

## Quantum electrodynamics in the presence of dielectrics and conductors. II. Theory of dispersion forces

G. S. Agarwal

*Tata Institute of Fundamental Research, Homi Bhabha Road, Bombay-5, India*

(Received 25 April 1974)

The different kinds of response functions introduced in a previous paper are used to calculate the dispersion forces. An exact expression for the interaction energy in a system of harmonic oscillators interacting with a second-quantized radiation field is obtained in terms of appropriate response functions. The radiation field may be either the field appropriate to entire free space or the field altered by the presence of the dielectric. The result is valid for arbitrary geometries involving dielectric and conducting surfaces. An expansion of our result in powers of  $e^2$  leads to the results of other authors. The calculation of the dispersion force for the case of excited states is also briefly discussed. Next the problem of the dispersion force between macroscopic bodies is considered. Lifshitz's expression for the dispersion force is rederived using the response functions and the two methods are compared. The role of surface modes in the determination of dispersion force is discussed. Finally the dispersion force between a spatially dispersive and spatially nondispersive dielectric is calculated exactly. When the spatial dispersion is weak, then it is found that the first-order term is repulsive in nature, in contrast to the zeroth-order term which is attractive. As a by-product of our analysis, surface-polariton dispersion relations are obtained.

### I. INTRODUCTION

In Paper I of this series of papers<sup>1</sup> I introduced various kinds of response functions and used them to study black-body fluctuations. In the present paper I show how these can be used in the calculation of dispersion forces in a system of many atoms as well as between macroscopic bodies. The dispersion forces between two atoms were originally calculated by London,<sup>2</sup> and later by Casimir and Polder,<sup>3</sup> taking fully into account the retardation effects. Aub and Zienau<sup>4</sup> carried out the calculation to order  $e^6$ . These methods used straightforward perturbation theory. Over the last decade, a number of other methods have been developed, which are basically of three types: (i) a method based on the S-matrix approach,<sup>5</sup> (ii) one based on calculating the normal modes of the coupled atom-field system and then summing over the energies of these modes to obtain the interaction energy,<sup>6,7</sup> and (iii) one based on a version of linear-response theory.<sup>8</sup> Of these, the method of Mclachlan appears to be specially attractive although he carried out calculations to various orders in  $e^2$ . In what follows linear-response theory will be used to obtain a closed-form expression valid to all orders in  $e^2$  for the interaction energy in a system of harmonic oscillators. Our result agrees with the result of Renne. Moreover, an expansion of our result to various orders in  $e^2$  leads to the results of Casimir and Polder, Aub and Zienau, and Mclachlan.

We next discuss the problem of Van der Waals

forces between macroscopic bodies, the calculation of which is much more involved. Lifshitz<sup>9,10</sup> introduced the idea of a fluctuating electric and magnetic currents in Maxwell equations. The correlation functions of the electromagnetic field variables can be calculated in terms of the correlation functions of the fluctuating current, and from the knowledge of the correlation functions the stress tensor was computed. Van Kampen *et al.*<sup>11</sup> calculated the interaction energy between macroscopic bodies by summing over the energies of the normal modes of the system. These normal modes were calculated using Maxwell equations. In a dielectric, which is finite in extent, one has two types of modes: (i) bulk modes and (ii) surface modes. The bulk modes are independent of the geometry whereas surface modes depend on shape of the dielectric. It can be shown easily that only surface modes contribute to the dispersion force. The modes of the dielectric are also known as the polariton modes, which are essentially coupled photon-material excitation modes. The method of Van Kampen *et al.* has received a good deal of attention<sup>12</sup>; however, if the damping of the dielectric function is included, then the method seems to fail. In presence of damping the normal-mode frequencies are complex, and it is not clear what one should sum over to obtain the interaction energy. Other problems associated with this method when retardation effects are included are discussed in Ref. 13.

In the present article I show how the response functions can be used in the calculation of Van der

Waals forces between macroscopic bodies. In a brief communication<sup>14</sup> I have discussed surface response functions and showed their use in the calculation of dispersion forces when retardation effects are unimportant. The outline of the present paper is as follows. I first derive an important relation between the equilibrium fluctuations and the response functions at purely imaginary frequencies. In Sec. II, I consider a system of harmonic oscillators. I start with a microscopic Hamiltonian and calculate the linear response function valid to all orders in the coupling constant between the field and matter oscillator. The fluctuation-dissipation theorem then leads to the interaction energy. The result is valid for any geometrical arrangement involving dielectric and conducting surfaces. The dispersion force is calculated in a system of harmonic oscillators. In Sec. III, I calculate using the result of Sec. II the dispersion force between an atom and a dielectric. The results for the case of a conductor follow as a special case. I also point out some of the problems associated with the normal-mode method. The results at each stage are compared with those of other authors. I also consider the dispersion force for the case of excited states. In Sec. IV, I use the response functions calculated in Paper I to obtain Lifshitz's formula for the dispersion force. A brief comparison of our method with Lifshitz's method is also given. In Sec. V, I calculate the dispersion force between a spatially dispersive and a spatially nondispersive body. The role of surface modes is clearly displayed in our method. For simplicity I treat only the case of zero temperature, although the method is equally applicable to finite temperatures.

Let us now discuss the relation between the equilibrium correlation  $S_{ij}(\vec{r}, \vec{r}', 0)$  and the response function at imaginary frequency. The response function  $\chi_{ij}(\vec{r}, \vec{r}', z)$  for  $\text{Im}z \geq 0$  is related to  $\chi''_{ij}(\vec{r}, \vec{r}', \omega)$  by

$$\chi_{ij}(\vec{r}, \vec{r}', z) = \frac{1}{\pi} \int_{-\infty}^{+\infty} d\omega (\omega - z)^{-1} \chi''_{ij}(\vec{r}, \vec{r}', \omega). \quad (1.1)$$

As mentioned in Paper I, the response function  $\chi''_{ij}$ , for the case when the variables  $A_i$  and  $A_j$  have the same parity under time reversal, has the symmetry property

$$\chi''_{ij}(\vec{r}, \vec{r}', \omega) = \chi''_{ji}(\vec{r}', \vec{r}, \omega) = -\chi''_{ij}(\vec{r}, \vec{r}', -\omega). \quad (1.2)$$

On using (1.1) and (1.2), it is easily shown that

$$\int_0^{\infty} \chi_{ij}(\vec{r}, \vec{r}', i\omega) d\omega = \int_0^{\infty} \chi''_{ij}(\vec{r}, \vec{r}', \omega) d\omega. \quad (1.3)$$

On using fluctuation dissipation theorem, Eq. (I.2.10), we find that the equilibrium correlation

function at zero temperature is given by

$$\begin{aligned} S_{ij}(\vec{r}, \vec{r}', 0) &= (1/2\pi) \int_{-\infty}^{+\infty} d\omega S_{ij}(\vec{r}, \vec{r}', \omega) \\ &= (\hbar/2\pi) \int_{-\infty}^{+\infty} d\omega \coth(\beta\omega\hbar/2) \chi''_{ij}(\vec{r}, \vec{r}', \omega) \\ &= (\hbar/2\pi) \int_0^{\infty} d\omega [\chi''_{ij}(\vec{r}, \vec{r}', \omega) - \chi''_{ij}(\vec{r}, \vec{r}', -\omega)], \\ &\qquad\qquad\qquad \beta = \infty, \end{aligned}$$

which on using (1.2) becomes

$$S_{ij}(\vec{r}, \vec{r}', 0) = (\hbar/\pi) \int_0^{\infty} d\omega \chi''_{ij}(\vec{r}, \vec{r}', \omega),$$

and then on using (1.3) we have the important relation

$$S_{ij}(\vec{r}, \vec{r}', 0) = (\hbar/\pi) \int_0^{\infty} d\omega \chi_{ij}(\vec{r}, \vec{r}', i\omega), \quad (1.4)$$

which has been obtained under the assumption that  $A_i$  and  $A_j$  have the same signature under time reversal.

