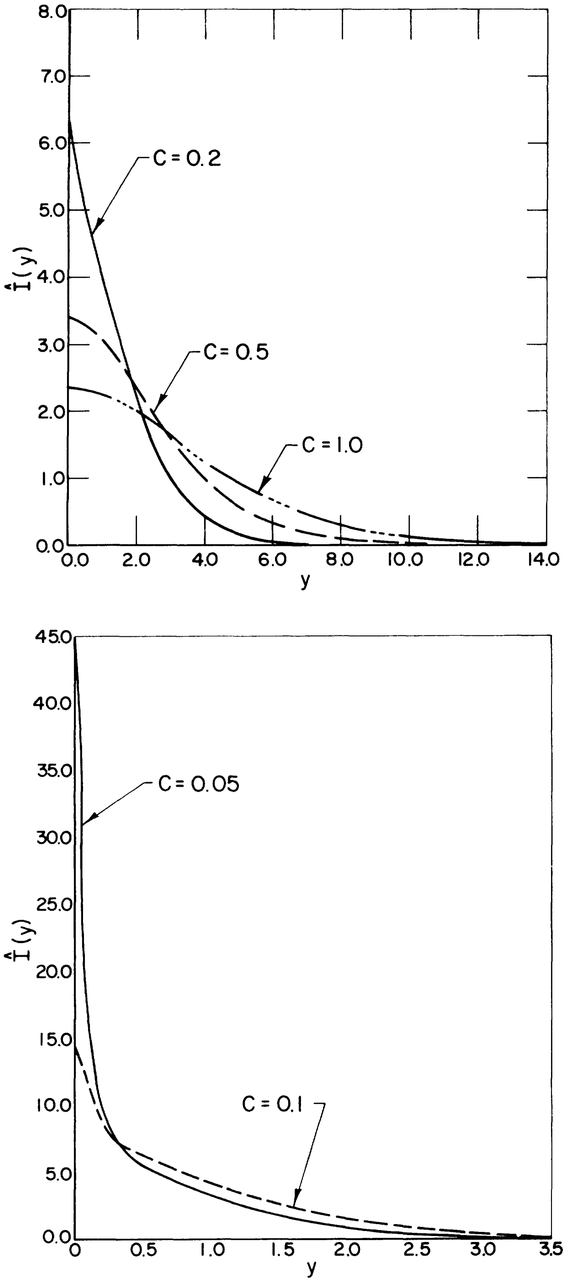

FIG. 1. $\omega = 2\omega_0$ ultrasonic magnetic resonance line shape in solid H_2, where $\hat{I}(y) = I(\omega - 2\omega_0)/(\frac{140}{27}\pi)^{1/2}\Gamma$ and $y = (\omega - 2\omega_0)/(\frac{140}{27}\pi)^{1/2}\Gamma$. (a) $c = 1.0$, 0.5, 0.2 and (b) $c = 0.1$, 0.05.

and

$$\alpha \sim c^{17/3}, \quad c \to 0 . \tag{18b}$$

Thus for small c, α will fall off extremely rapidly with c. Since current ultrasonic techniques can measure an attenuation of the order of 10^{-6} cm^{-1},[12] Eq. (18b) indicates that calculating α for $c \leq 0.05$ is probably not worthwhile.

In light of the above discussion, Eq. (15a) becomes, for ω *near* resonance,

$$\alpha(\omega) = \sum_{m=1}^{4} \alpha_m(\omega) , \tag{19a}$$

where

$$\alpha_4(\omega) = \frac{16\hbar\omega_0^2 h(c)}{MV^3kT} \sum_{\vec{r}} z^2 |\nabla_z J_{22}(\vec{r})|^2 I_{22}(\omega - 4\omega_0) , \tag{19b}$$

$$\alpha_3(\omega) = \frac{4\hbar\omega_0^2 h(c)}{MV^3kT} \sum_{\vec{r}} z^2 |\nabla_z J_{21}(\vec{r})|^2 I_{21}(\omega - 3\omega_0) , \tag{19c}$$

