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Many-body optics

IV. The total transverse response and $\epsilon_t(\mathbf{k}, \omega)$

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Abstract. It is shown that besides the optical modes which can be excited by light there can be additional transverse electromagnetic modes in a molecular fluid. These can be excited by incident transverse currents but are not then excited alone: it is shown that the total transverse optical response is composed of two parts, a translationally invariant part (the virtual response) in which \mathbf{k} and ω are free, and a second part (the real response) which depends on the surface geometry.

The virtual response can be expressed in terms of a single dielectric constant $\epsilon_t(\mathbf{k}, \omega)$. A formula for $\epsilon_t(\mathbf{k}, \omega)$ for a molecular fluid is given in terms of cluster integrals to all orders; it is a natural counterpart of the formula for $\epsilon_l(\mathbf{k}, \omega)$ given previously; but $\epsilon_t(\mathbf{k}, \omega)$ and $\epsilon_l(\mathbf{k}, \omega)$ coincide only in the complex dielectric constant approximation when they are both now identical with the square of the transverse refractive index. The surfaces of singularity (essentially poles for given \mathbf{k}) of the transverse virtual response are exactly the transverse dispersion relations obtained previously for all choices of ω .

The virtual transverse modes are *not* excited by incident light: the virtual response to light is zero. The additional real response to transverse currents is a consequence of the optical Extinction Theorem and the breakdown of translational invariance; it depends on \mathbf{k} , ω , the surface geometry and the transverse refractive index $m_t(\omega)$. When the transverse probe is light the real response coincides with the transverse optical response calculated previously. Thus the real response is an essential part of the total response.

All the results are generalized to the case when \mathbf{k} is not normal to the surface of the system: only the real response is changed and it changes only in so far as it depends on that changed surface geometry. The longitudinal response considered previously only for \mathbf{k} normal to the surface now also consists of two parts: the translationally invariant virtual longitudinal response is unchanged but is accompanied by a *transverse* real response. This sort of result must apparently mean that when \mathbf{k} is not normal to the surface (transverse) light can couple to those longitudinal modes which satisfy the longitudinal dispersion relation calculated previously.

We discuss the existence of optical normal modes in a molecular fluid. We conclude that there are no optical normal modes in any geometry within the precise terms of outgoing boundary conditions considered: all such modes would be real modes and these scatter light out of the surface. But we also show that we can at least find longitudinal normal modes within a rectangular dielectric cavity if acausal boundary conditions are admissible.

We extend the theory of the total virtual response in the following paper V.

1. Introduction

In the previous paper (Bullough 1970 a—to be referred to as III) we developed the theory of forced longitudinal modes in a molecular fluid confined to a region V . The forcing probe was an incident charge density and the region V was a parallel-sided slab. We could consider only those modes whose wave vector direction $\hat{\mathbf{k}}$ lay parallel or anti-parallel to the axis of the slab because of the constraint of the optical

Extinction Theorem. This constraint in general depended on the form of the surface Σ of V but vanished for longitudinal modes in the slab when $\hat{\mathbf{k}}$ was parallel or anti-parallel to the slab axis. Because of this the system could respond to the probing charge density with forced longitudinal modes and a linear response function independent of Σ could be defined in terms of a longitudinal dielectric constant $\epsilon_l(\mathbf{k}, \omega)$; acceptable normal modes with axial $\hat{\mathbf{k}}$ lay on the surfaces $\epsilon_l(\mathbf{k}, \omega) = 0$ and this was the longitudinal dispersion relation of the earlier paper (Bullough 1968—to be referred to as I).

In contrast the system could also respond to incident light with forced transverse modes. The wave vectors of these satisfied the transverse dispersion relation of I, but the modes nevertheless satisfied a linear response relation which depended on the refractive index $m_t(\omega)$ and very significantly on Σ even when \mathbf{k} was axial. This contrast sharply exposes the questions of whether a natural transverse dielectric constant $\epsilon_t(\mathbf{k}, \omega)$ can be defined and whether a (\mathbf{k}, ω) -dependent linear response relation for forced transverse modes exists.

It is easy to see how to proceed, however, once we acknowledge that there are two sorts of transverse probe and that there should therefore be two sorts of transverse response function.† The transverse response considered so far is a response to incident light: the longitudinal response is a response to a primary probe which is not an electric field but a free-charge density. We can only impose free-charge density on a molecular fluid by firing free charge into the system. A moving charge system is compounded of both longitudinal and transverse currents. It is the purpose of this paper IV, now, to investigate the response of the system to such an imposed transverse current density $\mathbf{j}_t(\mathbf{k}, \omega)$.

We shall find that the principal problem facing us in this program is that of satisfying the optical extinction theorem. For there is now no choice of wave vector direction $\hat{\mathbf{k}}$ which can eliminate this constraint from the theory. We shall nevertheless manage to show that we can use the surface Σ of V to couple external light to the system and still be able to introduce a surface-dependent (\mathbf{k}, ω) -dependent response to an arbitrary transverse field.

We shall now attack this complex of problems. We adopt the notation, results and definitions of III without additional remark. The plan of this paper IV is already outlined in § 1 of III and we shall finally summarize all of this material at the end of the following paper (Bullough 1970 b—to be referred to as V).

2. The transverse dielectric constant $\epsilon_t(\mathbf{k}, \omega)$

We impose an arbitrary external solenoidal (transverse) current density $\mathbf{j}_t(\mathbf{k}, \omega)$ on V and its exterior; its transform $\mathbf{j}_t(\mathbf{k}, \omega)$ has \mathbf{k} and ω as free variables. Since $\mathbf{j}_t(\mathbf{k}, \omega)$ must describe the motion of point charges there will ultimately be a condition describing that motion connecting \mathbf{k} and ω , but that condition need not concern us here.‡ These primary currents generate a secondary transverse field $\mathbf{E}_t(\mathbf{x}, \omega)$ with transform $\mathbf{E}_t(\mathbf{k}, \omega)$, with \mathbf{k} and ω free so that, although $\mathbf{E}_t(\mathbf{x}, \omega)$ satisfies III,

† Equations (2.19)–(2.23) below show that each response function is a different aspect of the total response function (2.22) which contains both of them.

‡ The Fourier transform of $e\delta(\mathbf{x} - \mathbf{v}t)$ is $2\pi e\delta(\omega - \mathbf{k} \cdot \mathbf{v})$ for a point charge e travelling with velocity \mathbf{v} ; the transverse current density mode labelled by (\mathbf{k}, ω) is $2\pi e\mathbf{v} \cdot (\mathbf{U} - \hat{\mathbf{k}}\hat{\mathbf{k}})\delta(\omega - \mathbf{k} \cdot \mathbf{v})$: the condition, which is a dispersion relation, is $\omega = \mathbf{k} \cdot \mathbf{v}$. By working in (\mathbf{k}, ω) space the Lienard–Wiechert potentials are built into the expressions for the probing fields without the complication which surrounds these potentials in (\mathbf{x}, t) space.

equation (2.9a), it does not satisfy III, equation (2.9b): indeed equations (2.9) of III are replaced by

$$\operatorname{div} \mathbf{E}_t(\mathbf{x}, \omega) = 0 \quad (2.1a)$$

$$(\nabla^2 + k_0^2) \mathbf{E}_t(\mathbf{x}, \omega) = -4\pi i k_0 c^{-1} \mathbf{j}_t(\mathbf{x}, \omega) \quad (2.1b)$$

with transforms

$$i\mathbf{k} \cdot \mathbf{E}_t(\mathbf{k}, \omega) = 0 \quad (2.2a)$$

$$(k^2 - k_0^2) \mathbf{E}_t(\mathbf{k}, \omega) = 4\pi i k_0 c^{-1} \mathbf{j}_t(\mathbf{k}, \omega). \quad (2.2b)$$

From III, equation (3.3), with $\mathbf{P}_t(\mathbf{k}, \omega)$ transverse to $\hat{\mathbf{k}}$, we have†

$$\begin{aligned} \mathbf{P}_t(\mathbf{k}, \omega) = \alpha(\omega) & \left\{ \mathbf{E}_t(\mathbf{k}, \omega) + (k_0^2 \mathbf{U} - \mathbf{k}\mathbf{k}) \cdot 4\pi n \mathbf{P}_t(\mathbf{k}, \omega) (k^2 - k_0^2)^{-1} + \frac{4}{3}\pi n \mathbf{P}_t(\mathbf{k}, \omega) \right\} \\ & + (k_0^2 \mathbf{U} - \mathbf{k}\mathbf{k}) \cdot \frac{n\alpha(\omega)}{(2\pi)^3} \int \frac{I(\mathbf{k}, -\mathbf{k}'; \omega)}{k'^2 - k_0^2} \mathbf{P}_t(\mathbf{k}', \omega) d\mathbf{k}'. \end{aligned} \quad (2.3)$$

Whilst there may be pathological possibilities in (2.3) we have only the following straightforward solution. The solution of (2.1b) is

$$\mathbf{E}_t(\mathbf{k}, \omega) = \frac{4\pi i k_0 c^{-1} \mathbf{j}_t(\mathbf{k}, \omega)}{k^2 - k_0^2} + \mathbf{E}_{t0}(\hat{\mathbf{k}}, \omega) k_0^{-2} \delta(k - k_0) \quad (2.4a)$$

in which we assume $\mathbf{j}_t(\mathbf{k}, \omega) = O(k - k_0)$ or better near $k = k_0$ for convenience. This equation we write in the form

$$\mathbf{E}_t(\mathbf{k}, \omega) = \mathbf{E}_{t1}(\mathbf{k}, \omega) + \mathbf{E}_{t0}(\hat{\mathbf{k}}, \omega) k_0^{-2} \delta(k - k_0). \quad (2.4b)$$

If we restrict $\mathbf{P}_t(\mathbf{k}, \omega)$ to modes with $\hat{\mathbf{k}}$ parallel to the axis of the slab V , the integral in (2.3) is transverse to $\hat{\mathbf{k}}$ and, from the definition in III, equation (3.4), $I(\mathbf{k}, -\mathbf{k}'; \omega)$ always has a factor $\delta(k - k_0)$: the whole last term in (2.3) is then transverse to $\hat{\mathbf{k}}$ and has $\delta(k - k_0)$ as a factor. We now choose $\mathbf{E}_{t0}(\hat{\mathbf{k}}, \omega)$ in (2.4b) to eliminate this last term in (2.3) essentially as the extinction theorem eliminates the free forcing field in III, § 2 (we look at the form of $\mathbf{E}_{t0}(\hat{\mathbf{k}}, \omega)$ again later). This choice now still leaves $\mathbf{E}_{t1}(\mathbf{k}, \omega)$, which is the secondary field generated by the current density $\mathbf{j}_t(\mathbf{k}, \omega)$ in the absence of the fluid, as the effective transverse forcing field.

Indeed the response with (\mathbf{k}, ω) free variables is now given by

$$\begin{aligned} 4\pi n \mathbf{P}_t(\mathbf{k}, \omega) = [4\pi n \alpha(\omega) \{1 - 4\pi n \alpha(\omega) k_0^2 (k^2 - k_0^2)^{-1} - \frac{4}{3}\pi n \alpha(\omega) \\ - n \alpha(\omega) J_t(\mathbf{k}, \omega)\}^{-1}] \mathbf{E}_{t1}(\mathbf{k}, \omega) \end{aligned} \quad (2.5)$$

in which we can include $J_t(\mathbf{k}, \omega)$ by noting, as in III, that it merely augments the Lorentz $4\pi/3$ term. We could of course define a (\mathbf{k}, ω) -dependent transverse response function by the square bracket in (2.5). This equation is in fact directly comparable with the longitudinal response relation III, equation (3.8), and indeed the square bracket has the property comparable with III, equation (3.10), that the zeros of its denominator are the roots of the transverse dispersion relation (2.5b) for m_t^2 . However, essentially the same considerations which led us to the expression III (3.14) for $\epsilon_t(\mathbf{k}, \omega)$ can lead us to a closely related form for $\epsilon_t(\mathbf{k}, \omega)$. We argue as follows.

