POSSIBILITY OF A RESONANCE CHANGE IN THE COHERENT PROPERTIES OF LIGHT SCATTERED BY A MULTILEVEL SYSTEM

É. Sh. TEPLITSKIĬ

Submitted December 7, 1967

Zh. Eksp. Teor. Fiz. 55, 1272–1276 (October, 1968)

The effect of statistical properties of an electromagnetic wave traversing a system of atoms on the statistical properties of another wave is considered.

COHERENT properties of an electromagnetic field play a definite role in the interaction of light with matter leading to certain specific features in the development of physical processes and to a change in the statistical characteristics of the medium; at the same time the statistical properties of the scattered light in turn depend on the properties and the physical state of the scatterer (e.g., in the presence of inhomogeneities and of fluctuating parameters[1,2]). Coherent properties can also be altered when light passes through homogeneous linear media, provided that strong absorption occurs in them at the frequencies of the incident light fields[3]. As is shown below in such a medium a characteristic interaction of light with light can occur in which only its statistical properties are altered. This interaction arises as a result of simultaneous excitation of a multilevel atomic system by both fields and, as it has turned out, has its greatest value when the frequencies ω_1 and ω_2 are equal to each other and coincide with the corresponding transition frequency Ω_1 in atoms. This process may be of interest in those phenomena where the intensity of the transmitted light plays no special role, and measurement is restricted merely to the response of one of the monochromatic light beams to the effect of a second beam whose coherent properties are known or are to be determined.

In the case under consideration the Hamiltonian of the system acted upon by two light beams of frequencies ω_1 and ω_2 has the form

$$H = \hbar\omega_1 a_1^+ a_1 + \hbar\omega_2 a_2^+ a_2 + \hbar \sum_r \Omega_r \sum_{n=1}^{N(r)} \sigma_n^+(r)\sigma_n^-(r) + \hbar \sum_{i=1}^{2} \sum_{rn} (\lambda_{ri}\sigma_n^-(r) a_i^+ + \lambda_{ri}^* \sigma_n^+(r) a_i), \tag{1}$$

where

$$\lambda_{ri} = (2\pi / \varepsilon\hbar\omega_i V)^{1/2} (\mathbf{M}_{ri}\eta_{ik}) e^{ikr}, \tag{2}$$

$\epsilon(\mathbf{r})$ is the dielectric permittivity of the medium, η_{ik} are the polarization vectors, $\mathbf{M}_{ri}$, are the matrix elements for the transitions in the dipole approximation, a_i^+, a_i, are the creation and annihilation operators for the quanta, $[a_i, a_j^+] = \delta_{ij}$, while σ_n^+, σ_n^- are the operators for the excitation of the atoms, for which the relations $\sigma_n^2 = 0$, $[\sigma_n^-, \sigma_m^+] = [\sigma_n^-, \sigma_m^-] = 0$, $n \neq m$, and $\{\sigma_n^-, \sigma_m^+\} = 1$ hold. In (1) it is assumed that the first beam of light interacts with the medium during a time $t_1 \leq t \leq t_2$, while the second beam interacts during a time $T_1 \leq t \leq T_2$, with $T_1 < T_2$. Dissipation is not included in the Hamiltonian (1) since its contribution does not alter in any essential manner the final formulas which characterize the process under consideration.

As is well known, the coherent properties of electromagnetic fields are determined by field averages of normally ordered products of positive and negative frequency parts of the electric field vector in the Heisenberg representation[4]. Generally speaking, it is not possible in the case of interacting fields to carry out the separation in terms of the sign of the frequencies, but scattered fields, since for them the scattering medium and the photon detector are spatially separated, can be described by asymptotic (for $t = \pm\infty$) states which do allow such a separation. For the construction of $a_{ki}(\pm\infty)$ we use the system of Heisenberg equations for all the operators appearing in (1):

$$a_j(t) = e^{-i\omega_j t}\left[a_j(-\infty) - i\int_{-\infty}^{t} \sum_r \lambda_{rj} C(r, t') e^{i\omega_j t'} dt' \right],$$

$$a_j(t) = e^{-i\omega_j t}\left[a_j(+\infty) + i\int_{t}^{\infty} \sum_r \lambda_{rj} C(r, t') e^{i\omega_j t'} dt' \right],$$

$$j = 1, 2, \tag{3}$$

$$C(r, t) = e^{-i\Omega_r t}\left[C(r, 0) + iS_r(j)\lambda_{r1}^* \int_{t_1}^{t} e^{i\Omega_r t'} a_1(t') dt' + iS_r(j)\lambda_{r_2}^* \int_{T_1}^{t} e^{i\Omega_r t'} a_2(t') dt' \right],$$

where

$$C(r, t) = \sum_{n=1}^{N(r)} \sigma_n^-(r, t)$$

and the following approximation[5] has been utilized

$$S_r = -[C(r, t),\ C^+(r, t)] = -\langle [C(r, 0), C^+(r, 0)] \rangle = N_+(r) - N_-(r),$$

which has enabled us to linearize the system (3) (N_+ and N_- are the numbers of excited and unexcited atoms in the initial state).

