XIX. ELECTRODYNAMICS OF MEDIA[*]

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RESEARCH OBJECTIVES

A major effort in this area came to a close with the publication, in August 1967, of a research monograph[1] based on this research work. This monograph constituted Special Technical Report No. 14 of the Research Laboratory of Electronics.

Further theoretical work in electrodynamics of media will proceed in several directions. Correct relativistic equations of a quadrupolar medium are not well understood, at present, and these will be derived and examined.[2] A further examination of power flow and energy as predicted in the previous work,[1] and its relation to energy and power flow as defined from the wave point of view, will be carried out. This is especially important for magnetic material. The relationship between small-signal power theorems and the principle of virtual power will be investigated.

The theoretical and experimental work on electrodynamics of nonlinear media in the infrared range of frequencies will continue along the lines of the past year's research.[3,4] The analysis of the linear and nonlinear response in inhomogeneously broadened media will be refined and put to experimental test by propagating pulses with nanosecond rise time through a CO_2 laser amplifier at 10.6 μ. Nonlinear interactions of short pulses (of duration short compared with the inverse linewidths of the transitions) will be studied experimentally and compared with theory.

L. J. Chu, H. A. Haus, P. Penfield, Jr.

References

1. P. Penfield, Jr. and H. A. Haus, Electrodynamics of Moving Media (The M. I. T. Press, Cambridge, Mass., 1967).

2. The nonrelativistic equations have been treated in Quarterly Progress Report No. 85, Research Laboratory of Electronics, M. I. T., April 15, 1967, pp. 51-55; also to be published in Annals of Physics, April 1967 issue.

3. K. T. Gustafson and H. A. Haus, "Nonlinear Amplifications," Quarterly Progress Report No. 84, Research Laboratory of Electronics, M. I. T., January 15, 1967, pp. 113-119.

4. K. T. Gustafson, "Pulse Distortion in Nonlinear Optics," Quarterly Progress Report No. 85, Research Laboratory of Electronics, M. I. T., April 15, 1967, pp. 56-68.

A. CO_2 LASER RESEARCH

Equipment has been built for studying the linear and nonlinear pulsed response of CO_2 laser amplifiers. A variety of laser oscillators (Q-switched, cw, single P transition) have been constructed, and one multipass amplifier giving 16 dB of unsaturated amplification is used in conjunction with oscillators. Gallium arsenide electro-optic modulators are used in conjunction with cw lasers to produce pulses $\leqslant 10^{-8}$ sec long. Q-switched CO_2 oscillators produce pulses approximately 10^{-7} sec long.

For fast detectors we are using a Ge:Au detector at 77° K (20 nsec rise time), and Ge:Cu detectors at 4.2° K (1 nsec rise time). A one-half meter spectrometer is used for detection of different transitions.

The necessary electrical equipment for nanosecond voltage pulses has also been purchased or constructed. This includes a Tektronix 556 dual-beam oscilloscope with 1A1 and 1S1 plug-in units, a marx-bank voltage generator, and other voltage generators.

T. J. Bridges, P. W. Hoff

B. INTERPRETATION OF ENERGY AND POWER IN UNIFORMLY MOVING MEDIA

Many years ago, Brillouin[1] presented an expression for the energy density in a linear, time-dispersive material that is describable by a dielectric tensor $\overline{\overline{\epsilon}}$ which, in general, is a complex function of frequency.

$$W_B = \frac{1}{2} \overline{E}^* \cdot \frac{\partial}{\partial \omega} (\omega \overline{\overline{\epsilon}}) \cdot \overline{E}. \tag{1}$$

This expression was supplemented[2] later by an expression for power-flow density in such a medium,

$$(\overline{S}_B)_i = \frac{1}{2} \text{Re} \ (\overline{E} \times \overline{H}^* + \overline{E}^* \times \overline{H}) + \frac{1}{2} \overline{E}^* \cdot \frac{\partial}{\partial k_i} (\omega \overline{\overline{\epsilon}}) \cdot \overline{E}, \tag{2}$$

where k is the propagation constant. These two obey the conservation relation

$$\nabla \cdot \overline{S}_B + \frac{\partial W_B}{\partial t} = 0. \tag{3}$$

Such expressions are useful for several reasons. For example, discussions of the activity of materials can be based on investigations of whether or not W_B is inherently positive or, on the other hand, can be negative.

These expressions are based upon a form of Maxwell's equations in which the constitutive laws of the material involved ω and $\overline{k}$. This dependence upon ω and $\overline{k}$ can arise not only because of non-local interactions, or the usual dispersion in stationary

material, but also because of motion of material that, when stationary, is nondispersive.