† We continue to omit $J_t(\mathbf{k}, \omega)$ here. Since $\mathbf{k} \cdot \mathbf{P}_t = 0$ we can simplify one of the terms in (2.3) but certainly not the integral since $\mathbf{k} \cdot \mathbf{P}_t(\mathbf{k}', \omega) \neq 0$ for general \mathbf{k}' .

We introduce a pseudo-macroscopic transverse Maxwell field vector $\bar{\mathbf{E}}_t(\mathbf{k}, \omega)$ which will satisfy

$$\{-k^2 + \epsilon_t(\mathbf{k}, \omega)k_0^2\}\bar{\mathbf{E}}_t(\mathbf{k}, \omega) = ik_04\pi c^{-1}\mathbf{j}_t(\mathbf{k}, \omega) \quad (2.6a)$$

and from (2.4a) and (2.4b) we find that

$$\bar{\mathbf{E}}_t(\mathbf{k}, \omega) = \{k^2 - k_0^2\}\{k^2 - \epsilon_t(\mathbf{k}, \omega)k_0^2\}^{-1}\mathbf{E}_{t1}(\mathbf{k}, \omega). \quad (2.6b)$$

We define $\epsilon_t(\mathbf{k}, \omega)$ by

$$\{\epsilon_t(\mathbf{k}, \omega) - 1\}\bar{\mathbf{E}}_t(\mathbf{k}, \omega) = 4\pi n\mathbf{P}_t(\mathbf{k}, \omega). \quad (2.6c)$$

With this definition and the response function (2.5) we now find after a little manipulation that

$$\epsilon_t(\mathbf{k}, \omega) - 1 = 4\pi n\alpha(\omega)\{1 - \frac{4}{3}\pi n\alpha(\omega) - n\alpha(\omega)J_t(\mathbf{k}, \omega)\}^{-1}. \quad (2.7)$$

We can also now rewrite the relation (2.5) in the form

$$4\pi n\mathbf{P}_t(\mathbf{k}, \omega) = \left[\frac{k^2 - k_0^2}{k^2 - \epsilon_t(\mathbf{k}, \omega)k_0^2} \right] \{\epsilon_t(\mathbf{k}, \omega) - 1\} \mathbf{E}_{t1}(\mathbf{k}, \omega) \quad (2.8)$$

a relation immediately evident from (2.6c) with (2.6b).

The quantity in square brackets is a transverse response function indicating the response to the secondary transverse field $\mathbf{E}_{t1}(\mathbf{k}, \omega)$: it is very different from III (2.15) which is the response to a primary transverse field given by III (2.9); and in particular III (2.15) depends on the geometry of the surface Σ of V whilst (2.8) does not. This is of course because (2.8) does not contain the response to the total transverse field: the surface geometry is concealed in what is necessarily the carefully chosen form of the second of the two fields on the right-hand side of (2.4b) if the response in \mathbf{P}_t is not to be changed.

In terms of the primary probing field $\mathbf{j}_t(\mathbf{k}, \omega)$, (2.8) takes the form

$$4\pi n\mathbf{P}_t(\mathbf{k}, \omega) = \left[\frac{\epsilon_t(\mathbf{k}, \omega) - 1}{k^2 - k_0^2\epsilon_t(\mathbf{k}, \omega)} \right] 4\pi ik_0c^{-1}\mathbf{j}_t(\mathbf{k}, \omega). \quad (2.9)$$

This is the form comparable with III (3.15b), especially when we use the continuity equation

$$-i\omega\rho(\mathbf{k}, \omega) + ikj_i(\mathbf{k}, \omega) = 0. \quad (2.10)$$

In each form of the response function in the square brackets of (2.5), (2.8) or (2.9) we see that the surfaces of singularity are the transverse dispersion relation (2.5b): in (2.8) and (2.9) these surfaces are obviously the surfaces

$$k^2 - \epsilon_t(\mathbf{k}, \omega)k_0^2 = 0. \quad (2.11)$$

The surfaces of singularity of $\epsilon_t(\mathbf{k}, \omega) - 1$ are *not* singularities of the response function: neither these surfaces nor the strict zeros of $\epsilon_t(\mathbf{k}, \omega)$ appear to have any physical significance although the zeros of $\epsilon_t(\mathbf{k}, \omega)$ are apparently very little different from the longitudinal dispersion relation of III (2.5a). Moreover when $c \rightarrow \infty$ the photon propagator in (2.5) is eliminated and the poles of the response function are close to the resonances of the refractive index expression $m_t^2(\omega) - 1$ and the response

III (2.15). But this is because k is effectively small when it has the value $m_t(\omega) k_0$ and $m_t(\omega) \simeq 1.5$.†

Comparison of (2.7) with III (3.14) shows that we have now reached the two relations

$$\epsilon_{t,i}(\mathbf{k}, \omega) - 1 = 4\pi n\alpha(\omega) \{1 - \frac{4}{3}\pi n\alpha(\omega) - n\alpha(\omega) J_{t,i}(\mathbf{k}, \omega)\}^{-1} \quad (2.11)$$

for the molecular fluid. This is one of the main results of the two papers III and IV. Observe that the $J_{t,i}(\mathbf{k}, \omega)$ are cluster expansions containing intermolecular correlation to all orders. Note also that‡

$$\epsilon_t(\mathbf{0}, \omega) = \epsilon_i(\mathbf{0}, \omega)$$

and for small k the two dielectric constants are little different. These differences are of vital importance in the theory of the binding energy of the molecular fluid, however, as is already implicit in paper II (Bullough 1969 a—to be referred to as II) of this series (and cf. also Bullough and Obada 1969 a,b and V which follows). This feature will be elaborated in more detail later.

In reaching the symmetrical transverse-longitudinal expressions (2.11) we have partly been motivated by the wish to find expressions for (\mathbf{k}, ω) -dependent dielectric constants for the molecular fluid comparable with those already obtained for other systems. For these are very suitable for establishing compact expressions for physical applications of the theory (cf. II, V and Bullough and Obada 1969 a,b). Helpful guidelines are implicit in some of the previous work on (\mathbf{k}, ω) -dependent dielectric constants: e.g. Lindhard (1954), Born and Huang (1955), Nozières and Pines (1958 a,b,c, 1959), Ehrenreich and Cohen (1959), Adler (1962), Pines (1963), Schultz (1963) and Mahan (1965) are all helpful for a comparison. In particular we can now easily establish the precise formal equivalence of our definitions (2.6c) and III (3.11b) to those of Nozières and Pines (1959) and Adler (1962).

For the molecular fluid we define induced current densities by

$$\mathbf{j}_{t,i}^{\text{ind}}(\mathbf{k}, \omega) = i\omega n \mathbf{P}_{t,i}(\mathbf{k}, \omega) \quad (2.12)$$

these are the changes in the ‘displacement currents’. This definition enables us to write (2.6c) immediately, and III (3.11b) after using the continuity equation, in the form

$$-ik_0 \{\epsilon_{t,i}(\mathbf{k}, \omega) - 1\} \bar{\mathbf{E}}_{t,i}(\mathbf{k}, \omega) = 4\pi c^{-1} \mathbf{j}_{t,i}^{\text{ind}}(\mathbf{k}, \omega). \quad (2.13)$$

This is exactly§ the definition used by Nozières and Pines and by Adler.

This being so it is important to notice that as dielectric constants which determine the longitudinal response of III (3.15) and the transverse response of (2.9) above the formulae (2.11) are not complete. Thus we must now develop the theory of the total response function: this means we must look next at the role of the second field in (2.4b) which satisfies the dispersion relation of the free field.

So far we have viewed this field as an imposed field just sufficient to eliminate the surface integral terms in (2.3). This situation is a physically possible one: we require

† Because the expression III (2.5b) for $m_t^2(\omega) - 1$ is only an implicit relation for $m_t^2(\omega)$ the resonances of $m_t^2(\omega) - 1$ will be rather complicated. Otherwise $m_t(\omega)k_0$ is obviously small for small enough k_0 except at a resonance of $m_t(\omega)$.

‡ Compare the brief remarks at the end of III § 3 and see the following paper V § 3 for the complete discussion. The result is already reported in II (7).

§ Note that the fields $\mathbf{E}_{t,i}$ in (2.13) have to be interpreted as the pseudo-Maxwell fields $\bar{\mathbf{E}}_{t,i}$ and are not the forcing fields: equation (2.13) is not an explicit response relation.

to eliminate a free field inside V ; and we do this by imposing a second free field inside V with just this property. However, a physical field which is a free field in V can only be imposed from outside V and analysis then shows that the equations can be solved only with at least one mode of the free field travelling into V . The physical picture is one of transverse currents imposed throughout space in the form of moving charges striking V and these currents are accompanied by incident light. This cannot be the only solution to the problem since there is no physical reason why we should not simply fire charge into V without additional incident light.†

The solution to this dilemma is the following: it is sufficient to set up the field \mathbf{E}_{t_0} inside V only, and this can be arranged by choosing as the total transverse field a linear combination of the two solutions (2.5), with wave number k , and III (2.4b), with wave number $m_t k_0$. We can if we like think of the incident field \mathbf{E}_{t_1} of wave vector \mathbf{k} inducing a response \mathbf{P}_{t_1} of wave vector \mathbf{k} by (2.5); then \mathbf{P}_{t_1} induces the free field \mathbf{E}_{t_0} in V with wave number k_0 ; and this free field induces a response \mathbf{P}_{t_0} with wave number $m_t k_0$ by a response relation like III (2.15).

We therefore first choose the trial solution‡

$$\mathbf{P}_{t_1}(\mathbf{k}_1, \omega) + (2\pi)^3 m_t^{-2} k_0^{-2} \mathbf{P}_{t_0}(\hat{\mathbf{k}}, \omega) \delta(k - m_t k_0) \delta(\hat{\mathbf{k}} - \hat{\mathbf{k}}_1) \tag{2.14}$$

where \mathbf{P}_{t_1} is the response to a single mode $\mathbf{E}_{t_1}(\mathbf{k}_1, \omega)$ according to (2.9), and $\mathbf{P}_{t_0}(\mathbf{k}, \omega)$ satisfies III (2.11). The field \mathbf{E} generated by \mathbf{P}_{t_0} according to III (2.11) is exactly to cancel the field \mathbf{E}_{t_0} which it is necessary to impose in V in order that $\mathbf{P}_{t_1}(\mathbf{k}_1, \omega)$ is an acceptable solution: thus the surface integral terms associated with \mathbf{P}_{t_1} and \mathbf{P}_{t_0} are simply to cancel each other and the condition for this should be sufficient to determine \mathbf{P}_{t_0} completely.

From III (2.11) the Fourier transform of the field \mathbf{E} generated by \mathbf{P}_{t_0} is

$$\begin{aligned} \mathbf{E}_0(\mathbf{k}, \omega) = & -(2\pi)^4 [\exp\{-i(m_t - 1)k_0 c\} \delta(\mathbf{k} - k_0 \hat{\mathbf{k}}_1) (1 + m_t) \\ & + \exp\{i(m_t + 1)k_0 d\} \delta(\mathbf{k} + k_0 \hat{\mathbf{k}}_1) (1 - m_t)] (m_t^2 - 1)^{-1} \mathbf{P}_{t_0}(\hat{\mathbf{k}}_1, \omega). \end{aligned} \tag{2.15a}$$

The free field generated by $\mathbf{P}_{t_1}(\mathbf{k}, \omega)$ has a transform which we write slightly symbolically in the form

$$\begin{aligned} (k_0^2 \mathbf{U} - \mathbf{k}_1 \mathbf{k}_1) \cdot n\alpha(\omega) I(\mathbf{k}_1, -\mathbf{k}_1; \omega) (k_1^2 - k_0^2)^{-1} \mathbf{P}_{t_1}(\mathbf{k}_1, \omega) \\ = k_0^2 n\alpha(\omega) I(\mathbf{k}_1, -\mathbf{k}_1; \omega) (k_1^2 - k_0^2)^{-1} \mathbf{P}_{t_1}(\mathbf{k}_1, \omega). \end{aligned} \tag{2.15b}$$

This form is so because \mathbf{P}_{t_1} is a single mode with wave vector \mathbf{k}_1 if \mathbf{E}_{t_1} is, and we continue to consider the slab V with axis in the direction of \mathbf{k}_1 .

