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Interference properties of coherent photons selectively reflected from resonance media

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***Abstract*. It is shown that, according to the quantum theory of light, the spatial period of an interference pattern formed by light incident on a medium and reflected from it is determined both by the wavelength of light and the number of coherent photons in a scattered mode. The scattered signal is assumed arbitrarily weak.**

Keywords: *quantum field, resonance radiation, interference.*

Selective reflection of resonance radiation from the interface between two media has revealed many surprises and has been permanently studied for many years. In 1909, R. Wood found experimentally that diffuse scattering upon reflection of resonance radiation from mercury vapour changed to specular scattering with increasing pressure of the mercury vapour [1]. This phenomenon was studied for the next half century [2 – 4]. In 1966, Koester found experimentally the amplification of resonance radiation upon reflection from an inverted medium [5]. This phenomenon is still being studied [6, 7].

This paper is devoted to a theoretical study of interference effects appearing upon selective reflection of resonance radiation from an unexcited medium. A standard quantum electrodynamics predicts the features of the interference pattern that are not described by the semiclassical theory of reflection, which deals with a nonquantized electromagnetic field. The case in point is a difference between the interference patterns appearing in a traditionally linear region of the interaction of light with matter. We call attention to the fact that mutually independent photons in the incident flux and photons that are mutually correlated in this flux produce different interference patterns after reflection. This difference is related to fourth-order interference processes [8 – 10], which are detected by the coincident output signals from two photodetectors located at different spatial points. In our case, the interference pattern is tested in a standard way by moving a single photodetector from one spatial point to another. We focus attention not on the quantum properties of light in vacuum but on the mechanism of formation of the interference pattern produced upon reflection a two-photon field from a resonantly scattering medium. While the authors of papers [8 – 10] studied the interference properties of two-photon fields, we are interested in their diffraction properties.

Let the scattered light be in a Fock state, and the functions φ_i form a complete system of the wave functions of a medium occupying the volume Ω. We assume also that the medium is in the state φ_0 before the interaction with light. After switching on the interaction, the total wave function of the system consisting of the medium and electromagnetic field be expanded into the series

$$\Psi = f_0\varphi_0 + \sum_{i\neq 0} f_i\varphi_i,$$

where the expansion coefficients f_i depend on the electromagnetic-field arguments.

Let a photodetector detecting the electromagnetic-field intensity be located at the point $\boldsymbol{r}$. The output signal from the photodetector irradiated by a photon is proportional to the probability of excitation of an atom in it, and therefore it is characterised by the operator, which is proportional to the density operator $\hat{n}^{vv'}$ for photons of the electromagnetic field:

$$J\hat{n}^{vv'}(\boldsymbol{r}) = J\sum_{\boldsymbol{k}_1,\lambda_1,\boldsymbol{k}_2,\lambda_2} e^{\lambda_1}_{\boldsymbol{k}_1 v}\frac{\hat{\alpha}^{+}_{\boldsymbol{k}_1\lambda_1}}{(2k_1)^{1/2}}\exp(-\mathrm{i}\boldsymbol{k}_1\boldsymbol{r}) \times e^{\lambda_2}_{\boldsymbol{k}_2 v'}\frac{\hat{\alpha}_{\boldsymbol{k}_2\lambda_2}}{(2k_2)^{1/2}}\exp(\mathrm{i}\boldsymbol{k}_2\boldsymbol{r}). \quad (1)$$

Here, J is a coefficient that characterises the photodetector sensitivity; $\hat{\alpha}_{\boldsymbol{k}\lambda}(\hat{\alpha}^{+}_{\boldsymbol{k}\lambda})$ are the annihilation (creation) operators for a photon in the state $(\boldsymbol{k},\lambda)$; $\boldsymbol{k}$ is the photon wave vector; and λ is the index of linear polarisation of the photon. The electromagnetic field is assumed transverse ($\lambda = 1, 2$). The unit vectors $\boldsymbol{e}^{\lambda}_{\boldsymbol{k}}$ are such that $k_v e^{\lambda}_{\boldsymbol{k}v} = 0$ for $\lambda = 1, 2$ and $k_v e^{\lambda}_{\boldsymbol{k}v} = k$ for $\lambda = 3$ (where v denotes summation). By moving the photodetector from one point to another, we can study the spatial distribution of the interference pattern. The average value of the operator (1) in the state Ψ, taking into account the mutual orthogonality of the functions φ_i, can written in the form

$$\langle\hat{n}^{vv'}(\boldsymbol{r})\rangle = \mathrm{Sp}\rho_{\mathrm{c}}\hat{n}^{vv'}(\boldsymbol{r}) + \mathrm{Sp}\rho_{\mathrm{n}}\hat{n}^{vv'}(\boldsymbol{r}), \quad (2)$$

