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The propagation of electromagnetic energy through an absorbing dielectric

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Abstract. The energy associated with an electromagnetic wave passing through a dielectric resides partly in the electromagnetic field and partly in the accompanying excitation of the dielectric. The theory of energy propagation through an absorbing classical dielectric having a single resonant frequency is presented in this paper. Simple expressions are derived for the velocity of energy transport associated with an electromagnetic wave, and for the finite energy relaxation time caused by the damping mechanism. The variations of these quantities, and of the absorption coefficient, with the relative values of the damping constant and dipole moment of the classical oscillator are investigated. This information is used to throw light on the basic mechanism of irreversible dissipation of energy by an electromagnetic wave in a dielectric. The similarities between the calculations of the dielectric constant by classical dispersion theory and by quantum mechanics are discussed.

1. Introduction

It is well known that the velocity of propagation of electromagnetic energy through a dispersive but non-absorbing dielectric is equal to the group velocity

$$v_G = d\omega/dk \tag{1}$$

where ω and k are the angular frequency and wave vector of the electromagnetic wave.

For an absorbing dielectric, on the other hand, difficulties arise in attempting to relate v_G to the velocity of energy propagation. The wave vector k is now complex, corresponding to attenuation of the wave in its passage through the material. If the real part of k is used in equation (1), it is found that v_G may be greater than the free-space velocity of light in certain frequency ranges, and may be negative in others. These features are incompatible with energy flow, and the concept of group velocity breaks down. The situation is discussed in several textbooks (for example, Stratton 1941, Panofsky and Philips 1962, Knox 1963, Born and Wolf 1965, Lipson and Lipson 1969) and has been treated in detail by Brillouin (1960).

The true energy velocity v_E for an electromagnetic wave can be defined as the rate of energy flow, determined by the Poynting vector, divided by the stored energy density of the wave. It will be shown below that v_E defined in this way is equal to v_G for a non-absorbing dielectric, but the two velocities differ in the presence of absorption.

The main aim of the present paper is to obtain expressions for v_E based on the classical oscillator model of a dielectric. The Poynting vector is easily calculated, and the significant step in deriving v_E is the calculation of the energy density associated with the wave. A previous calculation of v_E by Brillouin (1960) gives an incorrect result for the energy density, and more recently Neufeld (1966, 1969) has suggested

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that classical dispersion theory is incapable of producing a satisfactory expression for the energy density. After a review of classical theory in § 2, it will be shown in § 3 that an acceptable expression for the energy density can in fact be obtained. When the frequency of the electromagnetic wave is close to the oscillator frequency of the dielectric, a substantial part of the energy of the wave resides in the excited oscillators. This part of the energy must be added to the electromagnetic field energy to obtain a correct expression for the energy density.

The absorption coefficient is related to the energy velocity and is discussed in § 4. Expressions for v_E itself are presented in § 5. This quantity is of current interest in the theories of self-induced transparency (McCall and Hahn 1967, Courtens 1968), Raman scattering by polaritons (Loudon 1969) and resonance Raman scattering by phonons (Burstein *et al.* 1969). Use of v_G instead of v_E in the latter problem can lead to incorrect predictions, as pointed out by Loudon (1965). The energy velocity has also been discussed by Schulz-DuBois (1969).

Consideration is given in the following section to another related quantity known as the relaxation time, defined as the time taken for the energy flow to decrease to a fraction 1/e of its initial value due to the absorption. The attenuation of the electromagnetic wave is due to the dissipation of that part of the energy of the wave which resides in the dielectric oscillators. The rate of attenuation in time is proportional to the fraction of the wave's energy which resides in the oscillators.

The properties of the energy velocity, absorption coefficient and relaxation time in the limits of large and small oscillator damping are examined in § 7.

Finally, § 8 contains discussions of the relationship between the classical model and the quantum-mechanical treatment of absorption processes. It is shown that the relaxation time usually calculated quantum mechanically is simply related to the imaginary part of the dielectric constant and is not the quantum-mechanical analogue of the relaxation time defined in § 6.

2. The classical model

In classical theory, a dispersive and absorbing medium is represented by a collection of damped, non-interacting, harmonic oscillators of displacement $\boldsymbol{r}$, mass M, natural frequency ω_0 and effective charge e. Assuming a damping proportional to $\dot{\boldsymbol{r}}$, the equation of motion of an oscillator in the presence of an oscillating electric field $\boldsymbol{E}$ is:

$$M(\ddot{\boldsymbol{r}} + \Gamma\dot{\boldsymbol{r}} + \omega_0^2\boldsymbol{r}) = e\boldsymbol{E}. \tag{2}$$

It is assumed that the frequency ω of the electric field lies in the vicinity of ω_0, and that all the other resonances in the medium are at frequencies well removed from ω_0. The effect of the resonances which have frequencies greater than ω_0 can then be represented by a real constant background dielectric constant ϵ_∞. If there are no higher frequency resonances, then $\epsilon_\infty = 1$.