## II. INTERACTION ENERGY BETWEEN A SYSTEM OF HARMONIC OSCILLATORS AND THE ELECTROMAGNETIC FIELD

Before we calculate the dispersion forces in a system of harmonic oscillators and the atom and a dielectric, we present the result for the mean value of interaction energy in terms of the response functions. The interaction Hamiltonian for a system of identical oscillators and the radiation field is

$$H_1 = - \int \vec{P}(\vec{r}) \cdot \vec{E}(\vec{r}) d^3r, \quad (2.1)$$

where  $\vec{E}(\vec{r})$  is the second-quantized electric field operator and where  $\vec{P}(\vec{r})$  is the polarization operator,

$$\begin{aligned} \vec{P}(\vec{r}) &= \sum_i (a_i + a_i^\dagger) \delta(\vec{r} - \vec{r}_i) \vec{d} \\ &\equiv \sum_i \vec{d} p_i \delta(\vec{r} - \vec{r}_i). \end{aligned} \quad (2.2)$$

Here  $\vec{d}$  is the dipole moment matrix element and  $a$  and  $a^\dagger$  are the annihilation and creation operators satisfying the usual boson commutation relations. We assume that the system is further driven by an external electromagnetic field giving rise to a perturbation Hamiltonian

$$H_{\text{ext}} = - \int \vec{P}(\vec{r}) \cdot \vec{E}(\vec{r}, t) d^3r. \quad (2.3)$$

It is easily seen, from (2.1), (2.3), and the fact that  $a$  and  $a^\dagger$  are boson operators, that  $p_i$  satisfies the equation

$$\ddot{p}_i + \omega_0^2 p_i = 2\omega_0 \vec{d} \cdot [\vec{E}(\vec{r}_i, t) + \vec{G}(\vec{r}_i, t)]. \quad (2.4)$$

We will put  $\hbar = 1$  throughout this section. The electric field operator obeys the Heisenberg equation of motion and hence its time dependence is given by ( $t_0$  being the time when Schrödinger and Heisenberg pictures coincide)

$$\vec{E}(\vec{r}, t) = e^{i\mathcal{L}(t-t_0)} \vec{E}(\vec{r}, t_0), \quad (2.5)$$

where  $\mathcal{L}$  is the Liouville operator,

$$\mathcal{L} = [H, \dots], \quad H = H_0 + H_1 + H_{\text{ext}}. \quad (2.6)$$

We now use the identity<sup>15</sup>

$$e^{i\mathcal{L}(t-t_0)} = e^{i\mathcal{L}_0(t-t_0)} + \int_{t_0}^t e^{i\mathcal{L}_0(t-\tau)} i(\mathcal{L}_1 + \mathcal{L}_{\text{ext}}) e^{i\mathcal{L}_0(\tau-t_0)} d\tau \quad (2.7)$$

to rewrite (2.5) in the form

$$\begin{aligned} \vec{E}(\vec{r}, t) = & \vec{E}_0(\vec{r}, t) - i \int_{t_0}^t d\tau e^{i\mathcal{L}_0(t-\tau)} \\ & \times \left[ \int d^3r' \vec{P}(\vec{r}') \cdot \vec{E}(\vec{r}', \tau), \vec{E}_0(\vec{r}, t - \tau + t_0) \right], \end{aligned} \quad (2.8)$$

where  $\vec{E}_0$  is the field operator evolving according to the unperturbed Hamiltonian, i.e.,

$$\vec{E}_0(\vec{r}, t) = e^{i\mathcal{L}_0(t-t_0)} \vec{E}(\vec{r}, t_0). \quad (2.9)$$

Now if we use the fact that the commutator of the free field operators at two different space-time points is a  $c$  number, then (2.8) reduces to

$$\begin{aligned} \vec{E}(\vec{r}, t) = & \vec{E}_0(\vec{r}, t) + i \int_{t_0}^t \int d^3r' d\tau \\ & \times \sum_j [\vec{E}_0(\vec{r}, t - \tau), \vec{E}_{0j}(\vec{r}', 0)] \vec{P}_j(\vec{r}', \tau). \end{aligned} \quad (2.10)$$

In particular we have from (2.10)

$$\begin{aligned} \vec{d} \cdot \vec{E}(\vec{r}, t) = & \vec{d} \cdot \vec{E}_0(\vec{r}, t) \\ & + i \int_{t_0}^t \sum_j D(\vec{r}, \vec{r}_j, t - \tau) p_j(\tau) d\tau, \end{aligned} \quad (2.11)$$

where  $D$  is the free field propagator<sup>16</sup>

$$D(\vec{r}, \vec{r}', \tau) = [\vec{d} \cdot \vec{E}_0(\vec{r}, \tau), \vec{d} \cdot \vec{E}_0(\vec{r}', 0)]. \quad (2.12)$$

On taking the Fourier transforms and letting  $t_0 \rightarrow -\infty$  we obtain, from (2.4) and (2.11), the equations

$$\frac{\delta \langle \vec{d} \cdot \vec{E}(\vec{r}, \omega) \rangle}{\delta \mathcal{G}_j(\vec{r}', \omega)} = i \sum \mathcal{D}(\vec{r}, \vec{r}_j, \omega) \frac{\delta \langle p_j \rangle}{\delta \mathcal{G}_j(\vec{r}', \omega)}, \quad (2.13)$$

$$\begin{aligned} (-\omega^2 + \omega_0^2) \langle p_i(\omega) \rangle = & 2i\omega_0 \sum_j \mathcal{D}_{ij}(\omega) \langle p_j(\omega) \rangle \\ & + 2\omega_0 \vec{d} \cdot \vec{G}(\vec{r}_i, \omega), \end{aligned} \quad (2.14)$$

$$\mathcal{D}_{ij}(\omega) \equiv \mathcal{D}(\vec{r}_i, \vec{r}_j, \omega) \equiv \int_0^\infty d\tau D(\vec{r}_i, \vec{r}_j, \tau) e^{i\omega\tau}.$$

On inverting (2.14) we have

$$\langle p_i(\omega) \rangle = \sum_j 2\omega_0 \mathfrak{M}_{ij}^{-1} \vec{d} \cdot \vec{G}(\vec{r}_j, \omega),$$

or

$$\frac{\delta \langle p_i(\omega) \rangle}{\delta \vec{d} \cdot \vec{G}(\vec{r}_j, \omega)} = 2\omega_0 \mathfrak{M}_{ij}^{-1}, \quad (2.15)$$

where the matrix  $\mathfrak{M}$  is given by

$$\mathfrak{M}_{ij} = (-\omega^2 + \omega_0^2) \delta_{ij} - 2i\omega_0 \mathcal{D}_{ij}(\omega). \quad (2.16)$$

On combining (2.13) and (2.15), we obtain

$$\frac{\delta \langle \vec{d} \cdot \vec{E}(\vec{r}_i, \omega) \rangle}{\delta \vec{d} \cdot \vec{G}(\vec{r}_j, \omega)} = 2i\omega_0 \sum_l \mathcal{D}_{il}(\omega) \mathfrak{M}_{lj}^{-1}. \quad (2.17)$$