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where the summation in the right-hand side is performed over all the arguments of the electromagnetic field and

$$\rho_{\rm c} = f_0 f_0^*, \quad \rho_{\rm n} = \sum_{i\neq 0} f_i f_i^*.$$

The density matrix $\rho_{\rm c}$ describes inelastic scattering of photons, when the atoms in a scattering medium remain in the initial quantum state φ_0. We will call such scattering a coherent scattering. Upon coherent scattering, the photon energy does not change, while the change in the photon momentum occurs at the interface between two media. One should bear in mind that the atoms are not free but are localised within the volume Ω, which is manifested in their wave functions [11]. The matrix $\rho_{\rm n}$ describes the scattering processes in which the atoms in a medium change their initial state. We will call such scattering an incoherent scattering. This scattering produces, in particular, the diffuse scattering of light and also involves inelastic scattering of one of the photons upon absorption of another photon by the medium.

Expression (2) is remarkable in two respects. First, the average density of photons of some polarisation ($\nu = \nu'$) at the point $\boldsymbol{r}$ can be written as a sum of two terms, each of them being positive. We will consider the photon density, which is determined by the coherent ($\langle \hat{n}^{\nu\nu} \rangle_{\rm c}$) and incoherent ($\langle \hat{n}^{\nu\nu} \rangle_{\rm n}$) scattering channels. Second, the total photon density equals the sum of $\langle \hat{n}^{\nu\nu} \rangle_{\rm c}$ and $\langle \hat{n}^{\nu\nu} \rangle_{\rm n}$. This means that coherent and incoherent channels do not interfere with each other. Note that the disappearance of the interference is caused first of all not by the properties of light, as in the semiclassical theory, but by the orthogonality of the wave functions of the atoms in the medium in the initial (φ_0) and final (φ_i) states [12].

It is clear now that, if a photodetector consists of many atoms rather than of one, its output signal is again a sum of the positive contributions of the coherent and incoherent scattering channels.

Let us calculate the intensity of the interference pattern produced by incident radiation and radiation reflected from the medium. According to the above analysis, the interference pattern is produced only due to coherent scattering (which is important), when the wave function φ_0 of the medium does not change.

We assume that gas consisting of nonrelativistic atoms, which have one valence electron and the resonance-transition frequency $\omega_{m\mu}$, occupies the half-space $z > 0$. The quasi-resonance ($|k_0 - \omega_{m\mu}| \ll k_0$) radiation with the wave vector $\boldsymbol{k}_0$ and the polarisation index λ_0 is incident at some angle on the interface between two media. The radiation is assumed transverse ($\lambda = 1, 2$) and linearly polarised. The Schrödinger equation describing the system consisting of atoms and the electromagnetic field has the form ($\hbar = c = 1$)

$$\mathrm{i}\frac{\partial \Psi}{\partial t} = \hat{H}\Psi, \tag{3}$$

where

$$\hat{H} = \hat{H}_0 + \hat{H}_{\rm int};\ \hat{H}_0 = \hat{H}_{\rm a} + \hat{H}_{\rm ph};\ \hat{H}_{\rm a} = \sum_{i,\boldsymbol{p}} \varepsilon_i(p)\hat{b}^+_{i\boldsymbol{p}}\hat{b}_{i\boldsymbol{p}};$$

$$H_{\rm ph} = \sum_{\boldsymbol{k},\lambda} k\hat{\alpha}^+_{\boldsymbol{k}\lambda}\hat{\alpha}_{\boldsymbol{k}\lambda};\ H_{\rm int} = -\frac{e}{m}\int \hat{\psi}^+(\boldsymbol{r},\boldsymbol{R})\hat{\boldsymbol{p}}\hat{\boldsymbol{A}}(\boldsymbol{r})\hat{\psi}(\boldsymbol{r},\boldsymbol{R})\mathrm{d}\boldsymbol{r}\mathrm{d}\boldsymbol{R};$$