The symbol $I(\mathbf{k}_1, -\mathbf{k}_1; \omega)$ in (2.15b) as calculated from III (3.4) is \mathbf{k} -dependent, and symbolizes

$$\begin{aligned} I(\mathbf{k}_1, -\mathbf{k}_1; \omega) \equiv & (2\pi)^3 [-2\pi \exp\{i(k_1 - k_0)c\} \delta(\mathbf{k} - k_0 \hat{\mathbf{k}}_1) (1 + k_1 k_0^{-1}) \\ & - 2\pi \exp\{i(k_1 + k_0)d\} \delta(\mathbf{k} + k_0 \hat{\mathbf{k}}_1) (1 - k_1 k_0^{-1})]. \end{aligned} \tag{2.16}$$

The two free fields in (2.15) are to cancel and this is obviously impossible. We therefore replace the trial solution (2.14) by the new trial solution

$$\mathbf{P}_{t_1}(\mathbf{k}_1, \omega) + (2\pi)^3 m_t^{-2} k_0^{-2} \mathbf{P}_{t_0}(\hat{\mathbf{k}}_1, \omega) \delta(k - m_t k_0) \{\delta(\hat{\mathbf{k}} - \hat{\mathbf{k}}_1) + \Lambda(\mathbf{k}_1, \omega) \delta(\hat{\mathbf{k}} + \hat{\mathbf{k}}_1)\}. \tag{2.17}$$

† There is of course no *a priori* physical objection to light leaving the surface of V : this would be light generated at the surface by the moving charge.

‡ Which omits no factors. cf. III (2.8d) and the discussion of notation there.

We can now choose Λ so that the coefficients of $\delta(\mathbf{k} \pm k_0 \hat{\mathbf{k}}_1)$ in the sum of (2.15a) and (2.15b) vanish separately. Then, given $\mathbf{j}_t(\mathbf{k}_1, \omega)$ as primary single mode probe, $\mathbf{P}_t(\mathbf{k}_1, \omega)$ is fixed by (2.9) and $\Lambda(\mathbf{k}_1, \omega)$ and $\mathbf{P}_{t0}(\hat{\mathbf{k}}_1, \omega)$ are fixed by the two equations implicit in the elimination of the sum of the two expressions (2.15).

We have thus demonstrated that current forced transverse modes are acceptable modes in V if, and only if, every forcing field mode $\mathbf{j}_t(\mathbf{k}_1, \omega)$ of wave vector \mathbf{k}_1 (with $\hat{\mathbf{k}}_1$ along the axis of the slab V) generates the *three* modes which are described by (2.17): one of these, $\mathbf{P}_{t1}(\mathbf{k}_1, \omega)$, has wave vector \mathbf{k}_1 and is related to $\mathbf{j}_t(\mathbf{k}_1, \omega)$ by (2.9): the two additional modes have wave vectors $\pm m_t k_0 \hat{\mathbf{k}}_1$ and the wave numbers $m_t k_0$ are determined at the frequency ω by the dispersion relation III (2.4b) for the refractive index $m_t(\omega)$. Because $m_t(\omega)$ satisfies the transverse dispersion relation these two additional modes are real photon-like modes: we shall refer to them as real modes. Because the mode \mathbf{P}_{t1} has a free wave vector \mathbf{k}_1 not fixed by ω it is virtual, photon-like, and we refer to it as a virtual mode. We have thus shown that the response to a single mode transverse current probe $\mathbf{j}_t(\mathbf{k}_1, \omega)$ normally incident on V consists of one virtual mode of wave vector \mathbf{k}_1 and frequency ω and two real modes of wave vectors $\pm m_t k_0 \hat{\mathbf{k}}_1$ and frequency ω . More generally the real modes depend on the surface geometry whilst the virtual modes do not.

The results for \mathbf{P}_{t0} in terms of \mathbf{P}_{t1} and for Λ are

$$\begin{aligned} \mathbf{P}_{t0} = & \frac{-k_0^2(m_t^2 - 1)}{k_1^2 - k_0^2} \mathbf{P}_{t1} [\exp\{-i(k_1 c + m_t k_0 d)\}(1 + k_1 k_0^{-1})(1 + m_t) \\ & - \exp\{i(m_t k_0 c + k_1 d)\}(1 - k_1 k_0^{-1})(1 - m_t)] \\ & \times [\exp\{-i m_t k_0(c + d)\}(1 + m_t)^2 - \exp\{i m_t k_0(c + d)\}(1 - m_t)^2]^{-1} \end{aligned} \quad (2.18a)$$

and

$$\begin{aligned} \Lambda(\mathbf{k}_1, \omega) = & -[\exp\{-i(k_1 c - m_t k_0 d)\}(1 + k_1 k_0^{-1})(1 - m_t) \\ & - \exp\{-i(m_t k_0 c - k_1 d)\}(1 - k_1 k_0^{-1})(1 + m_t)] \\ & \times [\exp\{-i(k_1 c + m_t k_0 d)\}(1 + k_1 k_0^{-1})(1 + m_t) \\ & - \exp\{-i(m_t k_0 c + k_1 d)\}(1 - k_1 k_0^{-1})(1 - m_t)]^{-1}. \end{aligned} \quad (2.18b)$$

One immediate check is that when $k_1 \rightarrow k_0$

$$\Lambda = \left(\frac{m_t - 1}{m_t + 1} \right) \exp(2i m_t k_0 d)$$

in precise agreement with III (2.14a).

The total response to $\mathbf{j}_t(\mathbf{k}_1, \omega)$ can now be written in the form

$$\begin{aligned} 4\pi n \mathbf{P}_t(\mathbf{k}, \omega) \equiv & 4\pi n [\mathbf{P}_{t1}(\mathbf{k}, \omega) - (2\pi)^3 m_t^{-2} k_0^{-2} \mathbf{P}_{t0}(\hat{\mathbf{k}}_1, \omega) \\ & \times \delta(k - m_t k_0) \{\delta(\hat{\mathbf{k}} - \hat{\mathbf{k}}_1) + \Lambda \delta(\hat{\mathbf{k}} + \hat{\mathbf{k}}_1)\}] \\ = & \left\{ \frac{\epsilon_t(\mathbf{k}, \omega) - 1}{k^2 - \epsilon_t(\mathbf{k}, \omega) k_0^2} \right\} \left[(k^2 - k_0^2) - k_0^2 (m_t^2 - 1) \mathcal{S}(\Sigma; \mathbf{k}, \omega) \right. \\ & \left. \times \{\sigma^+(\mathbf{k}) + \Lambda(\mathbf{k}, \omega) \sigma^-(\mathbf{k})\} \right] \left(\frac{4\pi i k_0 c^{-1}}{k^2 - k_0^2} \right) \mathbf{j}_t(\mathbf{k}, \omega). \end{aligned} \quad (2.19)$$

Here $\mathbf{j}_t(\mathbf{k}, \omega)$ is supposed to be the single mode expression

$$\mathbf{j}_t(\mathbf{k}, \omega) = (2\pi)^3 k^{-2} \delta(k - k_1) \delta(\hat{\mathbf{k}} - \hat{\mathbf{k}}_1) \mathbf{g}_t(\mathbf{k}, \omega) \quad (2.20)$$

in which $g_t(\mathbf{k}, \omega)$ is a smooth function, and $\sigma^\pm(\mathbf{k})$ are the operators of III (2.15) now changing the wave number k_1 of j_t to $m_t k_0$ and in the case of $\sigma^-(\mathbf{k})$ also changing the direction of the wave vector from \hat{k}_1 to $-\hat{k}_1$.

The expression $\hat{S}(\Sigma; \mathbf{k}, \omega)$ is the combination of the two square brackets in (2.18a): when \mathbf{k} is $k_0 \hat{k}_1$, $\hat{S}(\Sigma; \mathbf{k}, \omega)$ is exactly the quantity $\hat{S}(\Sigma; \mathbf{k}, \omega)$ in the response relation of III (2.15) and which is determined by III (2.14b). Thus we have been able to generalize that response relation to include the response to a single mode transverse current probe of arbitrary wave number k and frequency ω but still with the special wave direction \hat{k}_1 .

Indeed we can now work consistently with a transverse single mode electric field probe of wave vector \mathbf{k}_1 :

$$E_t(\mathbf{k}, \omega) = \frac{4\pi i k_0 c^{-1} (2\pi)^3 k^{-2} \delta(k - k_1) \delta(\hat{\mathbf{k}} - \hat{\mathbf{k}}_1) g_t(\mathbf{k}, \omega)}{k^2 - k_0^2} \tag{2.21}$$

We arrange that $g_t(\mathbf{k}, \omega) = O(k - k_0)$ or better in the neighbourhood of $k = k_0$ so that (2.21) passes smoothly into the free-field probe. The response relation (2.19) now takes the form which is perhaps the principal result of the two papers III and IV:

$$4\pi n P_t(\mathbf{k}, \omega) = \left\{ \frac{\epsilon_t(\mathbf{k}, \omega) - 1}{k^2 - \epsilon_t(\mathbf{k}, \omega) k_0^2} \right\} \left[(k^2 - k_0^2) - k_0^2 (m_t^2 - 1) \hat{S}(\Sigma; \mathbf{k}, \omega) \right. \\ \left. \times \{ \sigma^+(\mathbf{k}) + \Lambda(\mathbf{k}, \omega) \sigma^-(\mathbf{k}) \} \right] E_t(\mathbf{k}, \omega) \tag{2.22}$$

with $\epsilon_t(\mathbf{k}, \omega)$ given by (2.7), m_t^2 given by (2.4b), $\hat{S}(\Sigma; \mathbf{k}, \omega)$ the combination of the two square brackets in (2.18a), $\Lambda(\mathbf{k}, \omega)$ given by (2.18b) and $\sigma^\pm(\mathbf{k})$ the operators described under (2.20). We have assumed that \mathbf{k} is in fact \mathbf{k}_1 parallel to the axis of V as (2.21) implies. We observe that when $k_1 \rightarrow k_0$ the total response relation (2.22) takes the form

$$4\pi n P_t(\mathbf{k}, \omega) = (m_t^2 - 1) \hat{S}(\Sigma; k_0 \hat{k}_1, \omega) \{ \sigma^+(\mathbf{k}) + \Lambda(k_0 \hat{k}_1, \omega) \sigma^-(\mathbf{k}) \} \cdot E_t(\mathbf{k}, \omega) \tag{2.23}$$

and, because \hat{S} and Λ are exactly the quantities in III (2.14), this result is indeed exactly the response relation III (2.15).

We have shown in (2.22) that the total transverse response to a transverse electric field probe is made up of two natural parts. One part describes the virtual response and is independent of the surface geometry of V : the other part describes the real response and depends very much on the surface geometry of V . When the probe is light satisfying the free-field dispersion relation there is no virtual response and the total response is the real response. But this limit (2.23) of (2.22) shows that the real response is an essential part of the total transverse response. It can be neglected only when $k = k_1 \gg k_0$ for the virtual contribution to the square bracket in (2.22) is $O(k_1 k_0^{-1})$ and the real contribution is $O(k_1^2 k_0^{-2})$ when $k_1 \gg k_0$: the contributions to the total response are then $O(k_0 k_1^{-1})$ and $O(1)$ respectively. For $k_1 \ll k_0$ the contributions are very comparable, and for $k_1 \simeq k_0$ the *virtual* response is negligible. This is easily understood: if the transverse probe is an incident charged particle of velocity v , $vc^{-1} \geq k_0 k_1^{-1}$ and $k_1 \gg k_0$ is the non-relativistic region. The real response is then a relativistic correction to the non-relativistic virtual response: when $k_1 \simeq k_0$ the real response dominates the situation as we expect. Thus the real response should be an important feature in the theory of external Čerenkov radiation:

only the contribution of the virtual response to this has been calculated so far and this is reported in II (II—equation (9)).