In the lowest approximation the relation between the asymptotic states of the first field which follows from (3) has the form

$$a_1(+\infty) = \Delta_1 a_1 + \Delta_2 a_1, \tag{4}$$

where $\Delta_1 a_1$ is the contribution of the interaction with the medium to the change in a_1:

$$\Delta_1 a_1 = a_1(-\infty)\left\{ 1 - i(t_2 - t_1) \sum_r \frac{|\lambda_{r1}|^2 S_r(j)}{\Omega_r - \omega_1} + \sum_r \frac{|\lambda_{r1}|^2 S_r(j)}{(\Omega_r - \omega_1)^2} (1 - e^{i(\omega_1 - \Omega_r)(t_2 - t_1)}) \right\}$$

$$+\sum_{r}\frac{\lambda_{r1}C(r,0)}{\Omega_r-\omega_1}\left(e^{i(\omega_1-\Omega_r)t_2}-e^{i(\omega_1-\Omega_r)t_1}\right), \quad (5)$$

while $\Delta_2 a_1$ is the corresponding contribution of the second field

$$\Delta_2 a_1 = a_2(-\infty)\sum_r \frac{\lambda_{r1}\lambda_{r2}^* S_r}{(\Omega_r-\omega_2)}\left[\frac{e^{i(\omega_1-\omega_2)t_2}-e^{i(\omega_1-\omega_2)t_1}}{\omega_2-\omega_1} + e^{i(\Omega_r-\omega_2)T_1}\frac{e^{i(\omega_1-\Omega_r)t_2}-e^{i(\omega_1-\Omega_r)t_1}}{\omega_1-\Omega_r}\right]. \quad (6)$$

The interaction of light with light in the sense indicated above is characterized by the second term in (4). We consider the change in the statistics of the first field associated with this assuming that it is possible to carry out an experiment in which all the other changes in it (as a result of fluctuations, inhomogeneities, etc.) can be compensated for. In particular, this can occur if the light beam under investigation is divided into two beams passing through identical samples of a given medium, and the statistics of one of these beams is compared with the statistics of the other one. If one of the samples is subjected to the action of a second light field then a difference will appear between the two statistics being compared which is associated only with the term $\Delta_2 a_1$ in (4).

The most convenient method for the investigation of coherence is the determination of the statistics of photoelectrons which, in principle, allows one to construct correlation functions for the field of arbitrary order since the factorial moments for the distribution of photon counts during a time (O, t) are determined by means of the coherence functions

$$G^{(n)}(\mathbf{r}_1t_1,\dots\mathbf{r}_nt_n;\mathbf{r}_n't_n',\dots,\mathbf{r}_1't_1') = \mathrm{Sp}\{\rho E^{(-)}(\mathbf{r}_1t_1)\dots E^{(-)}(\mathbf{r}_nt_n)E^{(+)}(\mathbf{r}_n't_n')\dots E^{(+)}(\mathbf{r}_1't_1')\} \quad (7)$$

by the expression[4]

$$\left\langle\frac{C!}{(C-n)!}\right\rangle = \sum_{\mathbf{k}_1\dots\mathbf{k}_{2n}} V_{\mathbf{k}_1\dots\mathbf{k}_{2n}}(t)\langle a_{\mathbf{k}_1}^+\dots a_{\mathbf{k}_n}^+ a_{\mathbf{k}_{n+1}}\dots a_{\mathbf{k}_{2n}}\rangle, \quad (8)$$

where

$$V_{\mathbf{k}_1\dots\mathbf{k}_{2n}}(t) = \int_0^t\dots\int_0^t \prod_{j=1}^n s(t_j'-t_j'')\,n(\mathbf{r}_j)\,\mathbf{e}_{\mathbf{k}_1}^*(\mathbf{r}_1t_1')\dots \dots\mathbf{e}_{\mathbf{k}_n}^*(\mathbf{r}_nt_n')\,\mathbf{e}_{\mathbf{k}_{n+1}}(\mathbf{r}_nt_n'')\dots\mathbf{e}_{\mathbf{k}_{2n}}(\mathbf{r}_1t_1'')\,d\mathbf{r}_j\,dt_j'dt_j'', \quad (9)$$

$s(t_j'-t_j'')$ is the sensitivity of the radiation detector, $n(\mathbf{r}_j)$ is the number of atoms per unit volume,

$$\mathbf{e}_{\mathbf{k}}(\mathbf{r},t) = i(\hbar\omega_k/2)^{1/2}\mathbf{u}_{\mathbf{k}}(\mathbf{r})e^{-i\omega_k t},$$

and the averaging is taken over the initial state of the field, $\mathrm{Sp}\{\rho\dots\} = \langle\dots\rangle$.

