Spontaneous Emission in a Periodic Structure

V. P. Bykov

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Spontaneous emission by an excited atom located in a periodic structure is considered for the case when the transition frequency is in the opacity band. Decay of the excited state may occur in two ways. The atom may go over to the ground state with emission of photons in the transparency band, or it may go over to the so-called "dynamic state," which is a combination of the excited atom and its associated electromagnetic field which cannot propagate in a periodic structure. The dependence of the dynamic-state energy on transition frequency, and also the spontaneous emission spectrum, are calculated.

THE probability of spontaneous emission of an atom in free space, as is well known[1], is equal to

$$w_{sp} = {}^2/_3\pi^2\alpha\rho\omega_0 c\,|\mathbf{r}_{nm}|^2, \tag{1}$$

where α is the fine-structure constant, $\rho = \omega^2/2\pi^2c^3$ the density of states of the electromagnetic field in a unit volume, ω_0 the transition frequency, c the velocity of light, and $\mathbf{r}_{nm}$ the matrix element of the transition. In a periodic structure made up of some dielectric layers, lattices, metallic surfaces, etc., the density of states ρ is not a simple function of the frequency ω and can vanish in definite segments of the spectrum (the so-called opacity bands) and become infinite at the boundaries of these bands. In this case, formula (1) "does not work." It is therefore of interest to investigate the spontaneous decay of an excited atom located in a periodic structure. We shall consider below, by way of a simple example, an atom located in a one-dimensional periodic structure made up of a two-conductor line filled with regularly spaced homogeneous dielectric washers. This example makes it possible to illustrate all the characteristic features of the problem.

We investigate first the properties of a two-conductor line and quantize the electromagnetic fields in it, after which we consider the interaction between this field and the excited atom.

THE PERIODIC STRUCTURE

We consider a two-conductor line of arbitrary cross section (Fig. 1), filled with periodically disposed dielectric washers. Since the natural waves of the two-conductor line with homogeneous filling are known, the natural waves of the line with the periodic washers can be obtained by joining the solutions for the vacuum and the dielectric lines on the boundary between the dielectric and the vacuum. The time dependence is taken in the form $e^{i\omega t}$. The waves of the vacuum line, which travel to the left, are described by the vector potential

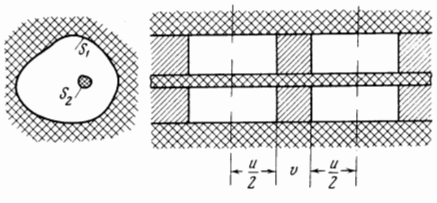

FIG. 1

$$\mathbf{A}_{vac} = A\ \mathrm{grad}\ \Pi e^{ikz}, \tag{2}$$

where $\Pi(x, y)$ is a function satisfying the equation

$$\partial^2\Pi/\partial x^2 + \partial^2\Pi/\partial y^2 = 0, \tag{3}$$

with boundary conditions $\Pi = \Pi_1$ on the surface S_1 and $\Pi = \Pi_2$ on the surface S_2. The waves traveling to the right are obtained by reversing the sign of the argument of the exponential in relation (2) and, generally speaking, replacing the coefficient A by the coefficient B.

We assume throughout that waveguide modes arise in the line under consideration at frequencies much higher than those of interest to us. This occurs when the transverse dimensions of the line are much smaller than the radiation wavelength. It is obvious, however, that these transverse dimensions should be much larger than the dimensions of the atom. Both requirements can be satisfied, since the wavelength in the optical band is larger than the atomic dimension by four orders of magnitude.

The waves of the dielectric line, which travel to the left, are described by a vector potential of the type

$$\mathbf{A}_{diel} = \mathbf{A}'\ \mathrm{grad}\ \Pi e^{ikn z}, \tag{4}$$

where n is the refractive index of the dielectric. The waves traveling to the right are obtained by reversing in (4) the sign in the argument of the exponential, and also by replacing the coefficient A′ by the coefficient B′.