Local field corrections do not need to be explicitly included in the equations since their effect can always be taken into account by suitable modification of the natural frequency ω_0 and the effective charge e (see § 9 of Born and Huang 1954).

The complex polarization $\boldsymbol{P}$ has a part due to the oscillators of natural frequency ω_0, and a part due to the higher frequency resonances

$$\boldsymbol{P} = \frac{e}{V}\boldsymbol{r} + \frac{\epsilon_\infty - 1}{4\pi}\boldsymbol{E} \tag{3}$$

where V is the volume occupied by a single oscillator. For an isotropic dielectric medium or for a cubic crystal, $\boldsymbol{r}$, $\boldsymbol{E}$ and $\boldsymbol{P}$ all point in the same direction, and the complex dielectric constant is

$$\epsilon = 1 + 4\pi \frac{P}{E}. \tag{4}$$

If $\boldsymbol{r}$, $\boldsymbol{E}$ and $\boldsymbol{P}$ have space and time dependence of the form $\exp\{\mathrm{i}(\boldsymbol{k} \cdot \boldsymbol{R} - \omega t)\}$, then equations (2), (3) and (4) give

$$\epsilon = \left(\frac{kc}{\omega}\right)^2 = \epsilon_\infty + \frac{4\pi e^2}{MV} \frac{1}{-\omega^2 - \mathrm{i}\omega\Gamma + {\omega_0}^2}. \tag{5}$$

It is convenient to define a frequency Λ by

$$\Lambda^2 = \frac{4\pi e^2}{MV\epsilon_\infty}. \tag{6}$$

This is the frequency of plasma oscillations of a collection of charge carriers of mass M, charge e and concentration $1/V$ in a medium of background dielectric constant ϵ_∞. The quantity Λ measures the strength of the interaction between the oscillators and the electromagnetic field, i.e. Λ is proportional to the dipole moment of the oscillator.

The index of refraction n and the extinction coefficient κ are defined by

$$\epsilon^{1/2} = \frac{kc}{\omega} = n + \mathrm{i}\kappa. \tag{7}$$

Separation of equation (5) into real and imaginary parts now yields the usual results of the classical model:

$$n^2 - \kappa^2 = \epsilon_\infty \left\{1 + \frac{\Lambda^2({\omega_0}^2 - \omega^2)}{({\omega_0}^2 - \omega^2)^2 + \omega^2\Gamma^2}\right\} \tag{8}$$

$$2n\kappa = \frac{\epsilon_\infty \Lambda^2 \omega\Gamma}{({\omega_0}^2 - \omega^2)^2 + \omega^2\Gamma^2}. \tag{9}$$

If we denote the dielectric constant at frequencies much smaller than ω_0 by ϵ_0, it follows from (5) that

$$\begin{aligned} \epsilon_0 &= \epsilon_\infty + \frac{4\pi e^2}{MV{\omega_0}^2} \\ &= \epsilon_\infty \left\{1 + \left(\frac{\Lambda}{\omega_0}\right)^2\right\}. \end{aligned} \tag{10}$$

A common application of the theory is in the interpretation of the absorption and dispersion caused by lattice vibrations (see e.g. Spitzer *et al.* 1959). The above analysis applies directly to the vibrations of a cubic diatomic lattice if M is taken to be the reduced mass of the two atoms in the unit cell and V is the unit cell volume. Equation (10) is then the well-known expression for the effective charge e in terms of $(\epsilon_0 - \epsilon_\infty)^{1/2}$ (Born and Huang 1954, Kittel 1966).

For zero damping, $\Gamma = 0$, it is seen from (8) that n vanishes for frequencies lying in the range ω_0 to $(\omega_0{}^2+\Lambda^2)^{1/2}$. This frequency range is known as the stop-band. For frequencies outside the stop-band, k is real and the group velocity determined from (1) and (8) is

$$v_G = \frac{c}{\epsilon_\infty^{1/2}}\left\{1+\frac{\Lambda^2}{\omega_0{}^2-\omega^2}\right\}^{1/2}\left\{1+\frac{\Lambda^2\omega_0{}^2}{(\omega_0{}^2-\omega^2)^2}\right\}^{-1}. \tag{11}$$

It follows from equation (11) that v_G is always positive and never exceeds $c/\epsilon_\infty{}^{1/2}$. These properties do not hold good if v_G is evaluated using equation (1) with $k = n\omega/c$ for the case of finite damping ($\Gamma \neq 0$) (Brillouin 1960).