These results are inevitable if we believe that (2.21) applies to arbitrary transverse electromagnetic probes not excluding light: for in the absence of the real response there is no response whatsoever to light when $k_1 \rightarrow k_0$ in (2.22). It should be possible to devise a theory in which the transverse probe can pass smoothly into a light wave: the methods of von Weizsäcker (1934) and Williams (1935), for example, invoke the idea that a relativistically fast charged particle approximates to a system of light waves (cf. Heitler 1954—p. 414). Equation (2.22) is the solution to this problem. Indeed for the simple geometry considered in this § 2 of $\mathbf{k} = k_1$ parallel to the axis of the slab we have completely solved the immediate problem posed in this paper IV. For we have constructed a transverse response theory for arbitrary electromagnetic waves which is compatible with the extinction theorem; and it is also compatible with the surface-dependent optical response as this was computed in § 2 of the previous paper III.

We extend the solution to arbitrary directions $\hat{\mathbf{k}}$ of \mathbf{k} in the following § 3; but the result of (2.22) already throws up a number of points which we can usefully discuss now. The main point is that (2.22) exhibits a natural division between a translationally invariant and a non-translationally invariant contribution to the total transverse electromagnetic response. The rigorous construction of a translationally invariant theory in many-body physics is a problem of interest in its own right, and we should like to discuss this. But the natural division exhibited by (2.22) is also important because we reported in II (cf. also Bullough and Obada 1969 b) closed expressions for the binding or free energies of a molecular fluid which depend solely on the total electromagnetic response function: if our understanding is correct the total virtual response should yield the translationally invariant (i.e. extensive) part of the total binding energy, whilst the surface-dependent part should yield a surface energy and permit an *ab initio* calculation of the surface tension. No analysis of this surface-dependent contribution has been made yet, but the results of II show a rather complete derivation of the fully retarded bulk binding energy of a molecular fluid from the translationally invariant part. This theory differs in important respects from the semi-phenomenological theories of Lifshitz (1956) and Dzyaloshinskii *et al.* (1961) and this is the subject matter of V. Now we look at the problem of constructing a translationally invariant theory. The rest of this § 2 is concerned with this problem.

One feature to note immediately is that response function theories are scattering theories and these are very sensitive to surface effects: binding energy theories are much less sensitive to these. Thus although the binding energy theory reported in II easily ignores the rather subtle surface effects the response theory shows that the real modes, certainly induced by the surface of V , nevertheless exist throughout the whole interior of V . They are little damped and then only by scattering from the interior of V .

On the other hand we might interpret the real modes as modes induced by breaking the continuous group of spacial translations. They are bosons (in the sense that the polaritons of Hopfield (1958) are bosons[†]); they exhibit a zero-frequency excitation ($\hbar\omega$ is a root of $m_t(\omega)\omega = ck$ for fixed k , and $\omega = 0$ is a root at $k = 0$); thus they have these features of the symmetry-breaking bosons of Goldstone (Goldstone *et al.* 1963, Brout 1965). These modes are not obviously ones which restore a feature of

[†] The polarization diagram approximation of Bullough *et al.* (1968) is equivalent to Hopfield's boson approximation (Bullough and Thompson 1970).

the broken symmetry, however. We shall find later that the real modes are the simplest ones induced by the surface of V : there are certain additional modes which appear in the complete analysis of the external scattering at optical frequencies.†

It is not of course a new result that we must break translational invariance in order to couple external light to a material system; this is also so in the Maxwell phenomenological scheme. The result (2.23) is actually a striking generalization of that scheme; but it is more than this since it involves actual reinterpretation of terms. To see this let us construct a translationally invariant theory by focusing attention on the virtual modes and dropping the real modes entirely. We shall find that modes like the real modes are normal modes in this theory. This is conventional many-body physics and important for this reason. For convenience of subsequent reference we call it 'virtual mode theory'.

We observe first that (2.7) and the transverse dispersion relation of III (2.5b) (and see II (6)) together show that

$$\epsilon_t(m_t k_0 \hat{\mathbf{k}}, \omega) \equiv m_t^2(\omega) \tag{2.24}$$

for any $\hat{\mathbf{k}}$. Then (2.6a) shows that if $\mathbf{j}_t(\mathbf{k}, \omega) = \mathbf{0}$

$$\{-k^2 + \epsilon_t(\mathbf{k}, \omega) k_0^2\} \bar{\mathbf{E}}_t(\mathbf{k}, \omega) = \mathbf{0} \tag{2.25}$$

and this has a solution (cf. I)

$$\bar{\mathbf{E}}_t(\mathbf{k}, \omega) = \mathbf{E}_t(\hat{\mathbf{k}}, \omega) \delta\{k - m_t(\omega) k_0\} \tag{2.26a}$$

provided that

$$k^2 k_0^{-2} = \epsilon_t(m_t k_0 \hat{\mathbf{k}}, \omega) = m_t^2(\omega). \tag{2.26b}$$

Then (2.6c) means that

$$n\mathbf{P}_t(\mathbf{k}, \omega) = \left(\frac{m_t^2(\omega) - 1}{4\pi} \right) \bar{\mathbf{E}}_t(\mathbf{k}, \omega) \tag{2.27}$$

and (2.6a) means that $\bar{\mathbf{E}}_t(\mathbf{k}, \omega)$ and $\mathbf{P}_t(\mathbf{k}, \omega)$ are normal modes which satisfy in (\mathbf{x}, ω) space‡

$$\{\nabla^2 + m_t^2(\omega) k_0^2\} \bar{\mathbf{E}}_t(\mathbf{x}, \omega) = 0 \tag{2.28a}$$

or

$$(\nabla^2 + k_0^2) \bar{\mathbf{E}}_t(\mathbf{x}, \omega) = -4\pi k_0^2 n\mathbf{P}_t(\mathbf{x}, \omega). \tag{2.28b}$$

The second form (2.28b) is a forced oscillator equation for the field $\bar{\mathbf{E}}$: the induced dipoles are the sources and drive the field as in Lamb's (1964) theory of the maser, for example. Then (2.6b) shows that the modes are normal modes in the sense that $\bar{\mathbf{E}}_t(\mathbf{k}, \omega)$ is a finite internal response for a zero external field $\mathbf{E}_{t1}(\mathbf{k}, \omega)$. Since the modes are normal they couple to no external field: in particular they do not couple to light. However, this coupling can now be achieved by breaking translational invariance and appealing to Maxwell's phenomenological boundary conditions. This theory completely agrees in its final consequences with the results of II § 2 for the strictly optical response. It is unsatisfactory only because it invokes the separate hypothesis of Maxwell's *phenomenological* boundary conditions across the surface. It is therefore

† These are associated with the surface terms of Bullough and Hynne (1968) and intuitively with the scattering of light across the surface.

‡ However, we already see the theory is incomplete: $m_t(\omega)$ determined by (2.26b) is complex through external scattering and at this delicate level we cannot obviously construct a translationally invariant theory.

remarkable that this theory is constructed from our virtual modes: for the equation (2.22) shows in a much more natural way that the microscopic theory, with no surface boundary conditions, is a limiting consequence of the *real* modes as $k \rightarrow k_0$. This is the reinterpretation we spoke of.

These remarks justify the choice of the virtual response (2.8) or (2.9) as the total translationally invariant transverse response and justify the virtual mode theory as a translationally invariant theory. However, this can be no more than a definition agreeing with less sophisticated methods unless we can *explicitly eliminate* the real response in the limit of a translationally invariant theory. All the evidence we have is that it is not possible to construct a translationally invariant theory in this sense without wholly rejecting outgoing boundary conditions.† A number of points in this connection may be worth recording here.

In order to eliminate external sources we can hope to set up a normal mode theory for a finite or infinite region V . This is the problem of the existence of solutions of the *homogeneous* fundamental integral equation III (2.1) and of interest in its own right. We look at this problem and the connected problem of eliminating the real response when V is infinite and translationally invariant now. We also return again to the problem of normal modes in a finite box in § 4 after we have considered the case of oblique modes in § 3.

At first sight (2.22) might suggest that modes which satisfy (2.26b) are always normal modes—just as they are by (2.8) or (2.9) in the virtual mode theory. However, analysis shows that, although the virtual responses (2.8) or (2.9) diverge when $k \rightarrow m_t(\omega)k_0$, the *total* response from (2.22) is a finite response for a finite applied field with this k ; the response is zero for zero applied field, and the limit of (2.22) actually exists when $k \rightarrow m_t(\omega)k_0$.‡

Thus modes which satisfy (2.26) are *not* normal modes when the total response is considered. Normal modes are modes possible in the absence of any probe: to find any of these we need now to look at the singularities of (2.22).

These occur at the singularities of $m_t^2 - 1$, $\hat{S}(\Sigma; \mathbf{k}, \omega)$ or of $\Lambda(\mathbf{k}, \omega)$: singularities of $\epsilon_t(\mathbf{k}, \omega)$ (if these are infinities) are not singularities of (2.22). Thus we need the infinities of $m_t^2 - 1$, the roots of

$$\left[1 - \frac{(m_t - 1)^2}{(m_t + 1)} \exp\{2im_t k_0(c + d)\}\right] = 0 \quad (2.29)$$

or the roots of

$$\begin{aligned} &[\exp\{-i(k_1 c + m_t k_0 d)\}(1 + k_1 k_0^{-1})(1 + m_t) - \exp\{-i(m_t k_0 c + k_1 d)\}(1 - k_1 k_0^{-1}) \\ &\quad \times (1 - m_t)] = 0. \end{aligned} \quad (2.30)$$

The last condition would correspond to creating a mode in the direction $-\mathbf{k}$ instead of \mathbf{k} for zero applied field: thus we do not consider it further. We can presumably also dismiss the infinities of $m_t^2(\omega) - 1$ since the wave number of the associated real modes would be undefined. They should in any case be in the lower half ω -plane since they are observable resonances with a finite scattering width. Thus only (2.29) remains.

This condition is actually the usual type of condition for finding normal modes in a finite box: one surrounds the box with perfectly conducting walls (for example) so

† cf. (2.32) below and the acausal conditions which admit normal cavity modes in § 4. A limiting sequence of dielectric cavities is translationally invariant.

‡ The limit continues to depend on c and d even though, for example, $\Lambda \rightarrow 0$.

m_t in (2.29) $\rightarrow \infty$. One then chooses $2m_t k_0(c+d) = 2k(c+d) = 2\nu\pi$ with ν an integer fixing k .† This argument cannot be applied here since we do not apply boundary conditions at the surface of V but only at infinity.

Equation (2.29) is actually satisfied by $m_t = 0$. However, this is not a condition for transverse normal modes. One can check that when $m_t \rightarrow 0$ (from above‡) a transverse light wave normally incident on the parallel-sided slab $-c \leq z \leq d$ is totally reflected. At the same time the roots of $m_t(\omega) = 0$ fix frequencies at (or certainly very close to) those of the acceptable longitudinal normal modes of I and III, § 2, and to this extent the normal modes are longitudinal.

From this discussion we infer that transverse normal modes are not admissible in any finite volume V without either the formal application of boundary conditions across the surface or the physical inclusion of additional external sources. We can infer that if V is the slab the homogeneous integral equation III (2.1) has no transverse solution and we confirm this again in § 4. We thus fall back on virtual mode theory for a normal mode theory. This is physically sensible: any transverse mode of wave number $m_t k_0$ approaching the surface of V radiates out of this surface (unless, for example, it is not normally incident, is totally internally reflected and still evanescent outside V). The longitudinal normally incident modes do not radiate and so can exist as normal modes. This completes our present discussion of possible normal modes in a 'box'.