We have investigated the physical meaning of these expressions for power in a linear isotropic dielectric material that is set in motion, and maintained in motion, at a constant, uniform velocity $\bar{v}$. In the Chu formulation of electromagnetism, the dielectric tensor $\bar{\bar{\epsilon}}$ then becomes a function of ω and $\bar{k}$, because of the motion. Thus the medium is dispersive when it is in motion, even if it is nondispersive in the rest frame. In this case, W_B and $\bar{S}_B$ do not coincide with the true power flow and energy of a moving dielectric, as reported by Penfield and Haus.[3] If certain relativistic terms are neglected, the analysis of Penfield and Haus leads to the following expressions for energy and power flow in a moving dielectric which in the rest frame is nondispersive.

$$W_T = \frac{1}{2}\epsilon_o E^2 + W_m^o \tag{4}$$

$$\bar{S}_T = \bar{E} \times \bar{H} - \bar{\bar{t}} \cdot \bar{v} - \bar{P}(\bar{E} \cdot \bar{v}) + \bar{v} W_m^o, \tag{5}$$

where W_m^o is the rest-frame energy density associated with the material. The subscript T in these expressions indicates that we consider these to be the "true" energy and power flow. These expressions obey the conservation relation,

$$\nabla \cdot \bar{S}_T + \frac{\partial W_T}{\partial t} = -\bar{f}_k \cdot \bar{v}, \tag{6}$$

where $\bar{f}_k$ is the force density acting upon the material,

$$\bar{f}_k = \nabla \cdot \bar{\bar{t}} + \bar{P} \cdot \nabla \bar{E}. \tag{7}$$

The term $-\bar{f}_k \cdot \bar{v}$ can therefore be interpreted as the power supplied, per unit volume, by the agent maintaining the material in constant, uniform motion. In these expressions, for simplicity, certain relativistic terms have been neglected.

It is obvious that W_T and $\bar{S}_T$ cannot be equal to W_B and $\bar{S}_B$. This can be seen because the conservation relation (3) has zero on the right-hand side, whereas the conservation relation (6) has something that, in general, is nonzero. Thus we are justified in concluding that W_B and $\bar{S}_B$ cannot be regarded as the true energy density and power-flow vector of a moving dielectric.

The relation between these energies and power flows can be found, however, by using the following identity which comes from Maxwell's equations.

$$\nabla \cdot \left[\bar{E} \times \bar{H} + \bar{E} \times (\bar{v} \times \bar{P})\right] + \frac{\partial}{\partial t}\left(\frac{1}{2}\epsilon_o E^2 + W_m^o\right) = 0, \tag{8}$$

where we have assumed that there is no free charge or free current, and the velocity

is uniform in space and time, as is the density of the material and its entropy. We have only retained terms to first order in the magnetic field $\overline{H}$, to be consistent with our nonrelativistic assumptions. The relativistic treatment is not difficult, however, and will be the subject of a future paper. Equation 8 can be found by taking the dot product of $\overline{v}$ with (7), and working with the resulting terms to put them in the form of perfect divergences or time rates of change. There is, however, no justification for considering the terms that appear in (8) as true power flow or energy, in any sense.

We are led to ask whether there is any relationship between the quantities in (8) and those in (1) and (2). In our simple case, for which we assume that the material is isotropic in the rest frame, and if (as before) certain relativistic terms are neglected, the dielectric tensor $\overline{\overline{\epsilon}}$ as a function of ω and $\overline{k}$ is of the form

$$\overline{\overline{\epsilon}} = \left[\epsilon + \frac{\overline{k} \cdot \overline{v}}{\omega} (\epsilon - \epsilon_0) \right] \overline{\overline{I}} - \frac{\overline{k}\,\overline{v}}{\omega} (\epsilon - \epsilon_0), \tag{9}$$

and it may be verified that the extra term in (2) reduces to $\overline{E} \times (\overline{v} \times \overline{P})$.

We see, therefore, that the extra term in (2) can be interpreted physically as arising from conversion of energy from electromagnetic form to other forms. In particular, if there is some agent that maintains the dielectric moving at a constant uniform velocity $\overline{v}$, then this agent in general must supply or accept power, and the extra term in (2) accounts for this in terms of the electromagnetic variables. This is its physical interpretation.

The energy expression (1) is often used to investigate possible activity of a physical system. For the moving dielectric, this may be negative, whereas ordinarily the true energy W_T is not. Thus we see that the physical source of the activity is, as is reasonable, the agent keeping the dielectric in motion, and indeed activity can be investigated by means of (6) by inquiring when $\overline{f}_k \cdot \overline{v}$ can be negative.

H. A. Haus, P. Penfield, Jr.

References

1. L. Brillouin, Wave Propagation and Group Velocity (Academic Press, Inc., New York, 1960).

2. P. L. Auer, H. Hurwitz, Jr., and R. D. Miller, "Collective Oscillation in a Cold Plasma," General Electric Report 58-RL-1958.

3. P. Penfield, Jr. and H. A. Haus, Electrodynamics of Moving Media (The M. I. T. Press, Cambridge, Mass., 1967), see Sec. 4.10.

