

Damping of longitudinal waves in colloidal crystals of finite size

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The damping of longitudinal waves in colloidal crystals, considered in the primitive plasmalike model (counterions and macroions in liquid), is investigated in the framework of viscoelastic theory in continuous approximation. The transition from the discrete lattice to the continuous approximation for longitudinal waves is possible, because the wavelength is much larger than the lattice parameter. The dispersion relations, numerical and analytical results for the bulk and surface modes are found for the infinite, semifinite, and finite size crystals. On this basis the experimental results obtained earlier by Hoppenbrouwers and van de Water (unpublished) in finite size crystals are compared with our theoretical results. [S1063-651X(96)08412-7]

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I. INTRODUCTION

Colloidal crystals (CC) are complex systems of particles, consisting of a few components in three phases: macroions in a crystal phase, small ions (counterions) in a plasma phase, and the neutral molecules of a liquid (usually water). The damping of waves in such systems is strong because of the Stokes friction exerted on the macroions and the diffusion of counterions in water. This means that the imaginary part of the complex frequency of the oscillations is at least one order larger than the real part. The lattice dynamics of CC is investigated by means of photon correlation spectroscopy. Hurd *et al.* [1,2] studied crystals in 38 μm -wide thin-film cells. Light scattering experiments by Derksen and van de Water on CC [3] suggest that the dynamics of long-wavelength transverse modes is influenced by the walls bounding the crystal (a series of measurements for a range of thin-film thicknesses $l = 27\text{--}128 \mu\text{m}$). Hoppenbrouwers and van de Water [4] provided experimental evidence for a longitudinal optical mode that is due to the relaxation of the Debye clouds that surround moving macroions. The results of Hurd led Felderhof and Jones [5] to extending the theory of infinite crystals with the effects of the diffusion of counterions (plasma effects). In [3] it was discovered that the size of the crystals plays an important role. Therefore the experiments [1–4] indicate that it is necessary to take into account both the real geometry of CC and plasma effects. In this paper we present a theory in order to account for these wall effects in the low-frequency ($\omega \leq 10^5$ Hz) and long-wavelength limit. Our analysis is based on a continuum description of the dynamics in colloidal systems. The collective excitations in a CC can be modeled either by a dynamical matrix within a lattice dynamics approach or by a differential form following from viscoelastic theory. Obtaining the dispersion curve $\omega(k)$ for a finite CC is a difficult problem. Even for a pure finite crystal phase the eigenfrequencies and vectors of oscillations are unknown. To take into account the boundary conditions we use the continuum model of CC [6]. The transition from the discrete lattice to the continuum model is possible in the long-wavelength approximation (the wavelength λ order $\sim 50\text{--}100 \mu\text{m}$ is much larger than the lattice parameter $R_0 \sim 1 \mu\text{m}$). In the limit R_0/λ tending to

zero the system of dynamic equations for three interacting phases was solved with the appropriate boundary conditions on the surface of a crystal. The dispersion equation was investigated in a plane geometry. The main results of this work are (a) an appearance of a term linear in $k = 2\pi/\lambda$ in the expansion in powers of k of the imaginary part of the eigenfrequency of the longitudinal waves in the case of a half-infinite CC; (b) a modification of the dispersion relation in the case of a thin CC (symmetric and antisymmetric branches); (c) an explanation of the nonzero value of the wave damping for $k \rightarrow 0$ in a finite crystal, which has been observed in the experiment [4]. This model leads to a better understanding of the discrepancies between theory and experiment.