For the q-th cell it is meaningful to introduce in place of the coordinate z the coordinate ζ connected with z by the relation

$$\zeta = z - (q-1)(u+v), \tag{5}$$

where u is the distance between washers and v the thickness of the washer. By making the field components that are tangent to the dielectric-vacuum interface continuous at $\zeta = u/2$ and $\zeta = u/2 + v$ we are able to change over from the fields in the q-th cell to the fields in the (q + 1)-st cell. This transition is described by the matrix

$$\|a_{ij}\| = \begin{Vmatrix} {}^1/_4\, n^{-1}e^{iku}\left[(n+1)^2 e^{iknv} - (n-1)^2 e^{-iknv}\right] & {}^1/_4\, n^{-1}(n^2-1)\left(e^{iknv} - e^{-iknv}\right) \\ -{}^1/_4\, n^{-1}(n^2-1)\left(e^{iknv} - e^{-iknv}\right) & {}^1/_4\, n^{-1}e^{-iku}\left[(n+1)^2 e^{-iknv} - (n-1)^2 e^{iknv}\right] \end{Vmatrix} \tag{6}$$

The eigenvectors of this matrix correspond to waves which, on going from cell to cell, are only multiplied by

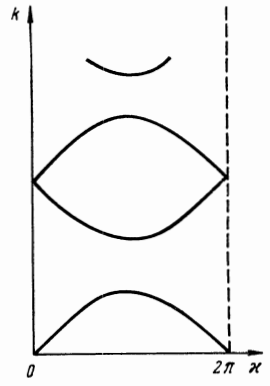

FIG. 2

a certain number (the eigenvalue X), equal to

$$X_{1,2} = Z \pm i(1 - Z^2)^{1/2} = e^{\pm i\varkappa}, \tag{7}$$

where 2Z is the trace of the matrix (6),

$$Z = {}^1/_2(a_{11} + a_{22}) = \cos\varkappa = {}^1/_4 n^{-1}[(n+1)^2 \cos k(u+nv) - (n-1)^2 \cos k(u-nv)]. \tag{8}$$

The dependence of the wave number $k = \omega/c = 2\pi/\lambda$ on the propagation constant κ defined by the relation (8) is shown in Fig. 2. It is seen from this dependence that there are frequency intervals in which wave propagation in the periodic structure is impossible. The lower and upper boundaries of these bands, called the opacity bands, are determined by the condition $\cos\kappa = -1$. The natural modes of the periodic structure are given by

$$\mathbf{A} = G \operatorname{grad} \Pi\{ae^{-i\omega t + iq\varkappa}[-a_{12}^* e^{-ikz} + (a_{11}^* - X_1^*)e^{ikz}] + a^* e^{i\omega t + iq\varkappa}[-a_{12} e^{ikz} + (a_{11} - X_1)e^{-ikz}]\}, \tag{9}$$

where κ varies from 0 to 2π. To quantize these waves, the quantities $ae^{-i\omega t}$ and $a^* e^{i\omega t}$ must be replaced respectively by the proton annihilation and creation operators a and a^+, and the normalization constant G must be chosen such that the wave energy is equal to $\hbar\omega a^+ a$[2]. From this condition we obtain for the constant G the relation

$$G^2 = \pi\hbar c^2 / QS(u+v)\omega g^2 \overline{(\operatorname{grad}\Pi)^2}, \tag{10}$$

where

$$g^2 = -\frac{-i(a_{11} - X_1)}{2n(u+v)} \times \{u[(n+1)^2 \sin k(u+nv) - (n-1)^2 \sin k(u-nv)] + nv[(n+1)^2 \sin k(u+nv) \cdot + (n-1)^2 \sin k(u-nv)]\} \tag{11}$$

and

$$\overline{(\operatorname{grad}\Pi)^2} = S^{-1} \int_S (\operatorname{grad}\Pi)^2 dS, \tag{12}$$

where S is the area of the cross section of the two-conductor line. Thus, the natural mode, when quantized, takes the following form:

$$\mathbf{A} = \mathbf{e}\left(\frac{2\pi}{QS(u+v)}\frac{\hbar}{\omega}\right)^{1/2}\frac{c}{g}\{ae^{-iq\varkappa}[-a_{12}^* e^{-ikz} + (a_{11}^* - X_1^*)e^{ikz}] + a^+ e^{iq\varkappa}[-a_{12} e^{ikz} + (a_{11} - X_1)e^{-ikz}]\}, \tag{13}$$

where

$$\mathbf{e} = (\operatorname{grad}\Pi) / [\overline{(\operatorname{grad}\Pi)^2}]^{1/2}. \tag{14}$$

