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Many-body optics
III. The optical extinction theorem and $\epsilon_l(k, \omega)$

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Abstract. It is shown that the optical modes which were investigated in the earlier paper I, which are either longitudinal or transverse, and which satisfy the dispersion relations of I, are acceptable solutions of the optical integral equation for a locally isotropic molecular fluid at normal temperatures. Acceptable solutions must satisfy the optical Extinction Theorem. This constraint is very much concerned with the boundary of the system and the system is necessarily finite: thus the response functions for the modes of I depend on the surface geometry. In simplest geometry the longitudinal modes prove to be normal modes; but the transverse modes are modes forced by incident light. The linear response of the system to light is calculated: it is shown that the existence of a surface to the system is essential to the existence of such a response. Light couples to the system through the surface.

It is shown that there are additional longitudinal modes which can be forced by incident free charge. In simplest geometry the extinction theorem now becomes irrelevant. It is then possible to express the longitudinal dipole response in terms of a $(\boldsymbol{k}, \omega)$-dependent longitudinal dielectric constant $\epsilon_l(\boldsymbol{k}, \omega)$: an explicit formula for $\epsilon_l(\boldsymbol{k}, \omega)$ in terms of cluster integrals for the fluid is obtained to all orders in intermolecular correlation but exhibition of the details of that correlation is deferred until later. The zeros of $\epsilon_l(\boldsymbol{k}, \omega)$ yield the dispersion relation for the longitudinal normal modes in the same simplest geometry. Only in the complex dielectric constant approximation does $\epsilon_l(\boldsymbol{k}, \omega)$ coincide with the square of the transverse refractive index $m_t^2(\omega)$.

The theory of the transverse dielectric constant $\epsilon_t(\boldsymbol{k}, \omega)$ for the molecular fluid is developed in paper IV.

1. Introduction

This paper is the third in a series devoted to the presentation of a unified theory of the microscopic optics of molecular fluids. In the first paper of the series (Bullough 1968—to be referred to as I) we introduced the fundamental integral equation of the theory: we obtained transverse and longitudinal dispersion relations in forms which implicitly took account of intermolecular multiple scattering processes at all orders, but the analysis remained incomplete because we did not show that an essential condition, a precise formulation of the optical Extinction Theorem of Ewald (1912, 1916) and Oseen (1915), was satisfied. This is the first problem we take up now in this paper III.

However, the problem proves to be only a part of the bigger problem posed by the extinction theorem in the very much more general context of electromagnetic response theory. Paper I was concerned with the response of a molecular fluid to light: we need also to consider the response of such a system to arbitrary electromagnetic probes, for apart from the intrinsic interest of such a theory we can expect from previous work in many-body theory (e.g. Nozières and Pines 1958, Pines 1963, Abrikosov *et al.* 1965) that the general linear electromagnetic response implicitly provides a binding energy theory. The second paper of this series (Bullough 1969—

to be referred to as II) already shows that this is actually the case; but it neither justifies this fact nor develops any of the details of the electromagnetic response theory. It is therefore precisely the problem of providing a comprehensive and consistent formulation of this most general electromagnetic response theory which is the subject of this paper III. Unfortunately the problem posed by the extinction theorem in this most general context is such that we must continue its study in two further papers: these will therefore follow as parts IV and V of this series (Bullough 1970 a—to be referred to as IV and Bullough 1970 b—to be referred to as V).

It was our intention as stated in I to devote the three parts I, II and III of this series to a preliminary mathematical investigation of the fundamental integral equation of the theory prior to the detailed derivation of a number of connected physical results in the theory of optical scattering, intermolecular binding, and so on. We have found it convenient in practice to depart a little from this program of publication. Thus in the part II which has now appeared we reported a series of physical results of the theory as results simply. These were primarily in the theory of binding energy, for the structure of the scattering theory has been summarized in brief elsewhere (Bullough *et al.* 1968 and Bullough and Hynne 1968).

These statements of the connected results of the theory will ease the presentation of the analysis of the fundamental integral equation which we now take up again from I. The integral equation is examined in the more general context and the analysis continues through the three papers III, IV and V. These constitute a connected analysis and should be read together: they are separated only by exigences of space and are therefore summarized together in the final section, § 5, of V.†

The main problem considered and solved in the three papers is the construction of the most general linear electromagnetic response theory specifically for the molecular fluid. At first sight this problem is an exercise in conventional many-body theory on the lines laid down by Lindhard (1954), for example, or by Nozières and Pines in a series of papers in the late 1950's (e.g. Nozières and Pines 1958—on the plasma). It is hard to see how this approach is consistent with the problem considered in I, however. There we considered the response of the molecular fluid to external light: dispersion relations emerged but no response theory. In fact the linear response of the system to light is exactly the content of the optical extinction theorem. Thus in § 2 of the present paper we develop this aspect of the extinction theorem: we show that the modes assumed as solutions of the fundamental integral equation in I are valid solutions; and we express the extinction theorem as a linear response relation.

Unfortunately the solution of this problem, already plain from previous work (e.g. Darwin 1924, Born and Wolf 1959, Bullough 1962) raises a second problem. The analysis of the optical response shows that the boundary of the material system plays an essential role in the way in which light couples to the system: on the other hand many-body systems are usually assumed to be translationally invariant (e.g.

† The material specifically intended for the parts II and III of the series as projected in I appears in fact in this paper III and the following paper IV. We are still concerned in these papers III and IV with the program envisaged in I, namely mathematical aspects of the solution of the fundamental integral equation, and that program now continues also in V. But we shall find it convenient to raise the status of the paper III, projected as a 'mathematical appendix' in I, to a fundamental analysis of the interaction of electromagnetic fields across the boundary of a many-body system—now the paper IV. Aspects of the binding energy theory projected in I to appear in II are now comprehensively summarized in II as it has appeared, and these results will be developed in detail later in the series. The paper V examines a semi-phenomenological binding energy theory as noted in II.

Nozières and Pines 1958) and admit arbitrary electromagnetic field modes running through the interior either as forced modes or (e.g. Hopfield 1958) as normal modes. We are therefore now faced with two problems: that of showing that our molecular fluid system can admit arbitrary electromagnetic fields in its interior, and that of showing that the existence of these modes is not incompatible with the essential role of the boundary in coupling the system to light.

That a solution of these two problems exists is evident from the brief report in II. The key to the solution is to show that the complete linear response breaks up into two parts: one part is surface-independent and translationally invariant: the other part is surface-dependent and it is this part which reproduces the optical linear response theory when the external probe is light. These two parts to the response functions enable us to make a natural distinction between real and virtual electromagnetic processes in the molecular fluid. This natural distinction should extend to many-body systems other than molecular fluids, for it is certainly applicable to the molecular crystal (Bullough and Obada 1969 a,b) and there is no reason to suppose it must be limited to *molecular* many-body systems.

The translationally invariant response enables us to define $(\boldsymbol{k}, \omega)$-dependent dielectric constants for the molecular fluid. This response has the usual structure of conventional linear response theory: normal mode solutions satisfying dispersion relations are possible solutions and the dispersion relations are singular surfaces of the response functions. This raises the question of whether such normal modes are normal modes of the homogenous form of the fundamental integral equation of I. They are not, in fact, and this leads us to search for normal modes in the more general context of the total linear response theory.

These several problems are very much complicated by the existence of the surface integral which is such an essential feature of the extinction theorem when the probe is light. In consequence the analysis of these problems occupies the third and final section of this paper and the whole of IV. This leaves us free in V to focus attention on the surface-independent translationally invariant part of the linear response. The conventional many-body theoretical structure this exhibits is subsumed under the title of 'virtual mode theory'. Virtual mode theory has a natural and physically important approximation: this is the complex dielectric constant approximation introduced in II in which intermolecular correlation is supposed of such limited range that the $(\boldsymbol{k}, \omega)$-dependent dielectric constants are effectively $\boldsymbol{k}$-independent. The virtual mode theory in this approximation is a natural microscopic expression of a translationally invariant form of a long-wavelength virtual photon theory of the type considered by Dzyaloshinskii *et al.* (1961). Thus we can compare the results of the virtual mode theory in the complex dielectric constant approximation with the semi-phenomenological response theory of these authors and with their theory of free energy: this is the subject matter of V.

The working plan of the papers is given section by section below. It will be indicative of the emphasis of the work if we recall now why molecular fluids are worthy of such exhaustive study. Many-body theorists have previously focused most attention on ground-state energies, free energies, and virial expansions: much of the binding energy work has been devoted to the plasma (cf. for example, Brout and Carruthers 1963) and the arguments based on the Coulomb interaction alone. Interest in real fluids of molecular type remains endemic however (e.g. Yvon 1937, Mayer and Mayer 1948, Kirkwood and Buff 1951, Frisch and Lebowitz 1964, Brout 1965) and new inelastic scattering techniques (Benedek 1966, Egelstaff 1967) and photon counting methods (Glauber 1963, Bertolotti *et al.* 1967) are both increasing the precision and range of the experimental analysis and focusing attention on scattering theory. The radiation field is an essential feature of any many-body optical scattering theory and early work by Casimir (Casimir and Polder 1948) established the significant role of the radiation field in the intermolecular potentials at an intermediate range (say 100 Å separation).