3. The energy density

It is straightforward to derive, from Maxwell's equations for the curls of $\boldsymbol{E}$ and $\boldsymbol{H}$, the result (Stratton 1941)

$$\frac{c}{4\pi}\int_\sigma \boldsymbol{E}\times\boldsymbol{H}\,.\,\mathrm{d}\boldsymbol{\sigma} = -\int_\tau\left\{\frac{\boldsymbol{E}\,.\,\dot{\boldsymbol{E}}+\boldsymbol{H}\,.\,\dot{\boldsymbol{H}}+4\pi\boldsymbol{E}\,.\,\dot{\boldsymbol{P}}}{4\pi}\right\}\mathrm{d}\tau \tag{12}$$

where τ is any volume and σ is the surface surrounding τ. The left-hand side represents the rate at which energy leaves τ by propagation across its surface, determined by the Poynting vector

$$\boldsymbol{S} = \frac{c}{4\pi}\boldsymbol{E}\times\boldsymbol{H}. \tag{13}$$

The integrand on the right-hand side of (12) can be re-expressed, using (2) and (3) to obtain

$$\begin{aligned}\boldsymbol{E}\,.\,\dot{\boldsymbol{P}} &= \frac{\epsilon_\infty-1}{4\pi}\boldsymbol{E}\,.\,\dot{\boldsymbol{E}}+\frac{M}{V}(\ddot{\boldsymbol{r}}+\Gamma\dot{\boldsymbol{r}}+\omega_0{}^2\boldsymbol{r})\,.\,\dot{\boldsymbol{r}}\\ &= \frac{\mathrm{d}}{\mathrm{d}t}\left\{\frac{\epsilon_\infty-1}{8\pi}E^2+\frac{M}{2V}(\dot{r}^2+\omega_0{}^2r^2)\right\}+\frac{M\Gamma\dot{r}^2}{V}.\end{aligned} \tag{14}$$

The integrand is thus the sum of a perfect time-differential and the dissipation term $M\Gamma\dot{r}^2/V$. If we define an energy density

$$W = \frac{M}{2V}(\dot{r}^2+\omega_0{}^2r^2)+\frac{\epsilon_\infty E^2+H^2}{8\pi} \tag{15}$$

and use (13) and (14), equation (12) can be written

$$\int_\sigma \boldsymbol{S}\,.\,\mathrm{d}\boldsymbol{\sigma}+\int_\tau\frac{M\Gamma\dot{r}^2}{V}\mathrm{d}\tau = -\int_\tau \dot{W}\,\mathrm{d}\tau. \tag{16}$$

Now $M\Gamma\dot{r}^2$ is the rate at which the oscillator dissipates energy owing to the damping mechanism. Equation (16) thus expresses conservation of energy in the dielectric, the terms on the left being the rate of energy loss in the volume τ by leakage across its surface and by dissipation, while the integral on the right is the rate of change of the energy stored within τ. Since $\dot{W}$ is a perfect differential, the expression (15)

for the energy density W is valid for all forms of time variation of the vector quantities and there is no restriction to harmonic oscillations in this paragraph. We note that W includes contributions from the kinetic and potential energies of the oscillators in addition to the contributions which depend on the electromagnetic field. Expressions similar to (15) have been derived for zero damping by Huang (1951) (see also Born and Huang 1954) and have been assumed by Brillouin (1960).

Returning now to the case of a harmonic time dependence for $\boldsymbol{r}$, $\boldsymbol{P}$, $\boldsymbol{E}$ and $\boldsymbol{H}$, we require expressions for the real Poynting vector $\boldsymbol{S}$ and energy density W averaged over a cycle of the oscillation. The time average of the product of the real parts of two harmonically time-varying complex numbers A and B is equal to $\frac{1}{2}\mathrm{Re}(AB^*)$. Thus, if we denote time averages by a bar, and use $H = (n+\mathrm{i}\kappa)E$, the magnitude of $\bar{\boldsymbol{S}}$ given by (13) is

$$\bar{S} = \frac{cn}{8\pi}|E|^2. \tag{17}$$

In a similar way, using equations (2), (8) and (9) to eliminate $\boldsymbol{r}$ and $\boldsymbol{H}$ from (15),

$$\begin{aligned}\overline{W} &= \frac{|E|^2}{16\pi}\left\{\frac{\epsilon_\infty \Lambda^2(\omega^2+\omega_0{}^2)}{(\omega_0{}^2-\omega^2)^2+\omega^2\Gamma^2}+\epsilon_\infty+n^2+\kappa^2\right\}\\ &= \frac{|E|^2}{16\pi}\left\{\frac{2n\kappa}{\omega\Gamma}(\omega^2+\omega_0{}^2)+\epsilon_\infty+n^2+\kappa^2\right\}\\ &= \frac{|E|^2}{8\pi}\left(\frac{2\omega n\kappa}{\Gamma}+n^2\right).\end{aligned} \tag{18}$$