II. THE DYNAMIC SHIELDING OF ELECTRIC FIELDS IN THE COLLOIDAL PLASMA

Longitudinal and transverse waves in CC are excited by the Brownian motion of the macroions. These waves are overdamped due to the friction forces exerted by the solvent fluid. The dynamics of the CC that includes electric and hydrodynamic interactions is described by the equations of motion of charged spherical particles (macroions: radius a , charge $-Ze$, mass M , displacements in harmonic approximation $\mathbf{s}(\mathbf{R}_j; t) = \mathbf{s}_j e^{-it\omega}$), the Navier-Stokes equations for incompressible low-Reynolds-number fluid flow, and the Poisson equation for the electric field $\mathbf{E}(\mathbf{r}\omega)$ with the charge densities ρ_+ (counterions) and ρ_- (macroions). The solvent is assumed structureless with dielectric constant ϵ_0

$$\text{div } \mathbf{E}(\mathbf{r}\omega) = 4\pi \delta \rho_+(\mathbf{r}\omega) - 4\pi \text{div}(\mathbf{P}_0 + \mathbf{P}_-);$$

$$\mathbf{P}_0 = \frac{\epsilon_0 - 1}{4\pi} \mathbf{E}; \quad (1)$$

P_0 , P_- are the dielectric polarization of water and macroions, respectively. Using the Dirac δ function one may write the perturbed charge density of the macroions as

$$\delta\rho_-(\mathbf{r}\omega) = \text{div } \mathbf{P}_- = Ze \sum_j (\mathbf{s}_j \cdot \nabla) \delta(\mathbf{r} - \mathbf{R}_j).$$

The solution of the Poisson equation in $(\mathbf{k}\omega)$ space,

$$\epsilon_0 k^2 \varphi(\mathbf{k}\omega) = 4\pi \delta\rho_+(\mathbf{k}\omega) + 4\pi \delta\rho_-(\mathbf{k}\omega) = 4\pi \frac{\delta\rho_-(\mathbf{k}\omega)}{\epsilon(\mathbf{k}\omega)};$$

$$\frac{1}{\epsilon(\mathbf{k}\omega)} - 1 = \frac{\delta\rho_+(\mathbf{k}\omega)}{\delta\rho_-(\mathbf{k}\omega)} = \frac{\rho_{\text{ind}}}{\rho_{\text{ext}}}$$

is given by

$$\begin{aligned} \varphi(\mathbf{r}\omega) &= \frac{4\pi}{\epsilon_0} \int \frac{d^3k}{(2\pi)^3} e^{i\mathbf{k}\cdot\mathbf{r}} \frac{\delta\rho_-(k\omega)}{\epsilon(\mathbf{k}\omega)k^2} \\ &= -\frac{Ze}{\epsilon_0 \Lambda^2 R_D^2} \sum_j (\mathbf{s}_j \cdot \nabla) \frac{1 - \Lambda^2 R_D^2 - e^{-\Lambda|\mathbf{r}-\mathbf{R}_j|}}{|\mathbf{r}-\mathbf{R}_j|}, \end{aligned} \quad (2)$$

$$\Lambda(\omega) = \frac{1}{R_D} \left(1 - i\omega\tau - \frac{\omega^2}{\omega_p^2} \right)^{1/2},$$

$$\omega_p^2 = \frac{4\pi e^2 n_+^0}{\epsilon_0 m} = \frac{v_+^2}{R_D^2}, \quad \frac{1}{R_D^2} = \frac{4\pi e^2 n_+^0}{\epsilon_0 k_B T},$$

$$v_+^2 = \frac{k_B T}{m}, \quad \frac{1}{\tau} = \frac{\omega_p^2}{\nu}.$$

Here ν is the effective collision frequency between counterions and neutrals of the solvent, R_D , n_+^0 and m are the Debye radius, the equilibrium density, and the mass of the counterions, respectively. In this paper we use the hydrodynamic model dielectric function $\epsilon(\mathbf{k}\omega)$ of the counterions

$$\epsilon(\mathbf{k}\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\nu) - v_+^2 k^2}. \quad (3)$$