RADIATION AND DYNAMIC STATE

Let the atom be located in the plane z = 0. The Hamiltonian of the complete atom plus electromagnetic field system is of the following form:

$$\hat{H} = \frac{\mathbf{p}^2}{2m} + V(\mathbf{q}) + \sum_k \hbar\omega_k a_k^+ a_k - \frac{e}{m}\sum_k \left[\frac{2\pi}{QS(u+v)}\frac{\hbar}{\omega_k}\right]^{1/2} \Lambda_k(\mathbf{pe})(a_k^+ + a_k) + \frac{\delta m}{m}\frac{\mathbf{p}^2}{m}, \tag{15}$$

where $\mathbf{p}$ and $\mathbf{q}$ are the momentum and coordinate of the electron, $\Lambda_k = g^{-1}(-a_{12} + a_{11} - X_1)$, and $\mathbf{p}^2\delta m/m^2$ is an increment corresponding to the renormalization of the mass δm[3]. We denote the states of the atoms by the Greek letters α, β, and γ. The symbol $|n\rangle$ will denote the state of the unperturbed Hamiltonian. We assume that the frequency of the $\beta \to \alpha$ transition falls in the opacity region of the periodic structure. The interaction of the electromagnetic field with the atom will be investigated by successive approximations.

Most convenient for our purposes is a variant of the method described by Heitler and Ma[4], which is close to the method of Wigner and Weisskopf[5]. This method consists of the following. It is assumed that the spectrum of the unperturbed Hamiltonian is discrete, and the stationary states of the complete Hamiltonian are sought. The problem then reduces to the stationary perturbation theory[6]. An arbitrary solution of the Schrödinger equation is presented in the form of a superposition of stationary states with corresponding time factors. If the p-th stationary state is of the form

$$\left|\psi_{st}\right\rangle = \exp\left(-\frac{i}{\hbar}E_p t\right)\sum_n \chi_n^{(p)}\left|n\right\rangle, \tag{16}$$

and the initial state $|i\rangle$ is one of the states of the unperturbed Hamiltonian, then the state at the instant t is given by

$$\left|\psi\right\rangle = \sum_n \sum_k \chi_n^{(k)} \chi_i^{(k)*} \exp\left(-\frac{i}{\hbar}E_k t\right)\left|n\right\rangle. \tag{17}$$

The initial state will henceforth be assumed to be one in which the atom is excited and there are no photons. The stationary state obtained from this initial state as a result of the action of the perturbation will be marked by the index 1.

To make the unperturbed-Hamiltonian spectrum discrete, it is necessary to introduce the fundamental region. In the case considered by us, however, we cannot confine ourselves to only one fundamental region. The reason is that both the walls of the two-con-

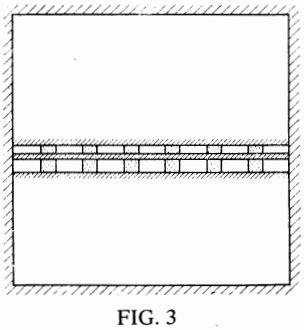

FIG. 3

ductor line and the dielectric filling the line have dispersion. At high frequencies they are completely transparent. We assume for simplicity that the optical properties of the line walls and of the dielectric vanish jumpwise at a certain high frequency ω_g. We introduce two fundamental regions for frequencies higher and lower than ω_g, respectively (3). The first fundamental region for the lower frequencies is bounded by the walls of the two-conductor line and by the boundary of the second fundamental region at the place where it intersects the two-conductor line. The second fundamental region, for high frequencies, takes the form of a cube with a side equal to the length of the two-conductor line.

There is a strong difference between the case when there is no periodic structure and the case when the structure exists. In the first case the spectrum of the oscillators of the electromagnetic field becomes more and more frequent when the fundamental region is increased and becomes continuous in the limit. The initial state then becomes one of the states of the continuous spectrum. The sum at n = 1 of the state (17) turns into an integral over the continuous spectrum, which is simply the Fourier representation of a certain time-damped function. That is to say, the initially excited state becomes damped with time. This, as is well known, is the usual process of spontaneous emission. On the other hand, if there is a periodic structure on the line, then there are opacity bands in the spectrum of the electromagnetic field, and these are not filled by the natural frequencies when the fundamental region is increased without limit. Therefore among the exponentials of the coefficient at $|n\rangle$ of the state (17) there will be an isolated exponential, say $\exp(-iE_1t/\hbar)$ corresponding to the unperturbed state when the atom is excited and there are no photons. The remaining exponentials form a continuous spectrum of single-photon states, which is the Fourier representation of a certain damped function. Consequently, in the presence of a periodic structure, the final state is a certain stationary state of the excited atom with an admixture of photons with summary energy E_1 coupled to it, and a group of emitted photons. The stationary state is a complicated mixture of the excited and unexcited states of the atom and of a certain electromagnetic field which is unable to propagate in the periodic structure. This state will henceforth be called electrodynamic or simply dynamic.