C. FORCE ON MAGNETIC DIPOLE AND ELECTRIC CURRENT LOOP

Tellegen[1] has pointed out that the force exerted on a pair of magnetic charges in a time-variant electromagnetic field is different from that exerted on a circulating current loop, with the same magnetic dipole moment. He suggested that an experiment could decide whether a magnetic dipole was in fact an entity made up of two magnetic charges or a circulating current loop.

The authors[2] have pointed out that a circulating current loop in an electric field acquires a momentum which, when changed, calls for a force. When this momentum is taken into account, no difference results between a formulation of electrodynamics based on magnetic dipoles (the Chu formulation) or one based on the model in which magnetic dipoles are produced by electric current loops (Amperian current formulation).

This report gives a simple model of a circulating current loop which takes into account properly the kinetics of the charges making up the current loop. On the basis of this model, we show that a Maxwell Demon, holding a magnetic dipole in an electromagnetic field, must exert the same force on a magnetic dipole made up of two stationary magnetic charges, as he would have to exert on a magnetic dipole made up of a circulating current and having the same magnetic dipole moment.

Consider the force $\overline{F}_m$ exerted upon a stationary, constant magnetic dipole made up of equal, opposite magnetic charges q^m spaced a vector distance $\overline{d}$ apart. This force in the most general situation of a time-variant electromagnetic field is given by

$$\overline{F}_m = q^m \overline{d} \cdot \nabla \overline{H} = \mu_o \overline{m} \cdot \nabla \overline{H}, \tag{1}$$

where $\overline{m} = q^m \overline{d}/\mu_o$ is the magnetic dipole moment. Next, consider the force exerted on a circulating current of magnitude i following around a contour C. If the current is made up of positive and negative charges such that the net charge is zero everywhere, the force is entirely magnetic in character and given by

$$\overline{F}_a = \oint_C i \, \overline{ds} \times \mu_o \overline{H} = \int_s i \, (\overline{da} \times \nabla) \times \mu_o \overline{H} \tag{2}$$

Here we have used a form of Stokes' theorem. Using the vector identity

$$(\nabla \times \overline{\overline{H}}) \times \overline{da} = \overline{da} \cdot \nabla \overline{H} - \nabla \overline{H} \cdot \overline{da} \tag{3}$$

and one of Maxwell's equations,

$$\nabla \times \overline{H} = \epsilon_o \frac{\partial \overline{E}}{\partial t}, \tag{4}$$

one may write Eq. 2 in the form

$$\overline{F}_a = \mu_0 \overline{m} \cdot \nabla \overline{H} - \frac{\partial \overline{E}}{\partial t} \times \frac{\overline{m}}{c^2},$$ (5)

where we have defined the magnetic dipole moment of the current loop,

$$\mu_0 \overline{m} = \int_S i \, d\overline{a}.$$ (6)

The forces (1) and (5) are not the same, a fact that made Tellegen[1] suggest the possibility of finding out experimentally whether a magnetic dipole is made up of two opposite magnetic charges or a circulating current loop. We suggest here, on the basis of a specific model, that such a test is impossible.

Consider, first, the magnetic dipole made up of magnetic charges. These are stationary and, in order to preserve the dipole moment constant, they must be rigidly connected by a rod to overcome the attractive forces between the two magnetic charges. A Maxwell Demon holding the dipole would have to exert upon the rod a force equal and opposite to that of (1).

Next, consider the current loop. In order to construct a current loop that is electromagnetically indistinguishable from the magnetic dipole, one must constrain the current flow in the closed contour. This can be accomplished by having charges flow through a thin rigid tube following the contour C. Furthermore, if the electromagnetic field is time-variant, the induced electric field would, in general, change the rate at which the particles constituting the current would circulate around the loop. One convenient way of ensuring that the circulating current is invariant under time-variant electromagnetic fields is to assume that it is made up of "particles" that circulate at the speed of light. An applied electric field then changes their momentum but not their speed, and hence the dipole moment will remain constant, independently of the applied electromagnetic field. Finally, in order to have no magnetic charge associated with the current, it is convenient to think of the current i as made up of $2\pi Rn$ particles of positive charge circulating around one way and of an equal number of negative charge circulating in the opposite direction, so that $i = 2nqc$. The net charge per unit length of the two streams of particles is zero.