Equations (17) and (18) form the basis for most of the subsequent discussion.

It is seen that $\overline{W}$ depends not only on n and κ but also on the parameters of the model used to describe the absorbing dielectric. Expression (18) is thus not completely general. The dependence of energy density on the model has been discussed by Ginzburg (1964) for the case of an electromagnetic wave interacting with a plasma. Despite this model dependence, we expect the qualitative properties of the equations derived to be independent of the details of the chosen model.

Note that the stored energy density $\overline{W}$ is always a positive quantity. Neufeld (1966, 1969) has incorrectly suggested that standard methods cannot lead to an acceptable expression for the energy density and has introduced a drastic revision of classical dispersion theory in order to surmount this supposed difficulty. In this revised theory the energy stored in the oscillators is independent of Γ. In the present theory the oscillator energy is represented by the first term in the bracket of the first line of (18), and when integrated over frequency is found to be proportional to $1/\Gamma$. This is in agreement with the predictions of quantum mechanics for the mean energy of excitation of atoms subjected to a beam of radiation.

In the limit of zero damping, where Γ and κ vanish, (18) can be written

$$\overline{W} = \frac{|E|^2}{8\pi} n \frac{\mathrm{d}}{\mathrm{d}\omega}(n\omega) \qquad \Gamma \to 0 \tag{19}$$

where (8) and (9) have been used. This is a special case of a general result derived for zero damping by several authors (Brillouin 1960, Pelzer 1951, Landau and Lifshitz 1960, Stern 1963, Erber 1964).

4. The absorption coefficient

The mean rate of dissipation of energy density is

$$\frac{M\Gamma|\dot{r}|^2}{2V} = \frac{2n\kappa\omega|E|^2}{8\pi}$$

$$= \frac{2\omega\kappa\bar{S}}{c}$$

$$= \text{oscillator kinetic-energy density} \times 2\Gamma \tag{20}$$

where equations (2), (9) and (17) have been used. The time-averaged form of (16) is thus

$$\int_\sigma \bar{\boldsymbol{S}} \cdot \mathrm{d}\boldsymbol{\sigma} + \int_\tau \frac{2\omega\kappa\bar{S}}{c}\,\mathrm{d}\tau = 0. \tag{21}$$

If $\bar{\boldsymbol{S}}$ is assumed to point in the z direction, this integral equation can be written in the differential form

$$\frac{\partial\bar{S}}{\partial z} = -\frac{2\omega\kappa\bar{S}}{c} = -\frac{\bar{S}}{L} \tag{22}$$

where

$$\frac{1}{L} = \frac{2\omega\kappa}{c} \tag{23}$$

is the absorption coefficient for the wave. That is, L is the distance after which the energy flow in the wave is reduced to $1/e$ of its original value.

The expression for the absorption coefficient derived by Seitz (1940, p. 631) differs from the above result by a factor n. This is due to his use of incorrect equations for the energy density $\overline{W}$ and for the velocity of energy propagation.

5. The energy velocity

The velocity v_E with which energy is transported through a dielectric medium is called the energy velocity. This velocity was first defined and studied theoretically by Brillouin (1960—the expression for the energy velocity obtained from equations (20) and (26) of chapter 5 of this reference is incorrect owing to errors in the equation at the top of p. 120). In terms of the quantities defined in § 3

$$v_E = \frac{\bar{S}}{\overline{W}} = \frac{c}{n+2\omega\kappa/\Gamma} \tag{24}$$

where (17) and (18) have been used. This compares with the velocity $v_P = c/n$ of the planes of constant phase.