The self-field acting on a macroion with center \mathbf{R}_j due to its own distorted Debye cloud is given by

$$\mathbf{E}_j(\omega) = -\frac{Ze}{3\epsilon_0 R_D^3} (1 - \Lambda R_D) \mathbf{s}_j \quad (4)$$

and gives rise to ionic friction. The electric force due to the positions of the other macroions is given by

$$\begin{aligned} \mathbf{F}_j &= -[-Ze \nabla \varphi(\mathbf{R}_j \omega)] - Ze \mathbf{E}_j(\omega) \\ &= -\frac{Z^2 e^2}{\epsilon_0 \Lambda^2 R_D^2} \sum_{n \neq j} (\mathbf{s}_n \cdot \nabla) \nabla \frac{1 - \Lambda^2 R_D^2 - e^{-\Lambda R_{nj}}}{R_{nj}}, \end{aligned} \quad (5)$$

$$R_{nj} = |\mathbf{R}_n - \mathbf{R}_j|.$$

The potential $\varphi(\mathbf{r}\omega)$ obtained above corresponds to the hydrodynamical approximation. On the basis of the Batmager, Gross, and Krook (BGK) model [7] for the collision integral it can be shown that the hydrodynamical model for $\epsilon(\mathbf{k}\omega)$ (3) is always valid for the case $\nu > \omega$ [8], which is of interest for realistic conditions of CC. For the opposite case $\omega > \nu$ the form (3) is satisfactory only far away from the resonance $\omega \cong kv_+$.

Felderhof and Jones [5] introduced the effects of an ionic double layer surrounding each particle moving through the fluid. The potential obtained above tends to the Felderhof-Jones one [5] for $\omega/\omega_p \ll 1$.

III. LONGITUDINAL COLLECTIVE MODES IN THE BULK COLLOIDAL CRYSTAL

The continuum approximation of the dynamic CC is expressed by the equations of motion of a viscoelastic material [9]

$$\begin{aligned} -\rho \omega^2 u_j &= -(\lambda_{jqlm} - i\omega \eta_{jqlm}) k_q k_l u_m + i\omega \frac{\rho}{M} \Gamma_{jq} u_q \\ &\quad - \frac{Ze\rho}{M} E_j, \quad \rho = Mn_-^0 \end{aligned} \quad (6)$$

—and the Poisson equation

$$i\epsilon_0 \epsilon(\mathbf{k}\omega) (\mathbf{k} \cdot \mathbf{E}) = 4\pi \delta\rho_-(\mathbf{k}\omega).$$

Here \mathbf{u} is the deformation profile, λ_{jqlm} is the Lamé tensor of elastic constants, η_{jqlm} is the tensor of the relaxation coefficients (representing the “internal” friction between macroions); the friction matrix Γ_{jq} reflects the “external” friction (between macroions and solvent).

By using the transition from lattice to continuous variables [9]

$$\begin{aligned} \frac{\delta V}{V_0} &= -\frac{\delta n_-}{n_-^0} = \text{div } \mathbf{u}, \quad \delta\rho_- = -Ze \delta n_- = Zen_-^0 \text{ div } \mathbf{u}, \\ V_0 n_-^0 &= V n_- = 1, \end{aligned} \quad (6a)$$

one may write the dispersion equation (A_{sj}^{-1} is the Green's matrix of phonons in the CC)

$$\begin{aligned} |\epsilon_{sj}^b(\mathbf{k}\omega)| &= 0, \quad \epsilon_{sj}^b = \epsilon(\mathbf{k}\omega) \delta_{sj} - \Omega^2 A_{sj}^{-1}, \\ A_{sm} &= \omega^2 \delta_{jm} + i \frac{\omega}{M} \Gamma_{jm} - \frac{1}{\rho} (\lambda_{jqlm} \\ &\quad - i\omega \eta_{jqlm}) k_q k_l, \quad A_{jm}^{-1} A_{ms} = \delta_{js}, \end{aligned} \quad (7)$$

$$\Omega^2 = 4\pi \frac{Z^2 e^2 n_-^0}{M \epsilon_0}, \quad \epsilon_0 \epsilon_{sj}^b(\mathbf{k}\omega) k_s E_j = 0.$$