We now look at the problem of eliminating the real response by going to infinity: we still use the slab and set c and $d \rightarrow \infty$. We find no transverse normal modes and cannot eliminate the real response, and this is clearly associated with our original choice of outgoing boundary conditions. These mean that J_t (and indeed J_l) are complex and m_t is complex. Then although $d \rightarrow \infty$ causes no difficulty the phase factor $\exp\{i(m_t - 1)k_0 c\}$ in III (2.14b), for example, tends to zero. This means that the probing field E_0 must oscillate infinitely as $c \rightarrow \infty$ in order that the optical dipole response P_{t0} is finite. We expect E_0 as a field imposed at infinity to diverge when there is scattering from an infinite medium; but that it oscillates infinitely seems rather meaningless. From this we infer the reasonable result that outgoing boundary conditions are consistent only with the finite system.§

We have also noticed the following point. We can argue that the source of logical difficulty in going to infinity is that we start initially with the Fourier transform of $\exp(ik_0 r)r^{-1}$ on a spatially finite region V and this is the source of the surface integral of the extinction theorem and of the unwanted integrals in III (3.3) and in (2.3) here, for example. Then we can argue that outgoing boundary conditions means ω is $(\omega + i\delta)$ with $\delta > 0$ and this eliminates all oscillatory terms from the surface of V when V is all space. This is correct except for one circumstance: the Fourier transform

$$\int_V \exp(ik_0 r)r^{-1} \exp\{i\mathbf{k} \cdot (\mathbf{x}' - \mathbf{x})\} d\mathbf{x}'$$

† Only one condition emerges from (2.29) since we set the radius R of the cylindrical slab V to ∞ rejecting oscillatory terms of modulus 1. A second condition is needed, for modes with $\mathbf{k} = (0, 0, k_z)$ are never possible modes of the box with conducting walls (see § 4).

‡ When $m_t^2 < 0$, m_t is purely imaginary: we can take $\text{Re}(m_t)$ as non-negative since $-\text{Re}(m_t)$ is associated with the reflected mode.

§ The slab is two-dimensionally infinite by the choice R , the radius, $\rightarrow \infty$. We believe that as long as we consider modes with $\hat{\mathbf{k}}$ parallel to the slab axis we need not concern ourselves too precisely with this limit for R . On the other hand the example in § 4 shows that this limit is very relevant when $\hat{\mathbf{k}}$ is oblique.

will not exist in general when $k = |\mathbf{k}|$ satisfies a dispersion relation. For, in particular, when $\omega \rightarrow \omega + i\delta$, $k \rightarrow m \times (\omega + i\delta)c^{-1}$ and even when m is real there will be a contribution from the surface of V when V becomes infinite (as actual analysis of the limit shows). This means that we cannot eliminate the real modes or accept transverse normal modes in the infinite system this way.

However, provided $\mathbf{E}_i(\mathbf{k}, \omega) \neq \mathbf{0}$ in the basic equation (2.3) the same argument actually implies that for real k the forced solutions (2.5) are the complete solution of (2.3) for all $\hat{\mathbf{k}}$ when V is all space and it implies that there are no additional modes generated by any surface (now supposedly at infinity). Thus in the infinite system with real k the only forced modes are the virtual modes, and this is indeed the implicit assumption of all previous work on current forced (= virtual) modes. This picture is very much more artificial and much less complete than the argument which is summarized in the total response of the finite system in equation (2.22). In particular the argument on which this picture of the infinite system is based does not consistently admit normal modes which necessarily satisfy a dispersion relation as we showed in the previous paragraph.

The ultimate source of the difficulties surrounding the existence of transverse normal modes lies of course in the surface integral of the extinction theorem: this is intimately connected with the boundary conditions and these as we shall show later in this series are connected with causality. We now show here how these difficulties and the complication surrounding the total transverse response can be eliminated by changing to acausal boundary conditions. We show explicitly how this choice can eliminate the surface integral. Then we could show that the virtual solutions (2.5) are the complete forced transverse solutions; the solutions III (3.8) are the forced longitudinal solutions; the surfaces of singularity of (2.5) are dispersion relations for transverse normal modes; the surfaces of singularity of III (3.8) continue to be dispersion relations for the longitudinal modes; and these results are indeed valid for all wave vector directions. This consistent scheme is achieved only by rejecting causality (and the Kramer-Kronig relations) and limiting the theory to the infinite system. It says nothing about the coupling of external light to the system.†

We replace the causal Green's function $\exp(ik_0r)r^{-1}$ by

$$\frac{1}{2}\{\exp(ik_0r) + \exp(-ik_0r)\}r^{-1} \quad (2.31)$$

and both ω and $k_0 = \omega c^{-1}$ are purely real. Nothing is now changed in the calculations of either the virtual transverse or the virtual longitudinal modes except that $J_t(k, \omega)$ and $J_l(k, \omega)$ both become real; then in particular $m_t^2(\omega)$ is given by III (2.4b) and is real when ω is real.‡ We now find, for example,§ that because of the phase factors in III (2.10), $\Sigma(\mathbf{x}, \omega) \rightarrow \mathbf{0}$ as c and $d \rightarrow \infty$ providing only that the frequency

† Mead (1960) has suggested that only by rejecting causality is it possible to accept the usual periodic boundary conditions imposed on crystalline modes (see also Deutsche and Mead 1965). We find agreement between the Hopfield (1958) polariton theory and linear response theory with outgoing boundary conditions only by rejecting contributions to the extinction theorem in the response theory and interpreting the polariton theory as a causal theory (Bullough and Thompson 1970).

‡ We work with the principal (physical) root of III (2.4b) for m_t^2 : this makes m_t real when $J_t(k, \omega)$ is when $m_t^2 > 0$ as we assume. We note the possibility of additional roots in III § 2.

§ The terms of III (2.10) are changed by ingoing boundary conditions. The change is obtained by $k_0 \rightarrow -k_0$ and $m_t \rightarrow -m_t$. For the slab the combination is not zero for mixed ingoing-outgoing boundary conditions.

ω has finite spread however small. That is

$$\lim_{\Delta\omega \rightarrow 0} \lim_{c,d \rightarrow \infty} \frac{1}{\Delta\omega} \int_{\omega - \frac{1}{2}\Delta\omega}^{\omega + \frac{1}{2}\Delta\omega} \Sigma(\mathbf{x}, \omega') d\omega' = \mathbf{0}. \quad (2.32)$$

It is intuitive that this argument and its result (2.32) must remain true however $V \rightarrow \infty$ in each of three linearly independent directions. Thus the surface integral of the extinction theorem vanishes from the whole theory and all the consequences noted above now follow.

This academic argument does not have the satisfactory physical content of that which leads to (2.22). We shall show later in this series that the acausal Green's function (2.31) does not emerge from the quantal theory whilst the causal one does—although this is admittedly a consequence of an adiabatically switched in interaction (cf. II and Bullough *et al.* 1968). We conclude the following: the total transverse response (2.22) is the proper physical description of the transverse response and the virtual modes are necessarily accompanied by the additional real modes. This is the most important physical result of the theory. The virtual modes constitute a translationally invariant theory by, but only by the expedient of neglecting the real modes; and this is 'virtual mode' theory. There are no normal transverse modes and no solutions of the *homogeneous* fundamental integral equation III (2.1), although the modes which satisfy the dispersion relation are normal modes within the restrictions of the 'virtual mode theory'.

We can now summarize this § 2 as follows. Current forced transverse modes can exist in a finite region V . In particular, if V is the parallel-sided slab a transverse current density mode with wave vector \mathbf{k} parallel to the axis of V will excite transverse modes with the same wave vector \mathbf{k} and polarization. Since \mathbf{k} and ω do not satisfy a dispersion relation they are virtual photon-like and we call them 'virtual' modes. However, because the virtual modes induce modes of wave number k_0 inside V (a free field), and because a free field cannot exist inside V , the virtual modes of wave vector \mathbf{k} and frequency ω are solutions of the fundamental integral equation III (2.1) only if these are accompanied by two modes of wave number $m_t k_0$: these are just sufficient to annihilate the free field inside V . Because $m_t k_0$ is determined by the transverse dispersion relation III (2.4*b*) at the frequency ω these two modes are real photon-like and we call them 'real' modes. There is no reason why an externally imposed free field (incident light) should not be used to annihilate the modes of wave number k_0 inside V instead. The equations are linear and in our picture this free field generates modes of wave number $m_t k_0$ inside V sufficient to eliminate the two real modes of that wave number which otherwise accompany each virtual mode labelled by \mathbf{k} and ω .

The total response function (2.22) therefore consists of two parts: the surface-independent virtual mode response which has the transverse dispersion relation as its surfaces of singularity; and the surface-dependent real response. The real response is negligible only in the non-relativistic region where $k \gg k_0 = \omega c^{-1}$ and becomes the total response when $k \rightarrow k_0$, the situation is relativistic, and the probe is light.

We find that there are no acceptable transverse normal modes in a finite region as long as we retain outgoing boundary conditions and that it is not possible to construct a translationally invariant theory. There are formally acceptable transverse normal modes in an infinite system if causality is rejected by using mixed ingoing-outgoing boundary conditions. A translationally invariant theory which is internally consistent at the expense of causality or neglect of scattering can be set up as 'virtual

mode theory'. This simply neglects the real modes in the total transverse response, and modes which satisfy the dispersion relation are now normal modes. Virtual mode theory is incapable of describing the interaction of a system with light since this couples to a system via the surface. The results of II show that virtual mode theory has a natural and fundamental importance in the theory of bulk binding energy but it is not a physical theory.

The results in this section for the finite system are proved only when $\hat{\mathbf{k}}$ is parallel to the axis of the slab V . We now complete our analysis of the solutions of the fundamental integral equation III (2.1) by investigating oblique modes in the slab V .

3. The oblique modes

We now remove the restriction which lay on all the previous work that \mathbf{k} was parallel to the axis of the slab V . We work with single-mode probes still and as in III § 2 it is now more convenient to work in \mathbf{x} space rather than in \mathbf{k} space. We have to consider all three types of probe, light (the free field) of III § 2, the charge-longitudinal current density probe of III § 3, and the transverse current density probe of § 2 here; and we have also to investigate the oblique modes which correspond to the longitudinal normal (i.e. unforced) modes of III § 2.

The theory of the transverse response to either transverse probe is very similar to that for the longitudinal modes which we give below. We shall merely quote the results therefore: we find that when the transverse probe is light the generalizations of III (2.14) and III (2.15) exhibit all the features of the macroscopic Maxwell theory without appeal to Maxwell boundary conditions on the surface of V : Snell's law emerges without any addition to the microscopic assumption of radiating boundary conditions and the Fresnel coefficients appear: the total internal reflection of two transverse real modes of wave number $m_t k_0$ with the emission of an evanescent wave and no need of a forcing field like III (2.9) occurs. The microscopic theory thus entirely reproduces the observable optics of the macroscopic theory.

It now follows that everything we have said about the current forced modes in the previous section still holds. A probing transverse current probe with wave vector \mathbf{k} induces a transverse virtual mode of wave vector \mathbf{k} : this mode induces a mode of wave number k_0 via the surface integral of the extinction theorem: this induced free field can be extinguished by imposing an external free field: alternatively it can be extinguished by inducing two real modes proportional to

$$\{\sigma^+(\mathbf{k}) + \Lambda(\mathbf{k}, \omega)\sigma^-(\mathbf{k})\}\mathbf{E}_t(\mathbf{k}, \omega) \quad (3.1)$$

as in the § 2.

It follows that formulae (2.19) and (2.22) are valid except that $\hat{S}(\Sigma; \mathbf{k}, \omega)$ and $\Lambda(\mathbf{k}, \omega)$ take forms appropriate to the passage of transverse waves with wave numbers k and $m_t k_0$ striking the surface of V obliquely: the details can be inferred from the treatment of the longitudinal modes below. The action of the operators $\sigma^+(\mathbf{k})$ is also slightly different: these 'refract' virtual modes of wave vector \mathbf{k} to appropriate real modes with wave number $m_t k_0$ and in addition $\sigma^-(\mathbf{k})$ creates the real mode reflected from the back face of V by reversing the sign of the component of the wave vector created by $\sigma^+(\mathbf{k})$ in the direction parallel to the axis of the slab. Thus there is nothing intrinsically new about the situation: the surface-dependent terms in (2.19) and (2.22) are more complicated; but an important result is that the response function (2.9) for the virtual response is *entirely unchanged*.