$$\hat{\psi}(\boldsymbol{r},\boldsymbol{R}) = \sum_{i,\boldsymbol{p}} \psi_i(\boldsymbol{r}-\boldsymbol{R})\Phi_{\boldsymbol{p}}(\boldsymbol{R})\hat{b}_{i,\boldsymbol{p}};\ \hat{\boldsymbol{p}} = -\mathrm{i}\nabla;$$

$$\hat{\boldsymbol{A}}(\boldsymbol{r}) = \sum_{\boldsymbol{k},\lambda} \frac{\boldsymbol{e}^{\lambda}_{\boldsymbol{k}}}{(2kV)^{1/2}}\left(\hat{\alpha}^+_{\boldsymbol{k}\lambda}\mathrm{e}^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}} + \hat{\alpha}_{\boldsymbol{k}\lambda}\mathrm{e}^{-\mathrm{i}\boldsymbol{k}\boldsymbol{r}}\right);$$

$V = L_x L_y L_z$ is the normalising volume; ψ_i are the wave functions describing the inner state of the atoms in the medium with the energy ε_i. The wave functions $\Phi_{\boldsymbol{p}}(\boldsymbol{R})$ determine the translational motion of the atoms localised within the volume Ω occupied by gas. Outside this volume, $\Phi_{\boldsymbol{p}}(\boldsymbol{R}) = 0$. The operators $\hat{b}^+_{i,\boldsymbol{p}}$ ($\hat{b}_{i,\boldsymbol{p}}$) describe the creation (annihilation) of atoms in the state $(i, \boldsymbol{p})$. In the absence of the temperature degeneration of the gas, they can be treated as Bose–Einstein operators.

Let us calculate an interference pattern in the region $z < 0$. For this purpose, we rewrite equation (3) in the integral form

$$\Psi = \Psi_0 + \mathrm{i}\frac{e}{m}\int_{t_0}^{t} \exp[-\mathrm{i}H_0(t-t')]\hat{\psi}^+\hat{\boldsymbol{p}}\hat{\boldsymbol{A}}\hat{\psi}\mathrm{d}\boldsymbol{r}\mathrm{d}\boldsymbol{R}\Psi(t')\mathrm{d}t', \tag{4}$$

where the function Ψ_0 describes the initial state of the system before switching on the interaction between the electromagnetic field and atoms in the medium, and $t_0 \to -\infty$.

We will seek the solution of equation (4) in the form of an iteration series by omitting the terms proportional to the odd powers of the charge e, which are responsible for incoherent scattering. The sum of the remaining series is equivalent to the solution of the integral equation

$$\Psi = \Psi_0 - \left(\frac{e}{m}\right)^2 \int_{-\infty}^{t} \mathrm{d}t' \int \exp[-\mathrm{i}H_0(t-t')]\hat{\psi}^+\hat{\boldsymbol{p}}\hat{\boldsymbol{A}}\hat{\psi}\mathrm{d}\boldsymbol{r}'\mathrm{d}\boldsymbol{R}'$$

$$\times \int_{-\infty}^{t'} \exp[-\mathrm{i}H_0(t'-t'')]\hat{\psi}^+\hat{\boldsymbol{p}}\hat{\boldsymbol{A}}\hat{\psi}\mathrm{d}\boldsymbol{r}''\mathrm{d}\boldsymbol{R}''\Psi(t'')\mathrm{d}t''. \tag{5}$$

Because the atoms of the medium remain in the initial state after coherent scattering, only the products

$$\hat{\psi}^+\hat{\psi}\hat{\psi}^+\hat{\psi} \sim \hat{b}^+_{\mu\boldsymbol{p}}\hat{b}_{m\boldsymbol{p}'}\hat{b}^+_{m\boldsymbol{p}'}\hat{b}_{\mu\boldsymbol{p}}$$

are nonzero among all the products of the operators $\hat{\psi}$ and $\hat{\psi}^+$. The indices m and μ refer to the Zeeman sublevels of the excited and ground state of an atom. The variables in equation (5) can be separated, i.e.,

$$\Psi = \chi\varphi_0,$$

if we take into account that they were certainly separated before switching on the interaction:

$$\Psi_0 = \chi_0\varphi_0 \equiv \rangle_0,\ \varphi_0 = \prod_{\mu,\boldsymbol{p}} c_{\mu\boldsymbol{p}}\hat{b}^+_{\mu\boldsymbol{p}}|0\rangle\mathrm{e}^{-\mathrm{i}\varepsilon_\mu(p)t}.$$