Thus a unified study of the collective effects of the radiation field in a many-body system has been lacking and is needed now. It is the purpose of this series of papers to provide this, and molecular systems are particularly suitable for a study of this type.

The choice of molecular systems for study may seem to limit the arguments in fact to a rather particular choice of material model. For we interpret a molecular fluid as one in which the wave functions of the free molecules form a good basis for the coupled system; then we can express collective macroscopic parameters like the complex refractive index and the dielectric constants in terms of microscopic parameters like the polarizability of the free molecules and the intermolecular correlation functions.

However, it is an aim of the theory to transcend the limitations of the molecular model by finding interrelations between macroscopic optical properties of the system. What can be done here is already evident in II, whilst V is specifically intended to examine the thesis of Dzyaloshinskii *et al.* that, although the wave functions in a real condensed system are so distorted that there is little hope of calculating them, these microscopic features can be concealed in the macroscopic dielectric constant in a calculation of the free energy. If this thesis is applicable to a real fluid it should apply to a consistent application of our model.

At the same time the microscopic processes which may be said to occur in a many-body system are important in themselves: the scattering theory reported (Bullough *et al.* 1968 and Bullough and Hynne 1968) which is based on the molecular model examplifies what can be achieved in relating the microscopic processes to macroscopic properties and shows where macroscopic results need explicit reinforcement by microscopic theory.

These are the reasons for formulating the microscopic many-body optics of a molecular fluid. The program of the next three papers devoted to this end is, in detail, as follows: in § 2 which follows we start from the extinction theorem as it emerged in I. There it appeared as a constraint on the polarization induced in an arbitrary macroscopic region V containing many molecules ($\sim 10^{23}$). In order to carry the analysis further here we particularize V to a parallel-sided slab of finite width $(c+d)$. We are then able first of all to characterize the particular modes of the types derived in I which propagate *along the axis of the slab* as forced or normal modes: in particular the longitudinal modes of I are normal modes in this case.

In § 3 we next show that in the presence of a longitudinal forcing field $\boldsymbol{E}$ parallel to the slab axis there are additional forced solutions. These enable us to introduce for the molecular fluid the formal $(\boldsymbol{k}, \omega)$-dependent dielectric constant $\epsilon_l(\boldsymbol{k}, \omega)$ analogous to that first introduced by Lindhard (1954).

This completes III.

After the brief Introduction, § 2 of IV introduces the transverse dielectric constant $\epsilon_t(\boldsymbol{k}, \omega)$: it offers obvious comparison with that of Nozières and Pines (1959), for example. There is an immediate difficulty associated with the extinction theorem which cannot be resolved by choosing particular directions for the mode wave vectors

$\boldsymbol{k}$. The solution of the difficulty leads naturally into a discussion of transverse normal modes: this is still limited to possible transverse modes with $\boldsymbol{k}$ along the slab axis.

In § 3 of IV we extend the earlier arguments for the forced modes to include all directions of $\boldsymbol{k}$.

In § 4 we extend the discussion of normal modes to both transverse and longitudinal normal modes and all directions of $\boldsymbol{k}$.

This completes IV.

After the brief Introduction, § 2 of V extends the theory, and especially the virtual mode theory, to the case of multicomponent fluid systems.

In § 3 we introduce the complex dielectric constant approximation and a complex dielectric constant $\epsilon(\omega)$. We derive the integral equation of Dzyaloshinskii *et al.* from the microscopic virtual mode theory in this approximation.

In § 4 we adopt the prescription of Dzyaloshinskii *et al.* for the free energy. We can do this by using $\epsilon(\omega)$ alone and all the microscopic processes are concealed in this quantity. We contrast the results with the exact† results of the microscopic theory of intermolecular binding energy reported in II. We can infer that the Dzyaloshinskii prescription so used does not correctly describe all the significant microscopic processes.

In § 5 we attempt a synthesis of the work of the three papers III, IV and V: we restate some of the more important results and summarize the arguments and conclusions.

The one problem still left outstanding from I, that arising from breaking the translational invariance of the correlation functions, will be treated later.

2. The extinction theorem

The fundamental integral equation considered in I was

$$\boldsymbol{P}(\boldsymbol{x}, \omega) = \alpha(\omega)\left\{\boldsymbol{E}(\boldsymbol{x}, \omega)+n\int_V \mathsf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega)g(\boldsymbol{x}, \boldsymbol{x}') \,.\, \boldsymbol{P}(\boldsymbol{x}', \omega)\,\mathrm{d}\boldsymbol{x}'\right\}. \tag{2.1}$$

The field $\boldsymbol{E}$ was a probe there taken to satisfy the free field dispersion relation‡

$$k = \omega c^{-1} \equiv k_0$$

for each mode of wave vector $\boldsymbol{k}$: ω is the angular frequency. The $\alpha(\omega)$ are the (supposed isotropic) frequency-dependent polarizabilities of the isolated particles coupled neither to the field nor to each other:

$$\alpha(\omega) = \frac{e^2}{3\hbar}\sum_k{}' \frac{2\omega_k|\boldsymbol{r}_{k0}|^2}{\omega_k{}^2-\omega^2} \tag{2.2}$$

and as discussed in I (cf. I—§ 2 and equations (2.3) (2.5)) ω^2 in (2.2) can be read as $(\omega+\mathrm{i}\delta)^2$ with δ a positive infinitesimal and ω real and positive, or as ω^2 simply with ω real or not. The quantity n is the average particle number density; the function $g(\boldsymbol{x}, \boldsymbol{x}')$ is a particle pair correlation function taken as a function of $r = |\boldsymbol{x}-\boldsymbol{x}'|$

† Exact within the approximations of the microscopic theory and the particular results under consideration.

‡ We use k for the magnitude of $\boldsymbol{k}$. We used $|\boldsymbol{k}|$ in I. In the free-field dispersion relation k is real when ω is. From (2.5*b*) $k \equiv m_t k_0$ is complex because m_t is. Thus if $\boldsymbol{k} = (k_1, k_2, k_3)$, $k^2 \equiv k_1{}^2+k_2{}^2+k_3{}^2$ and is not $\boldsymbol{k}^* \,.\, \boldsymbol{k}$ (see I). We assume we can always write $\boldsymbol{k} = k\hat{\boldsymbol{k}}$ with $\hat{\boldsymbol{k}}$ a real unit vector. This may be inadequate (cf. footnote ‡ on p. 742).

alone by translational invariance† and local isotropy. The tensor $\mathsf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega)$ is a function of r and ω alone:

$$\mathsf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega) = (\nabla\nabla + k_0^2\mathsf{U}) \exp(ik_0 r)r^{-1} \tag{2.3}$$

in which U is the unit tensor and $k_0 = \omega c^{-1}$. The quantity V is a *finite* region occupied by the molecular fluid.

In I it was shown that the Fourier transform of longitudinal and transverse solutions for $\boldsymbol{P}$ of the fundamental integral equation (2.1) were (I—equation (4.8))

$$\boldsymbol{P}_l(\boldsymbol{k}, \omega) = m_l^{-2}k_0^{-2}\boldsymbol{P}_{l0}(\hat{\boldsymbol{k}}, \omega)\delta(k-m_l k_0) \tag{2.4a}$$

$$\boldsymbol{P}_\mathrm{t}(\boldsymbol{k}, \omega) = m_\mathrm{t}^{-2}k_0^{-2}\boldsymbol{P}_{\mathrm{t}0}(\hat{\boldsymbol{k}}, \omega)\delta(k-m_\mathrm{t} k_0) \tag{2.4b}$$

with $\hat{\boldsymbol{k}}$ a (real) unit vector in the direction of $\boldsymbol{k}$ (which may be complex) if and only if

$$n\alpha(\omega)J_l(m_l k_0, \omega) = 1+\tfrac{8}{3}\pi n\alpha(\omega) \tag{2.5a}$$

$$m_\mathrm{t}^2-1 = 4\pi n\alpha(\omega)\{1-\tfrac{4}{3}\pi n\alpha(\omega)-n\alpha(\omega)J_\mathrm{t}(m_\mathrm{t}k_0, \omega)\}^{-1} \tag{2.5b}$$

and providing the constraint (2.7) below, which is the extinction theorem,‡ could be satisfied. In (2.4) $\delta(x)$ is the usual δ-function§ and the functions $\boldsymbol{P}_{l0}(\hat{\boldsymbol{k}}, \omega)$ and $\boldsymbol{P}_{\mathrm{t}0}(\hat{\boldsymbol{k}}, \omega)$ are so far unspecified except that they are respectively longitudinal and transverse:

$$\boldsymbol{P}_{l0}(\hat{\boldsymbol{k}}, \omega)\times\hat{\boldsymbol{k}} = \boldsymbol{0}, \qquad \boldsymbol{P}_{\mathrm{t}0}(\hat{\boldsymbol{k}}, \omega)\,.\,\hat{\boldsymbol{k}} = 0.$$

We shall find that in the situation envisaged in I the extinction theorem fixes these functions except in one singular case.