$$\alpha_2(\omega) = \frac{4\hbar\omega_0^2 h(c)}{MV^3kT} \sum_{\vec{r}} z^2 |\nabla_z J_{20}(\vec{r})|^2 \times \left[2I_{20}(\omega - 2\omega_0) + \left(\frac{C_{11}}{C_{20}}\right)^2 I_{11}(\omega - 2\omega_0)\right], \tag{19d}$$

and

$$\alpha_1(\omega) = \frac{2\hbar\omega_0^2 h(c)}{MV^3kT} \sum_{\vec{r}} z^2 |\nabla_z J_{10}(\vec{r})|^2 \times \left[I_{10}(\omega - \omega_0) + \left(\frac{C_{2,-1}}{C_{10}}\right)^2 I_{11}(\omega - \omega_0)\right]. \tag{19e}$$

Here the C_{nm} are the Clebsch-Gordan coefficients from Eq. (2b). The maximum attenuation is obtained from these equations by setting ω equal to the appropriate resonance frequency.

The line shape of the resonant attenuation for $\omega = m\omega_0$, $m = 1, 2, 3, 4$, has been obtained from Eqs. (19b)–(19e) for various concentrations in the range $0.01 \leq c \leq 1.0$ by the use of the functions computed in Refs. 13 and 14. The terminology "line shape" here means the ω dependent part [I_{22} in Eq. (19b), I_{21} in Eq. (19c) and the bracketed quantities in Eqs. (19d) and (19e)]. The results of this calculation are presented for $\omega = 2\omega_0$ in Fig. 1. In that figure the dimensionless variables

$$y = (\omega - 2\omega_0)/(\tfrac{140}{27}\pi)^{1/2}\Gamma , \tag{20a}$$

and

$$\hat{I}(y) = I(\omega - 2\omega_0)/(\tfrac{140}{27}\pi)^{1/2}\Gamma , \tag{20b}$$

$$I(\omega - 2\omega_0) = 2I_{20}(\omega - 2\omega_0) + (C_{11}/C_{20})^2 I_{11}(\omega - 2\omega_0) , \tag{20c}$$

have been used, where Γ is the EQQ coupling constant. The line shapes for the other resonances are, except for a change of scale, essentially the same in shape for a given c and have the same variation with c as the line shapes in Fig. 1. For this reason they will not be shown explicitly. As can be seen from the figure, the acoustic magnetic resonance line shape for c near unity is Gaussian like. As c gets smaller, it begins to get sharper near the center until finally for $c \lesssim 0.1$ the line shape is Lorentzian like. These line shapes have been calculated for c as low as 0.01 with the result that there is virtually no deviation

TABLE I. Resonant attenuation α as a function of c. α is in cm^{-1}, $h(c)$ is discussed in the text.

c	$\alpha(4\omega_0)$	$\alpha(3\omega_0)$	$\alpha(2\omega_0)$	$\alpha(\omega_0)$
1.0	$1.137\times10^{-6}h(c)=1.137\times10^{-6}$	$4.840\times10^{-5}h(c)=4.840\times10^{-5}$	$7.549\times10^{-4}h(c)=7.549\times10^{-4}$	$2.747\times10^{-6}h(c)=2.747\times10^{-6}$
0.5	$1.644\times10^{-6}h(c)=8.218\times10^{-7}$	$7.010\times10^{-5}h(c)=3.505\times10^{-5}$	$1.093\times10^{-3}h(c)=5.466\times10^{-4}$	$3.987\times10^{-6}h(c)=1.993\times10^{-6}$
0.2	$3.022\times10^{-6}h(c)=6.045\times10^{-7}$	$1.293\times10^{-4}h(c)=2.585\times10^{-5}$	$2.014\times10^{-3}h(c)=4.029\times10^{-4}$	$7.424\times10^{-6}h(c)=1.485\times10^{-6}$
0.1	$9.899\times10^{-6}h(c)=4.595\times10^{-13}$	$3.052\times10^{-4}h(c)=1.417\times10^{-11}$	$4.727\times10^{-3}h(c)=2.194\times10^{-10}$	$1.748\times10^{-5}h(c)=8.113\times10^{-13}$
0.05	$2.109\times10^{-5}h(c)=6.069\times10^{-15}$	$9.031\times10^{-4}h(c)=2.599\times10^{-13}$	$1.394\times10^{-2}h(c)=4.011\times10^{-12}$	$5.142\times10^{-5}h(c)=1.480\times10^{-14}$