Here, the coefficients $c_{\mu\boldsymbol{p}}$ characterise the distribution of atoms over the states $(\mu, \boldsymbol{p})$, and $\varepsilon_\mu(p)$ are the energies of these states. The wave function χ depends of the arguments of the electromagnetic field. If the initial mode $(\boldsymbol{k}_0, \lambda_0)$ of the Fock state contains N_0 photons, then

$$\chi_0(t) = \frac{(\hat{\alpha}^+_{\boldsymbol{k}_0\lambda_0})^{N_0}}{\sqrt{N_0!}}|0\rangle\mathrm{e}^{-\mathrm{i}k_0N_0t}.$$

We obtain from equation (5) the equation

$$\chi = \chi_0 + \int \Delta_{\rm r}^0(t-t')\hat{\mathscr{P}}_{\rm r}(t'-t'')\chi(t'')\mathrm{d}t''\mathrm{d}t', \tag{6}$$

for the wave function χ, where

$$\Delta_{\rm r}^0(t) = -\mathrm{i}\vartheta(t)\exp(-\mathrm{i}\hat{H}_0 t);\ \hat{\mathscr{P}}_{\rm r}(t) = \int_{-\infty}^{\infty} \mathrm{e}^{-\mathrm{i}Et}\hat{\mathscr{P}}_{\rm r}(E)\frac{\mathrm{d}E}{2\pi};$$

$$\hat{\mathscr{P}}_{\rm r}(E) = \sum_{\boldsymbol{k}_1,\lambda_1,\boldsymbol{k}_2,\lambda_2} \hat{\alpha}^+_{\boldsymbol{k}_1\lambda_1} C_{\rm r}^{(\boldsymbol{k}_1\lambda_1\boldsymbol{k}_2\lambda_2)}(E-\hat{H}_{\rm ph})\hat{\alpha}_{\boldsymbol{k}_2\lambda_2};$$

$$C_{\rm r}^{(\boldsymbol{k}_1\lambda_1\boldsymbol{k}_2\lambda_2)}(E) = \sum_{\boldsymbol{p}_1,\boldsymbol{p}_2} \frac{P^{\lambda_1 *}_{m\mu}(\boldsymbol{k}_1)P^{\lambda_2}_{m\mu}(\boldsymbol{k}_2)}{2V(k_1k_2)^{1/2}} N_\mu(\boldsymbol{p}_1)\times$$

$$\int \frac{\Phi^*_{\boldsymbol{p}_1}(\boldsymbol{R}_1)\Phi_{\boldsymbol{p}_2}(\boldsymbol{R}_1)\Phi^*_{\boldsymbol{p}_2}(\boldsymbol{R}_2)\Phi_{\boldsymbol{p}_1}(\boldsymbol{R}_2)\exp[-\mathrm{i}(\boldsymbol{k}_1\boldsymbol{R}_1-\boldsymbol{k}_2\boldsymbol{R}_2)]}{E-\varepsilon_m(p_2)+\varepsilon_\mu(p_1)+\mathrm{i}0}\mathrm{d}\boldsymbol{R}_1\mathrm{d}\boldsymbol{R}_2; \tag{7}$$

$\vartheta(t)$ is the Heaviside function; and

$$N_\mu(\boldsymbol{p}) = \langle \hat{b}^+_{\mu\boldsymbol{p}}\hat{b}_{\mu\boldsymbol{p}}\rangle_0;$$

$$P^\lambda_{m\mu}(\boldsymbol{k}) = \sum_v \mathrm{e}^\lambda_{\boldsymbol{k}v}P^v_{m\mu}(\boldsymbol{k});\quad P^v_{m\mu}(\boldsymbol{k}) = \frac{e}{m}\int \psi^*_m\hat{p}_v\mathrm{e}^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}}\psi_\mu\mathrm{d}\boldsymbol{r}.$$

In the two-level approximation, only those terms are taken into account that make an appreciable contribution upon the quasi-resonance interaction of photons with gas atoms. The appearance of the term i0 in the dominator in expression (7) is related to the causality principle. So far we assumed that the energy levels of the atoms are real quantities, in accordance with the adopted model. However, actually the gas atoms interact with each other and with foreign particles in the gas (reservoir), resulting in the collision broadening γ of the energy levels of the atoms.