The statistics of the field can be characterized by the parameter

$$\xi_{\mathbf{k}} = \langle(a_{\mathbf{k}}^+)^2(a_{\mathbf{k}})^2\rangle - \langle a_{\mathbf{k}}^+a_{\mathbf{k}}\rangle^2, \quad (10)$$

since for thermal and for completely coherent (single mode) fields we have respectively

$$\langle(a^+)^n(a)^n\rangle_{\text{ther}} = n!\langle a^+a\rangle^n, \quad \langle(a^+)^n(a)^n\rangle_{\text{coh}} = \langle a^+a\rangle^n. \quad (11)$$

This quantity according to (8) is directly related to the dispersion in the photoelectron counts.

Since at the initial instant both fields and the medium are statistically independent, then under the conditions of the experiment outlined above one can segregate the contribution of the second field

$$\Delta\xi^{(1)} = \xi^{(1)} - \xi_0^{(1)} = |A|^2(\xi^{(2)}|A|^2 + \langle a_2^+(-\infty)a_2(-\infty)\rangle\langle a_1^+a_1\rangle), \quad (12)$$

where $\xi^{(2)}$ is the parameter (9) for the initial (prior to incidence on the medium) statistics of the second field, A is a numerical coefficient in front of $a_2(-\infty)$ in (6). It follows from (11) and (12) that the greatest change in statistics is produced by a completely incoherent second field.

We now consider the quantity $|A|^2$ for certain extreme cases, assuming that the level Ω_2 is sufficiently far removed from Ω_1 and $S_1 \gg S_2$. It follows from (6) that for $\Omega_1 = 2\omega_1 - \omega_2$

$$|A|^2 = 4|\lambda_1|^2|\lambda_2|^2(N_+-N_-)^2(\omega_2-\omega_1)^{-4}\sin^4\left(\frac{(\omega_2-\omega_1)T}{2}\right), \quad (13)$$

for $\Omega_1 = \Omega_2 = \Omega$

$$|A|^2 = {}^1/_4|\lambda_1|^2|\lambda_2|^2(N_+-N_-)^2T^4, \quad t_1 = T_1 = 0,\ t_2 = T, \quad (14)$$

for $\omega_1 = \Omega_1$, $\omega_2 = \Omega_2$, $\omega_1 \neq \omega_2$ and $\omega_1 = \Omega_1$, $\omega_2 + \omega_1 = \Omega_2$

$$|A|^2 = |\lambda_1|^2|\lambda_2|^2(N_+-N_-)^2(\omega_2-\omega_1)^{-2}T^2. \quad (15)$$

Since $\omega T \gg 1$, then in the resonance case (14) the effect of the modulation of statistics is a maximum. The observed change in the dispersion in the distribution of the photoelectrons in accordance with (8) can now be written (e.g., for a fully coherent second field $\xi(2) = 0$) for the case (14) in the form

$$C_m = (sN_0)^2(\hbar\omega/2)^2t^2\Delta\xi = \frac{9}{64}\frac{\pi^2c^6\hbar^2}{\omega^2V^2\varepsilon^2}(sN_0)^2(N_+-N_-)^2T^4t^2W\langle n_1\rangle\langle n_2\rangle, \quad (16)$$

where N_0 is the number of atoms per unit volume of a homogeneous detector, s is its sensitivity, t is the time during which photons were counted, $\langle n_1\rangle$ and $\langle n_2\rangle$ are the average numbers of photons respectively for the first and the second fields, prior to the switching on of the second field under the conditions of the experiment outlined above, and W is the probability for the spontaneous dipole emission of a photon per unit time by an isolated molecule

$$W = \frac{2}{3}\frac{\omega^2V\varepsilon}{\pi c^3}|\lambda|^2.$$

The process described above can be regarded as a form of induced Raman scattering since the system of equations (3) under the assumptions made above can be set in correspondence with the Hamiltonian (with the medium parameters being independent of the time) in which the interaction of light with light corresponds to the term

$$iTS_2\lambda_1\lambda_2^*a_1^+(t)a_2(0)e^{-i\omega_2t} + iTS_1\lambda_1^*\lambda_2a_2^+(t)a_1(0)e^{-i\omega_1t} + \text{c.c.},$$

which is analogous to the Hamiltonian of the nonlinear effect[3] indicated above

$$H_{int} = \sum_i\{\eta_R\sigma\mathbf{E}_1^{(-)}(r_i)\mathbf{E}_2^{(+)}(r_i) + \text{c.c.}\},$$

where η_R is the matrix element for the absorption of one quantum and the emission of another one.

[1] N. Bertolotti, B. Crosignani, P. DiPorto and D. Sette, Phys. Rev. **157**, 146 (1957).
[2] F. Gori and D. Sette, Phys. Rev. Letters **17**, 361 (1966).
[3] Y. R. Shen, Phys. Rev. **155**, 921 (1967).
[4] R. Glauber, Optical Coherence and Statistics of Photons. In the collection of translated articles "Quantum Optics and Quantum Radiophysics," Mir, 1966, p. 91.
[5] A. E. Glassgold and D. Holliday, Phys. Rev. **139**, 1717 (1965).

Translated by G. M. Volkoff
142