We shall compute the force exerted by a Maxwell Demon who prevents the motion of the circulating loop. Since, according to (2), the force applied to any portion of the circulating current is perpendicular to the walls of the tube confining the current, the entire force $\overline{F}_a$ is transferred to the tube. This is not the force experienced by the Maxwell Demon holding the tube stationary. The circulating particles have a mass that is a function of the electric field. If the particles gain or lose mass, the centrifugal force acting upon the walls of the confining tube will not in general cancel, and the Maxwell Demon holding the tube will have to counterbalance the additional force that arises from the unbalance of the centrifugal forces. The change of mass (momentum divided
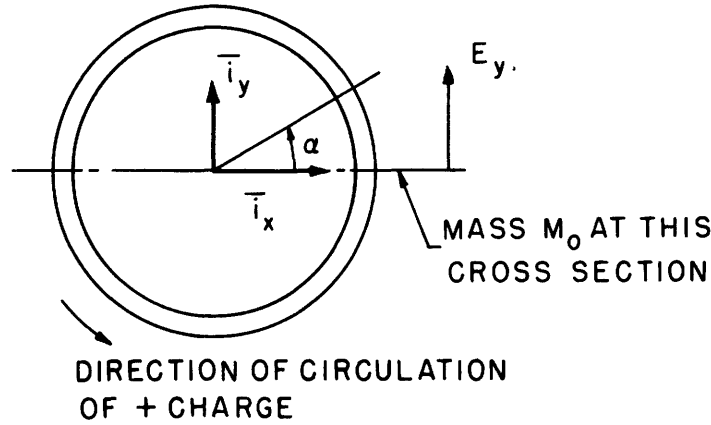
Fig. XIX-1. Magnetic dipole.

by the speed c) of a particle covering the distance $\overline{ds}$ is given by

$$dM_{\pm} = \pm \frac{q}{c^2} \overline{E} \cdot \overline{ds}, \tag{7}$$

where the upper and lower signs apply to positive and negative particles, respectively. Consider, first, a time-invariant electric field (see Fig. XIX-1). If the mass in the symmetry plane is given by M_o, the mass at any point ϕ along the contour is given by

$$M_{\pm} = M_o \pm \frac{q}{c^2} \int E_y \cos \phi\, R\, d\phi. \tag{8}$$

The net centrifugal force exerted by the counter-rotating positive and negative charges is given by

$$\overline{F}_c = \sum_{+,-} \int_0^{2\pi} \overline{i}_r (nM_{\pm}c) \frac{c}{R} R\, d\phi. \tag{9}$$

Using the fact that

$$\overline{i}_r = \overline{i}_x \cos \phi + \overline{i}_y \sin \phi, \tag{10}$$

one sees that the net centrifugal force is zero. Next, consider a time-variant field

$$\overline{E} = \overline{i}_y \left(E_y + \frac{\partial E_y}{\partial t} t \right) \qquad t > 0, \tag{11}$$

where the time t is assumed to be long enough so that several traversals around the circular contour are made by any particle, but it is short enough so that (11) is an adequate

description of the field as a function of time over the time interval considered. Since the time needed to cover an angular difference $\phi - \phi_o$, starting from the position ϕ_o at t = 0 is

$$t = \frac{\phi - \phi_o}{c} R, \tag{12}$$

one sees that a sufficiently small R allows t to be made arbitrarily small. Now consider the change $\Delta M_+(\phi)$ of the mass as caused by the time-variant field alone. Combining (8) and (12), one has

$$\Delta M_+(\phi) = \frac{q}{c^2} \frac{\partial E_y}{\partial t} \frac{R^2}{c} \int_{\phi_o}^{\phi} (\phi - \phi_o) \cos \phi \, d\phi. \tag{13}$$

This equation gives the mass of a particle at the time t positioned at the angle ϕ which was at time t = 0 at position ϕ_o. When computing the centrifugal force that is used at any particular time t (12) has to be kept fixed, and this provides a means for eliminating the starting angle ϕ_o.

$$\Delta M_+(\phi) = \frac{q}{c^2} \frac{\partial E_y}{\partial t} \frac{R^2}{c} \left[\frac{ct}{R} \sin \phi + \cos \phi - \cos \frac{ct}{R} \cos \phi + \sin \frac{ct}{R} \sin \phi \right] \tag{14}$$

Next, consider the change in mass caused by the time-variant portion of the magnetic field upon the circulating negative charges. To obtain this expression, one must simply realize that a change of mass of a negatively charged particle starting at $-\phi_o$ and ending at the angle $-\phi$ will be identical to the change of mass of a positive particle starting at $+\phi_o$ and ending at the angle $+\phi$. When one realizes that transit time of the negative particles is given by

$$t = \frac{\phi_o - \phi}{c} R, \tag{15}$$

one finds the expression

$$\Delta M_-(\phi) = \frac{\epsilon}{c^2} \frac{\partial E_y}{\partial t} \frac{R^2}{c} \left[-\frac{ct}{R} \sin \phi + \cos \phi - \cos \frac{ct}{R} \cos \phi + \sin \frac{ct}{R} \sin \phi \right]. \tag{16}$$

To compute the contribution to the centrifugal force attributable to mass changes produced by the time-variant electric field, one must evaluate

$$\Delta \overline{F}_c = \sum_{+,-} \int \overline{i}_r n \Delta M_{\pm} c^2 \, d\phi. \tag{17}$$