Using (23), the expression for v_E can be rewritten

$$\frac{1}{v_E} = \frac{1}{v_P}\left(1+\frac{2\omega\kappa}{n\Gamma}\right)$$

$$= \frac{1}{v_P} + \frac{1}{L\Gamma}. \tag{25}$$

For the case of exact resonance, where $\omega = \omega_0$ and $\kappa^2+\epsilon_\infty = n^2$, it is seen from (18)

that the factor $2\omega\kappa/n\Gamma$ is just the oscillator energy density divided by the electromagnetic energy density. In this form the expression (25) for v_E is identical with that derived for the velocity of transmission of a light pulse in self-induced transparency experiments (McCall and Hahn 1967, Courtens 1968). In fact, a rigorous treatment of the velocity of propagation of a *pulse* of electromagnetic radiation (sometimes called the signal velocity) requires complicated mathematics (Brillouin 1960, Baerwald 1930, Weber and Trizna 1966). However, the detailed theory shows that in certain limits the signal velocity is identical with the energy velocity discussed here.

It may also be mentioned that the theory of the propagation of ultrasonic waves in a region of resonant absorption closely parallels that given here for electromagnetic waves. An expression for the ultrasonic energy velocity similar to (25) is derived, and the theoretical predictions have been verified experimentally (Shiren 1962, 1965).

A property of the energy velocity which is required by relativity theory is that it should be smaller than the free-space velocity of light c at all frequencies ω. It is not immediately obvious from (24) that v_E has this property. The property can be verified, however, by substituting for n and κ obtained by solution of (8) and (9).

When $\Gamma \to 0$, the expression for v_E obtained from (24) (or from (17) and (19)) is identical with the group velocity v_G defined by (1).

6. The relaxation time

In addition to the absorption coefficient $1/L$, which measures the spatial rate of attenuation of the wave, it is also possible to define a relaxation time T which determines the time rate of decay of energy flow. Consider a point which moves with the wave at the energy velocity v_E. In the notation of hydrodynamics (see e.g. Coulson 1952, p. 61) the time rate of change of the energy flow $\bar{S}$ at this moving point is

$$\begin{aligned}\frac{D\bar{S}}{Dt} &= \frac{\partial \bar{S}}{\partial t} + v_E \frac{\partial \bar{S}}{\partial z} \\ &= -\frac{v_E \bar{S}}{L} = -\frac{\bar{S}}{T}\end{aligned} \tag{26}$$

using equation (22) and the fact that $\partial \bar{S}/\partial t = 0$. Thus T is the time at which the energy flow at a point moving with the energy velocity is reduced to 1/e of its original value. The quantities v_E, L and T are related by

$$v_E = \frac{L}{T}. \tag{27}$$

From equations (23), (24) and (27):

$$\begin{aligned}\frac{1}{T} = \frac{v_E}{L} &= \frac{\bar{S}}{\overline{W}} \frac{2\omega\kappa}{c} \\ &= \frac{2\omega\kappa}{n + 2\omega\kappa/\Gamma}.\end{aligned} \tag{28}$$

Thus, in words,

$$\begin{aligned}\frac{1}{T} &= \frac{\text{Rate of dissipation of energy density}}{\text{Total energy density}} \\ &= \frac{\text{Oscillator kinetic energy density}}{\text{Total energy density}} \times 2\Gamma\end{aligned} \tag{29}$$

where all the energy densities are assumed to be time-averaged. Finally, using (8) and (9), we can write down an explicit expression for the relaxation time in terms of Γ and Λ:

$$\frac{1}{T} = \frac{4\omega^2\Lambda^2\Gamma}{[\{(\omega_0{}^2-\omega^2)^2+\omega^2\Gamma^2\}^{1/2}+\{(\omega_0{}^2-\omega^2+\Lambda^2)^2+\omega^2\Gamma^2\}^{1/2}]^2+4\omega^2\Lambda^2-\Lambda^4}. \quad (30)$$

The *positive* square roots must always be used on the right-hand side of (30).

7. Discussion

The properties of wave propagation through a dielectric can be illustrated by some limiting cases. Consider first the zero-damping limit. When Γ is very much smaller than Λ and ω_0, equation (30) reduces to

$$\frac{1}{T} = \Gamma \qquad \text{for } \omega_0{}^2 < \omega^2 < \omega_0{}^2+\Lambda^2 \quad (31)$$

$$\frac{1}{T} = \frac{\omega^2\Lambda^2\Gamma}{(\omega_0{}^2-\omega^2)^2+\omega_0{}^2\Lambda^2} \qquad \text{for } \omega^2 < \omega_0{}^2 \text{ and } \omega^2 > \omega_0{}^2+\Lambda^2. \quad (32)$$