Colloidal crystals form a bcc lattice with the elastic constants $\lambda_{1111} = C_{11}$, $\lambda_{1122} = C_{12}$, $\lambda_{2323} = C_{44}$. The elastic constants C_i can be expressed through the constants A_i , B_i usually considered for the one-component model in the framework of the next nearest neighbor approximation

$$C_{11} = \frac{2}{R_0} (A_1 + A_2 + B_1/3 + B_2),$$

$$C_{44} = \frac{2}{R_0} (A_1 + B_1/3 + A_2),$$

$$C_{12} = \frac{2}{R_0} (B_1/3 - A_1 - A_2).$$

Longitudinal measurements [3,4] were done in the [110] direction:

$$\mathbf{u}(\mathbf{r}t) = \mathbf{u}_0 e^{i(k/\sqrt{2})(x+y) - i\omega t}, \quad \mathbf{u}_0 = \left(\frac{u_0}{\sqrt{2}}, \frac{u_0}{\sqrt{2}}, 0 \right),$$

$$\mathbf{k} = \left(\frac{k}{\sqrt{2}}, \frac{k}{\sqrt{2}}, 0 \right). \quad (7a)$$

We use simple [110] geometry and the approximations

$$\Gamma_{jm} = \Gamma_0 \delta_{jm}, \quad \eta_{jqlm} = 0,$$

where for Γ_0 there are three approximations: $\Gamma_0/M = \gamma_0 = 6\pi\eta a/M$ (Stokes law; η is the viscosity of the fluid)—without account of hydrodynamic interactions of macroions; $\Gamma_0/M = \gamma_1$ with hydrodynamic interactions in a random medium [10], and $\Gamma_0/M = \gamma_2$ for the discrete model crystal [11]

$$\gamma_1 = \frac{\gamma_0}{3 - \frac{\gamma_0}{\sqrt{2}} \Phi^{1/2}}, \quad \gamma_2 = \frac{\gamma_0}{1 - k_L \Phi^{1/3}}, \quad \Phi = \frac{4}{3} \pi n_0^0 a^3, \quad (8)$$

where Φ is the volume fraction occupied by the macroions. The function k_L weakly depends on k and $k_L = 1.792$ for $k=0$. For the values Φ used in the considered experiments the difference between γ_0 , γ_1 and γ_2 is negligible.

The total friction coefficient of the macroions γ taking into account the self-field effect can be obtained from the equations of motion. The total force of friction $\mathbf{f}(\omega)$ and γ is given by the expressions

$$\mathbf{f}(\omega) = i\omega M \Gamma_0 \mathbf{s} + \frac{Z^2 e^2 (1 - \Lambda R_D)}{3 \epsilon_0 R_D^3} \mathbf{s} = i\omega M \gamma(\omega) \mathbf{s},$$

$$\mathbf{f}(t) = -M \gamma \mathbf{s}(t), \quad (9)$$

$$\gamma(\omega) = \gamma_0 \left(1 + \alpha_0 \frac{1 - \sqrt{1-z}}{z} \right), \quad \alpha_0 = \frac{Z^2 e^2 \tau}{18\pi \epsilon_0 \eta a M R_D^3},$$

$$z = i\omega \tau.$$

The dispersion equation is given by

$$\epsilon_b(\mathbf{k}\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\nu) - v_+^2 k^2} - \frac{\Omega^2}{\omega(\omega + i\gamma) - v_-^2 k^2} = 0,$$

$$v_-^2 = \frac{C_{11} + C_{12} + 2C_{44}}{2\rho}. \quad (10)$$

In the approximation $\Gamma \approx \gamma_0$, $\omega \ll \nu$, γ_0 and $k \rightarrow 0$ there are two modes in Eq. (10): the bulk optical mode ($\sigma = -\text{Im } \omega$)

$$\sigma_1 = \frac{1}{\tau_0} + \frac{\gamma_0^2 \omega_p^2 v_+^2 + \nu^2 \Omega^2 v_-^2}{\nu \gamma_0^2 \omega_p^2 + \gamma_0 \nu^2 \Omega^2} k^2, \quad \frac{1}{\tau_0} = \frac{\omega_p^2}{\nu} + \frac{\Omega^2}{\gamma_0} \quad (10a)$$

and the acoustical mode

$$\sigma_2 = \frac{\omega_p^2 v_-^2 + \Omega^2 v_+^2}{\gamma_0 \omega_p^2 + \nu \Omega^2} k^2. \quad (10b)$$