We assume that the collision width is greater than the radiative width γ_r. The inclusion of the collision widths of the energy levels results in the replacement of the term i0 by $\mathrm{i}\gamma/2$ in expression (7). It is important to note that the signs in front of i0 and $\mathrm{i}\gamma/2$ should coincide because otherwise the causality principle will be violated. Below, we will always take the term $\mathrm{i}\gamma/2$ into account explicitly. Expressions (6) and (7), taking into account the term $\mathrm{i}\gamma/2$, can be rigorously obtained in the more developed but rather cumbersome theory [13].

Let us specify the integrals over $\boldsymbol{R}_1$ and $\boldsymbol{R}_2$. Expression (7) describes reemission of photons during two successive virtual processes: an atom located at the point $\boldsymbol{R}_2$ first absorbs a scattered photon in the $(\boldsymbol{k}_2,\lambda_2)$ state and then the same atom at the point $\boldsymbol{R}_1$ emits a new photon to the state $(\boldsymbol{k}_1,\lambda_1)$. We can assume that the centre of gravity of the atom during the scattering event is located at the point $(\boldsymbol{R}_1+\boldsymbol{R}_2)/2$. Because this centre of gravity cannot be located outside the volume Ω occupied by gas, the structural coefficient $C_{\rm r}^{(\boldsymbol{k}_1\lambda_1\boldsymbol{k}_2\lambda_2)}(E)$ will not change if we introduce the function

$$\vartheta\left(\frac{\boldsymbol{R}_1+\boldsymbol{R}_2}{2}\right) = \sum_{\boldsymbol{q}} \vartheta(\boldsymbol{q})\exp\left(\mathrm{i}\boldsymbol{q}\frac{\boldsymbol{R}_1+\boldsymbol{R}_2}{2}\right), \tag{8}$$

to the integrand in expression (7), where

$$\vartheta(\boldsymbol{q}) = \int_\Omega \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\boldsymbol{R}}\vartheta(\boldsymbol{R})\frac{\mathrm{d}\boldsymbol{R}}{V}; \tag{9}$$

$$\vartheta(\boldsymbol{R}) = \begin{cases} 1, & \boldsymbol{R}\in\Omega,\\ 0, & \boldsymbol{R}\notin\Omega.\end{cases}$$

The function $\Phi_{\boldsymbol{p}}(\boldsymbol{R})$ can be approximated by exponentials $1/\sqrt{V}\exp(\mathrm{i}\boldsymbol{p}\boldsymbol{R})$, describing a free translational motion of the atoms. Such an approximation (the Wigner approximation) assumes that the atoms are localised within the volume Ω where they are quasi-free. The quantity $\boldsymbol{p}$ acquires now the meaning of the momentum of the centre of gravity of an atom. The integrals over $\boldsymbol{R}_1$ and $\boldsymbol{R}_2$ can be explicitly calculated and expressed in terms of the Kronecker symbols:

$$\int \Phi^*_{\boldsymbol{p}_1}(\boldsymbol{R}_1)\Phi_{\boldsymbol{p}_2}(\boldsymbol{R}_1)\exp\left[-\mathrm{i}\left(\boldsymbol{k}_1+\frac{\boldsymbol{q}}{2}\right)\boldsymbol{R}_1\right]\mathrm{d}\boldsymbol{R}_1$$

$$= \delta\left(\boldsymbol{p}_2-\boldsymbol{k}_1+\frac{\boldsymbol{q}}{2},\boldsymbol{p}_1\right),$$

$$\int \Phi^*_{\boldsymbol{p}_2}(\boldsymbol{R}_2)\Phi_{\boldsymbol{p}_1}(\boldsymbol{R}_2)\exp\left[\mathrm{i}\left(\boldsymbol{k}_2+\frac{\boldsymbol{q}}{2}\right)\boldsymbol{R}_2\right]\mathrm{d}\boldsymbol{R}_2$$

$$= \delta\left(\boldsymbol{p}_1+\boldsymbol{k}_2+\frac{\boldsymbol{q}}{2},\boldsymbol{p}_2\right).$$

If the atoms occupy a plane–parallel layer of thickness l located in the plane xy, then, according to (8) and (9), we have

$$\vartheta(\boldsymbol{q}) = \delta(q_x)\delta(q_y)\vartheta_l(q_z),\ \vartheta_l(q_z) = \int_0^l \exp(-\mathrm{i}q_z z)\frac{\mathrm{d}z}{L_z}.$$