The mean value of  $H_1$

$$\langle H_1 \rangle = - \sum_i \langle p_i \vec{d} \cdot \vec{E}(\vec{r}_i) \rangle$$

is, on using (2.17) and the relation (1.4), given by

$$\langle H_1 \rangle = \frac{1}{\pi} \int_0^\infty d\nu \sum_{ij} [-2i\omega_0 \mathcal{D}_{ij}(\omega) \mathfrak{M}_{ij}^{-1}]_{\omega=i\nu}. \quad (2.18)$$

We recall that  $p_i$  and  $\vec{E}$  have the same parity under time reversal. It should be noted that  $\mathcal{D}$  is proportional to  $e^2$  (second order in the coupling constant). The total energy is obtained in the usual manner<sup>17</sup> by integrating over  $e^2$ , i.e.,

$$\begin{aligned} \delta E = \langle H \rangle - \langle H_0 \rangle \\ = \frac{1}{2\pi} \int_0^\infty d\nu \int_0^{e^2} \frac{de^2}{e^2} \text{Tr} \{ -2i\omega_0 \mathcal{D}(\omega) \\ \times [-\omega^2 + \omega_0^2 - 2i\omega_0 \mathcal{D}(\omega)]^{-1} \}_{\omega=i\nu}, \end{aligned} \quad (2.19)$$

where  $\langle H_0 \rangle$  is the ground-state energy in the absence of any interaction between the radiation field and matter. On integrating over  $e^2$ , we obtain

$$\delta E = \frac{1}{2\pi} \int_0^\infty dx \text{Tr} \ln [1 - 2i\omega_0 \alpha_0(i x) \mathcal{D}(i x)], \quad (2.20)$$

where  $\alpha_0$  is proportional to the bare polarizability

$$\alpha_0(\omega) = (\omega_0^2 - \omega^2)^{-1}. \quad (2.21)$$

An expansion of (2.20) in powers of  $e^2$  gives

$$\delta E = - \frac{1}{2\pi} \int_0^\infty dx \sum_{n=1}^\infty \frac{1}{n} \text{Tr} [2i\omega_0 \alpha_0(i x) \mathcal{D}(i x)]^n. \quad (2.22)$$

Equation (2.20) is our final expression for the interaction energy in a system of harmonic oscillators. The field propagator which appears in (2.20) is related to the response function. Let  $\chi_{ij}(\vec{r}, \vec{r}', \omega)$  be, as before, the response of the electric field to an applied polarization  $\mathcal{P}$

$$\chi_{iEE}(\vec{r}, \vec{r}', \omega) = \delta \langle E_i(\vec{r}, \omega) \rangle / \delta \mathcal{P}_j(\vec{r}', \omega), \quad (2.23)$$

then from Eq. (I.2.4) we have

$$\mathfrak{D}(\vec{r}, \vec{r}', \omega) = -i \sum_{ij} d_i d_j \chi_{ijEE}(\vec{r}, \vec{r}', \omega), \quad (2.24)$$

and hence

$$\delta E = \frac{1}{2\pi} \int_0^\infty dx \operatorname{Tr} \ln \left( 1 - 2\omega_0 \alpha_0(ix) \times \sum_{ij} d_i d_j \chi_{ijEE}(ix) \right). \quad (2.25)$$

The dispersion force is now obtained by substituting into (2.25) the appropriate response function. We first consider the case where there are no surfaces; then  $\chi_{ijEE}(\vec{r}, \vec{r}', \omega)$  is translationally invariant and is given by (I.4.7), i.e.,

$$\chi_{ijEE}(\vec{r}, \vec{r}', \omega) = \left( \frac{\omega^2}{c^2} \delta_{ij} + \frac{\partial^2}{\partial x_i \partial x_j} \right) \frac{e^{i(\omega/c)|\vec{r}-\vec{r}'|}}{|\vec{r}-\vec{r}'|}. \quad (2.26)$$

In this case (2.20) can also be written, on introducing the renormalized polarizability

$$\alpha(\omega) = [\omega_0^2 - \omega^2 - 2i\omega_0 \mathfrak{D}(\vec{r}_i, \vec{r}_i, \omega)]^{-1}, \quad (2.27)$$

as

$$\delta E = \delta E_0 + \frac{\hbar}{2\pi} \int_0^\infty dx \operatorname{Tr} \ln [1 - 2i\omega_0 \alpha(ix) \mathfrak{D}^{(0)}(ix)] \quad (2.28)$$

$$= \delta E_0 - \frac{1}{2\pi} \int_0^\infty dx \sum_1^n \frac{1}{n} \operatorname{Tr} [2i\omega_0 \alpha(ix) \mathfrak{D}^{(0)}(ix)]^n, \quad (2.29)$$

where

$$\delta E_0 = \frac{N}{2\pi} \int_0^\infty dx \ln [1 - 2i\omega_0 \alpha_0(ix) \mathfrak{D}_{ii}(ix)], \quad (2.30)$$

$$\mathfrak{D}_{ij}^{(0)}(ix) = (1 - \delta_{ij}) \mathfrak{D}_{ij}(ix), \quad (2.31)$$

and  $N$  is the number of oscillators. The term  $\delta E_0$  corresponds to self-interactions; i.e., it is the one-particle term. It should be noted that  $\delta E_0$  is not the entire one-body contribution—to this one should add the contribution coming from the contact term<sup>18</sup>  $2\pi \int |\vec{P}|^2 d^3r$ . The terms corresponding to  $n=2, 3, \dots$ , etc. yield respectively the two-particle, three-particle, ... contributions. The result (2.29) coincides with what Renne<sup>6</sup> obtained by using normal-mode method except for the presence of *one-body terms*  $\delta E_0$ . The results are also in agreement with those obtained by Casimir and Polder,<sup>3</sup> and Aub and Zienau<sup>4</sup> except that the bare polarizability of the oscillator is replaced by  $\alpha(\omega)$ , which contains the effects of radiation damping. The present derivation differs from that of McLachlan<sup>8</sup> in that we produced an exact expression for the interaction energy. McLachlan calculates the dispersion force to each order in  $e^2$ .

### III. DISPERSION FORCE BETWEEN AN ATOM AND A DIELECTRIC

In this section we consider the dispersion force between an atom and the dielectric. We treat the atom as an oscillator. The dispersion force between an oscillator and a perfect conductor was calculated to order  $e^2$ , by Casimir and Polder<sup>3</sup> using a straightforward perturbation theory. Renne<sup>6</sup> has calculated the force to all orders in  $e^2$  by summing over the energies of the normal modes of the system comprising the oscillator, conductor, and radiation field. The effect of the conductor was replaced by an image dipole. The problem of dielectric is more involved than the conductor case. It is not quite clear how to apply, say, the method of normal modes to calculate the dispersion force between the atom and dielectric, since with retardation included it is not at all obvious what to take, for example, for the image dipole. We have seen (Sec. II) how the linear-response theory leads to an exact expression for the dispersion force. For the case of a single oscillator located at  $\vec{r} = \vec{b}$ , (2.25) reduces to

$$\delta E = \frac{1}{2\pi} \int_0^\infty dx \ln \left( 1 - 2\omega_0 \alpha_0(ix) \times \sum_{ij} d_i d_j \chi_{ijEE}(\vec{b}, \vec{b}, ix) \right). \quad (3.1)$$

This result is valid for arbitrary geometries. It should be noted that the effect of the geometry is contained in  $\chi_{ijEE}(\vec{b}, \vec{b}, \omega)$ .