IV. SURFACE COLLECTIVE MODES IN BOUNDED COLLOIDAL PLASMA FOR OPTICAL MODES

Let us consider a half-space occupied by a two-component plasma. In the case of a semi-infinite plasma and specular reflection of counterions from the boundary between the colloidal plasma and a dielectric medium the dispersion equation for the surface mode has the form

$$1 + \frac{k}{\pi} \int_{-\infty}^{\infty} \frac{dq}{(k^2 + q^2) \epsilon_b(\mathbf{k}, \omega)} = 1 + \frac{1}{\alpha} + \frac{1}{\alpha} B(k, \omega) k = 0;$$

$$B(k, \omega) = \frac{b_{31} b_{32}}{b_{43} \sqrt{b_3 (b_3 k^2 - 1)}} + (3 \rightleftharpoons 4);$$

$$\alpha = 1 - \alpha_1 - \alpha_2; \quad \alpha_1 = \frac{\omega_p^2}{\omega(\omega + i\nu)}; \quad \alpha_2 = \frac{\Omega^2}{\omega(\omega + i\gamma)};$$

$$b_1 = \frac{v_+^2}{\omega(\omega + i\nu)}; \quad b_2 = \frac{v_-^2}{\omega(\omega + i\gamma)}; \quad (11)$$

$$b_{3,4} = -\delta \pm \sqrt{\delta^2 - \frac{b_1 b_2}{\alpha}};$$

$$\delta = \frac{1}{2} (b_1 + b_2 - \alpha_1 b_2 - \alpha_2 b_1); \quad b_{ij} = b_i - b_j;$$

where \mathbf{k} is a three-dimensional wave vector with components $k_z = q$ and \mathbf{k}_{\parallel} with $|k_{\parallel}| = k$, normal and parallel to the surface, respectively, and ϵ_b is the bulk dielectric function (10). Equation (11) gives the surface-plasmon dispersion relation for two-component half-space colloidal plasma (Fig. 1). For $k \rightarrow 0$ the dispersion relation is

$$1 + \alpha(\omega) + B(0, \omega) k + 0(k^2) = 0, \quad 1 + \alpha(\omega_0) = 0, \quad (12)$$

$$\omega(k) = \omega_0 - \frac{B(0, \omega_0)}{\alpha'(\omega_0)} k; \quad \omega_0 \approx -\frac{i}{2} \left(\frac{\omega_p^2}{\nu} + \frac{\Omega^2}{\gamma_0} \right).$$

A similar linear dependence $\omega(k)$ for the case of metallic semi-infinite plasmas has been obtained in [12]. The concrete form of the coefficients in Eq. (12) is, naturally, different from those in [12]. Our result (curve 6 in Fig. 1) is