The second-order corrections to the unperturbed state are

$$E_\beta'' = \frac{e^2}{\hbar c}\sum_{\gamma\neq\beta}\frac{E_{\gamma\beta}|(\mathbf{r}_{\gamma\beta}\mathbf{e})|^2 c}{S(u+v)}\int_0^{\omega_g}\frac{(\omega_\gamma-\omega_\beta)|\Lambda|^2 d\varkappa}{(\omega_\beta-\omega_\gamma-\omega)\omega}$$

$$-\frac{8\pi}{3}\frac{e^2}{\hbar c}\sum_{\gamma\neq\beta}\frac{E_{\gamma\beta}|(\mathbf{r}_{\gamma\beta}\mathbf{e})|^2}{\lambda_{\gamma\beta}^2(\omega_\gamma-\omega_\beta)}\int_0^{\omega_g}\frac{\omega d\omega}{\omega_\beta-\omega_\gamma-\omega}$$

$$-\frac{8\pi}{3}\frac{e^2}{\hbar c}\sum_{\gamma\neq\beta}\frac{E_{\gamma\beta}|(\mathbf{r}_{\gamma\beta}\mathbf{e})|^2}{\lambda_{\gamma\beta}^2(\omega_\gamma-\omega_\beta)}\int_0^{k}\frac{\omega d\omega}{\omega_\beta-\omega_\gamma-\omega}+\hat{\mathbf{H}}_{\beta\beta}''. \quad (18)$$

In this sum we can neglect all the terms except the first. Actually, the second term is $\sim 10^{18}\mathbf{E}_{\gamma\beta}$, and the last two terms produce a small Lamb shift. The principal part of the first term is made up of the residue at the point $\kappa = \kappa_0$ corresponding to $\omega = \omega_\beta - \omega_\gamma$. The function $\omega(\kappa)$ has an infinite number of branch points. The integration

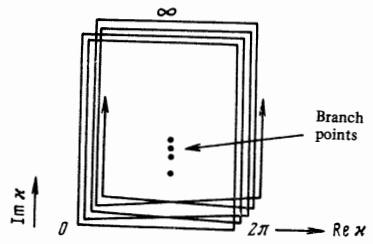

FIG. 4

contour is shown schematically in Fig. 4. With such a contour, the integral can be calculated immediately over all the branches of the dispersion curve. Since the pole $\kappa = \kappa_0$ lies inside the integration contour only for the state α, which lies lower in energy than the state β, it follows that the remaining states make no contribution to the integral. It should be noted that when taking the residue at the point $\kappa = \kappa_0$, we actually shift the upper limit of the integral to infinity. The ensuing error, however, is small, since the contribution of the high frequencies is small. We note also that it is precisely this residue which is characteristic of the periodic structure and depends on the position of the transition frequency in the opacity band.

The remaining contributions to the energy corrections will be approximately the same in the absence of a periodic structure. Thus, the correction to the energy is equal to

$$E_\beta'' = -\frac{e^2}{\hbar c}\frac{E_{\beta\alpha}|(\mathbf{r}_{\beta\alpha}\mathbf{e})|^2}{S(u+v)}\frac{|\Lambda_0|^2}{\operatorname{sh}\bar{\varkappa}_0}\frac{dZ}{dk}\Big|_{\omega=\omega_0}, \quad (19)$$

where $\bar{\kappa}_0$ is determined by the equations $\cosh\kappa_0 - \cosh\bar{\kappa}_0 = Z(\omega_0)$ and $\kappa_0 = \pi \pm i\bar{\kappa}_0$. When ω_0 approaches the end of the opacity band, $\sinh\bar{\kappa}_0 \to 0$, and consequently the corrections to the energy increase towards the edges of the opacity band. They have different signs at different edges of the opacity band, since the derivative dZ/dk reverses sign at the center of the band. The correction is positive near the lower edge of the band, and negative near the upper one. The increase of the corrections at the edges of the band is apparently connected with the fact that the formulation of the problem is not on a par at the two ends in the form presented here, for in this case one cannot investigate the radiation without considering the method of exciting the atom.