In the special case when the dielectric of dielectric constant  $\epsilon_0(\omega)$  occupies the domain  $-\infty \leq z \leq 0$  and the atom is situated in vacuum, the response function  $\chi_{ij}$  is obtained from (I.5.43), (I.5.42) by a trivial change of variables. We write it in the form

$$\chi_{ijEE}(\vec{r}, \vec{r}', \omega) = \chi_{ijEE}^{(0)}(\vec{r}, \vec{r}', \omega) + \chi_{ijEE}^{(1)}(\vec{r}, \vec{r}', \omega), \quad (3.2)$$

where  $\chi^{(0)}$  is given by (2.26) and  $\chi^{(1)}$  by

$$\chi_{ijEE}^{(1)}(\vec{r}, \vec{r}', \omega) = -\frac{i}{2\pi} \iint \frac{du dv}{w} \hat{\chi}_{ij}(u, v, \omega) \times \exp\{iu(x-x') + iv(y-y') + iw(z+z')\}, \quad (3.3)$$

$$w^2 = k_0^2 - k_{\parallel}^2, \quad w_0^2 = k_0^2 \epsilon_0 - k_{\parallel}^2,$$

$$k_{\parallel}^2 = u^2 + v^2, \quad k_0 = \omega/c,$$

with

$$\begin{aligned}
\hat{\chi}_{11}(u, v, \omega) &= k_0^2 \frac{w_0 - w}{w_0 + w} - u^2 \left( 1 - \frac{2w}{\epsilon_0 w + w_0} \right), \\
\hat{\chi}_{22}(u, v, \omega) &= \hat{\chi}_{11}(v, u, \omega), \\
\hat{\chi}_{33}(u, v, \omega) &= -\frac{k_0^2 (w \epsilon_0 - w_0)}{w \epsilon_0 + w_0}, \\
\hat{\chi}_{23}(u, v, \omega) &= \frac{vw (w \epsilon_0 - w_0)}{w \epsilon_0 + w_0}, \\
\hat{\chi}_{32}(u, v, \omega) &= -\hat{\chi}_{23}(u, v, \omega), \\
\hat{\chi}_{12}(u, v, \omega) &= -uv \left( 1 - \frac{2w}{w \epsilon_0 + w_0} \right), \\
\hat{\chi}_{13}(u, v, \omega) &= \hat{\chi}_{21}(u, v, \omega), \\
\hat{\chi}_{31}(u, v, \omega) &= \frac{uw (w \epsilon_0 - w_0)}{w \epsilon_0 + w_0} = -\hat{\chi}_{31}(u, v, \omega).
\end{aligned} \tag{3.4}$$

The interaction energy is obtained by substituting (3.2)–(3.4) into (3.1). Usually the orientation of the dipole moment is not known and hence (3.1) is to be averaged over the orientation of  $d_i$ . It does not appear possible to obtain a closed form expression from (3.1) once this averaging is done.

To lowest order in the coupling constant, we have from (3.1)

$$\delta E_0 = -\frac{\omega_0}{\pi} \sum d_i d_j \int_0^\infty dx \alpha_0(ix) \chi_{ijEE}(\vec{b}, \vec{b}, ix),$$

which on averaging over  $d_i$  reduces to

$$\begin{aligned}
\delta E_0 &= -\frac{\omega_0}{3\pi} |d|^2 \int_0^\infty dx \alpha_0(ix) \sum \chi_{iiEE}(\vec{b}, \vec{b}, ix) \\
&\equiv \delta E^{(0)} + \delta E^{(1)}(b),
\end{aligned}$$

where  $\delta E^{(0)}$  is the usual shift in the ground-state energy and  $\delta E^{(1)}$  is given by

$$\delta E^{(1)}(b) = -\frac{\omega_0 |d|^2}{3\pi} \int_0^\infty dx \alpha_0(ix) \sum \chi_{iiEE}^{(1)}(\vec{b}, \vec{b}, ix). \tag{3.5}$$

On substituting (3.3) and (3.4) into (3.5) and after some algebra, we obtain

$$\begin{aligned}
\delta E^{(1)}(b) &= -\frac{2\omega_0 |d|^2}{3\pi c^3} \int_0^\infty dx \alpha_0(ix) x^3 \\
&\times \int_1^\infty dp \left( \frac{p_0 - p}{p_0 + p} + \frac{(p^2 - 1)p(\epsilon_0 - 1)}{\epsilon_0 p + p_0} \right) e^{-2pbx/c},
\end{aligned} \tag{3.6}$$

where  $\epsilon_0$  is at pure imaginary frequency  $ix$  and

$$p_0^2 = p^2 + \epsilon_0 - 1. \tag{3.7}$$

For large distances, (3.6) reduces to

$$\begin{aligned}
\delta E^{(1)}(b) &\cong -\frac{4\omega_0 |d|^2}{\pi c^3} \alpha_0(0) \left( \frac{c}{2b} \right)^4 \\
&\times \int_1^\infty \frac{dp}{p^4} \left( \frac{p_0 - p}{p_0 + p} + \frac{(p^2 - 1)p(\epsilon_0 - 1)}{\epsilon_0 p + p_0} \right),
\end{aligned} \tag{3.8}$$

which is equivalent to the result of Dzyaloshinskii *et al.*<sup>10</sup> To show this, we write (3.6) as

$$\begin{aligned}
\delta E^{(1)}(b) &= -\frac{\omega_0 |d|^2}{3\pi c^3} \int_0^\infty dx \alpha_0(ix) x^3 \\
&\times \int_1^\infty dp e^{-2pbx/c} \left\{ \frac{p_0 - p}{p_0 + p} + \frac{(2p^2 - 1)(\epsilon_0 p - p_0)}{\epsilon_0 p + p_0} \right. \\
&\quad \left. + \left[ \frac{p_0 - p}{p_0 + p} - 1 \right. \right. \\
&\quad \left. \left. + \frac{2p(p p_0 - p^2 + 1)}{p \epsilon_0 + p_0} \right] \right\},
\end{aligned}$$

and use the identity  $(p p_0 - p^2 + 1)(p + p_0) = p \epsilon_0 + p_0$  to show that the terms in square brackets reduce to zero and then (3.6) reduces to

$$\begin{aligned}
\delta E^{(1)}(b) &= -\frac{\omega_0 |d|^2}{3\pi c^3} \int_0^\infty dx \alpha_0(ix) x^3 \\
&\times \int_1^\infty dp e^{-2pbx/c} \left( \frac{p_0 - p}{p_0 + p} + \frac{(2p^2 - 1)(\epsilon_0 p - p_0)}{\epsilon_0 p + p_0} \right).
\end{aligned} \tag{3.9}$$

For the case of a conductor, on taking the limit of infinite conductivity, we obtain from (3.3) and (3.4)

$$\begin{aligned}
\chi_{ijEE}^{(1)}(\vec{r}, \vec{r}_0, \omega) &= \left( (2\delta_{j3} - 1) \frac{\partial^2}{\partial x_i \partial x_j} + k_0^2 (2\delta_{i3} - 1) \delta_{ij} \right) \\
&\times \frac{e^{ik_0 R_i}}{R_i},
\end{aligned} \tag{3.10}$$

$$\vec{R}_i = (x - x_0), (y - y_0), (z + z_0),$$

which should be compared with the translationally invariant response (2.26) and this is what one expects from the considerations of image dipole. The exact result for the interaction energy is obtained by substituting (3.10) into (3.1). The resulting expression does not appear to coincide with the result (21) of Renne.<sup>6</sup> The difference is presumably associated with how the averaging over the orientation is done. However the lowest-order result coincides with his expression (22) as well as with Casimir and Polder's result<sup>2</sup>

$$\begin{aligned}
\delta E^{(1)}(b) &= -\frac{2\omega_0 |d|^2}{3\pi c^3} \int_0^\infty dx \alpha_0(ix) \\
&\times x^3 \left( \frac{\partial^2}{\partial \alpha^2} \frac{e^{-\alpha}}{\alpha} \right)_{\alpha=2bx/c}.
\end{aligned} \tag{3.11}$$