Taking this into account, the structural coefficient can be written in the form

$$C_{\rm r}^{(\boldsymbol{k}_1\lambda_1\boldsymbol{k}_2\lambda_2)}(E) = \sum_{m,\mu,\boldsymbol{p}} \frac{P^{\lambda_1 *}_{m\mu}(\boldsymbol{k}_1)P^{\lambda_2}_{m\mu}(\boldsymbol{k}_2)}{2V(k_1k_2)^{1/2}}N_\mu(\boldsymbol{p})$$

$$\times \frac{\delta(k_{1x},k_{2x})\delta(k_{1y},k_{2y})\vartheta_l(k_{1z}-k_{2z})}{E-\omega_{m\mu}-\boldsymbol{p}(\boldsymbol{k}_1+\boldsymbol{k}_2)/2+\mathrm{i}\gamma/2}.$$

Here, the nonrelativistic approximation is used, and $\varepsilon_i(p) = \varepsilon_i + p^2/2M,$, where M is the atom mass. If a gas layer occupies a half-space, then $l\to\infty$.

If gas atoms are distributed over the Zeeman sublevels of the ground state uniformly, it is possible to perform summation over m and μ. In the dipole approximation, we obtain [14]

$$\sum_{m,\mu} P^{\lambda_1 *}_{m\mu}(\boldsymbol{k}_1)P^{\lambda_2}_{m\mu}(\boldsymbol{k}_2) = \frac{\pi(2j_m+1)}{\omega_{m\mu}}\gamma_{\rm r}\boldsymbol{e}^{\lambda_1}_{\boldsymbol{k}_1}\boldsymbol{e}^{\lambda_2}_{\boldsymbol{k}_2},$$

where j_m is the quantum number of an excited state of an atom. For simplicity, we assume that the Doppler broadening is negligible. Then, we have

$$C_{\rm r}^{(\boldsymbol{k}_1\lambda_1\boldsymbol{k}_2\lambda_2)}(E) = \frac{\pi(2j_m+1)}{2\omega^2_{m\mu}}\gamma_{\rm r}\boldsymbol{e}^{\lambda_1}_{\boldsymbol{k}_1}\boldsymbol{e}^{\lambda_2}_{\boldsymbol{k}_2}n_\mu$$

$$\times \frac{\delta(k_{1x},k_{2x})\delta(k_{1y},k_{2y})\vartheta_l(k_{1z}-k_{2z})}{E-\omega_{m\mu}+\mathrm{i}\gamma/2}, \qquad (10)$$

where $n_\mu = N_\mu/V$. We assume that the scattered radiation contains one photon:

$$\chi_0(E) = \hat{\alpha}^+_{\boldsymbol{k}_0\lambda_0}|0\rangle 2\pi\delta(E-k_0).$$

According to (6), the wave function describing the incident and reflected light in the lowest-order perturbation theory has the form

$$\chi = (1+\Delta^0_\mathrm{r}\hat{\mathscr{P}}_\mathrm{r})\chi_0.$$

This means that the interference pattern produced by the incident and reflected light is described at any instant of time by the expression

$$\langle J\hat{n}^{\lambda_0\lambda_0}(\boldsymbol{r})\rangle = \langle J\hat{n}^{\lambda_0\lambda_0}(\boldsymbol{r})\Delta^0_\mathrm{r}\hat{\mathscr{P}}_\mathrm{r}\rangle_0 + \text{c. c.}$$

$$= J\sum_{\boldsymbol{k}_z}\frac{\exp[-\mathrm{i}(\boldsymbol{k}_0-\boldsymbol{k}_1)\boldsymbol{r}]}{2(k_0k_1)^{1/2}}\int\frac{\delta(E-k_0)}{E-k_1+\mathrm{i}0}$$

$$\times C_\mathrm{r}^{(\boldsymbol{k}_1\lambda_0\boldsymbol{k}_0\lambda_0)}(E)\mathrm{d}E + \text{c. c.}$$

The sum over $\boldsymbol{k}_1$ can be calculated by using the asymptotic equality

$$\sum_{k_z}\frac{\mathrm{e}^{\mathrm{i}k_z z}}{k_0-k+\mathrm{i}0}f(k_z) \underset{L_z\to\infty}{\longrightarrow} \frac{L_z}{2\pi}\int_{-\infty}^{\infty}\frac{\mathrm{e}^{\mathrm{i}k_z z}f(k_z)\mathrm{d}k_z}{k-(k_z^2+k_{0x}^2+k_{0y}^2)^{1/2}+\mathrm{i}0}$$

$$\underset{z\to-\infty}{\longrightarrow} -\mathrm{i}\frac{L_z k_0}{k_{0z}}f(-k_{0z})\mathrm{e}^{-\mathrm{i}k_{0z}z}.$$

Here, $f(k_z)$ is a function that has no singularity at the point $k=-k_{0z}$.