$$\sigma = C_1 + C_2 k, \quad C_2 < 0.$$

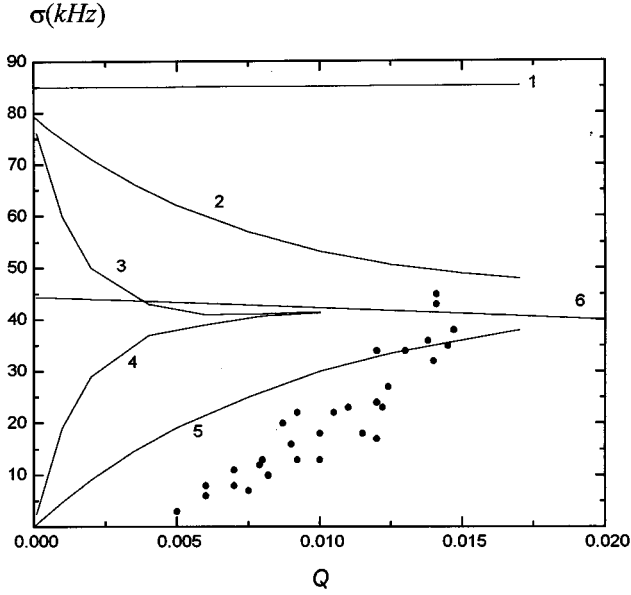


FIG. 1. Theoretically calculated longitudinal collective damping rates on basis of Eqs. (10a), (11), and (13): 1-bulk optical mode; 2,5-thin-crystal geometry, with $l=11 \mu\text{m}$ (symmetric or antisymmetric); 3,4-thickness $l=55 \mu\text{m}$; solid line 6-half-space. The circles-experiment in [4]. Crystal parameters: $R_0=0.75 \times 10^{-4} \text{ cm}$, $Z=250$, $a=0.51 \times 10^{-5} \text{ cm}$; $Q=R_0 k v \sqrt{2}/4\pi$.

Our calculation may be easily extended to deal with thin-crystal geometries (thickness $l=2L$)

$$\int_{-\infty}^{\infty} \frac{dq l}{2\pi} (\dots) \rightarrow \sum_{\substack{m=2n \\ m=2n+1}} (\dots); \quad q \rightarrow \frac{\pi}{l} m.$$

Let us consider a slab of colloidal plasma bounded by the planes $z=\pm L$ and surrounded on both sides by a medium with dielectric constant ϵ_0 (water). We seek solutions of the equations for the electric field that have space oscillations in the plasma, but which decay exponentially in the medium going away from the slab. The boundary conditions lead to the following equations (which can be solved numerically) for the eigenvalues:

$$\begin{aligned} 1 + \frac{k}{L} \sum_{\substack{m=2n \\ m=2n+1}} \frac{1}{\left[k^2 + \left(\frac{\pi m}{l} \right)^2 \right] \epsilon_b} \\ = 1 + \frac{1}{\alpha} \left(\frac{\coth kL}{\tanh kL} \right) + \frac{b_{31} b_{32} k}{\alpha b_{43} \sqrt{b_3 (b_3 k^2 - 1)}} \\ \times \left(\frac{\coth kL \sqrt{1 - \frac{1}{b_3 k^2}}}{\tanh kL \sqrt{1 - \frac{1}{b_3 k^2}}} \right) + (3 \Leftrightarrow 4) = 0. \end{aligned} \quad (13)$$

The upper (lower) equation, which includes \coth (\tanh), was obtained when the sum runs over all even (odd) integers m . The dispersion relation (13) is a natural generalization for

finite L of the result (11). A similar result has been obtained for one-component metal plasmas in Ref. [13]. In the limit $k \rightarrow 0$ ($kL \sim 1$) we find the relations for the roots of equation (13)

$$\begin{aligned} \omega_{\pm}(k) = \omega_{\pm}^0 - \frac{B(0, \omega_{\pm}^0)}{\alpha'(\omega_{\pm}^0)} (1 \pm e^{-2kL}) k; \\ 1 + \alpha(\omega_{\pm}^0) \pm 2e^{-2kL} = 0; \end{aligned} \quad (14)$$

$$\omega_{\pm}^0 \approx -\frac{i}{2} \left(\frac{\omega_p^2}{\nu} + \frac{\Omega^2}{\gamma_0} \right) (1 \mp e^{-2kL}).$$

The interference effects, which include the influence of the second surface, lead to a modification of the surface-wave dispersion relation. Surface modes in thin colloidal system are symmetric or antisymmetric character with respect to the plane $z=0$. It should be noted that in the used approximation ($\omega \ll \nu$, γ_0 and $k \rightarrow 0$) Eq. (10) coincides with the results of kinetic considerations.