There is one apparent difficulty as soon as we wish to remove the restriction to single mode probes: in the case that the probe is light we are concerned only with the real response, but even so the integral over modes \mathbf{k} in (2.3) might appear to couple different modes. However, what now happens in the case of light is that every incident mode has the wave number k_0 of the free field and every induced real mode has wave number $m_t k_0$. Then the directions of the induced modes of wave number $m_t k_0$ are 'refracted' by the integral in (2.3), assigned wave number k_0 , and matched direction by direction against the modes of the incident field. Thus there is no difficulty but merely additional complication in the response consequent upon the greater complication of the probe.

In the case of the current forced response we first find $\mathbf{P}_{t1}(\mathbf{k}, \omega)$ in response to $\mathbf{E}_t(\mathbf{k}, \omega)$: this induces modes of wave number k_0 in 'refracted directions' via the integral of (2.3). This induced free field induces the real modes of wave number $m_t k_0$ and the problem reduces as before to the solution of the integral equation III (2.7) which is the extinction theorem.

The case of the longitudinal modes needs explicit examination because of the situation surrounding the longitudinal normal modes. We consider the single longitudinal mode of wave vector \mathbf{k} in \mathbf{x} space:

$$\mathbf{P}_l \exp(i\mathbf{k} \cdot \mathbf{x}), \quad \mathbf{P}_l \times \mathbf{k} = \mathbf{0}. \tag{3.2}$$

We use the notation

$$\mathbf{k} = (m_1, m_2, m_3)k_0, \quad m = (m_1^2 + m_2^2 + m_3^2)^{1/2} \tag{3.3}$$

m may or may not be the root of the longitudinal dispersion relation III (2.4a), but when it is it is our quantity m_l . In III (2.7a) the terms

$$\frac{(\nabla\nabla + k_0^2 \mathbf{U}) \cdot \boldsymbol{\Sigma}(\mathbf{x}, \omega)}{k^2 - k_0^2} \rightarrow \frac{(\nabla\nabla + k_0^2 \mathbf{U}) \cdot \boldsymbol{\Sigma}(\mathbf{x}, \omega)}{(m^2 - 1)k_0^2} \tag{3.4a}$$

with†

$$\begin{aligned} \boldsymbol{\Sigma}(\mathbf{x}, \omega) = & \frac{2\pi \mathbf{P}_l}{(1 - m_1^2 - m_2^2)^{1/2}} [\{m_3 - (1 - m_1^2 - m_2^2)^{1/2}\} \exp\{ik_0(m_1 x_1 + m_2 x_2 + m_3 d) \\ & + (1 - m_1^2 - m_2^2)^{1/2}(d - x_3)\} - \{m_3 + (1 - m_1^2 - m_2^2)^{1/2}\} \\ & \times \exp\{ik_0(m_1 x_1 + m_2 x_2 - m_3 c + (1 - m_1^2 - m_2^2)^{1/2}(c + x_3))\}]. \end{aligned} \tag{3.4b}$$

We have assumed that V is the parallel-sided slab with axis along the 3-direction (the z -axis of the previous work). The result reduces to that of III (2.10) (longitudinal part only) when \mathbf{k} lies along the axis of the slab now called the 3-axis.

If we set‡

$$\begin{aligned} \boldsymbol{\eta} &= \{m_1, m_2, (1 - m_1^2 - m_2^2)^{1/2}\} = (\eta_1, \eta_2, \eta_3) \\ \boldsymbol{\eta}' &= \{m_1, m_2, -(1 - m_1^2 - m_2^2)^{1/2}\} = (\eta_1, \eta_2, -\eta_3) \end{aligned} \tag{3.5a}$$

† The integral involved has been evaluated by Darwin (1924). We suppose $m \neq \pm 1$.

‡ If m is complex (and if it is m_l , III (2.4a) seems to make it complex) it is not possible to make $\boldsymbol{\eta}$ a real unit vector or to set $\boldsymbol{\eta} = \eta \hat{\boldsymbol{\eta}}$ with $\hat{\boldsymbol{\eta}}$ a real unit vector. This means that $\boldsymbol{\Sigma}(\mathbf{x}, \omega)$ does not really have the wave number of the free field—a complication we do not understand at this time.

then, compactly,

$$\begin{aligned} \Sigma(\mathbf{x}, \omega) = & \frac{2\pi P_l}{\eta_3} [(m_3 - \eta_3) \exp(ik_0 \boldsymbol{\eta}' \cdot \mathbf{x}) \exp\{ik_0(m_3 + \eta_3)d\} \\ & - (m_3 + \eta_3) \exp(ik_0 \boldsymbol{\eta} \cdot \mathbf{x}) \exp\{-ik_0(m_3 - \eta_3)c\}]. \end{aligned} \quad (3.5b)$$

We now observe that if we are prepared to match this field of wave number k_0 which has been generated by the *longitudinal* field (3.2) inside V by an imposed free (transverse) field inside and outside V there is no reason why we should not do it.†

The important point in this observation is the following: $\Sigma(\mathbf{x}, \omega)$ in (3.5b) is neither longitudinal nor transverse since $\boldsymbol{\eta}$ is not parallel to \mathbf{k} (and $\boldsymbol{\eta}'$ is certainly not along $-\mathbf{k}$). However, the field (3.4a) inside V is transverse and of wave number k_0 because the tensor operator $\nabla \nabla + k_0^2 \mathbf{U}$ ensures that every mode of wave number k_0 is transverse. Thus the field of (3.4a) inside V is a free field; and this could in principle be extinguished by an externally imposed free field. Thus there is actual coupling between incident (transverse) light and the longitudinal modes $P_l \exp(i\mathbf{k} \cdot \mathbf{x})$ inside V . This seems to mean that longitudinal modes can in principle be excited by light providing that light arrives obliquely to the surface of the region V .

It is now clear why the extinction theorem vanished from the problem of longitudinal modes when \mathbf{k} lay along the axis of the slab. For this corresponds to $m_1 = m_2 = 0$, $m_3 \neq 0$ in (3.4b), and in this case P_l is along the 3-axis so that the field (3.4a) is zero. This is a unique result for the different directions of $\hat{\mathbf{k}}$ and the axis of the slab is therefore a singular direction for \mathbf{k} .

If the mode (3.2) is a charge-longitudinal current forced mode as was considered in III § 3 the situation is now clear. The primary probe $\rho(\mathbf{k}, \omega)$ induces the secondary probe $\mathbf{E}_l(\mathbf{k}, \omega)$ (cf. III (3.2a)): this longitudinal field induces $P_l(\mathbf{k}, \omega)$ according to III (2.8). Then the longitudinal $P_l(\mathbf{k}, \omega)$ (viewed here as a single mode of wave vector \mathbf{k} in \mathbf{x} space) induces a transverse field in V determined by equations (3.4). Since this is transverse and of wave number k_0 ,‡ it is a free field and the argument of § 2 applies. It follows that a transverse mode of wave number $m_t k_0$ is induced inside V and (presumably, because we have not checked the details) is accompanied by a reflected wave: since \mathbf{k} is oblique, $\boldsymbol{\eta}$ and $\boldsymbol{\eta}'$, the direction of the reflected wave vector, are oblique, and we can expect the two oblique modes with wave vectors

$$m_t k_0 = \{m_1, m_2, (m_t^2 - m_1^2 - m_2^2)^{1/2}\} k_0, \quad m_t' k_0 = \{m_1, m_2, -(m_t^2 - m_1^2 - m_2^2)^{1/2}\} k_0$$

Thus it follows that an oblique longitudinal probe of wave vector \mathbf{k} stimulates a virtual longitudinal mode response with wave vector \mathbf{k} and two *transverse* real modes with wave number $m_t k_0$. The longitudinal response relation III (3.15) is therefore replaced by an expression of the form of (2.22), namely

$$\begin{aligned} 4\pi n P_l(\mathbf{k}, \omega) = & \left\{ 1 - \frac{1}{\epsilon_l(\mathbf{k}, \omega)} \right\} \left[1 - \frac{k_0^2 (m_t^2 - 1)}{(k^2 - k_0^2)} \hat{S}_l(\Sigma; \mathbf{k}, \omega) \{ \sigma_l^+(\mathbf{k}) + \Lambda_l \sigma_l^-(\mathbf{k}) \} \right] \\ & \times \mathbf{E}_l(\mathbf{k}, \omega). \end{aligned} \quad (3.6)$$

† In this case m is m_t and III (2.4a) applies: longitudinal modes with \mathbf{k}, ω free do not occur at all without current forcing.

‡ Compare the footnote ‡ on p. 741 and note here that $k \equiv m_t k_0$ is a free variable and so real here *providing* it can still be matched, again very much as in (3.4), with the m_t of the real modes. This m_t is complex through scattering.

Here $\sigma_l^\pm(\mathbf{k})$ convert \mathbf{k} to $m_t \mathbf{k}_0$ and refract \mathbf{k} simply (+), or both refract it and reflect it (-); but they also introduce transverse polarizations perpendicular to $m_t \mathbf{k}_0$ (+) and $m_t' \mathbf{k}_0$ (-). The polarizations lie in the planes of the wave vectors and the surface normal and are coplanar: $m_t \mathbf{k}_0$, $m_t' \mathbf{k}_0$ and the surface normal are coplanar. The case $\mathbf{k} = \mathbf{k}_0$ needs special treatment which we have not given: the field (3.4a) diverges there.† The formula does not apply to the zeros of $\epsilon_i(\mathbf{k}, \omega)$ for the reasons we describe in the next paragraph.

We have not evaluated the terms $\hat{S}_i(\Sigma; \mathbf{k}, \omega)$ and $\Lambda_i(\mathbf{k}, \omega)$ in (3.6) explicitly; but $\hat{S}_i(\Sigma; \mathbf{k}, \omega) = 0$ when \mathbf{k} is axial and the details of the oblique case are implicit in the following analysis. This concerns the case of the longitudinal modes which satisfy the longitudinal dispersion relation III (2.4a). Here it is not possible to create transverse modes to eliminate the surface integral accompanying all but axial normal modes: the reason for this is that frequencies ω which are acceptable longitudinal frequencies are zeros of $\epsilon_i(\mathbf{k}, \omega)$ and hence very close to the zeros of $m_t(\omega) = 0$; when $m_t(\omega) = 0$ no transverse mode with wave number $m_t(\omega) \mathbf{k}_0$ can propagate. Obviously we have not quite excluded the possibility of a very long-wavelength transverse excitation of the proper energy eliminating the surface integral and so accompanying the longitudinal modes which satisfy the dispersion relation; but this apart we are driven to the inescapable conclusion that the oblique longitudinal modes satisfying III (2.4a) are acceptable modes only when there is an externally imposed free field. For the free field generated by these modes inside V does not consist simply of light leaving V outside V and some light must enter V . We avoided this situation in § 2 by introducing the real modes but cannot do this here. The physics of the situation seems to be that oblique longitudinal modes can radiate from the surface of V : the longitudinal dipoles are not in phase in a plane normal to the dipolar axis. Unfortunately we have not yet understood the processes of energy transport here.