The final expression for the intensity of the interference pattern has the form

$$\langle J\hat{n}^{\lambda_0\lambda_0}(\boldsymbol{r})\rangle = -\mathrm{i}\frac{JL_z}{2k_{0z}}\mathrm{e}^{-2\mathrm{i}k_{0z}z}C_\mathrm{r}^{(\boldsymbol{K}\lambda_0\boldsymbol{k}_0\lambda_0)}(k_0) + \text{c. c.},$$

$$\boldsymbol{K} = \{k_{0x}, k_{0y}, -k_{0z}\}. \qquad (11)$$

Expression (11) can be rewritten using the standard refractive index of a medium:

$$\varkappa^{\lambda_0}(\boldsymbol{k}_0) = 1 + \frac{L_z}{k_0}C_\mathrm{r}^{(\boldsymbol{k}_0\lambda_0\boldsymbol{k}_0\lambda_0)}(k_0). \qquad (12)$$

It follows from (11) and (12) that

$$\langle J\hat{n}^{\lambda_0\lambda_0}(\boldsymbol{r})\rangle\big|_{z\to-\infty} = -\frac{Jk_0}{4k_{0z}}\mathrm{e}^{-2\mathrm{i}k_{0z}z}$$

$$\times[\varkappa^{\lambda_0}(\boldsymbol{k}_0)-1] + \text{c. c.} \qquad (13)$$

Expression (13) can be obtained from the semiclassical emission theory. We have presented in detail its electrodynamic derivation because other expressions will be derived below in the same way.

The dependence of the intensity of the interference pattern on coordinates is described, according to (13), by the function $\cos(2k_{0z}z)$ for $|k_0-\omega_{m\mu}| \gg \gamma$ and by the function $\sin(2k_{0z}z)$ for $|k_0-\omega_{m\mu}| \ll \gamma$. It follows from (10)–(13) that the interference pattern at the point $k_0=\omega_{m\mu}$ is determined by the dimensionless parameter

$$B = n_\mu \Lambda^3\frac{\gamma_\mathrm{r}}{\gamma} < 1, \qquad (14)$$

where $\Lambda = 2\pi/k$. Inequality (14) represents the condition of applicability of the perturbation theory and expression (13). The parameter B was considered for the first time in paper [15], where it was obtained based on phenomenological reasoning.

Consider now an incident mode containing two photons in the $(\boldsymbol{k}_0,\lambda_0)$ state, i.e.,

$$\chi_0(E) = \frac{(\hat{\alpha}^+_{\boldsymbol{k}_0\lambda_0})^2}{\sqrt{2!}}|0\rangle 2\pi\delta(E-2k_0).$$

In this case, two situations are possible. In the first case, a medium reflects one photon, which corresponds to the first iteration of equation (6). Another photon is still propagating in the initial direction. In the second case, two photons are reflected from the medium, and, therefore, equation (6) should be iterated twice.

Consider the second case, when the wave function of the electromagnetic field has the form

$$\Psi = (1+\Delta^0_\mathrm{r}\hat{\mathscr{P}}_\mathrm{r}\Delta^0_\mathrm{r}\hat{\mathscr{P}}_\mathrm{r})\Psi_0. \qquad (15)$$

We will omit for the time being the term corresponding to the first order of the perturbation theory. The interference pattern produced by the incident and reflected light can no longer be described by averaging the operator (1) over the state (15) because this average vanishes. The interference pattern can be now detected only when two atoms of a photodetector are simultaneously excited, which is described by the operator $(J\hat{n}^{\lambda_0\lambda_0})^2$. Similarly to (13), we obtain

$$\langle[J\hat{n}^{\lambda_0\lambda_0}(\boldsymbol{r})]^2\Delta^0_\mathrm{r}\hat{\mathscr{P}}_\mathrm{r}\Delta^0_\mathrm{r}\hat{\mathscr{P}}_\mathrm{r}\rangle_0 + \text{c. c.}$$

$$= 2\left\{\frac{Jk_0}{4k_{0z}^2}\mathrm{e}^{-2\mathrm{i}k_{0z}z}[\varkappa^{\lambda_0}(\boldsymbol{k}_0)-1]\right\}^2 + \text{c. c.} \qquad (16)$$