V. ANALYTICAL RESULTS FOR THE ACOUSTICAL MODES

Now let us consider the analytical results for the acoustic mode in a colloidal film for parameters of the system such that

$$\Omega^2 v_+^2 > \omega_p^2 v_-^2, \quad \gamma_0 \omega_p^2 > \nu \Omega^2.$$

These conditions have been realized particularly in the experiments [4] ($v_-^2 \sim 2.5 \times 10^3 \text{ cm}^2/\text{sec}$, $v_+^2 \sim 7 \times 10^{10} \text{ cm}^2/\text{sec}$, $\Omega^2 \sim 2 \times 10^{13} \text{ sec}^{-2}$, $\omega_p^2 \sim 3 \times 10^{18} \text{ sec}^{-2}$, $L \sim 10^{-2} \text{ cm}$, $\gamma_0 \sim 2 \times 10^9 \text{ sec}^{-1}$, $\nu \sim 10^{13} \text{ sec}^{-1}$). As is easy to see the approximate dielectric function for the description of such a mode in an infinite system can be taken to be

$$\epsilon_b(\mathbf{k}\omega) = 1 + \frac{\omega_p^2}{v_+^2 k^2} - \frac{\Omega^2}{i\omega \gamma_0} = 0. \quad (15)$$

From Eq. (13) we can find two dispersion relations [for the odd and even summation in Eq. (10), respectively],

$$\epsilon_{\text{odd}} = 1 + \frac{k}{1 - \frac{\Omega^2}{\gamma_0 \sigma}} \frac{1}{d} \tanh(Ld) = 0; \quad (16)$$

$$\epsilon_{\text{even}} = 1 + \frac{2k}{1 - \frac{\Omega^2}{\gamma_0 \sigma}} \frac{1}{d} \left\{ \coth(Ld) - \frac{1}{2} \tanh(Ld) - \frac{1}{2Ld} \right\} = 0, \quad (17)$$

$$d = \left[k^2 + \frac{\omega_p^2}{v_+^2 (1 - \Omega^2 / \sigma \gamma_0)} \right]^{1/2}. \quad (18)$$

From Eq. (16) we obtain the solutions only in the region $\sigma < \Omega^2 / \gamma$.

For $Ld \gg 1$ we have

$$\sigma = \frac{\Omega^2}{\gamma_0 \left\{ 1 + \frac{\omega_p^2}{2k^2 v_+^2} \left[1 + \left(1 + 4 \frac{k^4 v_+^4}{\omega_p^4} \right)^{1/2} \right] \right\}}. \quad (19)$$

This surface mode is valid for values of k such that

$$\frac{2kLk^2 v_+^2}{\omega_p^2 \left\{ 1 + \left(1 + 4 \frac{k^4 v_+^4}{\omega_p^4} \right)^{1/2} \right\}} \gg 1. \quad (20)$$

This inequality is always valid for $L \rightarrow \infty$, of course.

For $Ld \ll 1$ we obtain

$$\sigma = \frac{\Omega^2}{\gamma_0(1+kL)}. \quad (21)$$

This solution is valid for

$$kL > \frac{\omega_p^2}{k^2 v_+^2}, \quad kL \left(kL - \frac{\omega_p^2}{k^2 v_+^2} \right) \ll 1. \quad (22)$$

These solutions were considered for the case $d^2 > 0$. For the case $d^2 < 0$ we find for small k

$$\sigma = \frac{\Omega^2}{\gamma_0 \left(1 + \frac{\omega_p^2}{v_+^2 k_n^2} \right)},$$

$$k_n^2 = k^2 + \left(\frac{\pi}{2L} + \frac{\pi n}{L} \right)^2 = k^2 + \frac{\pi^2}{4L^2} (2n+1)^2. \quad (23)$$

For large L and $k \rightarrow 0$ the solution (23) reduces to a particular case of the volume acoustic mode (10b)

$$\sigma = \frac{\Omega^2 v_+^2}{\gamma_0 \omega_p^2} k_n^2,$$

for the conditions $\Omega^2 v_+^2 > \omega_p^2 v_+^2$ and $\gamma_0 \omega_p^2 > \nu \Omega^2$.