In this section we have considered the interaction energy between an oscillator and a dielectric. Using the same technique one could, of course, also consider, the problem of several oscillators in the presence of a dielectric. It should also be noted that we have been able to obtain an exact expression for the dispersion force because we treated the atom as an oscillator. If this assumption is to be relaxed then the expression for the interaction energy is very involved. However the lowest-order result (to order  $e^2$ ) is more or less identical to (3.5). Using the perturbation theory it is straightforward to compute the interaction energy for the ground state as well as for excited states. One, for example, finds for a two-level atom

$$\delta E^{(+)}(b) = - \left( \delta E^{(-)}(b) + \text{Re} \sum \chi_{ijEE}^{(1)}(\vec{b}, \vec{b}, \omega_0) d_i d_j \right), \quad (3.12)$$

where  $\delta E^{(+)}$  and  $\delta E^{(-)}$  are the interaction energies for the excited state and the ground state, respectively.  $\delta E^{(-)}$  is given by (3.5) and  $\chi_{ij}$  by (3.3). For a conductor  $\delta E^{(+)}$  is also proportional to  $b^{-3}$  at short distances. However  $\delta E^{(+)} - \delta E^{(-)}$  is not proportional to  $b^{-3}$ . It is clear that the large distance behavior of  $\delta E^{(+)}$  is determined by the term  $\chi^{(1)}$  and hence is very different from that of  $\delta E^{(-)}$ .

#### IV. DISPERSION FORCE BETWEEN TWO ISOTROPIC SPATIALLY NONDISPERSIVE DIELECTRICS

We now calculate the dispersion force between two isotropic spatially nondispersive dielectrics of dielectric constant  $\epsilon(\omega)$  and separated by vacuum. The dielectrics are assumed to occupy the volumes  $0 \leq z \leq \infty$  and  $-\infty \leq z \leq -d$ ,  $d$  being the separation between two bodies. The dispersion force is given by the  $zz$  component of the stress tensor at a point just outside the medium occupying volume  $0 \leq z \leq \infty$ , i.e., at the point  $z=0^-$ . It is given by

$$F_{zz} = (1/4\pi) [\langle E_z(\vec{r}, t) E_z(\vec{r}, t) \rangle + \langle H_x(\vec{r}, t) H_x(\vec{r}, t) \rangle - \frac{1}{2} \langle \vec{E}(\vec{r}, t) \cdot \vec{E}(\vec{r}, t) \rangle - \frac{1}{2} \langle \vec{H}(\vec{r}, t) \cdot \vec{H}(\vec{r}, t) \rangle ]_{z=0}. \quad (4.1)$$

Note that  $\vec{E}$  ( $\vec{H}$ ) is even (odd) under time reversal and hence the correlations of the form  $\langle \vec{E} \vec{E} \rangle$  or  $\langle \vec{H} \vec{H} \rangle$  would be given by (1.4). The appropriate response functions have already been computed in Paper I. We must also subtract from (4.1) the contribution to the stress tensor if the second medium were absent ( $d \rightarrow \infty$ ). So the effective response functions would be given by

$$\chi_{ijEE}^{\text{eff}}(\vec{r}, \vec{r}', \omega) = \chi_{ijEE}(\vec{r}, \vec{r}', \omega) - \lim_{d \rightarrow \infty} \chi_{ijEE}(\vec{r}, \vec{r}', \omega) \quad (4.2)$$

$$= \chi_{ijEE}^{(1)}(\vec{r}, \vec{r}', \omega) - \lim_{d \rightarrow \infty} \chi_{ijEE}^{(1)}(\vec{r}, \vec{r}', \omega). \quad (4.3)$$

The translationally invariant part does not contribute to  $\chi^{\text{eff}}$ .  $\chi_{ijEE}^{(1)}$ , etc. [ $\lim_{d \rightarrow \infty} \chi_{ijEE}^{(1)}$ ] are to be obtained from equations (I.5.18)–(I.5.21) [(I.5.42)–(I.5.44)]. Similarly the effective response functions involving the magnetic field would be

$$\chi_{ijHH}^{\text{eff}}(\vec{r}, \vec{r}', \omega) = \chi_{ijHH}^{(1)}(\vec{r}, \vec{r}', \omega) - \lim_{d \rightarrow \infty} \chi_{ijHH}^{(1)}(\vec{r}, \vec{r}', \omega). \quad (4.4)$$

Subtracting (I.5.43) from (I.5.18) and (I.5.44) from (I.5.20), we obtain for the effective electric fields

$$(\vec{K}_{\parallel} \times \vec{\mathcal{E}}_{\parallel}^{(-)}) = \frac{ik_0^2}{2\pi w_0} D_2^{-1} (\vec{K}_{\parallel} \times \vec{p}_{\parallel}) \left( \frac{w - w_0}{w + w_0} e^{-i\vec{k}_0 \cdot \vec{r}_0} - e^{-i\vec{k}'_0 \cdot \vec{r}_0} \right), \quad (4.5)$$

$$(\vec{K}_{\parallel} \cdot \vec{\mathcal{E}}_{\parallel}^{(-)}) = - \frac{iw_0}{2\pi w_0} D_1^{-1} \left( \frac{w_0 \epsilon - w}{w_0 \epsilon + w} (k_{\parallel}^2 p_{\perp} - w_0 \vec{K}_{\parallel} \cdot \vec{p}_{\parallel}) e^{-i\vec{k}_0 \cdot \vec{r}_0} + (k_{\parallel}^2 p_{\perp} + w_0 \vec{K}_{\parallel} \cdot \vec{p}_{\parallel}) e^{-i\vec{k}'_0 \cdot \vec{r}_0} \right). \quad (4.6)$$

Similarly the effective magnetic fields are obtained by subtracting (I.5.45) and (I.5.46) from (I.5.29) and (I.5.27), respectively:

$$(\vec{K}_{\parallel} \times \vec{\mathcal{H}}_{\parallel}^{(-)}) = - \frac{ik_0^2}{2\pi w_0} D_1^{-1} (\vec{K}_{\parallel} \times \vec{m}_{\parallel}) \times \left( e^{-i\vec{k}'_0 \cdot \vec{r}_0} + \frac{w_0 \epsilon - w}{w_0 \epsilon + w} e^{-i\vec{k}_0 \cdot \vec{r}_0} \right), \quad (4.7)$$

$$(\vec{K}_{\parallel} \cdot \vec{\mathcal{H}}_{\parallel}^{(-)}) = \frac{iw_0}{2\pi w_0} D_2^{-1} \left( \frac{w - w_0}{w + w_0} \times (k_{\parallel}^2 m_{\perp} - w_0 \vec{K}_{\parallel} \cdot \vec{m}_{\parallel}) e^{-i\vec{k}_0 \cdot \vec{r}_0} - (k_{\parallel}^2 m_{\perp} + w_0 \vec{K}_{\parallel} \cdot \vec{m}_{\parallel}) e^{-i\vec{k}'_0 \cdot \vec{r}_0} \right). \quad (4.8)$$

No subtraction is needed for the fields  $\mathcal{E}^{(+)}$ ,  $\mathcal{H}^{(+)}$  because of (I.5.42). Let us now see what the vanishing of  $D_1$  and  $D_2$  implies. From (I.5.18)–(I.5.21) it is seen that in the absence of any probe

$$(\vec{K}_{\parallel} \times \vec{\mathcal{E}}_{\parallel}^{(+)}) D_2 = 0, \quad (\vec{K}_{\parallel} \cdot \vec{\mathcal{E}}_{\parallel}^{(+)}) D_1 = 0, \quad (4.9)$$

which implies the following.