When (14) and (16) are substituted in (17) and the integration is carried out one finds

$$\left\langle \Delta \overline{F}_c \right\rangle = \frac{2nq}{c} \, \pi R^2 \, \frac{\partial E_y}{\partial t} \, \overline{i}_x,$$

(18)

where we have averaged the expression over several transits of the particles and have set

$$\left\langle \cos \frac{ct}{R} \right\rangle = 0.$$

(19)

With the definition

$$\overline{i}_z \, 2nqc\pi R^2 \equiv \overline{m},$$

(20)

Eq. 18 can be cast into vector form:

$$\left\langle \Delta \overline{F}_c \right\rangle = -\frac{\overline{m}}{c^2} \times \frac{\partial \overline{\overline{E}}}{\partial t}.$$

(21)

Thus far, Eq. 21 has been proved only for the special orientation of the electric field with respect to the magnetic dipole of Fig. XIX-1. One can, however, go through a similar derivation for an x-directed electric field. Since the changes produced in the mass of the circulating particles, because of the two field components, are additive, one can go independently through such an evaluation. A z-directed field does not produce such an effect. Adding contributions from the derivatives of the three field components, one obtains (21) with no restriction upon the orientation of $\partial \overline{E}/\partial t$. The time-average-centrifugal force of (21), is exerted by the particles upon the tubes confining them. A Maxwell Demon holding the circulating current loop must overcome, in addition to the forces exerted electromagnetically, the forces produced by the mass unbalance. Hence the net force that must be exerted by a Maxwell Demon is equal to the sum of Eqs. 5 and 21:

$$\overline{F}_a + \left\langle \Delta \overline{F}_c \right\rangle = \mu_o \overline{m} \cdot \nabla \overline{H}.$$

(22)

Thus we have shown that the force needed to hold a circulating current loop in a time-variant electromagnetic field is equal to the force needed to hold a magnetic dipole of equal magnetic moment made up of positive and negative magnetic charges. The proof has been conducted by using a special case. General arguments have been given else-where[2] for the equality of forces exerted upon a magnetic dipole or circulating current loops of equal magnetic dipole moment. The exercise presented here gives a picture of the reasons for this equality.

The authors are indebted to Dr. Pezaris, whose searching questions led to the model presented here.

H. A. Haus, P. Penfield, Jr.

References

1. B. D. H. Tellegen, "Magnetic-Dipole Models," Am. J. Phys. Vol. 30, No. 9, pp. 650-652, September 1962.

2. P. Penfield, Jr. and H. A. Haus, Electrodynamics of Moving Media (The M.I.T. Press, Cambridge, Mass., 1967), pp. 214-216.

D. TIME-AVERAGED ENERGY FUNCTIONS OF NONLINEAR OPTICAL MEDIA

1. Introduction

In a continuing study of the interaction of intense short electromagnetic pulses with nonlinear optical media, we have investigated the conditions under which thermodynamic principles can be employed. In general, aside from the complications arising from nonlinearity, there is the possibility of frequency dependencies of the susceptibility tensors that describe the nonlinear interaction. It is necessary to realize, in this case, that the field is established in some manner. That is, the frequency dependence introduces memory into the problem. This can be included by a consideration of specific time evolutions of the field amplitudes from their initially zero values to their final values. If one knows a priori that the "time-averaged" electric work done on the medium is a function of the state of the medium, then the choice of method for field amplitude establishment must be arbitrary.

This criterion, then, allows one to determine general conditions for which such state functions exist. These conditions will be shown to be less stringent than the steady-state power conservation requirement.

2. General Formulation

We begin with series expansions for the polarization components. A general derivation is straightforward and follows from the work of Bloembergen.[1] The analysis will be carried out for a situation in which three frequencies are present, two of which are independent. The isotropic case will also only be considered, so that all the fields are scalars. The results can easily be generalized to more complicated situations.

The electric field components are given by

$$\overline{E}_{a_k} = \frac{1}{2}\left(E_{a_k} e^{i\left(\omega_{a_k} t\right)} + c.c.\right); \qquad k = 0, 1, 2 \tag{1}$$

in which the frequencies satisfy

$$\omega_{a_0} = \omega_{a_1} + \omega_{a_2}. \tag{2}$$

The expansion of the polarization component P_{a_0} in terms of the time derivatives of the field amplitudes, is given by

$$P_{a_0} = \sum_{\ell=0}^{\infty} (-i)^\ell \left(\frac{\partial}{\partial \omega_{a_0}} \bigg|_{\substack{\omega_{a_1} \\ \omega_{a_2}}} \left(s_{a_1} + s_{a_2} \right) + \frac{\partial}{\partial \omega_{a_1}} \bigg|_{\substack{\omega_{a_0} \\ \omega_{a_2}}} \left(s_{a_1} \right) + \frac{\partial}{\partial \omega_{a_2}} \bigg|_{\substack{\omega_{a_0} \\ \omega_{a_1}}} \left(s_{a_2} \right) \right)^\ell \chi_{a_0 a_1 a_2} E_{a_1} E_{a_2}, \tag{3}$$