As follows from Eq. (23) for finite L there is a finite value σ for $k \rightarrow 0$

$$\sigma = \frac{\Omega^2}{\gamma_0 \left(1 + \frac{4L\omega_p^2}{\pi^2(2n+1)^2 v_+^2} \right)} < \frac{\Omega^2}{\gamma_0}. \quad (24)$$

The condition for the existence of these modes is

$$\frac{kLk_n^2 v_+^2}{\omega_p^2 \left(\frac{\pi}{2} + \pi n \right)} \ll 1. \quad (25)$$

For $n=0$ (the mode with minimal damping) the solution (24) for thicknesses of the colloidal crystals $l_1=55 \mu\text{m}$ and $l_2=11 \mu\text{m}$, gives the values $\sigma_1(k=0)=3.5 \text{ Hz}$ and $\sigma_2(k=0)=90 \text{ Hz}$, respectively. As follows from Fig. 2 the

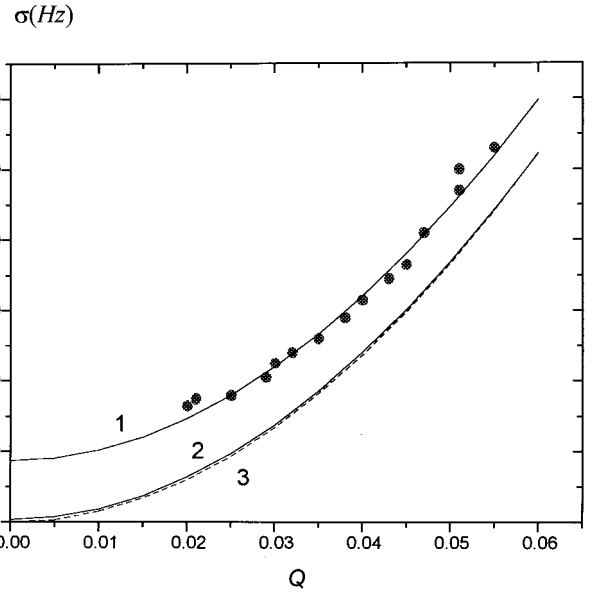


FIG. 2. Theoretically calculated longitudinal collective damping rates on basis of Eqs. (10b) and (23): 1-thin-crystal with $l=11 \mu\text{m}$; 2-thickness $l=55 \mu\text{m}$; dashed line 3-bulk acoustical mode. The circles-experiment [4]. Crystal parameters: $R_0=0.75 \times 10^{-4}$ cm, $Z=250$, $a=0.51 \times 10^{-5}$ cm; $Q=R_0 k v \sqrt{2}/4\pi$.

value $l_2=11 \mu\text{m}$ is a good one for the fitting of the entire experimental curve obtained from the light scattering experiments [4], in which the crystal thickness was not determined exactly. Now let us consider Eq. (17). In the case of $d^2 > 0$ and $Ld \gg 1$ the result is equivalent to Eq. (16). In the case $d^2 > 0$ and $Ld \ll 1$ we have

$$\sigma = \frac{\Omega^2}{\gamma_0 \left(1 + \frac{1}{kL} + \frac{\omega_p^2}{k^2 v_+^2} \right)} \quad (26)$$

and the limitation

$$\frac{k^2 L^2}{1 + \frac{\omega_p^2}{k^2 v_+^2} kL} \ll 1. \quad (27)$$

For the case $d^2 < 0$ and $k \rightarrow 0$ we find the solution from one of two conditions

$$\sin Ld' = 0, \quad \cos Ld' = 0, \quad d'^2 = -d^2 > 0. \quad (28)$$

The second of these equations corresponds to the consideration above. The first of them gives a similar spectrum which also leads to the volume acoustic mode.

VI. CONCLUSIONS

A system of dynamical equations for colloidal crystals in the continuum approximation is investigated and the results for the wave damping of the longitudinal modes for infinite and finite CC, obtained in [14], are systematically derived and analyzed. Collective modes in CC-bulk optical and

acoustic are found from this theory. The surface effects are taken into account. We find and analyze the dispersion relations for finite and half-infinite CC numerically and analytically. A comparison with the acoustical longitudinal mode found in the experiments in [4] is made.

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