Equation (2.5*b*) is a generalized form of the Lorentz–Lorenz expression for the refractive index $m_\mathrm{t}(\omega)$ of transverse polarization modes excited by light: equation (2.5*a*) is the condition for longitudinal modes, a condition equivalent in the simplest (long wavelength) case to the condition $m_\mathrm{t}^2(\omega) = 0$. The generalization is contained in the local field terms $J_{l,\mathrm{t}}(m_{l,\mathrm{t}}k_0, \omega)$. These quantities were initially defined in terms of the solution of (2.1) and hence in terms of the pair correlation function $g(r)$: the definitions were in effect (I—equations (4.4), (4.5) and (4.12), and cf. II)‖

$$J_l(m_l k_0, \omega) = \hat{\boldsymbol{k}}\hat{\boldsymbol{k}} : \int_{-v} \mathsf{F}(r, \omega)\exp(i\boldsymbol{k}\,.\,\boldsymbol{r})\{g(r)-1\}\,\mathrm{d}\boldsymbol{r}; \qquad k = m_l k_0 \tag{2.6a}$$

$$J_\mathrm{t}(m_\mathrm{t} k_0, \omega) = \boldsymbol{u}(\hat{\boldsymbol{k}})\boldsymbol{u}(\hat{\boldsymbol{k}}) : \int_{-v} \mathsf{F}(r, \omega)\exp(i\boldsymbol{k}\,.\,\boldsymbol{r})\{g(r)-1\}\,\mathrm{d}\boldsymbol{r}; \quad k = m_\mathrm{t} k_0. \tag{2.6b}$$

† Nevertheless we break translational invariance (see I, §§ 2 and 4).

‡ The extinction theorem is due in the first instance to Ewald (1912, 1916) and Oseen (1915). It was used in detail, but without explicit identification by Darwin (1924). It forms an essential feature of the arguments of Hoek (1939), Rosenfeld (1951), Mazur (1958) and other papers, Born and Wolf (1959), and Bullough (1962) and other papers. It has been studied by Osborne (1966) in a purely quantal context.

§ Since m_t is complex by (2.5*b*) (and m_l is apparently complex by (2.5*a*)), the δ-functions move into the complex k plane. We assume any contour from $k = 0$ to $k = +\infty$ passes through $k = m_\mathrm{t}k_0$ in a simple way; if the contour starts at $\mathrm{Re}\{k\} > \mathrm{Re}\{m_\mathrm{t}k_0\}$ the δ-function does not contribute; if it starts with $\mathrm{Re}\{k\} < \mathrm{Re}\{m_\mathrm{t}k_0\}$ the δ-function does contribute in the same simple way.

‖ As later in I we now distinguish between the refractive indices m_t and m_l: m_t is the usual refractive index as (2.5*b*) suggests.

In these $\boldsymbol{u}(\hat{\boldsymbol{k}})$ is a unit vector orthogonal to $\hat{\boldsymbol{k}}(\boldsymbol{u}\,.\,\boldsymbol{k} = 0)$. A vanishingly small sphere of volume v is taken out about the origin of the $\boldsymbol{r}$-space of integration: otherwise the integrals are taken over all space.

These quantities J_l and J_t were later generalized to include many-particle interactions (multiple scattering processes) of all orders (I—equations (4.17), (4.18) and (4.19), II—equations (3) and (4)) and then to two-component systems (I—equations (4.26) and (4.27)).† In cases of rigorous mathematical argumentation, as for example in this § 2, we shall assume that the J_l and J_t are defined by (2.4) in terms of the particle pair correlation function $g(r)$ alone. However, when we come in § 3, in IV § 2 and in V to discuss the physics of the forced modes of the virtual mode theory (designated 'virtual modes' in IV) rather more, we shall assume that J_l and J_t have been given their generalization to include all the many-particle interactions. The reason for this is the technical one that a rigorous discussion of the generalization introduces considerable complication to the solutions like (2.4) and to the extinction theorem which is equation (2.7): we conveniently defer discussion of this complication until much later when we present the theory of external scattering. It is there that we shall make a complete analysis of the cluster integrals which appear in J_l and J_t.‡

This completes a brief introduction to the optical integral equation (2.1) which was studied in I. We have yet to examine the constraint on the solutions (2.4) which is also a consequence of that integral equation and which is the optical extinction theorem. This is the problem of this section.

The constraint on the solutions (2.4) which is the extinction theorem for the incident (i.e. applied external) electric field $\boldsymbol{E}(\boldsymbol{x}, \omega)$ is (I—equation (3.13)).

$$\boldsymbol{0} = \boldsymbol{E}(\boldsymbol{x}, \omega) + \frac{(\nabla_x \nabla_x + k_0^2 \mathbf{U})\,.\,n\boldsymbol{\Sigma}(\boldsymbol{x}, \omega)}{(m^2-1)k_0^2} \tag{2.7a}$$

for any set of modes with wave number $k = mk_0$. The vector field $\boldsymbol{\Sigma}(\boldsymbol{x}, \omega)$ is defined for *all* points $\boldsymbol{x}$ as the surface integral

$$\boldsymbol{\Sigma}(\boldsymbol{x}, \omega) \equiv \int_\Sigma \{\boldsymbol{P}(\boldsymbol{x}', \omega)\,\mathrm{d}\boldsymbol{S}\,.\,\nabla \exp(\mathrm{i}k_0 r)r^{-1} - \exp(\mathrm{i}k_0 r)r^{-1}\,\mathrm{d}\boldsymbol{S}\,.\,\nabla \boldsymbol{P}(\boldsymbol{x}, \omega)\} \tag{2.7b}$$

in which $\boldsymbol{P}(\boldsymbol{x}, \omega)$, the induced polarization in V, is the solution of (2.1), and is in general made up of both transverse and longitudinal parts:

$$\boldsymbol{P}(\boldsymbol{x}, \omega) = \boldsymbol{P}_t(\boldsymbol{x}, \omega) + \boldsymbol{P}_l(\boldsymbol{x}, \omega) \tag{2.8a}$$

and $\boldsymbol{P}_t$ and $\boldsymbol{P}_l$ have the Fourier transforms on the variable $\boldsymbol{x}$ given by (2.4):

$$\boldsymbol{P}(\boldsymbol{k}, \omega) = \boldsymbol{P}_t(\boldsymbol{k}, \omega) + \boldsymbol{P}_l(\boldsymbol{k}, \omega). \tag{2.8b}$$

The problem to be solved in this section is therefore this: the induced polarization $\boldsymbol{P}(\boldsymbol{x}, \omega)$ of (2.8*a*) with Fourier transform given by (2.8*b*) together with (2.4) is already proved to be a potential solution of the fundamental equation (2.1): this form for $\boldsymbol{P}(\boldsymbol{x}, \omega)$ is consistent with but also demands the dispersion relations (2.5), for this

† The solution of (2.1) when $g(r) \equiv 1$, the case first considered in I, is the 'molecular field' approximation: equation (2.1) includes additional two-body correlations: the solution of the most general integral equation includes many-particle correlation of all orders (compare Brout 1965—p. 72).

‡ The complication is that of the surface terms of Bullough *et al.* (1968) and Bullough and Hynne (1968).

result was the main result of I. The problem now is to show that the potential solution (2.8*a*) satisfies the extinction theorem (2.7). Such a demonstration would show that the potential solution (2.8*a*) of (2.1) is indeed an actual solution of (2.1); and this would show that the physically important result, the dispersion relations (2.5), are valid relations.