We now give a precise analysis of the situation when the internal longitudinal modes satisfy the dispersion relation III (2.4a), V is the slab, and a free field is imposed both inside and outside V . We consider a single oblique mode of the free field; and this induces two longitudinal modes inside V as follows. We combine the single mode (3.2) with a second mode of vector $\mathbf{k}' = (m_1, m_2, -m_3) \mathbf{k}_0$. Since $|\mathbf{k}'| = |\mathbf{k}| = m \mathbf{k}_0$, the structure-dependent quantity $J_i(\mathbf{k}, \omega)$ is unchanged (a result of course implicit in all of this discussion of oblique modes). The second mode augments the proposed solution $\Sigma(\mathbf{x}, \omega)$ in (3.4) by

$$\Sigma'(\mathbf{x}, \omega) = \frac{2\pi \mathbf{P}_i'}{\eta_3} [(m_3' - \eta_3) \exp(i\mathbf{k}_0 \boldsymbol{\eta}' \cdot \mathbf{x}) \exp\{i\mathbf{k}_0(m_3' + \eta_3)d\} - (m_3' + \eta_3) \times \exp(i\mathbf{k}_0 \boldsymbol{\eta} \cdot \mathbf{x}) \exp\{-i\mathbf{k}_0(m_3' - \eta_3)c\}]. \tag{3.7}$$

If we include the transversality factor we find we can eliminate the mode $\exp(i\mathbf{k}_0 \boldsymbol{\eta}' \cdot \mathbf{x})$ by setting

$$\frac{2\pi}{\eta_3} (\mathbf{U} - \boldsymbol{\eta}' \boldsymbol{\eta}') \cdot [\mathbf{P}_i(m_3 - \eta_3) \exp\{i\mathbf{k}_0(m_3 + \eta_3)d\} + \mathbf{P}_i'(m_3' - \eta_3) \exp\{i\mathbf{k}_0(m_3' + \eta_3)d\}] = 0. \tag{3.8}$$

Since \mathbf{P}_i lies along $\hat{\mathbf{k}}$ and \mathbf{P}_i' along $\hat{\mathbf{k}}'$ we need

$$P_i(\mathbf{U} - \boldsymbol{\eta}' \boldsymbol{\eta}') \cdot \hat{\mathbf{k}}(m_3 - \eta_3) \exp(2i\mathbf{k}_0 m_3 d) = P_i'(\mathbf{U} - \boldsymbol{\eta}' \boldsymbol{\eta}') \cdot \hat{\mathbf{k}}'(m_3 + \eta_3). \tag{3.9}$$

† If $\rho(\mathbf{k}, \omega) = 2\pi e \delta(\omega - \mathbf{k} \cdot \mathbf{v})$, $k_0 \leq kv c^{-1} < k$, and no problem arises.

Since $\hat{\mathbf{k}}, \hat{\mathbf{k}}', \boldsymbol{\eta}, \boldsymbol{\eta}'$ all lie in the same plane,† both vectors in (3.9) are in that plane and orthogonal to $\boldsymbol{\eta}'$: hence (3.9) can be satisfied. We can therefore define a reflection coefficient Λ_l by

$$\Lambda_l = \frac{|(\mathbf{U} - \boldsymbol{\eta}'\boldsymbol{\eta}') \cdot \hat{\mathbf{k}}|(m_3 - \eta_3)\exp(2im_3k_0d)}{|(\mathbf{U} - \boldsymbol{\eta}'\boldsymbol{\eta}') \cdot \hat{\mathbf{k}}'|(m_3 - \eta_3)} \quad (3.10)$$

and $P_l' = \Lambda_l P_l$ in conformity with previous usage.

We now have remaining in the expression (3.4a) just

$$\begin{aligned} & \frac{(\mathbf{U} - \boldsymbol{\eta}\boldsymbol{\eta})}{(m^2 - 1)} \cdot \frac{2\pi P_l}{\eta_3} [-(m_3 + \eta_3)\exp\{-ik_0(m_3 - \eta_3)c\}\hat{\mathbf{k}} \\ & + \Lambda_l(m_3 - \eta_3)\exp\{ik_0(m_3 + \eta_3)c\}\hat{\mathbf{k}}'] \exp(ik_0\boldsymbol{\eta} \cdot \mathbf{x}). \end{aligned} \quad (3.11)$$

This is a wave in the plane of $\hat{\mathbf{k}}, \hat{\mathbf{k}}'$ and $\boldsymbol{\eta}$ transverse to $\boldsymbol{\eta}$. Then the longitudinal mode pair

$$P_l = P_l\{\hat{\mathbf{k}} \exp(i\mathbf{k} \cdot \mathbf{x}) + \Lambda_l \hat{\mathbf{k}}' \exp(i\mathbf{k}' \cdot \mathbf{x})\} \quad (3.12)$$

is a valid mode pair if and only if there is a transverse electric field \mathbf{E} incident on the slab. This we write as

$$\mathbf{E} = E_0\{(\mathbf{U} - \boldsymbol{\eta}\boldsymbol{\eta}) \cdot \hat{\mathbf{k}}\} \exp(ik_0\boldsymbol{\eta} \cdot \mathbf{x}) \quad (3.13)$$

which is transverse to $\boldsymbol{\eta}$ and lies in the plane of $\boldsymbol{\eta}$ and $\hat{\mathbf{k}}$.† Then (3.12) is a valid mode pair if k and ω satisfy the longitudinal dispersion relation III (2.4a) and

$$\begin{aligned} & (\mathbf{U} - \boldsymbol{\eta}\boldsymbol{\eta}) \cdot [E_0\hat{\mathbf{k}} - 2\pi P_l\{(m_l^2 - 1)\eta_3\}^{-1}[(m_3 + \eta_3)\exp\{-ik_0(m_3 - \eta_3)c\}\hat{\mathbf{k}} \\ & + \Lambda_l(m_3 - \eta_3)\exp\{ik_0(m_3 + \eta_3)d\}\hat{\mathbf{k}}']] = 0. \end{aligned} \quad (3.14)$$

We have shown that the oblique modes of the type considered in I, which satisfy the longitudinal dispersion relation first obtained in that paper and which is equation III (2.4a), can be maintained by an externally imposed (transverse) free field (light). We have scrutinized the argument from (3.7) to (3.14) very carefully and find by trial of alternatives that none but necessary assumptions have been made in finding this solution. Hence we cannot avoid from this analysis the conclusion that the longitudinal oblique modes will run if they are forced by incident *and only if* they are forced by incident light. The oblique longitudinal modes are not normal modes therefore, and the normal modes along the axis of V are singular normal modes.

The argument from (3.7) to (3.14) would seem to lead to the important physical result that the longitudinal oblique modes can be stimulated by external light and could be observed this way. However, there are a number of points which remain obscure. We have not investigated the energy transport (where problems arise in defining the fields carrying the energy); but we note that since the singular normal modes cannot carry energy across the surface of V it is not clear how the oblique modes can either. Further, if the oblique longitudinal modes can be stimulated by external light the question of uniqueness arises since there may be no reason to choose the longitudinal excitations in preference to the transverse ones even if the energies and polarization are right.‡

† The plane lies perpendicular to the direction $(m_1, -m_2, 0)$ and includes the 3-axis.

‡ According to (3.13) the polarization is in the plane of $\hat{\mathbf{k}}$ and $\boldsymbol{\eta}$ (and the 3-axis) and is transverse to $\boldsymbol{\eta}$. The refraction of $\boldsymbol{\eta}$ to $\hat{\mathbf{k}}$ depends on m_l . Snell's law holds: reflection of the incident wave and evanescence inside V seems possible if m_l is small enough.

In the case that the longitudinal excitations have characteristic energies at the zeros of $m_i(\omega)$ (so that the roots ω do not depend on \mathbf{k}) there is no problem of this kind since the transverse modes cannot run, as we have already noted, and the longitudinal modes are the only possible response to light of the characteristic energy. Indeed if $m_i = 0$ we would find from an expression like (5.4b) in the transverse case that we need a field normally incident on the surface of V since $m_1 = m_2 = 0$. In this case we know already (see III § 2) that the light is totally externally reflected from the surface of V . Thus light stimulates neither oblique or axial transverse modes, but from our analysis, can stimulate longitudinal modes. The problem of uniqueness is eliminated therefore, and we also see that the axial direction is a special direction and to that extent see why the axial longitudinal mode is normal.

However this argument cannot be quite correct since equation III (3.4a) is a dispersion relation and not an equation for characteristic energies, even though it does not depend strongly on $\mathbf{k} = m_i \mathbf{k}_0$. Indeed this is absolutely essential, since otherwise \mathbf{k} is arbitrary. However, if $\mathbf{k} \equiv m_i \mathbf{k}_0$, ω determines \mathbf{k} by the dispersion relation and then $\mathbf{k}_0 \equiv \omega c^{-1}$ and \mathbf{k} fix Λ_i by (3.10) with (3.5a), and then (3.14) fixes P_i and P_i' . This of course assumes that the root (or roots) for m_i are not incompatible with the argument of (3.7) to (3.14) and indeed that there are physically sensible roots for m_i when ω is real.

There is obviously a very intricate situation here which we have not quite unravelled. It looks as though the longitudinal modes can be stimulated by incident light and indeed must be stimulated by light to run at all. But we can accurately summarize the arguments of this section by saying only that they do not exclude the possibility of stimulating the longitudinal modes by light providing the incident wave vector is oblique to the surface and the polarization is right.

The conclusions of this § 3 are that as far as transverse modes are concerned neither the theory of the current forced modes, nor of those forced by light is changed in essence from the work of § 2 and III § 2 respectively. In particular, when the probe is light all the results of applying Maxwell's macroscopic boundary conditions at the surface of V (like Snell's law, for example) are obtained from the microscopic theory with outgoing boundary conditions. And when the probe is a transverse current the form of the total response (equation (2.22)) is unchanged but $\hat{S}(\Sigma, \mathbf{k}, \omega)$ and $\Lambda(\mathbf{k}, \omega)$ are changed in detail to take into account the changed surface geometry: both real and virtual modes are induced as before.

The oblique longitudinal modes are more complicated than those considered in § 2 and in § 3 of III. The oblique, charge-current induced, virtual photon-like (virtual) longitudinal modes are accompanied by two *transverse* real modes of wave number $m_i \mathbf{k}_0$: the total longitudinal response therefore takes the form of the total transverse response and can be exhibited as in equation (3.6). In so far as the modes which satisfy the longitudinal dispersion relations are modes of characteristic energy and frequency (and this is apparently a good approximation) these longitudinal modes cannot be accompanied by transverse real modes and can be maintained only if an external free field is imposed. Thus the theory strongly suggests that the oblique longitudinal modes can be stimulated by light and certainly does not exclude this possibility. If this is so both the (oblique) longitudinal modes and the transverse modes of I are forced modes satisfying dispersion relations. Thus they are now placed on the same conceptual footing.

Our knowledge of the oblique modes now enables us to look quickly at the normal mode problem again. This we do next.

4. Modes in cavities

The main point of this section is to show how far we can reproduce in present terms the conventional macroscopic theory of normal electromagnetic modes in a box of finite dimensions. The theory is important because electromagnetic cavities are important both in laser theory, for example, (Lamb 1964), and as a device for quantizing the fields. Although the arguments so far in this series are classical, a quantal basis has been reported (Bullough *et al.* 1968 and Bullough and Hynne 1968). We shall find that the theory is a field theory quantized in a box much larger than the region V containing matter. In this section we are interested in a box completely filled by matter.†

A set of normal modes in a macroscopic box of dielectric of *real* refractive index m_t satisfies the macroscopic boundary condition $\mathbf{E}_{\text{tan}} = \mathbf{0}$ where \mathbf{E}_{tan} is the Maxwell \mathbf{E} field component tangential to the surface Σ of the box.‡ For a rectangular box with sides perpendicular to the coordinate axes such transverse normal modes are, component wise and with appropriate phases fixed by chosen constants $c_i (i = 1, 2, 3)$

$$\mathbf{E}_t = \begin{cases} E_x = E_1^0 \cos k_1(x+c_1) \sin k_2(y+c_2) \sin k_3(z+c_3) \\ E_y = E_2^0 \sin k_1(x+c_1) \cos k_2(y+c_2) \sin k_3(z+c_3) \\ E_z = E_3^0 \sin k_1(x+c_1) \sin k_2(y+c_2) \cos k_3(z+c_3) \end{cases} \quad (4.1)$$

the vector $\mathbf{k} = (k_1, k_2, k_3)$ is real. This field satisfies an equation like (2.28b) namely

$$\nabla^2 \mathbf{E}_t(\mathbf{x}, \omega) + k_0^2 \mathbf{E}_t(\mathbf{x}, \omega) = -4\pi k_0^2 n \mathbf{P}_t(\mathbf{x}, \omega) \quad (4.2)$$

if, and only if, $k = |\mathbf{k}| = m_t k_0$ and $(m_t^2 - 1)\mathbf{E}_t = 4\pi n \mathbf{P}_t$; and it satisfies $\text{div} \mathbf{E} = 0$ if, and only if, it satisfies the transversality condition $\mathbf{k} \cdot \mathbf{E}^0 = 0$.