in which the following operator notation has been employed

$$s_{a_1} \left(E_{a_1} E_{a_2} \right) \triangleq \left(\frac{\partial E_{a_1}}{\partial t} \right) E_{a_2} \tag{4a}$$

$$s_{a_2} \left(E_{a_1} E_{a_2} \right) \triangleq E_{a_1} \left(\frac{\partial}{\partial t} E_{a_2} \right) \tag{4b}$$

$$s_a \left(E_{a_1} E_{a_2} \right) \triangleq \left(s_{a_1} + s_{a_2} \right) E_{a_1} E_{a_2}. \tag{4c}$$

Henceforth, the frequency constraints on the various partial derivatives will not be indicated explicitly. It is important, however, to notice the specific constraints, since a transformation of these derivatives will be carried out eventually.

If the notation of Bloembergen is adopted, then

$$\chi_{a_0 a_1 a_2} = \chi \left(\omega_{a_0} = \omega_{a_1} + \omega_{a_2} \right), \tag{5}$$

so, for instance,

$$\chi_{2\,1\,3} = \chi(\omega_2 = \omega_1 - \omega_3), \tag{6}$$

and thus

$$\omega_{a_0} \triangleq \omega_2 \tag{7a}$$

$$\omega_{a_1} \triangleq \omega_1 \tag{7b}$$

$$\omega_{a_2} \triangleq -\omega_3. \tag{7c}$$

The time-averaged differential work done on the nonlinear medium is given by a Legendre transform of the function G, which is simpler to deal with.

$$\frac{\partial G}{\partial t} = \left\langle \overline{P} \cdot \frac{\partial \overline{E}}{\partial t} \right\rangle = \sum_{a_o} \frac{1}{4} \left\{ P_{a_o} \left(s_{a_o} E^*_{a_o} - i\omega_{a_o} E^*_{a_o} \right) \right\},$$ (8)

where for a given a_o, $\omega_{-a_o} \triangleq -\omega_{a_o}$ and $E_{-a_o} \triangleq E^*_{a_o}$, the sum over a_o contains the three possible values of the a's and their negatives. The angular brackets denote the time average, which is taken over a few optical periods.

We wish to determine the conditions that must be obeyed in order for G to be a function of the state of the system.

First, the initial expression for P allows $\partial G/\partial t$ to be written

$$\frac{\partial G}{\partial t} = \frac{1}{4} \sum_{a_o} \sum_{\ell=0}^{2} \frac{(-i)^\ell}{\ell!} \left(s_a \frac{\partial}{\partial \omega_{a_o}} + s_{a_1} \frac{\partial}{\partial \omega_{a_1}} + s_{a_2} \frac{\partial}{\partial \omega_{a_2}} \right)^\ell \chi_{a_o a_1 a_2} E_{a_1} E_{a_2} \times \left(s_{a_o} - i\omega_{a_o} \right) E^*_{a_o}.$$ (9)

In this expression we shall investigate the first-order terms ($\ell=1$) involving ω_{a_o} and s_{a_1} or s_{a_2}, which can be written as follows:

$$\sum_{a_o} \sum_{j=1}^{2} s_{a_j} \frac{\partial}{\partial \omega_{a_j}} \chi_{a_o a_1 a_2} E_{a_1} E_{a_2} \left(-i\omega_{a_o} \right)$$

$$= \sum_{a_o} \sum_{j=1}^{2} \left(-s_{a_o} \frac{\partial}{\partial \omega_{a_o}} \chi_{a_o a_1 a_2} E_{a_1} E_{a_2} \left(i\omega_{a_j} \right) \right.$$

$$\left. + s_{a_o} \frac{\partial}{\partial \omega_{a_o}} \left(\chi_{a_o a_1 a_2} - \chi^*_{a_j a_o a_i} \right) E_{a_1} E_{a_2} \left(i\omega_{a_j} \right) \right).$$ (10)

It is apparent that $\chi_{a_o a_1 a_2}$ can be symmetrized in the pair of indices a_1, a_2, whereas this is not necessarily true for the pair a_o, a_1, or a_2. To proceed let us define the deviations from such a symmetry:

$$\chi_{a_o a_1 a_2} - \chi_{a_i a_o a_j} = X^{a_o a_i}_{a_i a_j},$$ (11)

where a_i equals either a_1 or a_2, and a_j equals the other. We then seek differential constraints involving these χ's.