The expressions (31) and (32) are continuous at the two ends of the stop-band. Comparison of (29) with (31) shows that, for very small Γ and for a frequency in the stop-band, half the total energy density of the wave resides in the kinetic energy of the oscillators. Evaluating κ from (8) and (9) in the limit of small Γ, the absorption coefficient given by (23) is

$$\frac{1}{L} = \frac{2\omega\epsilon_\infty{}^{1/2}}{c}\left(\frac{\Lambda^2+\omega_0{}^2-\omega^2}{\omega^2-\omega_0{}^2}\right)^{1/2} \qquad \text{for } \omega_0{}^2 < \omega^2 < \omega_0{}^2+\Lambda^2 \quad (33)$$

$$\frac{1}{L} = \frac{\omega^2\Gamma\Lambda^2\epsilon_\infty{}^{1/2}}{c|\Lambda^2+\omega_0{}^2-\omega^2|^{1/2}\,|\omega^2-\omega_0{}^2|^{3/2}} \qquad \text{for } \omega^2 < \omega_0{}^2 \text{ and } \omega^2 > \omega_0{}^2+\Lambda^2 \quad (34)$$

where only the leading term in a power series in Γ has been retained. These expressions are incorrect over small regions of extent $\sim\Gamma$ at the two ends of the stop-band.

The interpretation of the results of the last paragraph is as follows. In the limit where the damping Γ is reduced to zero, the relaxation time T tends to infinity as $1/\Gamma$. However, within the stop-band, the velocity v_E of energy transport tends to zero linearly with Γ, so that the product $v_E T$ is finite, and the absorption coefficient $1/L$ does not vanish. Outside the stop-band, where v_E remains finite as Γ tends to zero, both the inverse relaxation time and the absorption coefficient vanish in the limit. The integrated absorption coefficient for $\Gamma = 0$ obtained from (33) and (34) is

$$\int \frac{1}{L}\,d\omega = \frac{\pi\epsilon_\infty{}^{1/2}\Lambda^2}{2c}. \quad (35)$$

Indeed, this sum rule on the absorption coefficient is a general result which can be shown to hold irrespective of the magnitude of Γ (Stern 1963). The above result is in disagreement with Pekar (1959) who claims that the proportionality between the integrated absorption coefficient and the strength of the dispersion (proportional to Λ^2) is not valid in the limit of zero damping.

Now consider the limit where Γ is much larger than Λ^2/ω_0. The solutions of (8) and (9) show that $2n\kappa$ is small compared with unity and that κ is much smaller

than n. Thus if we neglect κ^2 on the left-hand side of (8) and substitute into (28):

$$\frac{1}{T} = \frac{\omega^2\Lambda^2\Gamma}{(\omega_0{}^2-\omega^2)^2+\omega^2\Gamma^2+\omega_0{}^2\Lambda^2}. \tag{36}$$

Further, if we set n equal to $\epsilon_\infty{}^{1/2}$ on the left-hand side of (9) the absorption coefficient given by (23) is

$$\begin{aligned}\frac{1}{L} &= \frac{\epsilon_\infty{}^{1/2}\omega^2\Lambda^2\Gamma/c}{(\omega_0{}^2-\omega^2)^2+\omega^2\Gamma^2} \\ &\simeq \frac{\epsilon_\infty{}^{1/2}\Lambda^2\Gamma/4c}{(\omega_0-\omega)^2+(\Gamma/2)^2}\end{aligned} \tag{37}$$

since $\Gamma \ll \omega_0$. The final approximate expression in this equation is the Lorentzian line shape. The integrated absorption coefficient given by (37) is identical with the result (35) obtained for the $\Gamma = 0$ limit.

When Γ is large, not only compared with Λ^2/ω_0 but also compared with Λ, the final term in the denominator of (36) can be neglected. Equations (36) and (37) show that v_E is equal to the phase velocity $c/\epsilon_\infty{}^{1/2}$ in this limit.

For phonons in polar diatomic cubic crystals, typical values are $\Gamma/\omega_0 = 10^{-3}$ to 10^{-2} and $\Lambda/\omega_0 = 10^{-1}$ to 1. Thus phonons correspond quite closely to the $\Gamma \ll \omega_0$ and $\Gamma \ll \Lambda$ limit discussed at the beginning of this section. For rare-earth doped crystals (e.g. Sm^{2+} in CaF_2) of the type used as laser rods, the electronic transitions of the rare-earth ions may have Γ/ω_0 of order 10^{-5} to 10^{-1} and Λ/ω_0 of order 10^{-5} to 10^{-2} with Λ generally an order of magnitude smaller than Γ. Thus the $\Gamma \gg \Lambda$ limit is usually more appropriate in this case. Exciton transitions in semiconductors show a wide variety of values of Γ and Λ. However, in many important cases, the inequality $\Gamma \gg \Lambda^2/\omega_0$ is satisfied, and Γ and Λ differ by an order of magnitude or less. The results (36) and (37) therefore apply.