(i) If  $D_1 \neq 0$ , then  $(\vec{K}_{\parallel} \cdot \vec{\mathcal{E}}_{\parallel}^{(+)}) = 0$  implying that  $\mathcal{E}_z^{(+)} = 0$  and hence  $(\vec{K}_{\parallel} \times \vec{\mathcal{E}}_{\parallel}^{(+)}) \neq 0$  and then the first of equations (4.9) leads to  $D_2 = 0$ . Therefore the vanishing of  $D_2$  corresponds to those modes for which the  $z$  component of the electric field vanishes. Such modes are usually referred to as TE modes. The dispersion relation for such modes, from (I.5.22), is given by

$$\left(\frac{w+w_0}{w-w_0}\right)^2 e^{-2iw_0d} = 1. \quad (4.10)$$

(ii) If  $D_2 \neq 0$ , then  $(\vec{K}_{\parallel} \times \vec{E}^{(*)}) = 0$  implying that  $(\vec{K}_{\parallel} \cdot \vec{E}^{(*)}) \neq 0$  which leads to  $D_1 = 0$ . Such modes are the so-called TM modes, and the dispersion relation for such modes is therefore

$$\left(\frac{w_0\epsilon + w}{w_0\epsilon - w}\right)^2 e^{-2iw_0d} = 1. \quad (4.11)$$

The dispersion relations (4.10) and (4.11) coincide with the well-known ones obtained by conventional methods.<sup>19-21</sup> The response function contains the information about these surface modes.

We will now use the response functions obtained from (4.5)–(4.8) and (I.5.19), (I.5.21), (I.5.28), (I.5.30) to calculate the dispersion force. From (4.6) and (I.5.21) the response function  $\chi_{33EE}^{\text{eff}}$  is

$$\chi_{33EE}^{\text{eff}}(\vec{p}, \vec{p}, \omega) = -i \iint \frac{du dv}{2\pi w_0} D_1^{-1} \frac{4k_1^2 w_0^2 \epsilon^2}{(w_0\epsilon)^2 - w^2}, \quad (4.12)$$

where  $\vec{p}$  is the two-dimensional vector  $\vec{p} = (x, y, 0)$ . Similarly using (4.5), (4.6), (I.5.19), and (I.5.21), we find

$$\chi_{11EE}^{\text{eff}}(\vec{p}, \vec{p}, \omega) + \chi_{22EE}^{\text{eff}}(\vec{p}, \vec{p}, \omega) = i \iint \frac{du dv}{2\pi w_0} \left( \frac{D_2^{-1} k_0^2}{w^2 - w_0^2} + \frac{w^2 D_1^{-1}}{w_0^2 \epsilon^2 - w^2} \right) 4w_0^2, \quad (4.13)$$

and on using (4.7), (4.8), (I.5.28), (I.5.30),

$$\chi_{33HH}^{\text{eff}}(\vec{p}, \vec{p}, \omega) = i \iint \frac{du dv}{2\pi w_0} D_2^{-1} \frac{4k_1^2 w_0^2}{w^2 - w_0^2}, \quad (4.14)$$

$$\chi_{11HH}^{\text{eff}}(\vec{p}, \vec{p}, \omega) + \chi_{22HH}^{\text{eff}}(\vec{p}, \vec{p}, \omega) = -i \iint \frac{du dv}{2\pi w_0} \left( \frac{w^2 D_2^{-1}}{w^2 - w_0^2} + \frac{k_0^2 \epsilon^2 D_1^{-1}}{w_0^2 \epsilon^2 - w^2} \right) 4w_0^2. \quad (4.15)$$

The dispersion force is equal to

$$F_{zz} = \frac{\hbar}{8\pi^2} \int_0^\infty d\omega \left( 2\chi_{33EE}^{\text{eff}}(\vec{p}, \vec{p}, i\omega) + 2\chi_{33HH}^{\text{eff}}(\vec{p}, \vec{p}, i\omega) - \sum_i \left[ \chi_{iiEE}^{\text{eff}}(\vec{p}, \vec{p}, i\omega) + \chi_{iiHH}^{\text{eff}}(\vec{p}, \vec{p}, i\omega) \right] \right). \quad (4.16)$$

On substituting (4.12)–(4.15) into (4.16), we find that

$$F_{zz} = \frac{\hbar}{8\pi^2} \int_0^\infty d\omega \iint du dv \left( \frac{4iw_0^2}{2\pi w_0} (D_1^{-1} + D_2^{-1}) \right)_{i\omega},$$

which by change of variable reduces to

$$F_{zz} = \frac{\hbar}{2\pi^2 c^3} \int_0^\infty d\omega \omega^3 \int_1^\infty p_0^2 dp_0 \times \left\{ \left[ \left( \frac{p+p_0}{p-p_0} \right)^2 e^{2p_0\omega d/c} - 1 \right]^{-1} + \left[ \left( \frac{p+p_0\epsilon}{p-p_0\epsilon} \right)^2 e^{2p_0\omega d/c} - 1 \right]^{-1} \right\}, \quad (4.17)$$

where

$$p^2 = \epsilon(i\omega) - 1 + p_0^2. \quad (4.18)$$

The expression (4.17) coincides with Lifshitz's expression.<sup>9</sup> We see how the zeros of  $D_1$  and  $D_2$  (which give the surface modes) contribute to the dispersion force. We could similarly consider the case of more complicated geometries or the magnetic bodies. The generalization to the case of finite temperatures is trivial.

We now briefly compare our method with Lifshitz's method. Maxwell's equations, in a medium, should be regarded as equations for the mean values. We solved such mean-value equations to obtain the appropriate response function and then used the fluctuation dissipation theorem to calculate the correlations. Lifshitz, on the other hand, augmented Maxwell's equations with fluctuating forces, thereby making them Langevin equations. The situation is similar to what one does in the theory of Brownian motion. For example, for a free particle, the macroscopic equation is given by

$$\dot{v} + \int_0^t \gamma(t-\tau)v(\tau) d\tau = 0, \quad (4.19)$$

which is replaced in the theory of Brownian motion by

$$\dot{v} + \int_0^t \gamma(t-\tau)v(\tau) d\tau = F(t). \quad (4.20)$$

The correlation of  $F(t)$  is calculated from the requirement that the equilibrium value of  $\langle v^2 \rangle$  as predicted by (4.20) must be equal to the one given by the law of equipartition of energy (*second* fluctuation-dissipation theorem<sup>22</sup>). Once the correlation functions of  $F(t)$  are known, all other correlation functions of  $v(t)$  can be calculated. Lifshitz's calculation is analogous to this procedure. The other method is to solve (4.19) for the linear response which then enables one to obtain equilibrium correlation functions via the *first* fluctuation-dissipation theorem.<sup>22</sup> The final results are, of course, identical.