Before we attack the problem we make two additional preliminary remarks. The first concerns notation. We shall find it convenient to work sometimes in $\boldsymbol{k}$ space and sometimes with *single modes* in $\boldsymbol{x}$ space. Although the two things are the same the technical connection between the two is slightly cumbersome. A single mode of polarization of wave vector $\boldsymbol{k}'$ and with longitudinal and transverse parts is

$$\boldsymbol{P}(\boldsymbol{x}, \omega) = \boldsymbol{P}_{\mathrm{t}0}(\hat{\boldsymbol{k}}', \omega)\exp(\mathrm{i}\boldsymbol{k}' \cdot \boldsymbol{x}) + \boldsymbol{P}_{l0}(\hat{\boldsymbol{k}}', \omega)\exp(\mathrm{i}\boldsymbol{k}' \cdot \boldsymbol{x}). \tag{2.8c}$$

The Fourier transform of this is

$$\boldsymbol{P}(\boldsymbol{k}, \omega) = (2\pi)^3\, \boldsymbol{P}_{\mathrm{t}0}(\hat{\boldsymbol{k}}', \omega)\delta(\boldsymbol{k}-\boldsymbol{k}') + (2\pi)^3\, \boldsymbol{P}_{l0}(\hat{\boldsymbol{k}}', \omega)\delta(\boldsymbol{k}-\boldsymbol{k}'). \tag{2.8d}$$

It is usually convenient to write

$$\delta(\boldsymbol{k}-\boldsymbol{k}') = k^{-2}\delta(k-k')\delta(\hat{\boldsymbol{k}}-\hat{\boldsymbol{k}}').$$

Even so $\boldsymbol{P}_{\mathrm{t}0}(\hat{\boldsymbol{k}}, \omega)$ and $\boldsymbol{P}_{l0}(\hat{\boldsymbol{k}}, \omega)$ in (2.8*d*) differ by extraction of the $(2\pi)^3$ and the angular δ-function from these same symbols in (2.4) when $\boldsymbol{k}' = mk_0\hat{\boldsymbol{k}}$. In practice, however, we shall use (2.8*c*) for an arbitrary single mode of wave vector $\boldsymbol{k}'$ in $\boldsymbol{x}$ space; and we shall also use expressions like (2.4) in $\boldsymbol{k}$ space. This omission or re-interpretation of factors will cause no confusion in practice. Note that because $\boldsymbol{k}'$ is the wave vector of both longitudinal and transverse parts in (2.8*c*) we must have

$$k' = mk_0 = m_{\mathrm{t}}k_0 = m_l k_0$$

in this particular case: usually we treat the two sorts of modes separately.

The second preliminary remark stems from the first. The reason why we wish to work in both $\boldsymbol{k}$ space and $\boldsymbol{x}$ space is the following: $\boldsymbol{k}$ space is ultimately the more convenient for the development of response functions—particularly in terms of the dielectric constants $\epsilon_{l,\mathrm{t}}(\boldsymbol{k}, \omega)$; but the method of Born and Wolf (1959), which was developed in I and made applicable to the solution of the fundamental integral equation now exhibited in (2.1) and to the more general form of that equation considered in I, was applied directly to the polarization $\boldsymbol{P}(\boldsymbol{x}, \omega)$ induced in $\boldsymbol{x}$ space. This quantity is probably the most convenient one for a first discussion of the extinction theorem.

This is so for the following reason: the surface integral $\boldsymbol{\Sigma}$ in (2.7) is a consequence of finite V and the break of translational invariance; but we must break translational invariance until we can show we can consistently do otherwise, and the considerations of § 2 of IV show that this is not straightforward. Because of the breaking of translational invariance $\boldsymbol{\Sigma}$ depends both on the field point $\boldsymbol{x}$ inside or outside V and is a functional of the induced polarization $\boldsymbol{P}(\boldsymbol{x}', \omega)$ at all points $\boldsymbol{x}'$ on the surface Σ of V. Thus the Fourier transform on $\boldsymbol{x}$ of $\boldsymbol{\Sigma}$ depends on the wave vector $\boldsymbol{k}$ and on all the modes $\boldsymbol{k}'$ in the Fourier resolution of $\boldsymbol{P}(\boldsymbol{x}', \omega)$ (as equations (3.3) below, and IV—equation (2.3), later explicitly show). This seems to imply coupling between all the modes: however, we shall see that the extinction theorem is such that in the most general case (considered in IV) there is at most coupling between one mode of wave vector $\boldsymbol{k}$ and two other modes with wave number $m_{\mathrm{t}}k_0$. With these two preliminary remarks out of the way we can now take up our problem.

Equations (2.7) are integro-differential equations for $\boldsymbol{P}(\boldsymbol{x}, \omega)$ when $\boldsymbol{E}(\boldsymbol{x}, \omega)$, the probe, is given: alternatively we can choose $\boldsymbol{P}(\boldsymbol{x}, \omega)$ the response and show that there is a physically reasonable $\boldsymbol{E}(\boldsymbol{x}, \omega)$ determined from it by (2.7*a*). Because we presently choose the single mode solution (2.8*c*) we implicitly adopt the second point of view; fortunately, because the response to a given probe is the important physical quantity, we find that after a suitable correction of the one-mode solution (2.8*c*) we do not really prejudice the solution of (2.7) for given $\boldsymbol{E}(\boldsymbol{x}, \omega)$ by using just that one mode.

We recall from I that so far $\boldsymbol{E}(\boldsymbol{x}, \omega)$ is an incident light (i.e. free) field and so is itself not quite arbitrary: it satisfies (I—equation (3.3))

$$\operatorname{div} \boldsymbol{E}(\boldsymbol{x}, \omega) = 0 \tag{2.9a}$$

$$(\nabla^2 + k_0^2)\boldsymbol{E}(\boldsymbol{x}, \omega) = \boldsymbol{0}. \tag{2.9b}$$

Equation (2.9*b*) implies the free-field dispersion relation $k = \omega c^{-1}$ for every mode $\boldsymbol{k}$ of $\boldsymbol{E}(\boldsymbol{x}, \omega)$.

Since (2.7*b*) is a surface integral taken over the boundary Σ of the region of volume V containing the fluid, we can expect to be able to choose two only of V, $\boldsymbol{E}$ and $\boldsymbol{P}$. In addition to $\boldsymbol{P}$ we therefore choose V and take it to be the very definite form of the parallel-sided slab $-c \leqslant z \leqslant d$, $x^2+y^2 \leqslant R^2$: the coordinates (x, y, z) are cartesian components of the vector $\boldsymbol{x}$: the slab is infinite in any two linearly independent directions orthogonal to the z axis in the sense that we take the limit $R \to \infty$. Thus V is not strictly finite now but is still adequate for our purpose.

For such a region V, providing we can assume that we can neglect any finitely oscillating terms from the surface $x^2+y^2 = R^2$ as $R \to \infty$, we find easily (cf. Bullough 1962—Appendix 1) that for all wave vector directions along the positive z axis (the direction $\hat{\boldsymbol{k}}_1$ (say) which is the axial direction of the slab) equation (2.8*c*) implies from the definition (2.7*b*) that

$$\begin{aligned}\boldsymbol{\Sigma}(\boldsymbol{x}, \omega) = {} & [-2\pi \exp\{-\mathrm{i}(m_\mathrm{t}-1)k_0 c\} \exp(\mathrm{i}k_0 z)(1+m_\mathrm{t}) - 2\pi \exp\{\mathrm{i}(m_\mathrm{t}+1)k_0 d\} \\ & \times \exp(-\mathrm{i}k_0 z)(1-m_\mathrm{t})]\boldsymbol{P}_\mathrm{t}(\hat{\boldsymbol{k}}_1, \omega) + [-2\pi \exp\{-\mathrm{i}(m_l-1)k_0 c\} \\ & \times \exp(\mathrm{i}k_0 z)(1+m_l) - 2\pi \exp\{\mathrm{i}(m_l+1)k_0 d\} \exp(-\mathrm{i}k_0 z)(1-m_l)]\boldsymbol{P}_l(\hat{\boldsymbol{k}}_1, \omega)\end{aligned} \tag{2.10}$$

for all $\boldsymbol{x}$ inside V.

Since $\boldsymbol{P}_\mathrm{t}$ and $\boldsymbol{P}_l$ are respectively transverse and longitudinal and their wave vectors are confined to the axial direction $\hat{\boldsymbol{k}}_1$ of the slab, we have by definition that

$$\boldsymbol{P}_\mathrm{t} \cdot \hat{\boldsymbol{k}}_1 = 0, \qquad \boldsymbol{P}_l \times \hat{\boldsymbol{k}}_1 = \boldsymbol{0}.$$

When (2.10) is substituted into (2.7*a*) we then simply get

$$\begin{aligned}\boldsymbol{0} = {} & (m_\mathrm{t}^2-1)\boldsymbol{E} + [-2\pi \exp\{-\mathrm{i}(m_\mathrm{t}-1)k_0 c\} \exp(\mathrm{i}k_0 z)(1+m_\mathrm{t}) - 2\pi \exp\{\mathrm{i}(m_\mathrm{t}-1)k_0 d\} \\ & \times \exp(-\mathrm{i}k_0 z)(1-m_\mathrm{t})]n\boldsymbol{P}_{\mathrm{t}0}(\hat{\boldsymbol{k}}_1, \omega).\end{aligned} \tag{2.11}$$

The important point is that $\boldsymbol{P}_{l0}(\hat{\boldsymbol{k}}_1, \omega)$ does not appear at all and so there is no constraint upon it.† The reason for this is that the differential operator operating on $\boldsymbol{\Sigma}(\boldsymbol{x}, \omega)$ in (2.7*a*) ensures that every mode of wave vector direction $\pm\hat{\boldsymbol{k}}_1$ and wave

† Except only that the wave vector direction is $\hat{\boldsymbol{k}}_1$.

number k_0 is strictly transverse to z and $\hat{\boldsymbol{k}}_1$ so that (2.11) contains examples of each of these modes and no others. It follows that the extinction theorem has now no relevance to the longitudinal modes: it is in this 'particular and limited sense' that the extinction theorem 'becomes irrelevant to the longitudinal modes.'†

A consequence of (2.11) is that $\boldsymbol{E}$ is also transverse to $\hat{\boldsymbol{k}}_1$ and has wave number k_0 and this is wholly consistent with (2.9): the point here however is that (2.11) implies that $\boldsymbol{E}$ *must* satisfy (2.9) if (2.8*c*) satisfies (2.7*a*) and so satisfies (2.1). Thus the probe must be light.