from the Lorentzian-like shape as c gets very small.

In order to compute the resonant attenuation from Eqs. (19), the lattice sums in those equations have been evaluated numerically for the hcp lattice of solid H_2. The sums were carried out over the first 140 shells of the lattice. This result was then combined with the results of the evaluation of the line shape functions for various c to obtain the maximum resonant attenuation for the various resonances as a function of c. A summary of this calculation is given in Table I. In that table the attenuation is expressed both in terms of the function $h(c)$ and in terms of the result obtained by evaluating it using the limiting values of $h(c)$ given by Eqs. (17a) and (17b) with the change from one form to the other being set arbitrarily at $c=0.2$ as is discussed in the sentences following those equations. In obtaining the numbers in Table I the parameters $V=2.4\times10^5$ cm/sec,[28] $M=3.34\times10^{-24}$ g, $\omega_0=2\pi\times0.6717\ H_0 kHz$,[2] $H_0=10^4$ G, $T=10$ K, and $\Gamma=0.712$ K,[13,14] have been used.

As can be seen from the table, if one uses $10^{-6}\ \text{cm}^{-1}$ as a lower limit on the possibility of detecting the resonance signal,[12] the feasibility of the experimental observation of ultrasonic magnetic resonance in solid H_2 looks very promising for all of the four resonances considered, at least for c not too small. Of those four resonances the resonance at $\omega=2\omega_0$ has the largest attenuation amplitude and thus has the greatest possibility of being detected. Physically, this is easily understood because the particles under consideration have rotational angular momentum $J=1$ and the $\omega=2\omega_0$ resonance corresponds to a $\Delta M_J=2$ transition induced by the phonon modulation of the EQQ interaction. This particular transition is the most likely transition to occur when two $J=1$ particles, coupled via the EQQ interaction are perturbed by phonons. It is clear from Table I, however, that all four resonances have amplitudes such that they might be observable for c near unity.

Of course, as the temperature is lowered below $T=10$ K, the attenuation is enhanced due to the T^{-1} dependence. Thus, at first glance it might appear as if the predicted attenuation for $T<10$ K would be larger than that in Table I. However, this is not necessarily the case since then the high-temperature limit for the attenuation and the infinite-temperature limit for the correlation functions[13,14] become less accurate and a more general temperature dependence for these functions must be taken into account. In particular both the high-temperature approximation for the attenuation and the $T=\infty$ approximation for the correlation functions are only expected to be accurate for $T\gg T_c$, where T_c is the quadrupolar-ordering temperature. T_c for solid H_2 is of the order of 1 K, thus for $T<10$ K, both approximations become questionable.

The attenuation, and thus the numbers in Table I also depend linearly on the EQQ coupling parameter Γ. The value used here $\Gamma=0.712$ K, was chosen to be consistent with the value found to give the best fit to T_1 data in the Erratum of Ref. 13. As is discussed in Refs. 2, 13, 14, and 29, the different experiments obtain values of Γ for solid H_2 which differ by as much as 10%. Thus, due to these uncertainties in Γ, the numbers in Table I have a built in uncertainty of at least 10%. The attenuation for other values of Γ (in °K) can be obtained from those in Table I by multiplying the numbers in that table by $\Gamma/0.712$ K.