#### V. DISPERSION FORCE BETWEEN A SPATIALLY DISPERSIVE AND A SPATIALLY NONDISPERSIVE BODY

As a final example of the calculation of dispersion force using the response function formalism, we consider a spatially nondispersive body of dielectric constant  $\epsilon_3(\omega)$  and occupying the domain  $-\infty \leq z \leq -d$  and a spatially dispersive body occupying  $0 \leq z \leq \infty$ . We assume that the two bodies are separated by vacuum and that the spatially dispersive body is characterized by the dielectric func-

tion (I.6.1), viz.,

$$\epsilon(\vec{K}, \omega) = \epsilon_0 + \chi / (k^2 - \mu^2). \quad (5.1)$$

For the meaning of various symbols we refer to Sec. VI of Paper I. We will assume that the bodies are separated by a distance  $d$  at which the retardation effects can be ignored. We have discussed (Sec. VI of Paper I) briefly the structure of the electromagnetic fields inside the medium characterized by (5.1). The dispersion force in the present case is given by

$$F_{zz} = (1/8\pi) [ \langle E_z(\vec{r}, t) E_z(\vec{r}, t) - E_x(\vec{r}, t) \times E_x(\vec{r}, t) - E_y(\vec{r}, t) E_y(\vec{r}, t) \rangle_{z=0} ]. \quad (5.2)$$

The correlation functions appearing in (5.2) are

$$\chi_{ijEE}^{(1)}(\vec{r}, \vec{r}_0, \omega) = \frac{i}{2\pi} \frac{\partial^2}{\partial x_i \partial x_j} \iint \frac{du dv}{w_1} D^{-1} e^{iu(\alpha-x_0) + iv(y-y_0)} \left( e^{i w_1(z-z_0)} + e^{-i w_1(z-z_0)} - \frac{\epsilon_3 + 1}{\epsilon_3 - 1} e^{-i w_1(z+z_0+2d)} - \frac{\beta_1 - \beta_2 \alpha + w_1(1-\alpha)}{\beta_1 - \beta_2 \alpha - w_1(1-\alpha)} e^{+i w_1(z+z_0)} \right). \quad (5.6)$$

As in Sec. V, we must subtract the contribution to (5.3) if the second isotropic dielectric were absent. The corresponding response function is given by (I.6.30), i.e.,

$$\lim_{d \rightarrow \infty} \chi_{ijEE}(\vec{r}, \vec{r}_0, \omega) = \chi_{ijEE}^{(0)}(\vec{r}, \vec{r}_0, \omega) + \frac{i}{2\pi} \frac{\partial^2}{\partial x_i \partial x_j} \iint \frac{du dv}{w_1} \left( \frac{\beta_1 - \beta_2 \alpha - w_1(1-\alpha)}{\beta_1 - \beta_2 \alpha + w_1(1-\alpha)} \right) e^{iu(\alpha-x_0) + iv(y-y_0) + i w_1(z+z_0)}. \quad (5.7)$$

The vanishing of the denominators in (5.6) and (5.7) yields the surface-polariton dispersion relations (with retardation effects ignored); i.e., (i) the surface-polariton dispersion relation, for the case when the spatially dispersive medium occupies  $0 \leq z \leq \infty$  and is surrounded by vacuum, is

$$\beta_1 - \beta_2 \alpha + w_1(1-\alpha) = 0, \quad (5.8a)$$

which can be shown to be equivalent to the standard dispersion relation.<sup>21,23</sup> (ii) The dispersion relation for surface polaritons in the geometry, when the spatially dispersive medium occupies  $0 \leq z \leq \infty$  and isotropic dielectric occupies  $-\infty \leq z \leq -d$  and the two are separated by vacuum, is

$$\frac{\beta_1 - \beta_2 \alpha + w_1(1-\alpha)}{\beta_1 - \beta_2 \alpha - w_1(1-\alpha)} \frac{\epsilon_3 + 1}{\epsilon_3 - 1} e^{-2i w_1 d} = 1, \quad (5.8b)$$

which can be shown to be equivalent to the dispersion relation obtained by standard methods.<sup>19-21</sup> The effective response function is therefore

$$\chi_{ijEE}^{\text{eff}}(\vec{r}, \vec{r}_0, \omega) = \frac{i}{2\pi} \frac{\partial^2}{\partial x_i \partial x_j} \iint \frac{du dv}{w_1} D^{-1} e^{iu(\alpha-x_0) + iv(y-y_0)} \times [ e^{i w_1 z} (e^{-i w_1 z_0} - e^{i w_1 z_0} \alpha) - e^{-i w_1 z} (\alpha^{-1} e^{-i w_1 z_0} - e^{i w_1 z_0}) ], \quad (5.9)$$

related to the response functions via (1.4) and therefore

$$F_{zz} = \frac{\hbar}{8\pi^2} \int_0^\infty d\omega \left( 2\chi_{33EE}(\vec{r}, \vec{r}, i\omega) - \sum_i \chi_{iiEE}(\vec{r}, \vec{r}, i\omega) \right)_{z=0^-}. \quad (5.3)$$

The response function from (I.6.27), (I.6.28), and (I.6.29) is

$$\chi_{ijEE}(\vec{r}, \vec{r}_0, \omega) = \chi_{ijEE}^{(0)}(\vec{r}, \vec{r}_0, \omega) + \chi_{ijEE}^{(1)}(\vec{r}, \vec{r}_0, \omega), \quad (5.4)$$

where

$$\chi_{ijEE}^{(1)}(\vec{r}, \vec{r}_0, \omega) = - \frac{\partial^2}{\partial x_i \partial x_j} |\vec{r} - \vec{r}_0|^{-1}, \quad (5.5)$$

where

$$\alpha = \frac{\beta_1 - \beta_2 \alpha + w_1(1-\alpha)}{\beta_1 - \beta_2 \alpha - w_1(1-\alpha)}. \quad (5.10)$$

On combining (5.3) and (5.9) we obtain

$$F = - \frac{\hbar}{4\pi^2} \int_0^\infty d\omega \iint du dv k_{\parallel} D^{-1} |_{i\omega} \quad (5.11)$$

$$= \frac{\hbar}{2\pi^2} \int_0^\infty d\omega \int_0^\infty \kappa^2 d\kappa \left( \frac{\epsilon_3 + 1}{\epsilon_3 - 1} \frac{\psi + 1}{\psi - 1} e^{2\kappa d} - 1 \right) |_{i\omega}, \quad (5.12)$$

where

$$\psi = \frac{\beta_2 \alpha - \beta_1}{(\alpha - 1) w_1}$$

$$= \epsilon_1 + \frac{\epsilon_1 w_1 (w_2 - w_\mu)}{(w_1 - w_2)(w_1 + w_\mu)} + \frac{\chi w_1}{2 w_\mu \mu^2}. \quad (5.13)$$

Here  $\epsilon_1$  is the local dielectric function

$$\epsilon_1 = \epsilon_0 - \chi / \mu^2, \quad (5.14)$$

and

$$w_1 = i\kappa, \quad w_2^2 = \mu^2 \epsilon_1 / \epsilon_0 - \kappa^2, \quad w_\mu^2 = \mu^2 - \kappa^2. \quad (5.15)$$

We first note that in the local limit of the dielectric function (5.1) ( $m^* \rightarrow \infty$ ),  $w_2 \propto \sqrt{m^*}$ ,  $w_\mu \propto \sqrt{m^*}$ , and hence  $\psi = \epsilon_1 + O(1/\sqrt{m^*})$ . Therefore in the lo-



cal limit, (5.12) becomes

$$F_0 = \frac{\hbar}{2\pi^2} \int_0^\infty d\omega \int_0^\infty \kappa^2 d\kappa \left( \frac{(\epsilon_3 + 1)(\epsilon_1 + 1)}{(\epsilon_3 - 1)(\epsilon_1 - 1)} e^{2\kappa d} - 1 \right)_{i\omega}^{-1} \\ = \frac{\hbar}{16\pi^2 d^3} \int_0^\infty d\omega \int_0^\infty x^2 dx \\ \times \left( \frac{[\epsilon_3(i\omega) + 1][\epsilon_1(i\omega) + 1]}{[\epsilon_3(i\omega) - 1][\epsilon_1(i\omega) - 1]} e^x - 1 \right)^{-1}, \quad (5.16)$$

which is the well-known expression for the dispersion force between two isotropic dielectrics at short distances and which displays the  $d^{-3}$  dependence on the separation between the two bodies.<sup>9, 10</sup>