We can therefore conclude the following: the longitudinal part of (2.8*c*) which can now take the form

$$\boldsymbol{P}_{l0}(\hat{\boldsymbol{k}}_1, \omega) \exp(im_l k_0 z)$$

is not a forced mode; the associated $\boldsymbol{P}_l(\boldsymbol{k}, \omega)$ of (2.4*a*) is not a forced solution of (2.1); and these longitudinal modes with $\hat{\boldsymbol{k}}$ along the axis of V are normal mode solutions of arbitrary vector amplitude. Obviously it does not matter whether $\hat{\boldsymbol{k}}$ lies along $\hat{\boldsymbol{k}}_1$ or $-\hat{\boldsymbol{k}}_1$: it is sufficient for a normal longitudinal mode that $\hat{\boldsymbol{k}}$ is either parallel or antiparallel to $\hat{\boldsymbol{k}}_1$.‡ At the same time, since $\boldsymbol{E}$ is necessarily light transverse to $\hat{\boldsymbol{k}}_1$, these longitudinal normal modes are never excited by light: we can choose $\boldsymbol{E} \equiv \boldsymbol{0}$, and although this implies $\boldsymbol{P}_{t0} \equiv \boldsymbol{0}$ by (2.11), this does not imply $\boldsymbol{P}_{l0} \equiv \boldsymbol{0}$. It is 'well-known' that longitudinal modes are not excited by light (compare e.g. Pines 1963—pp. 201–2) and this demonstration from the extinction theorem is very satisfying. We show in IV, § 3, however, that this simple situation is a consequence of the choice of $\hat{\boldsymbol{k}}$ parallel or antiparallel to the axis $\hat{\boldsymbol{k}}_1$ of the slab V.

Further since by (2.11) $\boldsymbol{E}$ is light and $\boldsymbol{E} \equiv \boldsymbol{0}$ implies $\boldsymbol{P}_{t0} \equiv \boldsymbol{0}$ the transverse solutions $\boldsymbol{P}_{t0}$ are forced solutions; and if $\boldsymbol{P}_{t0}$ is a single mode of wave vector $\boldsymbol{k}'$, $\hat{\boldsymbol{k}}'$ is along $\hat{\boldsymbol{k}}_1$ and $k' = m_t k_0$. In addition (2.11) shows that, at all points $\boldsymbol{x}$ inside V, $\boldsymbol{E}$ must consist of two modes with wave vectors $\pm k_0 \hat{\boldsymbol{k}}_1$ parallel and antiparallel to the slab axis. Then on physical grounds if $\boldsymbol{E}$ is an imposed field we must impose this field both inside and outside V so that it becomes a physical probe.§ With this choice of $\boldsymbol{E}(\boldsymbol{x}, \omega)$, the transverse part of (2.8*c*) is also a valid solution of (2.1); and since $\boldsymbol{E} \not\equiv \boldsymbol{0}$ this solution is a forced solution.

The most usual and certainly most desirable situation is one in which $\boldsymbol{E}(\boldsymbol{x}, \omega)$ is a single plane monochromatic wave travelling (say) in the direction $\hat{\boldsymbol{k}}_1$ of the *positive* z axis:

$$\boldsymbol{E}(\boldsymbol{x}, \omega) = \boldsymbol{E}_0(\hat{\boldsymbol{k}}_1, \omega) \exp(ik_0 z), \qquad \hat{\boldsymbol{k}}_0 \,.\, \boldsymbol{E}_0 = 0, \qquad \hat{\boldsymbol{k}}_0 = \hat{\boldsymbol{k}}_1. \tag{2.12}$$

In this case we can satisfy (2.11) if, but only if, (2.8*c*) is extended to include a second transverse mode travelling in the direction opposite to $\hat{\boldsymbol{k}}_1$. If this is done we easily find (Bullough 1962—Appendix 1) that, if now

$$\boldsymbol{P}_{t0}(\hat{\boldsymbol{k}}', \omega) \exp(i\boldsymbol{k}' \,.\, \boldsymbol{x}) \rightarrow \boldsymbol{P}_{t0}(\hat{\boldsymbol{k}}_1, \omega)\{\exp(im_t k_0 z) + \Lambda \exp(-im_t k_0 z)\}$$

then

$$\Lambda = \left(\frac{m_t - 1}{m_t + 1}\right) \exp(2im_t k_0 d) \tag{2.14a}$$

† See the Introduction to I.

‡ We find in IV, § 3 that this condition is also necessary.

§ $\boldsymbol{\Sigma}(\boldsymbol{x}, \omega) \neq \boldsymbol{0}$ outside V and then describes waves additional to the probe reflected from the surface Σ and outside V.

and

$$\boldsymbol{P}_{t0}(\hat{\boldsymbol{k}}_1, \omega) = \left\{\exp\{i(m_t-1)k_0c\}\frac{m_t^2-1}{4\pi n} \times \left(\frac{2/(1+m_t)}{[1-\{(m_t-1)/(m_t+1)\}^2\exp\{2im_tk_0(c+d)\}]}\right)\right\}\boldsymbol{E}_0. \quad (2.14b)$$

If we write equation (2.14*b*) in the form

$$\boldsymbol{P}_{t0}(\hat{\boldsymbol{k}}_1, \omega) = \frac{m_t^2-1}{4\pi n}\hat{S}(\Sigma; m_tk_0\hat{\boldsymbol{k}}_1, \omega)\boldsymbol{E}_0$$

we have the response relation in $\boldsymbol{k}$ space

$$\boldsymbol{P}_t(\boldsymbol{k}, \omega) = \frac{m_t^2-1}{4\pi n}\hat{S}(\Sigma; \boldsymbol{k}, \omega)\{\sigma^+(\boldsymbol{k})+\Lambda\sigma^-(\boldsymbol{k})\}\boldsymbol{E}(\boldsymbol{k}, \omega) \quad (2.15)$$

in the particular case when $\boldsymbol{k} = k_0\hat{\boldsymbol{k}}_1$. The quantities $\sigma^{\pm}(\boldsymbol{k})$ are operators: $\sigma^+(\boldsymbol{k})$ changes the wave number k_0 to m_tk_0 (by replacing $k_0^{-2}\delta(k-k_0)$ in $\boldsymbol{E}(\boldsymbol{k}, \omega)$ by $m_t^{-2}k_0^{-2}\delta(k-m_tk_0)$): $\sigma^-(\boldsymbol{k})$ does the same and in addition reverses the direction of the wave vector. The part of the response $\hat{S}(\Sigma; \boldsymbol{k}, \omega)$ depends on the surface Σ as (2.14*b*) shows.

The response relation (2.15) is useful for a later comparison. Here we need observe only that the results (2.14) are the exact results of the phenomenological Maxwell theory in the case of a single plane wave satisfying (2.9) incident normally upon an infinite parallel-sided slab of width $(c+d)$. However, we must emphasize that this result is obtained here not by imposing boundary conditions at the surface of V; for the boundary conditions are those of I, namely sources of $\boldsymbol{E}$ at infinity and an outgoing wave condition on the optical Green's function (2.3) of the theory. It is now plain on physical grounds that the transverse solution must depend very heavily on the form of V and its surface Σ and that we can use (2.8*c*) with (2.12) only because $\hat{\boldsymbol{k}}_1$ is the axis of the simple slab we chose for V. Even so we had to extend (2.8*c*) to include a wave reflected from the interior surface of the slab.