Another criterion for the observation of ultrasonic magnetic resonance in solid H_2 is that the resonant attenuation linewidth should be within observable range. These linewidths are essentially the reciprocals of the functions $I_{mm'}(\omega)$ evaluated at $\omega=0$. Using the numbers relevant for solid[1,2] H_2 along with the numerical results for the $I_{mm'}(\omega)$ discussed above, these linewidths are found to be of the order of 10^4 G for $c=1$. Furthermore, they vary as $c^{1/2}$ for c near unity and as $c^{5/3}$ for small c. Unfortunately, a line shape 10^4 G wide is too wide to be measurable with most laboratory fields. On the other hand, the linewidth will get smaller as c does, so that for some intermediate value of c the linewidth and the amplitude as given in Table I both might be observable by ordinary techniques. Also, superconducting magnets do exist which can reach fields of 10^5 G or more, and thus ultrasonic magnetic resonance ought to be observable in solid H_2 for c near unity if one uses such a magnet.

It should also be noted that since the numbers in Table I were calculated for a field of $H_0=10^4$ G, and the resonant attenuation is proportional to $(H_0)^2$, if a larger field were used, those numbers

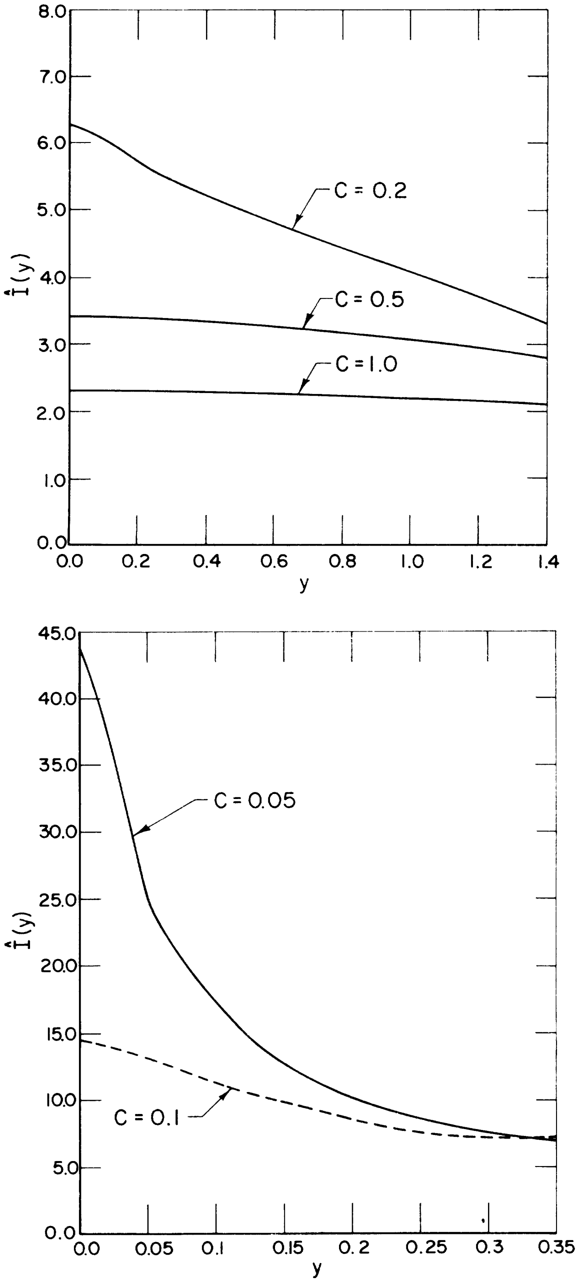

FIG. 2. Same curves as in Fig. 1 except for a limited frequency range near $y=0$. Here, $\hat{I}(y)=I(\omega-2\omega_0)/(\frac{140}{27}\pi)^{1/2}\Gamma$, and $y=(\omega-2\omega_0)/(\frac{140}{27}\pi)^{1/2}\Gamma$. (a) $c=1.0$, 0.5, 0.2 and (b) $c=0.1$, 0.05.

could increase significantly. Thus, a large field is desirable both for obtaining a large amplitude and for being able to measure the linewidth. It is also possible that other effects, such as dipole-dipole interactions,[1,2] might narrow the line sufficiently so that observation would be possible at large c for ordinary magnetic fields.