If one of the bodies is spatially dispersive, the dispersion force, at short distances, is no longer proportional to  $d^{-3}$  because of the dependence of  $\psi$  on  $\kappa$ . For a weakly spatially dispersive medium, one can carry out an expansion in powers of  $(m^*)^{-1/2}$  and it is clear that next term will be proportional to  $d^{-4}$ . A straightforward algebra shows that

$$F = F_0 + F_1 + \dots, \quad (5.17)$$

where  $F_0$  is given by (5.16) and  $F_1$  by

$$F_1 = -\frac{\hbar\omega_p}{32\pi^2 d^4 \omega_p} \left( \frac{\hbar}{m^* \omega_0} \right)^{1/2} \int_0^\infty d\omega \int_0^\infty x^3 dx \left( \frac{\epsilon_3 + 1}{\epsilon_3 - 1} \right) e^x \\ \times \epsilon_0 (\epsilon_1 - \epsilon_0)^{1/2} \left[ \left( \frac{\epsilon_1}{\epsilon_0} \right)^{1/2} - 1 \right] \left[ 3 \left( \frac{\epsilon_1}{\epsilon_0} \right)^{1/2} + 1 \right] \\ \times \left( \frac{(\epsilon_3 + 1)(\epsilon_1 + 1)}{(\epsilon_3 - 1)(\epsilon_1 - 1)} e^x - 1 \right)^{-2}, \quad (5.18)$$

where  $\omega_p$  is the plasma frequency  $\omega_p^2 = 4\pi\alpha\omega_0^2$ . It is clear that  $F_1$  is negative and hence, in contrast to  $F_0$ , it is repulsive in nature. Therefore, it seems

that if spatial dispersion is too strong, then the dispersion force may change its character from attractive to repulsive.

The present derivation clearly shows how the surface modes contribute to the dispersion force. Because of the special combination in which  $\psi$  and  $\epsilon_3$  appear in (5.12), it is thought that the dispersion force between two spatially dispersive bodies will be

$$F_{zz} = \frac{\hbar}{2\pi^2} \int_0^\infty d\omega \int_0^\infty \kappa^2 d\kappa \left( \frac{(\psi^{(1)} + 1)(\psi^{(2)} + 1)}{(\psi^{(1)} - 1)(\psi^{(2)} - 1)} e^{2\kappa d} - 1 \right)_{i\omega}^{-1}, \quad (5.19)$$

where  $\psi^{(1)}$  and  $\psi^{(2)}$  are the  $\psi$  functions, for each of the two spatially dispersive dielectrics, defined by a relation of the form (5.13). We again point out that  $\psi + 1 = 0$  gives the dispersion relation (5.8a), and hence if the spatially dispersive dielectric is treated in an approximation other than (I.6.3), then the dispersion force would still be given by (5.19) with a new function which is such that  $\psi + 1 = 0$  gives the dispersion relation for surface polaritons under this new approximation.<sup>24</sup>

Finally it should be noted that Lifshitz's method can be extended, to the case of nonlocal dielectric functions, presumably by taking the following as the correlation function for the fluctuating currents (notation is same as that of Lifshitz), in an isotropic medium,

$$\langle K_i(\vec{r}, t) K_j(0, 0) \rangle = \delta_{ij} \frac{1}{(2\pi)^4} \int d^3k d\omega \Gamma(\vec{k}, \omega) e^{i\vec{k}\cdot\vec{r} - i\omega t},$$

with

$$\Gamma(\vec{k}, \omega) = 2\hbar \coth(\beta\omega\hbar/2) \text{Im}\epsilon(\vec{k}, \omega).$$

We have, however, not tried this prescription to obtain (5.12).

<sup>1</sup>G. S. Agarwal, preceding paper, Phys. Rev. A **10**, 230 (1974). This paper will hence forth be referred to as I and the numbers of equations from this paper will be preceded by I [e.g., (I.2.10)].

<sup>2</sup>F. London, Z. Phys. **60**, 491 (1930).

<sup>3</sup>H. B. Casimir and D. Polder, Phys. Rev. **73**, 360 (1948).

<sup>4</sup>M. R. Aub and S. Zienau, Proc. Roy. Soc. A **257**, 464 (1960).

<sup>5</sup>C. Mavroyannis, Mol. Phys. **6**, 593 (1963).

<sup>6</sup>M. J. Renne, Physica **53**, 193 (1971); **56**, 125 (1971).

<sup>7</sup>R. K. Bullough and B. V. Thomson, J. Phys. C **3**, 1780 (1970).

<sup>8</sup>A. D. McLachlan, Proc. Roy. Soc. A **271**, 387 (1963); Mol. Phys. **6**, 423 (1963); **7**, 381 (1963).

<sup>9</sup>E. M. Lifshitz, Zh. Expt. Teor. Fiz. **29**, 94 (1955) [Sov. Phys.—JETP **2**, 73 (1956)].

<sup>10</sup>I. E. Dzyaloshinskii, E. M. Lifshitz, and L. P. Pitaevskii, Adv. Phys. **10**, 165 (1961).

<sup>11</sup>N. G. Van Kampen, B. R. A. Nijboer, and K. Schram, Phys. Lett. A **26**, 307 (1968).

<sup>12</sup>E. Gerlach, Phys. Rev. B **4**, 393 (1971); P. Richmond and B. W. Ninham, J. Phys. C **4**, 1988 (1971); B. W. Ninham, V. A. Parsegian, and G. H. Weiss, J. Stat. Phys. **2**, 323 (1970); see also an earlier paper by G. D. Mahan, J. Chem. Phys. **43**, 1569 (1965).

<sup>13</sup>K. Schram, Phys. Lett. A **43**, 282 (1973).

<sup>14</sup>G. S. Agarwal, Phys. Lett. A **43**, 447 (1973).

<sup>15</sup>Cf. R. M. Wilcox, J. Math. Phys. **8**, 962 (1967).

<sup>16</sup>The role of the free-field propagator in the calculation of binding energies is discussed by R. K. Bullough [J. Phys. A **2**, 477 (1969); A **3**, 751 (1970)] rather in detail.

<sup>17</sup>See, for example, D. Pines and P. Nozieres, *The Theory of Quantum Liquids* (Benjamin, New York, 1966), p. 297.

<sup>18</sup>For the significance of this term see E. A. Power and

S. Zienau, *Phil. Trans. Roy. Soc.* 251, 427 (1959).

<sup>19</sup>For the dispersion relations of the surface modes, see, for example, R. Fuchs and K. L. Kliewer, *Phys. Rev.* 140, A2076 (1965); *Phys. Rev.* 144, 495 (1966).

<sup>20</sup>A. A. Maradudin, E. W. Montroll, G. H. Weiss, and I. P. Ipatva, *Theory of Lattice Dynamics in Harmonic Approximation* (Academic, New York, 1971), Chap. 9, Sec. 2 e.

<sup>21</sup>G. S. Agarwal, *Phys. Rev. B* 8, 4768 (1973).

<sup>22</sup>R. Kubo, *Rep. Prog. Phys.* 29, 255 (1966).

<sup>23</sup>A. A. Maradudin and D. L. Mills, *Phys. Rev. B* 7, 2787 (1973).

<sup>24</sup>One may for example like to treat spatially dispersive dielectric in the approximation that excitons (electrons) are specularly reflected from the boundary. Then the Green's function which appears in (I. 6.3) will be different.