It is noteworthy that the two modes with wave vectors $\pm m_tk_0\hat{\boldsymbol{k}}_1$ correspond to the two obvious roots of (2.5*b*)†: the extinction theorem fixes both the amplitudes of these two modes by (2.14). However, as noted in I, it is by no means clear that (2.5*b*) does not have additional roots for m_t^2 since $J_t(m_tk_0, \omega)$ depends on m_t^2.† If this is so the most general solution of (2.1) is a linear combination of modes with wave numbers $\pm m_tk_0$ made from the different roots m_t, perhaps heavily damped, but still existing near the surface of V. There is no sign that more than two amplitudes can be fixed by the extinction theorem. This point is not understood, but it may be associated with the additional solutions which arise through the breakdown of the translational invariance of the correlation functions: where there is no correlation there are precisely two roots for m_t (see I—equation (3.7)). The problem associated with the breakdown of the translational invariance of the correlation functions will be looked at later in this series.

This completes our analysis of the validity of the modes (2.4) as solutions of the integral equation (2.1): the modes (2.4) *are* valid solutions.

† $J_t(m_tk_0, \omega)$ contains only even powers of m_t.

Thus in summary of this section we conclude that providing that $\hat{\boldsymbol{k}}$ lies parallel or antiparallel to the axis of the slab V all the modes (2.4) which were derived in the paper I are acceptable modes. The longitudinal modes (2.4*a*) are normal modes; and because there is no field $\boldsymbol{E}$ outside V associated with these modes† there is no energy flux across the surface of V and these modes cannot transmit energy.‡ It is hard to see, however, whether or not the longitudinal modes are still true normal modes at the delicate theoretical level at which we are obliged to discuss the scattering and whether there is any inconsistency there; for it is not clear whether the important physical roots of (2.5*a*) for m_l can be the purely real roots necessary for true normal modes and consistency. The quantities $J_l(k, \omega)$ on which $m_l(\omega)$ depends are complex for arbitrary k and ω and are very much concerned with external scattering processes (cf. II, Bullough *et al.* 1968 and Bullough and Hynne 1968) but this does not of itself eliminate the chance of purely real roots for the physical roots of (2.5*a*) when $k = m_l k_0$. This delicate question is thus still open.

In contrast with the longitudinal modes, the transverse modes (2.4*b*) with wave vector direction $\hat{\boldsymbol{k}}$ parallel to the axis of the slab V are forced modes excited by a strictly transverse free field (light). This free field contains precisely two modes with wave vectors parallel and anti-parallel to the axis of the slab. In the case when the imposed free field contains a single mode incident along the axis of the slab two distinct modes like (2.4*b*) with wave vectors parallel and antiparallel to the axis of the slab are induced in that slab. In both cases the most important physical roots for the refractive index m_t of these forced modes are certainly complex due to external scattering.§ The microscopic theory otherwise exhibits all the features of the Maxwell phenomenological theory but the boundary conditions are only outgoing boundary conditions on the fundamental microscopic integral equation (2.1).

The contrast between the characters of the two classes of modes raises the question of the existence of forced longitudinal solutions and of normal transverse solutions. There is also the question of the possibility of additional modes when the probing field is not light. We consider these problems in § 3 and the earlier sections of the following paper IV still keeping the parallel-sided slab for the region V and still restricting the wave vector directions $\hat{\boldsymbol{k}}$ parallel to the axis of the slab: in § 3 of IV we finally relax this condition on $\hat{\boldsymbol{k}}$.

3. Forced longitudinal modes and $\epsilon_l(k, \omega)$

We now want to investigate whether the integral equation (2.1) will admit longitudinal solutions forced by a longitudinal field $\boldsymbol{E}_l$. The forcing fields so far considered in I were light fields satisfying (2.9) and were necessarily transverse. Now $\boldsymbol{E}_l$ cannot satisfy (2.9*a*), free charge density $\rho(\boldsymbol{x}, \omega)$ must be present, continuity then implies a free longitudinal current density $\boldsymbol{j}_l(\boldsymbol{x}, \omega)$ and $\boldsymbol{E}_l$ will not satisfy (2.9*b*). We have instead of (2.9)

$$\operatorname{div} \boldsymbol{E}_l(\boldsymbol{x}, \omega) = 4\pi\rho(\boldsymbol{x}, \omega) \tag{3.1a}$$

$$\operatorname{curl} \boldsymbol{E}_l(\boldsymbol{x}, \omega) = \boldsymbol{0} \tag{3.1b}$$

† The transversality condition eliminates $\boldsymbol{\Sigma}(\boldsymbol{x}, \omega)$ both inside and outside V.

‡ There is no indication that the modes must be coupled into standing waves in order to transmit no energy.

§ We have yet to prove this here; but the report (Bullough *et al.* 1968 and Bullough and Hynne 1968) already shows how complete the scattering theory which depends on this fact actually is.

and the longitudinal condition (3.1*b*) means that there is at most a constant magnetic field associated with the probe. We shall choose this field to be zero everywhere.

It is the aim of this section to derive a longitudinal response function and it is now desirable to work wholly in $\boldsymbol{k}$ space. This introduces the complication associated with the extinction theorem noted in § 2, but this is soon eliminated for the longitudinal modes by working with single modes like the longitudinal mode of (2.8*c*) (but in $\boldsymbol{k}$ space), with the slab V, and with the single mode wave vector $\boldsymbol{k}$ along the slab axis.

The Fourier transform of equations (3.1) is straightforward: it is

$$\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{E}_l(\boldsymbol{k},\,\omega) = 4\pi\rho(\boldsymbol{k},\,\omega) \tag{3.2a}$$

$$\mathrm{i}\boldsymbol{k}\times\boldsymbol{E}_l(\boldsymbol{k},\,\omega) = \mathbf{0}. \tag{3.2b}$$

We must now go back to equation (3.11) of I. That equation had not yet included the local field terms $J_l(k,\,\omega)$ and $J_t(k,\,\omega)$: since they always add to the Lorentz field $(4\pi/3)n\alpha$ term it is clear where they go in the theory and we shall also omit them from the argument here for the moment.

The Fourier transform on the position variable $\boldsymbol{x}$ of the equation (3.11) of I is otherwise quite generally

$$\begin{aligned}\boldsymbol{P}(\boldsymbol{k},\,\omega) = {} & \alpha(\omega)\{\boldsymbol{E}(\boldsymbol{k},\,\omega)+(k_0{}^2\mathbf{U}-\boldsymbol{kk})\,.\,4\pi n\boldsymbol{P}(\boldsymbol{k},\,\omega)(k^2-k_0{}^2)^{-1}+(4\pi/3)n\boldsymbol{P}(\boldsymbol{k},\,\omega)\}\\ & +(k_0{}^2\mathbf{U}-\boldsymbol{kk})\,.\,\frac{n\alpha(\omega)}{(2\pi)^3}\int\frac{I(\boldsymbol{k},\,-\boldsymbol{k}';\,\omega)}{k'^2-k_0{}^2}\boldsymbol{P}(\boldsymbol{k}',\,\omega)\,\mathrm{d}\boldsymbol{k}'\end{aligned} \tag{3.3}$$

providing $\boldsymbol{P}(\boldsymbol{k}',\,\omega) = \mathrm{O}(k'-k_0)$ or better in the neighbourhood of $k' = k_0$.† In (3.3)

$$\begin{aligned}I(\boldsymbol{k},\,-\boldsymbol{k}',\,\omega) = {} & \int\exp(-\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{x})\,\mathrm{d}\boldsymbol{x}\int_\Sigma[\exp(\mathrm{i}\boldsymbol{k}'\,.\,\boldsymbol{x}')\,\mathrm{d}\boldsymbol{S}\,.\,\nabla_{x'}\\ & \times\{\exp(\mathrm{i}k_0|\boldsymbol{x}-\boldsymbol{x}'|)/|\boldsymbol{x}-\boldsymbol{x}'|\}-\{\exp(\mathrm{i}k_0|\boldsymbol{x}-\boldsymbol{x}'|)/|\boldsymbol{x}-\boldsymbol{x}'|\}\,\mathrm{d}\boldsymbol{S}'\,.\,\nabla_{x'}\\ & \times\exp(\mathrm{i}\boldsymbol{k}'\,.\,\boldsymbol{x}')]\end{aligned} \tag{3.4}$$

and is thus the scalar magnitude of the Fourier transform on $\boldsymbol{x}$ of the vector quantity $\boldsymbol{\Sigma}(\boldsymbol{x},\,\omega)$ of (2.7*b*) now evaluated for a single mode of wave vector $\boldsymbol{k}'$ and unit amplitude.

Careful inspection of (3.3) shows that it is the Fourier transform of the fundamental integral equation (2.1) in which $g(r)$ is replaced by unity: this introduces divergence at $\boldsymbol{x}' = \boldsymbol{x}$ in the integrand there, and this is the source of the $(4\pi/3)n\boldsymbol{P}(\boldsymbol{k},\,\omega)$ term. It is, however, the Fourier transform of the Green's function $\exp(\mathrm{i}k_0r)r^{-1}$ on the finite region V (breaking translational invariance so that $r = |\boldsymbol{x}-\boldsymbol{x}'|$ must be treated as a function of $\boldsymbol{x}$ and $\boldsymbol{x}'$ separately) which introduces all the complication of the extinction theorem: as noted in § 2 this complication now takes the form of the two wave vectors $\boldsymbol{k}$ and $\boldsymbol{k}'$ in (3.3). We cannot avoid this complication until we can see we need not break translational invariance: so far the argument of § 2 shows that we actually *need* this complication if the external probe is light since otherwise the equations cannot be satisfied.