According to expression (16), the interference pattern for resonance radiation is determined by the square of the parameter B (14), i.e., its intensity is lower than that of the interference pattern produced upon one-photon scattering. Note that the spatial period of this interference pattern is two times smaller than that in the case of one-photon scattering. In other words, in two limiting cases for $|k_0-\omega_{m\mu}| \gg \gamma$ and $|k_0-\omega_{m\mu}| \ll \gamma$, the interference pattern is described by the function $\cos(4k_{0z}z)$. The semiclassical theory of reflection cannot give this result.

It is important to note that when the scattered mode contains two photons, one-photon scattering is absent in the coherent channel [16]. Because such scattering is related to the incoherent channel, it does not affect the interference pattern, as shown above.

Let us explain the absence of one-photon reflection of two-photon incident radiation in the coherent channel. It

seems that the formal application of the perturbation theory to equation (6) allows one-photon scattering. The intensity of this scattering should exceed that of the two-photon scattering by a factor of B^{-1}. However, actually this does not occur. This is explained by the fact that upon one-photon reflection of the two-photon field, one of the photons will propagate without scattering deep inside an absorbing semi-infinite medium. However, such a process cannot exist. A photon propagating in a medium will be either multiply scattered and then reflected by the medium with the 100 % probability, contributing to the reflected field in the high orders of the perturbation theory, or absorbed by the medium, contributing to the incoherent scattering channel. In any case, one-photon reflection of the incident two-photon field is absent in the coherent channel [16, 17]. It follows from the above that only Feynman diagrams describing photons reflected to vacuum but not photons scattered to the medium are retained in the coherent channel.

The results obtained above can be obviously generalised. Let us assume that the incident mode contains N_0 photons in a Fock state. In this case, the interference pattern at large distances ($z \to -\infty$) from the interface between two media is produced only by the state of the photon field in which all N_0 photons are coherently reflected by the medium. Here, the interference pattern is described by the function $\cos(2N_0 k_{0z} z)$ or $\sin(2N_0 k_{0z} z)$, depending on the parity of the number N_0.

We considered above the interference pattern at the asymptotic distances $z \to -\infty$. In the description of the near-field zone of scattering, the extinction theorems [17] are violated, and the interference pattern is different.

Note that the difference between the interference patterns produced by one-photon and two-photon fluxes can be confirmed experimentally by studying the selective reflection of light emitted by a black-body radiation source. The density matrix of this radiation in the mode $(\boldsymbol{k}, \lambda)$ can be written in the form

$$\rho = |N_{\boldsymbol{k}\lambda}\rangle\langle N_{\boldsymbol{k}\lambda}| \left(1 - \mathrm{e}^{-k/\Theta}\right)^{-1} \exp\left(-\frac{kN_{\boldsymbol{k}\lambda}}{\Theta}\right),$$

where Θ is the statistical temperature and $N_{\boldsymbol{k}\lambda}$ are the occupation numbers of the mode $(\boldsymbol{k}, \lambda)$. If $k > \Theta$, then the most probable number of photons in the occupied mode equals unity, and the interference pattern described by expression (13) will be observed in the experiment. If $k < \Theta$, then the probability of two-photon occupation of the mode will be significant, and the interference pattern described by expression (16) will be observed against the background of the previous interference pattern. The intensity of other interference patterns corresponding to $N_{\boldsymbol{k}\lambda} > 2$ decreases exponentially with increasing $N_{\boldsymbol{k}\lambda}$.

For the resonance line of mercury, inequality (14) is valid up to the concentration $n_\mu \sim 10^{17}$ cm^{-3}. For $n_\mu \sim 10^{18}$ cm^{-3}, the reflection coefficient in the coherent channel is of the order of unity [1]. The interference patterns discussed above can be readily detected with a photodetector distinguishing the mechanisms of one-photon, two-photon, and multiphoton excitations. This means that the study of interference patterns by means of the photocount statistics opens up wide possibilities for analysis of scattering media and the statistical properties of the scattered radiation itself.

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