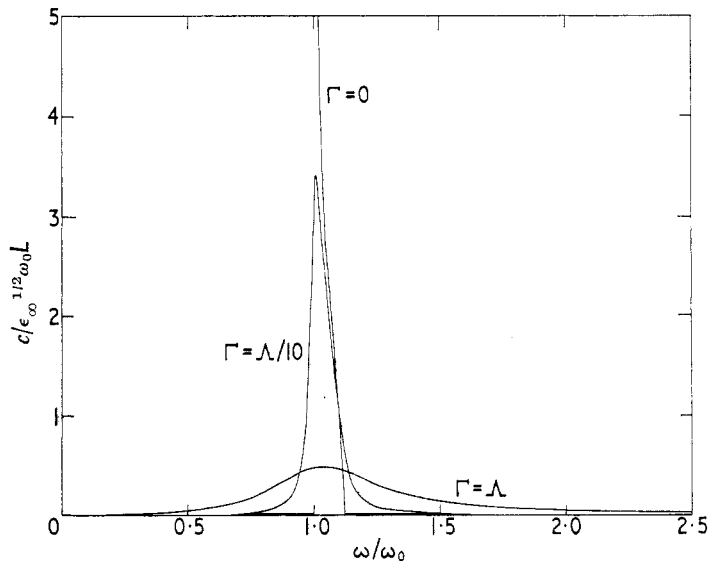

Figure 1. Frequency dependence of the absorption coefficient for $\Lambda = \omega_0/2$ and the three values of Γ indicated against the appropriate curves.

In figures 1, 2 and 3 we show the forms of $1/L$, $1/T$ and v_E as functions of frequency for $\Gamma = 0$, $\Lambda/10$ and Λ, and for $\Lambda = \omega_0/2$. This is a realistic value of Λ for the case of a phonon. In each of the three figures the quantity plotted is multiplied by a constant to produce a convenient scale and to avoid the necessity of choosing a particular value of ϵ_∞. For larger values of Γ relative to Λ the frequency dependences all become simpler, as discussed above, and we do not give any graphs for such values of Γ.

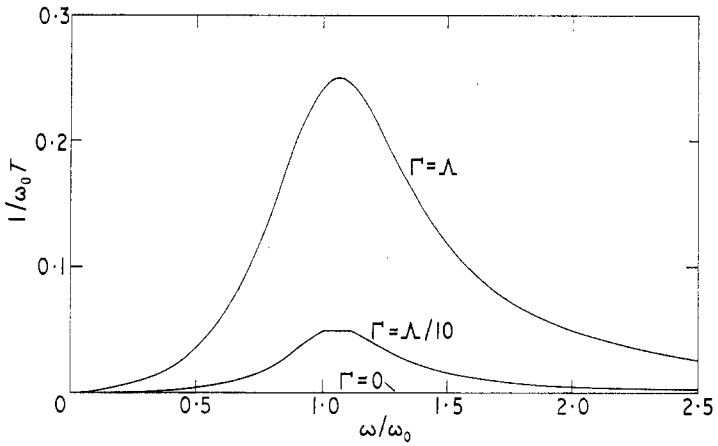

Figure 2. Frequency dependence of the relaxation rate. The rate is zero at all frequencies for $\Gamma = 0$.

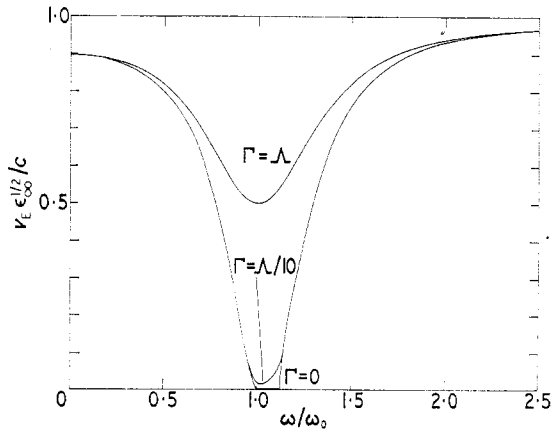

Figure 3. Frequency dependence of the energy velocity.