No parameter m appears in (3.3) since there are not yet any conditions on the induced polarization $\boldsymbol{P}(\boldsymbol{x},\,\omega)$ in V: in particular we do *not* assume as we did in I that

† It is mathematically convenient and physically sensible to exclude the case $k = k_0$ from $\boldsymbol{P}(\boldsymbol{k},\,\omega)$: we believe the theory for the longitudinal modes can be carried through without this restriction at the expense of an increase in complication: what happens to the transverse response of IV, § 2 for $k = k_0$ is considered there.

$\boldsymbol{P}(\boldsymbol{x}, \omega)$ necessarily satisfies in $\boldsymbol{x}$ space the condition

$$(\nabla^2 + m^2 k_0{}^2)\boldsymbol{P}(\boldsymbol{x}, \omega) = \boldsymbol{0}. \tag{3.5}$$

If this condition is to apply it must emerge now from its Fourier transform in (3.3).†

We restrict the polarization induced by $\boldsymbol{E}_l$ to a single set of modes with wave vector $\boldsymbol{k}$ parallel to the axis of the slab V and we are concerned only with its longitudinal part. Thus $\boldsymbol{P}(\boldsymbol{k}, \omega)$ is $\boldsymbol{P}_l(\boldsymbol{k}, \omega)$, longitudinal with a δ-function restricting the direction $\hat{\boldsymbol{k}}$ of $\boldsymbol{k}$ to the slab axis; since (3.5) does not apply there is no restriction on $k = |\boldsymbol{k}|$ (except that $k \neq k_0$). In this case the surface integral in (3.4) yields for every $\boldsymbol{k}'$ parallel to the slab axis two modes with wave vectors $\pm \boldsymbol{k}_0$ parallel to the slab axis (and wave number k_0) and (3.4) is a δ-function restricting the directions $\hat{\boldsymbol{k}}$ parallel or antiparallel to the slab axis: it is also a δ-function restricting the wave number k to the value k_0. Since $\hat{\boldsymbol{k}}$ and $\boldsymbol{P}_l$ are parallel to the slab axis the factor $(\boldsymbol{kk} - k_0{}^2\mathbf{U})$ is a transversality factor and eliminates the whole term. This argument exactly parallels the argument for the elimination of the surface integral for longitudinal modes in § 2.

Thus it follows that with $\boldsymbol{P}_l(\boldsymbol{k}, \omega)$ and hence $\hat{\boldsymbol{k}}$ parallel to the slab axis equation (3.3) reduces to

$$\boldsymbol{P}_l(\boldsymbol{k}, \omega) = \alpha(\omega)\{\boldsymbol{E}_l(\boldsymbol{k}, \omega) - (8\pi/3)n\boldsymbol{P}_l(\boldsymbol{k}, \omega)\}. \tag{3.6}$$

The field $\boldsymbol{E}_l$ is to satisfy (3.2) (and because we choose $\boldsymbol{P}$ to vanish as $\mathrm{O}(k - k_0)$ conveniently at $k = k_0$, $\boldsymbol{E}_l(\boldsymbol{k}, \omega)$ and hence $\rho(\boldsymbol{k}, \omega)$ should have the same property).‡ Because $\boldsymbol{P}_l(\boldsymbol{k}, \omega)$ is restricted to a single-mode direction $\boldsymbol{E}_l$ is also.

It follows from (3.6) that $\boldsymbol{P}_l(\boldsymbol{k}, \omega)$ is a forced solution of the integral equation (2.1) with $g(r) \equiv 1$ now if and only if

$$\boldsymbol{P}_l(\boldsymbol{k}, \omega) = [\alpha(\omega)\{1 + (8\pi/3)n\alpha(\omega)\}^{-1}]\boldsymbol{E}_l(\boldsymbol{k}, \omega) \tag{3.7}$$

and it is plain that the proper generalization to obtain a forced solution of that fundamental integral equation with $g(r)$ a typical pair correlation function is

$$\boldsymbol{P}_l(\boldsymbol{k}, \omega) = [\alpha(\omega)\{1 + (8\pi/3)n\alpha(\omega) - n\alpha(\omega)J_l(k, \omega)\}^{-1}]\boldsymbol{E}_l(\boldsymbol{k}, \omega). \tag{3.8}$$

For present purposes it is not necessary to restrict J_l to the two-body interaction of the definition (2.6*a*) and we can now think of it as having the much greater generality achieved in I in which J_l became a sum of many-particle interactions (multiple-scattering processes) of all orders. (I – equations (4.17), (4.18) and (4.19)).

Since $\rho(\boldsymbol{x}, \omega)$ is an arbitrary imposed free charge density its Fourier transform is arbitrary: it follows from (3.2) that

$$\boldsymbol{E}_l(\boldsymbol{k}, \omega) = \frac{\hat{\boldsymbol{k}}4\pi\rho(\boldsymbol{k}, \omega)}{\mathrm{i}k} \tag{3.9}$$

and no dispersion relation connects the variables $\boldsymbol{k}$ and ω (although some connection may be implied by the particular choice of $\rho(\boldsymbol{x}, \omega)$).§ Thus the quantity in square brackets in (3.8) is a linear response function connecting the polarization $\boldsymbol{P}_l$ induced by the probing charge density $\rho(\boldsymbol{k}, \omega)$, and $\boldsymbol{k}$ and ω are essentially free variables. This response function has the property we expect, namely that normal mode solutions

† It does of course when the probe is *light*: see IV—§ 2.

‡ Recall that an incident fast particle approximates to a *transverse* electromagnetic wave (Williams 1935).

§ The Fourier transform of $\delta(\boldsymbol{x} - \boldsymbol{v}t)$ is $2\pi\delta(\omega - \boldsymbol{k} \cdot \boldsymbol{v})$ for a point charge travelling with velocity $\boldsymbol{v}$ for example.

are possible whenever $\boldsymbol{k}$ and ω lie on the surfaces of zeros of its denominator: these surfaces are the roots of

$$1+(8\pi/3)n\alpha(\omega)-n\alpha(\omega)J_l(k,\omega) = 0 \tag{3.10}$$

and this is precisely the longitudinal dispersion relation (2.5*a*).

If we define the response function implicit in (3.8) as the significant analytical quantity we do not conform to current usage (e.g. Nozières and Pines 1958, Shultz 1963—chap. 3, § H) in the many-body theory of the electron gas. We therefore argue as follows: we first introduce a *formal* pseudo-Maxwell macroscopic electric field vector $\bar{\boldsymbol{E}}_l(\boldsymbol{x},t)$ with transform $\bar{E}_l(\boldsymbol{k},\omega)$. This is defined to satisfy the following pseudo-macroscopic equations:

$$\mathrm{i}\boldsymbol{k}\,.\,\epsilon_l(\boldsymbol{k},\omega)\bar{\boldsymbol{E}}_l(\boldsymbol{k},\omega) = 4\pi\rho(\boldsymbol{k},\omega) \tag{3.11a}$$

$$\mathrm{i}\boldsymbol{k}\{\bar{\boldsymbol{E}}_l(\boldsymbol{k},\omega)+4\pi n\boldsymbol{P}_l(\boldsymbol{k},\omega)\} = 4\pi\rho(\boldsymbol{k},\omega) \tag{3.11b}$$

in which $\epsilon_l(\boldsymbol{k},\omega)$ is a formal scalar $(\boldsymbol{k},\omega)$-dependent dielectric constant. We emphasize that the equations (3.11) are formal definitions: it does not follow that $\epsilon_l(\boldsymbol{k},\omega)$ has any of the physical significance associated with $m_t^2(\omega)$ for example. At the end of this section we note however that in the long-wavelength low-frequency limit both ϵ_l and m_t^2 become the frequency-dependent dielectric constant $\epsilon(\omega)$ identifiable with that of Maxwell phenomenological theory; and we elaborate on this (as the complex dielectric constant approximation of II) in V. The first equations (3.11) yields (for scalar ϵ_l $(\boldsymbol{k},\omega)$)

$$\mathrm{i}\boldsymbol{k}\,.\,\bar{\boldsymbol{E}}_l(\boldsymbol{k},\omega) = \frac{4\pi\rho(\boldsymbol{k},\omega)}{\epsilon_l(\boldsymbol{k},\omega)} \tag{3.12a}$$

in contrast with (3.2*a*); the second yields

$$\mathrm{i}\boldsymbol{k}\,.\,\bar{\boldsymbol{E}}_l(\boldsymbol{k},\omega) = 4\pi\{\rho(\boldsymbol{k},\omega)-\mathrm{i}\boldsymbol{k}\,.\,n\boldsymbol{P}_l(\boldsymbol{k},\omega)\}. \tag{3.12b}$$