Alternatively, since the amplitude of the resonant attenuation is clearly observable for some concentrations, even for fields of 10^4 G one could conceive of an experiment with an ordinary laboratory magnet where the field is swept, but the entire line shape is not seen. Rather, in such an experiment, one could visualize measuring only the shape of the curves of Fig. 1 around their peaks. A similar situation is predicted for paramagnets in Ref. 5. These peak areas for $\omega=2\omega_0$ are shown in Fig. 2 for $c=1$, 0.5, 0.2, 0, and 0.05. This figure is really nothing more than a small portion of Fig. 1 near $y=0$ on a scale where the shape on and near the peak can be clearly seen. It is hoped that the predictions of this section might stimulate experimental interest in the observation of these ultrasonic magnetic resonance effects.

In conclusion, in this paper a general expression was derived for the high-temperature ultrasonic attenuation in a quadrupolar solid. This expression is proportional to a four-point angular momentum correlation function. The most crucial (and probably the weakest) assumption of this paper is that this function is well approximated by a sum of products of two-point functions. With this factorization assumption, the attenuation becomes proportional to the Fourier transform of products of two of the angular momentum correlation functions which were calculated from first principles for solid H_2 in Refs. 13 and 14. The results of those references were used to calculate the ultrasonic attenuation for solid H_2 in the special case where it is magnetic field dependent. A new kind of ultrasonic magnetic resonance is thus predicted with the result that this phenomenon might be observable in solid H_2.

Finally, the general, magnetic-field-independent ultrasonic attenuation given by Eq. (14) could have been evaluated using the results of Refs. 13 and 14. This attenuation will have a broad peak as a function of frequency and will attain its maximum value at a frequency of $\nu\sim10^{11}$ Hz, which is roughly two orders of magnitude larger than is obtainable by current ultrasonic techniques. Thus, this general attenuation will not be evaluated here.

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APPENDIX A: COORDINATE TRANSFORMATIONS

It is convenient to define two coordinate systems (one denoted by superscript c) in which the Z axis coincides with the c axis of the hcp lattice, and one (denoted by superscript h) whose Z axis is along the direction of the external magnetic field $\vec{H}_0$. If the field direction is given by the angles (θ, ϕ), the transformation of the spherical tensor operators[17–19] from one system to another takes the form

$$A^h_{2m} = \sum_n \Gamma_{mn}(\theta, \phi) A^c_{2n} , \tag{A1}$$

where $\Gamma_{mn}(\theta, \phi)$ is in general a complicated function of (θ, ϕ). These rotation matrices Γ_{mn} are the ones used in Eq. (15b).

APPENDIX B: MAGNETIC FIELD DEPENDENCE OF THE FUNCTION $I_{mm'}(\omega)$

From Eq. (14c), the definition of $I_{mm'}(\omega)$ is

$$I_{mm'}(\omega) = \int_{-\infty}^{\infty} \frac{d\omega'}{\pi}\, g_{2m}(\omega') g_{2m'}(\omega - \omega') . \tag{B1}$$