We now investigate the spontaneous-emission spectrum. To this end we find the spectral density of the energy flux far from the radiating atom. Recognizing that the electric and magnetic fields in the q-th cell at $\zeta = 0$ are equal to

$$\mathbf{E}_q = \sum_j ie\left[\frac{2\pi\hbar\omega_j}{QS(u+v)}\right]^{1/2}\Lambda_j(a_j^+\exp(iq\varkappa_j) - a_j\exp(-iq\varkappa_j)),$$

$$H_{xq} = -\sum_j ie_y\left[\frac{2\pi\hbar\omega_j}{QS(u+v)}\right]^{1/2}\Lambda_j'(a_j^+\exp(iq\varkappa_j) - a_j\exp(-iq\varkappa_j)), \quad (20)$$

$$\Lambda_j' = g^{-1}(a_{12}^* + a_{11}^* - X_1^*), \quad (21)$$

and substituting these relations into the expression for the Poynting vector, we obtain for the energy flux the expression

$$|\mathbf{S}| = \frac{c}{4\pi}|[\mathbf{EH}]| = \frac{c}{4\pi}\sum_{j,l}\mathbf{e}^2\frac{2\pi\hbar(\omega_j\omega_l)^{1/2}}{QS(u+v)}\Lambda_j\Lambda_l'$$

$$\times[\exp[iq(\varkappa_j-\varkappa_f)]+\exp[-iq(\varkappa_j-\varkappa_f)]]a_j^+a_f. \quad (22)^*$$

The terms proportional to the product of two production operators or two annihilation operators have been omitted, since their matrix elements are equal to zero for the investigated state.

We represent the state (17) in the form

$$|\psi\rangle = A(t)\,|\beta,0\rangle + B\exp\left(-\frac{i}{\hbar}E_1 t\right)\left(|\beta,0\rangle + \sum_k \chi_k^{(1)}\,|\alpha,1_k\rangle\right)$$

$$+\sum_k C_k(t)\,|\alpha,1_k\rangle, \quad (23)$$

where the first term is the damped part of the initial state and the second term the dynamic state, and the third term represents the emitted photons. Far from the radiating atom (large q) only the third term contributes to the energy flux. We shall assume that the spectral instrument separates during the measurement a narrow frequency band $\pm\Delta\omega$ near the frequency ω. Then the summation in (20) should be carried out only over this narrow interval; all the coefficients can then be made constant. Thus, the energy flowing in the entire time in the spectral interval $\Delta\omega$ or in the equivalent interval $\Delta\varkappa = (d\varkappa/d\omega)\Delta\omega$ is equal to

$$\Delta W = e^2\frac{e^2c\,|\mathbf{r}_{\alpha\beta}\mathbf{e}|^2\,\omega_{\alpha\beta}{}^2}{\pi S^2(u+v)^2(\omega_0-\omega)^2}\Lambda\Lambda'\int dt\iint d\varkappa\,d\varkappa'$$

$$\times\left\{\exp\left[i\left(q+\frac{d\omega}{d\varkappa}t\right)(\varkappa-\varkappa')\right]+\exp\left[-i\left(q-\frac{d\omega}{d\varkappa}t\right)(\varkappa-\varkappa')\right]\right\}. \quad (24)$$

Integrating first with respect to κ and then with respect to t, we obtain

$$\Delta W = 4e^2\frac{e^2c\,|\mathbf{r}_{\alpha\beta}\mathbf{e}|^2\,\omega_{\alpha\beta}{}^2}{\pi S^2(u+v)^2(\omega_0-\omega)^2}\Lambda\Lambda'\int\frac{\sin^2(q-td\omega/d\varkappa)\Delta\varkappa}{(q-td\omega/d\varkappa)^2}dt$$

$$= 4e^2\frac{e^2c\,|\mathbf{r}_{\alpha\beta}\mathbf{e}|^2\,\omega_{\alpha\beta}{}^2}{S^2(u+v)^2(\omega_0-\omega)^2}\frac{d\varkappa}{d\omega}\Delta\varkappa\Lambda\Lambda'. \quad (25)$$