Note that

$$\left(\chi^*_{a_j a_o a_i} - \chi_{a_i a_o a_j}\right) = X^{a_o a_1}_{a_1 a_o} - X^{a_o a_j}_{a_j a_o}, \tag{12}$$

then (10) can be written

$$\sum_{a_o} \sum_{j=1} \left(-s_{a_o} \frac{\partial}{\partial \omega_{a_o}} \chi_{a_o a_1 a_2}\left(i\omega_{a_o}\right) + s_{a_o} \frac{\partial}{\partial \omega_{a_o}} \left(X^{a_o a_j}_{a_j a_o}\right)\left(i\omega_{a_j}\right)\right) E^*_{a_o} E_{a_1} E_{a_2}. \tag{13}$$

Combining (13) with the terms in $\frac{\partial G}{\partial t}$ in (9) which involve ω_{a_o} and $s_a \frac{\partial}{\partial \omega_{a_o}}$, one obtains the total time derivative

$$\sum_{a_o} \frac{\partial}{\partial t} \left(-\omega_{a_o} \frac{\partial}{\partial \omega_{a_o}} \chi_{a_o a_1 a_2} E_{a_1} E_{a_2} E^*_{a_o}\right), \tag{14}$$

since $s_a + s_{a_o}$ is the total time derivative operator. If the $\ell = 0$ terms of (9) are included, this leaves the remainder

$$\sum_{a_o} \left(\chi_{a_o a_1 a_2} + \frac{\partial}{\partial \omega_{a_o}}\left[\sum_{j=1}^{2} \omega_{a_j} X^{a_o a_j}_{a_j a_o}\right]\right) E_{a_1} E_{a_2} \left(\frac{\partial}{\partial t} E^*_{a_o}\right). \tag{15}$$

This quantity must be integrable in order for the electric work done to be a state function. This in turn demands that the Maxwell relations be obeyed. These are both necessary and sufficient conditions. From (15),

$$\left(\chi_{a_o a_1 a_2} + \frac{\partial}{\partial \omega_{a_o}} \sum_{j=1}^{2} \left(\omega_{a_j} X^{a_o a_j}_{a_j a_o}\right)\right) = \chi^*_{a_j a_i a_o} - \frac{\partial}{\partial \omega_{a_j}} \sum_{i \neq j} \left(\omega_{a_i} X^{a_j a_i}_{a_i a_j}\right) \tag{16}$$

for all $a_o \neq a_i \neq a_j$. These equations constitute a system of coupled linear partial differential equations, whose solution is immediate if the frequency constraint is employed. This constraint implies that the partial derivatives used up to this point can be transformed according to, for a value of a_o,

$$\left.\frac{\partial}{\partial \omega_{a_i}}\right|_{\substack{\omega_{a_o} \\ \omega_{a_j} \\ j \neq i}} = -\left.\frac{\partial}{\partial \omega_{a_o}}\right|_{\substack{\omega_{a_1} \\ \omega_{a_2}}} + \left.\frac{\partial}{\partial \omega_{a_i}}\right|_{\omega_{a_j}} \tag{17a}$$

$$\frac{\partial}{\partial \omega_{a_o}} \equiv \frac{\partial}{\partial \omega_{a_o}} \tag{17b}$$

for any function $f(\omega_0, \omega_1, \omega_2) = f\left(\sum_{j=1}^{2} \omega_{a_j}, \omega_{a_1}, \omega_{a_2}\right)$. The derivatives on the left are those which have appeared up to this point; whereas, those on the right are directional derivatives. The derivative $\dfrac{\partial f}{\partial \omega_{a_i}}\bigg|_{\omega_{a_j}}$ lies within the plane specified by the frequency constraint. The derivative $\dfrac{\partial f}{\partial \omega_{a_o}}\bigg|_{\omega_{a_1}, \omega_{a_2}}$ is taken in a direction pointing out of the plane at some angle.

If (12) is employed in the right-hand side of (16) and (17a) and (17b) are used to transform the derivatives, the two independent integrability conditions are

$$\frac{\partial}{\partial \omega_{a_k}} \sum_{j=1}^{2} \left(\omega_{a_j} X_{a_j a_o}^{a_o a_j}\right) = 0; \qquad k = 1, 2, \tag{18}$$

where the derivatives occurring are those arising out of the last term in (16).

Consequently, the integrability condition, at least to lowest order in the amplitude time derivatives, is

$$\sum_{j=1}^{2} \left(\omega_{a_j} X_{a_j a_o}^{a_o a_j}\right) = \text{const.} \tag{19}$$

The arbitrary constant is immediately determined, since the conservation of steady-state power yields

$$E_{a_o}^{*} E_{a_1} E_{a_2} \left(-i\omega_{a_o} X_{a_o a_1 a_2} + i\omega_{a_1} X_{a_1 a_o a_2}^{*} + i\omega_{a_2} X_{a_2 a_1 a_o}^{*}\right) + \text{c.c.} = 0. \tag{20}$$

This is equivalent to (19) with the constant set equal to zero. Consequently, the conservation of power requirement, in this case, is a stronger restriction than the integrability conditions. This is in contrast to the results for nondispersive media, in which case the integrability conditions show directly that all of the X's are zero, which is more stringent than (19).