In Refs. 4, 16, and 17 it is shown that in the absence of an external magnetic field the spectral functions $g_{2m}(\omega)$ may be written

$$g_{2m}(\omega) = \frac{\Gamma_m(\omega)}{[\omega - \pi_m(\omega)]^2 + [\Gamma_m(\omega)]^2} , \tag{B2}$$

where $\Gamma_m(\omega)$ and $\pi_m(\omega)$ depend on $g_{2m}(\omega)$ self-consistently and are shown explicitly in those references. Furthermore, in the presence of an external field, it is shown in Refs. 4, 16, and 17 that Eq. (B2) is replaced by

$$g_{2m}(\omega) = \frac{\Gamma_m(\omega)}{[\omega - m\omega_0 - \pi_m(\omega)]^2 + [\Gamma_m(\omega)]^2} , \tag{B3}$$

where $\omega_0 = \gamma H_0$, H_0 is the external magnetic field and γ is the gyromagnetic ratio. If Eq. (B3) is substituted into Eq. (B1) and some manipulation is done, it can be seen that the expression for $I_{mm'}(\omega)$ in the presence of an external magnetic field is

$$I_{mm'}(\omega) = \int_{-\infty}^{\infty} \frac{d\omega'}{\pi}\, g_{2m}(\omega') g_{2m'}[\omega - (m + m')\omega_0 - \omega'] . \tag{B4}$$

Thus, the field dependence of $I_{mm'}(\omega)$ is obtained from the field-independent function by the replacement $\omega \to \omega - (m + m')\omega_0$, as claimed in the text.

APPENDIX C: CONCENTRATION DEPENDENCE OF THE LATTICE SUM IN EQ. (15b)

In this Appendix, the limiting cases for $h(c)$ [Eqs. (17a) and (17b)] will be justified. As discussed in Ref. 25, for a random distribution of particles interacting via multipolar forces, let the probability distribution function for the square of the interaction strength F be denoted as $p(F; c)$, where c is the concentration of particles. In general, $p(F; c)$ must be evaluated numerically. However, for $1 - c \ll 1$ and $c \ll 1$, it is possible to find approximate analytic expressions which are exact in the appropriate limit.

For $1 - c \ll 1$, the distribution function may be approximated by[25]

$$p(F; c) = [\pi(1 - c)F_2]^{-1/2} \exp[-(F - cF_1)^2/(1 - c)F_2] , \tag{C1}$$

where $F_1 = \sum_j' g(i,j)$ and $F_2 = \sum_j' g^2(i,j)$, and $g(i,j)$ is proportional to the square of the interaction. Equation (C1) holds in the large c limit, independent of the details of the interaction.[30] For large c, it can thus be seen that the most probable value of the square of the interaction is cF_1. Therefore, if the $\vec{r}_{ij}$ dependent part of the summand in Eq. (15b) is defined to be $g(i,j)$ [the "interaction" is then proportional to $\nabla_{ij} J_{nn'}(i,j)$] the most probable large c value for the function $h(c)$ defined in Eq. (16) is c and Eq. (17a) is thus justified.

On the other hand, for $c \ll 1$, it can be shown[30] that

$$p(F; c) = \frac{n}{c^2b^2}\left(\frac{2n}{3\pi(2n - 3)}\right)^{1/2}\left(\frac{3c^2b^2}{2nF}\right)^{(4n-3)/(4n-6)} \times \exp\left[-\left(1 - \frac{3}{2n}\right)\left(\frac{3c^2b^2}{2nF}\right)^{3/(2n-3)}\right] , \tag{C2}$$

where it has been assumed that the square of the potential $g(i,j)$ is proportional to $(|\vec{r}_{ij}|)^{-2n}$ and b is an n-independent constant. Differentiation of Eq. (C2) with respect to F shows that the most probable value of F occurs at $F = B(c)^{(2n/3)}$, where B is a constant. If the $\vec{r}_{ij}$ part of Eq. (15b) is again defined as $g(i,j)$ and the function $J_{nn'}(i,j)$ is assumed to be of the EQQ form, Eq. (2b), some manipulation will show that $g(i,j)$ is proportional to $|\vec{r}_{ij}|^{-22}$. Therefore, using the above result with Eq. (16) it can be seen that the most probable small c value for the function $h(c)$ is $B(c)^{22/3}$ and Eq. (17b) is thus justified.

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