The quantity $\Lambda\Lambda'$ can be represented in the form

$$\Lambda\Lambda' = 2g^{-2}\sin\varkappa(\sin\varkappa - \operatorname{Im} a_{11}) \quad (26)$$

and consequently we obtain for the density of the radiated energy the expression

$$W_\varkappa' = 8e^2\frac{e^2c\,|(\mathbf{r}_{\alpha\beta}\mathbf{e})|^2\,\omega_{\alpha\beta}{}^2}{S^2(u+v)^2(\omega_0-\omega)^2}g^{-2}(\sin\varkappa - \operatorname{Im} a_{11})\frac{dZ}{d\omega}. \quad (27)$$

Thus, the radiation energy per unit interval of the propagation constant κ has no singularities whatever on the boundary of the band. Accordingly, the spectral density of the radiation becomes infinite like $d\varkappa/d\omega$.

CONCLUSION

We see that spontaneous emission of an atom located in a periodic structure has a number of unique features. After the atom is excited, the process can proceed in two ways, either emission of photons in the allowed band, with low probability, or transition of the excited atom into a special state, which we call dynamic, with high probability (~ 1). This state is relatively long-lived. Its lifetime is determined by the small absorption in the dielectric or by two-quantum decays[7].

The spontaneous emission and the dynamic state can be observed experimentally. Let us consider two variants of such experiments. In the radio band, the most convenient spectrum for the investigation is the well-studied inversion spectrum of ammonia. The construction of a periodic structure for a wavelength of 1.25 cm entails no difficulty. The ammonia molecules can be excited optically in accordance with a three-level scheme. By varying the period of the periodic structure we can change the position of the investigated transition in the opacity band. The intensity of the spontaneous emission then decreases and becomes of second order of smallness in perturbation theory, so that the probability of production of the dynamic state is equal to $|\chi_1^{(1)}|_2$ and differs from unity in second order of perturbation theory. Pumping to saturation is easy in such a system, since practically all the ammonia molecules go over into the dynamic state and the ground-state level will be depleted. This saturation is thus proof of a slowing down of a spontaneous decay and of formation of a dynamic state.

Greater interest attaches to observation of the same phenomenon in the optical band. In view of the small wavelength ($\sim 1\ \mu$), it is difficult to produce a periodic structure in this band. Such a structure can be created, however, with the aid of standing acoustic waves. Since the speeds of light and sound differ by more than five orders of magnitude, the ultrasonic picture for light will be practically unchanged. The device as a whole has the following form. An ultrasonic wave whose length is half the optical wavelength is fed to the end of a light pipe whose diameter is smaller than the optical wavelength. If luminescent ions are introduced into the material of the light pipe (say chromium or neodymium ions), then their luminescence, which is directed along the light pipe, will decrease sharply when the ultrasound is excited at those instants of time when the deviations of the density from the mean value are maximal. The excitation of the luminescence can be carried out optically through the side surface of the light pipe. Since the relative width of the luminescence line does not exceed 10^{-2}, it is necessary to ensure in the periodic structure $\Delta n/n \sim 10^{-2}$, and consequently the ratio of the change of density in the ultrasonic wave to the density itself, $\epsilon = \Delta\rho/\rho$, should also be of the same magnitude. The energy flux in the ultrasonic wave can be represented in the form

$$s = {}^1/_2\rho u^3\epsilon^2,$$

where u is the speed of sound and ρ the density of the light-pipe material. Assuming $\rho \cong 1\ g/cm^3$, $u \cong 10^5$ cm/sec, and $\epsilon = 10^{-2}$, we obtain $s \cong 5\ kW/cm^2$. This quantity is appreciable, but it must be taken into account that the total radiation power is small. It is equal to $s_{tot} \cong s\lambda^2 = 5\times10^{-5}$ W. The frequency of the ultrasound is approximately 10^9 Hz, and is experimentally attainable.

Another possibility of investigating spontaneous emission in periodic structures in the optical region is to produce in a sample, with the aid of ultrasound, a three-dimensional periodic structure such that the opacity band exists in all three directions. It is of interest to measure the dynamic state directly, say the distribution of the electromagnetic energy in it. Such experiments, however, are complicated and call for a separate analysis.

*$[\mathbf{EH}] \equiv \mathbf{E}\times\mathbf{H}$.

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