The time derivative $\left(\dfrac{\partial G}{\partial t}\right)$ can be readily determined, to first order, by adding to (14) the total time derivative form of (15), which is

$$\frac{\partial}{\partial t}\left(X_{a_o a_1 a_2} + \frac{\partial}{\partial \omega_{a_o}}\left[\sum_{j=1}^{2} \omega_{a_j} X_{a_j a_o}^{a_o a_j}\right]\right) E_{a_1} E_{a_2} E_{a_o}^{*}. \tag{21}$$

If one now uses (17) to transform the derivatives with respect to ω_{a_1} and ω_{a_2} which arise in (14), then the derivatives with respect to ω_{a_0} cancel identically and one is left with

$$G = \frac{1}{2} \text{Re} \left\{ E_{a_0}^* \left(\chi_{a_0 a_1 a_2} - \omega_{a_1} \frac{\partial}{\partial \omega_{a_1}} \chi_{a_1 a_0 a_2}^* - \omega_{a_2} \frac{\partial}{\partial \omega_{a_2}} \chi_{a_2 a_0 a_1}^* \right) \right\} E_{a_1} E_{a_2}. \quad (22)$$

To gain insight into the physical interpretation of the nonsymmetric character of the susceptibility tensor components, we consider the time-averaged electric work done, F, which is given by

$$F = <\bar{P} \cdot E> - G$$

$$= F_s + \Delta F. \quad (22a)$$

F_s is the result that would be obtained from symmetric tensor components

$$F_s = \frac{1}{2} E_{a_0}^* \left[\chi_{a_0 a_1 a_2} + \frac{1}{2} \left(\omega_{a_1} \frac{\partial}{\partial \omega_{a_1}} + \omega_{a_2} \frac{\partial}{\partial \omega_{a_2}} \right) \right] E_{a_1} E_{a_2} + \text{c. c.} \quad (23)$$

ΔF can be written as a divergence in frequency, within the plane of constraint

$$\Delta F = \frac{i}{4} \sum_{\substack{m=-2 \\ m \neq 0}}^{+2} \frac{\partial}{\partial \omega_{a_m}} \left(R_{a_m} \right) = \frac{i}{4} \nabla \cdot R, \quad (24)$$

where R_{a_m} is given by

$$R_{a_m} = \left(i \omega_{a_m} \chi_{a_m a_0}^{a_0 a_m} E_{a_0}^* E_{a_1} E_{a_2} \right) \quad (25)$$

for m equal to 1 or 2 and the respective complex conjugate for m equal to -1 or -2.

Associated with the state function, F, are two independent frequency-power formulas.[2,3] For ω_{a_1} and ω_{a_2} independent, it is easily seen that

$$\frac{P_{a_0}}{\omega_{a_0}} + \frac{P_{a_1}}{\omega_{a_1}} = \frac{\mathcal{R}_{a_1}}{\omega_{a_1}} \quad (26a)$$

$$\frac{P_{a_0}}{\omega_{a_0}} + \frac{P_{a_2}}{\omega_{a_2}} = \frac{\mathcal{R}_{a_2}}{\omega_{a_2}} \quad (26b)$$

in which

$$P_{\alpha_i} = \frac{1}{2} \operatorname{Re} \left(i\omega_{\alpha_i} P_{\alpha_i} E^*_{\alpha_i} \right) \tag{27}$$

$$\mathfrak{R}_{\alpha_i} = \frac{1}{2} \operatorname{Re} R_{\alpha_1} = \frac{1}{4} \left(R_{\alpha_i} + R_{-\alpha_i} \right). \tag{28}$$

Equations 22a, and 26 show that the vector R in frequency space, acts as a set of dependent source terms, each component giving the complex power delivered at the respective frequency. There is, however, no net power delivered by these "pseudo-sources" so that it would be more appropriate to interpret them as internal pumps acting to transfer power among the various frequency components.

The second-order terms have also been investigated. Similar results follow. In particular, the conservation of steady-state power is once again both necessary and sufficient to ensure that the time-averaged electric work done on the medium is a function of state. In this case the state depends not only upon the field amplitudes, but also upon the time derivatives.

For the particular case in which no internal sources exist $\Delta F \equiv 0$, and all the X's must be zero. The power-frequency formulas reduce to the Manley-Rowe relations, and the nondispersive and the dispersive portions of F are integrable separately. The latter portion is the Pershan F function.[4-7]

3. Conclusion

The primary result deduced is the fact that the symmetry relations for the susceptibility tensor components of a dispersive medium are not necessary in order for the time-averaged electric work done on the medium to be a function of state of the system. Since this time-averaged work is related to the internal energy increase or the free-energy increase, this result also applies to the thermodynamic state functions.

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