The transversality condition means that there are two arbitrary independent transverse polarizations. The field satisfies the boundary conditions on the box $-c_1 \leq x \leq d_1$, $-c_2 \leq y \leq d_2$, $-c_3 \leq z \leq d_3$ if, and only if

$$k_i \equiv m_i k_0 = 2\pi \nu_i (c_i + d_i)^{-1}, \quad i = 1, 2, 3 \quad (4.3)$$

and the ν_i are integers. The modes are trivial normal modes if any two of the ν_i are zero.

A set of longitudinal normal modes satisfying the same boundary conditions is

$$\mathbf{E}_l = \begin{cases} E_x = k_1 \cos k_1(x+c_1) \sin k_2(y+c_2) \sin k_3(z+c_3) \\ E_y = k_2 \sin k_1(x+c_1) \cos k_2(y+c_2) \sin k_3(z+c_3) \\ E_z = k_3 \sin k_1(x+c_1) \sin k_2(y+c_2) \cos k_3(z+c_3). \end{cases} \quad (4.4)$$

These satisfy $\text{curl} \mathbf{E}_l = \mathbf{0}$, $\text{div} \mathbf{E}_l = -4\pi n \text{div} \mathbf{P}_l$, § and the surface conditions if (4.3) holds and if \mathbf{P}_l is appropriately defined. This is the Maxwell phenomenological theory.

In the microscopic theory it is the modes of \mathbf{P}_l and \mathbf{P}_t which are the natural modes.

† Remarks on this appear in the Appendices of Bullough (1969 b).

‡ We consider this choice of boundary conditions rather than periodic ones.

§ This condition ensures that the Maxwell \mathbf{D} vector is identically zero and that the dielectric constant ϵ is identically zero: the condition $\epsilon(\omega) = 0$ fixes the possible ω as characteristic frequencies. Equation (4.3) fixes the \mathbf{k} .

For present purposes we shall therefore *define* formal Maxwell fields by

$$\mathbf{E}_{l,t}(\mathbf{x}, \omega) = \mathbf{E}_0(\mathbf{x}, \omega) + n \int_v \mathbf{F}(\mathbf{x}, \mathbf{x}'; \omega) \cdot \mathbf{P}_{l,t}(\mathbf{x}', \omega) d\mathbf{x}'. \quad (4.5)$$

So far this definition is incomplete in two ways: we have not yet defined $\mathbf{E}_0(\mathbf{x}, \omega)$ and the integral does not automatically exist.

We now propose the natural transverse normal \mathbf{P} modes

$$\mathbf{P}_t(\mathbf{x}, \omega) = \begin{cases} P_{1t}^0 \cos m_{1t}k_0(x+c_1) \sin m_{2t}k_0(y+c_2) \sin m_{3t}k_0(z+c_3) \\ P_{2t}^0 \sin m_{1t}k_0(x+c_1) \cos m_{2t}k_0(y+c_2) \sin m_{3t}k_0(z+c_3) \\ P_{3t}^0 \sin m_{1t}k_0(x+c_1) \sin m_{2t}k_0(y+c_2) \cos m_{3t}k_0(z+c_3) \end{cases} \quad (4.6a)$$

with the transverse condition $\sum_{i=1}^3 m_{it}P_{it} = 0$. In order to make

$$m_t = (m_{1t}^2 + m_{2t}^2 + m_{3t}^2)^{1/2}$$

real we shall choose

$$\mathbf{F}(\mathbf{x}, \mathbf{x}'; \omega) = \frac{1}{2}(\nabla\nabla + k_0^2 \mathbf{U})[\{\exp(ik_0r) + \exp(-ik_0r)\}r^{-1}].$$

Thus we assume acausal boundary conditions as in (2.31). Then the fluctuations contributing to m_t in the J_t do not scatter.

We also propose the natural longitudinal normal \mathbf{P} modes

$$\mathbf{P}_l(\mathbf{x}, \omega) = \begin{cases} P_l^0 m_{1l}k_0 \cos m_{1l}k_0(x+c_1) \sin m_{2l}k_0(y+c_2) \sin m_{3l}k_0(z+c_3) \\ P_l^0 m_{2l}k_0 \sin m_{1l}k_0(x+c_1) \cos m_{2l}k_0(y+c_2) \sin m_{3l}k_0(z+c_3) \\ P_l^0 m_{3l}k_0 \sin m_{1l}k_0(x+c_1) \sin m_{2l}k_0(y+c_2) \cos m_{3l}k_0(z+c_3). \end{cases} \quad (4.6b)$$

The acausal boundary conditions ensure that the fluctuations in the J_l do not scatter and we can expect real roots for m_l .

We have still to *define* the integral in (4.5) and this we do as follows. We set

$$\begin{aligned} \int_v \mathbf{F}(\mathbf{x}, \mathbf{x}'; \omega) \cdot \mathbf{P}_{l,t}(\mathbf{x}', \omega) d\mathbf{x}' &= \int_{v-v} \mathbf{F}(\mathbf{x}, \mathbf{x}'; \omega) \cdot \mathbf{P}_{l,t}(\mathbf{x}', \omega) d\mathbf{x}' \\ &+ \int_v \mathbf{F}(\mathbf{x}, \mathbf{x}'; \omega) \cdot \mathbf{P}_{l,t}(\mathbf{x}', \omega) d\mathbf{x}' \end{aligned} \quad (4.7)$$

where (as in paper I) v is a vanishingly small sphere about \mathbf{x} . The first integral on the right-hand side of (4.7) is now defined as a conditionally convergent integral, but the second is not. We therefore define this integral in the generalized function sense by defining

$$\int_v \mathbf{F}(\mathbf{x}, \mathbf{x}'; \omega) d\mathbf{x}' = -\frac{4}{3}\pi \mathbf{U}. \quad (4.8)$$

If the tensor integral exists component by component its trace must be the trace of $-(4\pi/3)\mathbf{U}$; and if it exists it can only be a scalar multiple of the unit tensor \mathbf{U} since v is spherically symmetrical. This definition is employed systematically and apparently without internal inconsistency throughout the microscopic theory: we use it in the following paper V and elsewhere (cf. Bullough *et al.* 1968).

With this definition we find that

$$\mathbf{E}_t(\mathbf{x}, \omega) = \frac{4\pi n \mathbf{P}_t(\mathbf{x}, \omega)}{m_t^2(\omega) - 1} \quad (4.9a)$$

$$\mathbf{E}_l(\mathbf{x}, \omega) = -4\pi n \mathbf{P}_l(\mathbf{x}, \omega). \quad (4.9b)$$

Thus these choices (4.6) in the microscopic theory reproduce the whole of the macroscopic theory of (4.1)–(4.4) providing $\mathbf{k} = m_t \mathbf{k}_0$ in the transverse case and $\mathbf{k} = m_l \mathbf{k}_0$ in the longitudinal case. We do not demand $m_t(\omega) = 0$ as we would in the macroscopic theory and indeed the dispersion relation for m_t , III (2.4a), shows that the condition $m_t(\omega) = 0$ is irrelevant. However the important point of difference is that we have still to satisfy the extinction theorem. If

$$\nabla \times \nabla \times \mathbf{\Sigma}(\mathbf{x}, \omega) = (\nabla \nabla + k_0^2 \mathbf{U}) \cdot \mathbf{\Sigma}(\mathbf{x}, \omega)$$

of § 3 does not vanish at all points \mathbf{x} inside the cavity V , we would need to choose $\mathbf{E}_0(\mathbf{x}, \omega)$ in (4.5) to be a non-vanishing free field just sufficient to eliminate this other vector field everywhere in V .

Unfortunately the only example we have been able to handle term by term in detail is that of an approximation to a cavity consisting of a superposition of three orthogonal parallel-sided slabs. In this case $\nabla \times \nabla \times \mathbf{\Sigma}(\mathbf{x}, \omega)$ certainly does not vanish everywhere inside V for either of the dipole fields (4.6). This must be a consequence of the approximation, however, for the following argument shows that $\nabla \times \mathbf{\Sigma}(\mathbf{x}, \omega)$ itself vanishes for the longitudinal modes (4.6b) when the cavity is the actual cavity $-c_1 \leq x \leq d_1$, etc.

The longitudinal dipole field (4.6b) satisfies $\nabla \times \mathbf{P}_l = \mathbf{0}$ everywhere inside V and \mathbf{P}_l is everywhere normal to the surface Σ of V . By taking the operator $(\nabla \nabla + k_0^2 \mathbf{U})$ outside the integral of (4.5) we immediately find that the curl of this integral is proportional to

$$\int_V \nabla_{\mathbf{x}} \frac{\exp(ik_0 r)}{r} \times \mathbf{P}_l(\mathbf{x}', \omega) d\mathbf{x}'$$

and this volume integral vanishes since $d\mathbf{S} \times \mathbf{P}_l$ vanishes everywhere on Σ and $\nabla \times \mathbf{P}_l = \mathbf{0}$ inside V . On the other hand the arguments for the extinction theorem also show that the integral of (4.5) is also proportional to

$$(\nabla \nabla + k_0^2 \mathbf{U}) \cdot \{A \mathbf{P}_l(\mathbf{x}, \omega) + \mathbf{\Sigma}(\mathbf{x}, \omega)\}$$

with A independent of \mathbf{x} . Then since $\nabla \times \mathbf{P}_l(\mathbf{x}, \omega) = \mathbf{0}$ it follows that $\nabla \times \mathbf{\Sigma}(\mathbf{x}, \omega) = \mathbf{0}$ which was to be proved. It follows that \mathbf{E}_l defined by (4.5) satisfies both (4.9b) and $\text{curl} \mathbf{E}_l \equiv \nabla \times \mathbf{E}_l = \mathbf{0}$ when but only when $\mathbf{E}_0(\mathbf{x}, \omega) \equiv \mathbf{0}$. Then \mathbf{P}_l and \mathbf{E}_l together reproduce the conventional macroscopic theory of longitudinal normal modes inside a cavity.

Two points are these: the result is achieved by adopting acausal boundary conditions which (we suppose) are sufficient to make m_t real; further the result demands that the tangential component of \mathbf{P}_l , and hence implicitly of \mathbf{E}_l , vanishes on the surface Σ of V and does not apply to the case of cyclic boundary conditions. We have not been able to show a comparable result for transverse modes, namely that $(\nabla \nabla + k_0^2 \mathbf{U}) \cdot \mathbf{\Sigma}(\mathbf{x}, \omega)$ vanishes when $\mathbf{P}_t(\mathbf{x}, \omega)$ is given by (4.6a), but the result is plausible since $\mathbf{P}_t(\mathbf{x}, \omega)$ is everywhere normal to the surface Σ of V and should not radiate from it.

We can therefore conclude that there are no optical normal modes (other than the singular axial longitudinal mode) in the parallel-sided slab and that the solutions we have obtained in this case are always unique. We also (very probably) have the important result that there are no optical normal modes inside a proper three-dimensional cavity if outgoing boundary conditions are assumed. (This result is not actually proved but certainly the longitudinal modes (4.6*b*) are not normal as soon as m_l is complex). Thus all solutions of the fundamental inhomogeneous integral equation III (2.1) which invokes outgoing boundary conditions are unique. We have also shown that there are longitudinal normal modes in such a cavity if acausal mixed ingoing-outgoing boundary conditions are assumed. These results all seem very plausible and wholly support the findings of § 2.

It is interesting to note that physical boundary conditions are presumably causal ones. We show in a following paper that these are outgoing. Thus real cavity modes exist only by adjusting to their own radiation back-scattered from the surface Σ of the cavity. The interesting question of how acausal boundary conditions emerge this way has not been solved.

The Fabry-Perot cavity (cf. Heavens 1964—pp. 31–7) is a special case since this is exactly our case of the parallel-sided slab. A small energy loss occurs here as radiative scattering but the dominant dissipative mechanism is via the surface integral of the optical extinction theorem. This emphasizes the importance of the theorem to cavity theory.

In the following paper V we focus entirely on the virtual modes and the ‘virtual mode theory’ of § 2 which simply omits the surface integral of the extinction theorem from the theory. We summarize the results of the papers III, IV and V there since the paper V completes our analysis of the classical integral equation which is equation III (2.1).

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