8. Connection with quantum mechanics

For a wide range of experiments, the simple classical theory accounts for the main features of the observed absorption and dispersion in the region of a resonance line. More detailed investigations of optical spectra sometimes reveal departures from the

simple classical formulae and require a more complex quantum-mechanical treatment. Nevertheless, it is common for quantum-mechanical treatments to lead to formulae of the classical type in some low order of calculation. It is therefore possible to make some comments on the quantum-mechanical treatment of absorption and dispersion, using as a basis the classical results of the preceding sections.

In the quantum-mechanical treatment of dispersion, in a dielectric having a single resonance, the physical system is regarded as having three components: the electromagnetic field, an oscillation (or excited state) of the dielectric which is coupled to the electromagnetic field, and a large number of excited states of the dielectric which are *not* directly coupled to the electromagnetic field. We call the excited state of the dielectric which interacts with the electromagnetic field (i.e. has a non-vanishing dipole moment) the *fundamental* state, while the large assembly of non-interacting states is called the *reservoir*. For example, in the theory of the optical properties of lattice vibrations, the long-wavelength transverse-optic vibration is the fundamental, and all the remaining vibrations of the lattice constitute the reservoir.

In an idealized measurement of the optical properties of a dielectric, a monochromatic plane-parallel beam of radiation is passed perpendicularly through a parallel slab of the dielectric and the amount of radiation transmitted through, or reflected from, the slab, parallel to the direction of incidence, is measured. Thus ideally the radiation has a single frequency ω with a well-defined wave vector k, and only a single mode of the electromagnetic field is excited. In the absence of any interaction between the fundamental state and the reservoir, the problem is simply that of a single mode of the electromagnetic field interacting with a dielectric having a single type of transition, from the ground state to the fundamental state. This problem can be solved exactly (see Landau and Lifshitz 1965, p. 139) and it is found that the dielectric oscillates in time between its ground and fundamental states, but that the time-rate of irreversible transfer of energy from the electromagnetic field to the dielectric is zero.

This situation is analogous to a classical treatment based on equation (2) but with the damping Γ neglected from the outset. However, in all real systems the fundamental is coupled to a reservoir. For example, the long-wavelength optic lattice vibration is coupled to the remaining vibrations by anharmonic interactions. The quantum-mechanical problem can in general no longer be solved exactly, and some approximate method must be used.

The most familiar approximation uses time-dependent perturbation theory (Heitler 1954), and what is calculated is the probability per unit time $1/\tau$ that the dielectric makes a transition from the ground state to an excited state accompanied by an irreversible extraction of a single quantum of energy from the radiation field. In this theory the fundamental state acts as a bridge between the electromagnetic field and the reservoir. Energy is first transferred reversibly from the electromagnetic field to the fundamental state and is then transferred irreversibly from the fundamental state to the reservoir state. The fundamental state is a virtual intermediate state in the absorption process, and the real final state of the dielectric is always one of the reservoir states. The energy carried by the electromagnetic field and the fundamental contributes to the energy density of the wave, in the spirit of § 3 above. Energy transferred to the reservoir states is irreversibly removed from the wave.

The quantum-mechanical rate at which electromagnetic energy density is dissipated in the dielectric is $\hbar\omega/V\tau$. Equating this to the analogous classical expression

in the first line of equation (20), it is found that

$$2n\kappa = \frac{8\pi\hbar}{V|E|^2\tau}. \tag{38}$$

Now $1/\tau$ calculated quantum-mechanically (using semi-classical radiation theory) is proportional to $V|E|^2$, and the right-hand side of (38) is of course independent of V and $|E|^2$. This equation can be established rigorously using the quantum-mechanical expression for the complex dielectric constant (Kubo 1957) and comparing the imaginary part with the quantum-mechanical expression for $1/\tau$ (see e.g. p. 147 of Landau and Lifshitz 1965).

The proportionality of $1/\tau$ to the imaginary part of the dielectric constant $2n\kappa$ is not usually made clear in textbook treatments. It should be emphasized that the classical relaxation rate $1/T$ introduced in § 6 refers to decay of energy flow at a point moving with the energy velocity, and is not the classical analogue of $1/\tau$.

Finally we note from (9) that, in the limit of zero Γ, the imaginary part of the dielectric constant tends to a delta function of strength $\pi\epsilon_\infty\Lambda^2/2\omega_0$ placed at $\omega = \omega_0$. This differs from the result of vanishing imaginary part at all frequencies which is obtained when Γ is neglected from the beginning. General causality requirements in the theory of the dielectric constant (Stern 1963) show that in the presence of dispersion there must always be a non-vanishing imaginary part. The correct way to treat the limit of zero damping both in classical and quantum theory is to derive expressions for the dielectric constant in the presence of damping, and then to let the damping strength tend to zero.

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