With $\hat{\boldsymbol{k}}$ restricted to the axis of the slab V we can use (3.8) to reduce the second equation to

$$\begin{aligned}\mathrm{i}\boldsymbol{k}\,.\,\bar{\boldsymbol{E}}_l(\boldsymbol{k},\omega) &= 4\pi\rho(\boldsymbol{k},\omega)-4\pi n\alpha(\omega)\{1+(8\pi/3)n\alpha(\omega)-n\alpha(\omega)J_l(k,\omega)\}^{-1}\\ &\quad\times\{\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{E}_l(\boldsymbol{k},\omega)\}\end{aligned} \tag{3.13a}$$

and from (3.2*a*) we then get

$$\begin{aligned}\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{E}_l(\bar{\boldsymbol{k}},\omega) &= 4\pi\rho(\boldsymbol{k},\omega)\{1-(4\pi/3)n\alpha(\omega)-n\alpha(\omega)J_l(k,\omega)\}\{1+(8\pi/3)n\alpha(\omega)\\ &\quad-n\alpha(\omega)J_l(k,\omega)\}^{-1}.\end{aligned} \tag{3.13b}$$

Equation (3.12*a*) now means that

$$\epsilon_l(\boldsymbol{k},\omega)-1 = \frac{4\pi n\alpha(\omega)}{1-(4\pi/3)n\alpha(\omega)-n\alpha(\omega)J_l(k,\omega)} \tag{3.14}$$

in which $\boldsymbol{k}$ and ω are free variables.

With this result the response function takes the compact form

$$\{1-1/\epsilon_l(\boldsymbol{k},\omega)\}(4\pi n)^{-1}$$

for equation (3.7) becomes

$$4\pi n \boldsymbol{P}_l(\boldsymbol{k}, \omega) = \{1 - 1/\epsilon_l(\boldsymbol{k}, \omega)\}\boldsymbol{E}_l(\boldsymbol{k}, \omega) \tag{3.15a}$$

$$= \{1 - 1/\epsilon_l(\boldsymbol{k}, \omega)\}\frac{\hat{\boldsymbol{k}}4\pi\rho(\boldsymbol{k}, \omega)}{\mathrm{i}k}. \tag{3.15b}$$

Note that the singularities of $\epsilon_l(\boldsymbol{k}, \omega)$ (if these are poles) are not singularities of the response function: indeed the singularities of $\epsilon_l(\boldsymbol{k}, \omega)$ appear to have no physical significance and this illustrates the point that $\epsilon_l(\boldsymbol{k}, \omega)$ is simply a formal construction. On the other hand the surfaces of singularity of the response function are the zeros of $\epsilon_l(\boldsymbol{k}, \omega)$ and these are the dispersion relations for longitudinal modes (2.5*a*). This well-accepted property of the macroscopic dielectric constant $\epsilon(\omega)$ has therefore been taken over by $\epsilon_l(\boldsymbol{k}, \omega)$.†

In contrast (3.14) is almost exactly the *transverse* dispersion relation (2.5*b*) for $m_t^2(\omega)$. It differs only in the usually very small numerical difference between $J_l(k, \omega)$ and $J_t(m_t k_0, \omega)$ when $k \simeq m_t k_0$ (recall from I that $J_l(0, \omega) = J_t(0, \omega)$ and $J_t(k, \omega)$ and $J_l(k, \omega)$ depend weakly on k). Conceptually (3.14) and (2.5*b*) are quite different of course: equation (2.5*b*) is a dispersion relation relating k and ω for transverse modes and, as (2.15) shows, $m_t^2 - 1$ is part of a rather complicated response function relating in terms of specific surface-dependent quantities the single transverse forcing field mode $\boldsymbol{E}(\boldsymbol{k}, \omega)$ (light) normally incident upon the slab V to the two transverse modes of dipole moment it induces: equation (3.14) is a formal expression which expresses the relation of a formal Maxwell field $\bar{E}_l(\boldsymbol{k}, \omega)$ to the probing field $\rho(\boldsymbol{k}, \omega)$ by (3.12*a*), it is independent of surface effects (which vanished with the surface integral of the extinction theorem) and is strictly longitudinal. Equations (3.15) show the response in longitudinal dipole moment.

Equation (3.15*b*) is formally comparable with the usual definition of the $(\boldsymbol{k}, \omega)$-dependent dielectric constant in the form

$$\rho_{\text{total}}(\boldsymbol{k}, \omega) \equiv \rho(\boldsymbol{k}, \omega) - \mathrm{i}\boldsymbol{k} \,.\, n\boldsymbol{P}_l(\boldsymbol{k}, \omega)$$

$$= \frac{\rho(\boldsymbol{k}, \omega)}{\epsilon_l(\boldsymbol{k}, \omega)} \tag{3.16}$$

(compare, e.g. Schultz 1963—p. 81). This demonstrates the firm structural connection between the work on $\epsilon(\boldsymbol{k}, \omega)$ for the plasma and the theory presented here for the molecular fluid. The connection is no idle one: formulae for electron stopping power and longitudinal contributions to the binding energy carry straight over and can be extended to include the radiation field as reported in II. But this does not mean that the extension from the plasma is trivial: the point of course is that complicated features specific to the theory of the molecular fluid are merely concealed in $\epsilon_l(\boldsymbol{k}, \omega)$ in (3.14). There is even some contribution in $J_l(k, \omega)$ from the intermolecular radiation field concealed there.

In the following paper IV we derive the natural transverse analogue of (3.14):

$$\epsilon_t(\boldsymbol{k}, \omega) - 1 = \frac{4\pi n\alpha(\omega)}{1 - (4\pi/3)n\alpha(\omega) - n\alpha(\omega)J_t(k, \omega)}. \tag{3.17}$$

† We observed in I that the zeros of $m_t^2(\omega)$ do not quite yield what is in fact the *dispersion relation* which is (2.5*a*).

This means that we then have the following results:

$$\epsilon_l(\mathbf{0}, \omega) = \epsilon_t(\mathbf{0}, \omega) \tag{3.18a}$$

always, and

$$\epsilon_l(\mathbf{0}, \omega) = \epsilon_t(\mathbf{0}, \omega) = m_t^2(\omega) \tag{3.18b}$$

when but only when $k_0 = \omega c^{-1}$ is much less than a reciprocal correlation length l^{-1}.† The result (3.18*b*) is that of the physically important complex dielectric constant approximation (see II, and V—§ 3) and it is this alone which enables us to ascribe any physical significance to $\epsilon_l(\boldsymbol{k}, \omega)$ or $\epsilon_t(\boldsymbol{k}, \omega)$. It is not even true (cf. Pines 1963—equation (4.66)) that $\epsilon_{l,t}(\mathbf{0}, \omega) = m_t^2(\omega)$ when we wish to consider the $\boldsymbol{k}$-dependent external scattering of light from a molecular fluid at optical frequencies (Bullough and Hynne 1968). Thus the dielectric constants must not be confused with either $m_t^2(\omega)$ or its low-frequency analogue the complex frequency-dependent dielectric constant $\epsilon(\omega)$.

We shall find in the following paper IV, however, that the most significant difference between the response function (3.15) in which $\boldsymbol{k}$ and ω are free variables, and the response function (2.15) which depends on ω alone, is rooted in the extinction theorem. We have reached (3.15) only because we have restricted the theory of longitudinal modes to wave vectors $\boldsymbol{k}$ which eliminate this awkward feature from the argument. But we find in IV that no such simplification is possible when we try to introduce the transverse dielectric constant $\epsilon_t(\boldsymbol{k}, \omega)$. There is still a natural way to reach (3.17), however.

The main conclusions of this § 3 are, first, that as well as the normal modes considered so far there can be in the molecular fluid additional forced longitudinal modes: the existence of both types of mode has been demonstrated only for those modes with wave vector $\boldsymbol{k}$ parallel to the axis of the slab V. Second, (for such $\boldsymbol{k}$ at least) there is a natural $(\boldsymbol{k}, \omega)$-dependent longitudinal dielectric constant $\epsilon_l(\boldsymbol{k}, \omega)$ for the molecular fluid: it satisfies the relation (3.14). The longitudinal response function, which is $1-1/\epsilon_l(\boldsymbol{k}, \omega)$ has the longitudinal normal mode dispersion relations (2.5*a*) as its surfaces of singularity.

We now go on to develop the theory of the natural transverse dielectric constant $\epsilon_t(\boldsymbol{k}, \omega)$ for the molecular fluid in the following paper IV.

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† $2\pi l$ is the typical dimension of the clusters in the cluster integrals of $J_{t,l}(k, \omega)$.

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