GENERATION OF THE SECOND HARMONIC OF LASER RADIATION DUE TO LIGHT SCATTERING BY AN ENSEMBLE OF ATOMS DRIFTING IN THE MARKOV THERMOSTAT

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We present here a theoretical treatment of the nonlinear effect that occurs when the atoms scattering the light are disturbed by foreign particles that fly upon them from one side. We show that a quasi-constant multipole moment is induced in these atoms because of the anisotropy of such a medium. The interaction between this moment and the external radiation should yield the harmonics of the fundamental frequency in the spectrum of scattered radiation.

In this paper we study theoretically the nonlinear effect that takes place when light-scattering atoms are disturbed by particles flying upon them mostly from one side. Such a situation can be observed, for instance, when an atomic ensemble is irradiated by an electron beam.

It is obvious that, if the disturbing particles move along a preferred direction, the atomic shells are deformed mostly from one side. As a result, the multipole moment should be induced in atoms, and its ensemble-averaged value must differ from zero. If the medium of such atoms is irradiated with a coherent radiation the polarization plane of the latter may rotate due to the interaction with the multipole moment. Moreover, under certain conditions, the radiation may, in addition, change its frequency. Let us consider this question in a more detail.

The main problem that arises when trying to take into account some anisotropy of the environment (below, we treat it as a thermostat with Markov stochastic properties) is how one should introduce corresponding operators into the Schrödinger or Neumann equations. Such operators should take into account not only anisotropy of the medium, but the collision nature of the interaction between the atoms and disturbing particles as well.

For this purpose, we use the method which is based on averaging the wave functions over the influence from the thermostat already at the stage of constructing the Schrödinger equation.[1,2] We construct equation for the ψ-function using the method of integrals over the Feynman[3] paths, in which the propagator is averaged over all possible virtual paths in the medium surrounding the quantum subsystem considered. In the general case this equation is nonlinear. However, a solution to it can be obtained only if we take into account some relations valid for the wave functions $\psi(\mathbf{r}, t)$ satisfying it. These relations are as follows:

$$\psi(\mathbf{r}, t) = \tilde{\psi}(\mathbf{r}, t) / \sqrt{\langle\tilde{\psi}(\mathbf{r}, t) | \tilde{\psi}(\mathbf{r}, t)\rangle}\,; \tag{1}$$

$$\tilde{\psi}(\mathbf{r}, t) = \exp\left(-\frac{i}{\hbar}\frac{\varkappa}{1+i\alpha}\,t + \frac{i}{\hbar}\frac{e}{c}\left(\left(\mathbf{A} + \frac{c}{e}(1-i\alpha)\frac{m\mathbf{V}}{2}\right)\mathbf{r}\right) + \frac{im}{8\hbar}(1+i\alpha)\int \mathbf{V}^2\,\mathrm{d}t\right)\psi_1(\mathbf{r}, t); \tag{2}$$

$$i\hbar\frac{\partial\psi_1}{\partial t} = \frac{1}{1+i\alpha}(\hat{H} - U)\,\psi_1 + \left(U + \frac{e}{2}\left(\mathbf{V}\int \mathbf{E}\mathrm{d}t\right)\right)\psi_1 - \frac{m}{2e}(1-i\alpha)\left(\frac{\partial\mathbf{V}}{\partial t}\,\mathbf{d}\right) - \frac{m}{8}(1+i\alpha)\,\mathbf{V}^2\,\psi_1 + (\mathbf{E}\,\mathbf{d})\,\psi_1. \tag{3}$$

Here, $\hat{H}$ is the Hamiltonian of an undisturbed atom; U is the potential energy operator; $\mathbf{A}$ and $\mathbf{E}$ are the vector potential and the intensity of the electric component of the external electromagnetic field; $\mathbf{d}$ is the dipole moment of the atom; e and m are the electron charge and mass. Besides, Eqs. (2) and (3) contain two non-negative parameters α and $\varkappa$. The larger the thermostat density, the larger are their values. The vector $\mathbf{V}$ describes the velocity of the directed drift of an individual atom with respect to the ambient medium. In the general case $\mathbf{V}$ may depend on time.

The velocity of a quantum subsystem (atom) with respect to the thermostat, entering the above expressions, allows one to take into account the thermostat anisotropy, or in other words the presence of a preferred direction of the particle ensemble motion. Indeed, from the mathematical viewpoint, it

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makes no difference whether the considered quantum subsystem moves in the thermostat or the thermostat particles move with respect to it. The presence of a preferred direction from which disturbing particles fly upon an atom can lead, as it was already noted, to inducing an additional multipole moment in atoms.

Let us find it. For clearness, let us restrict ourselves by a two-level approximation.

We assume that a system of atoms of the same kind that interact with a thermostat is irradiated by a monochromatic radiation with the frequency close to the frequency of a quantum transition from the second level to the first one

$$\mathbf{E}(\mathbf{R}, t) = \mathbf{E}_0 \cos(\omega t - \mathbf{K}\,\mathbf{R}), \tag{4}$$

where $\mathbf{E}_0$ is the field amplitude (we assume that it has a fixed direction); ω is the frequency; $\mathbf{K}$ is the wave vector; $\mathbf{R}$ is the radius-vector of the atom.

The solution of the equation for the auxiliary function $\psi_1(\mathbf{r}, t)$, in the resonance approximation, has the following form:

$$\psi_1(\mathbf{r}, t) = (C_1 \exp(\beta_1 t) + C_2 \exp(\beta_2 t)) \times$$
$$\times \exp\left(-\frac{i}{\hbar} E_1\, t - \frac{\alpha}{\hbar}(E_1 - U_{11})\, t\right) \psi_1(\mathbf{r}) +$$
$$+ (C_1\beta_1 \exp(-\beta_2 t) + C_2\beta_2 \exp(-\beta_1 t)) \times$$
$$\times \exp\left(-\frac{i}{\hbar} E_2\, t - \frac{\alpha}{\hbar}(E_2 - U_{22})\, t\right) \psi_2(\mathbf{r}) \times$$
$$\times \frac{i2\hbar}{e\,(\mathbf{E}_0\,\mathbf{r}_{12})} \exp(i\mathbf{K}\,\mathbf{R}). \tag{5}$$

Here $\psi_i(\mathbf{r})$ $(i = 1, 2)$ are the coordinate eigenfunctions of the upper and lower atomic levels. The constants C_i are determined by the initial conditions. The values β_i are approximately equal to

$$\beta_1 \approx (i\varepsilon - \gamma_{21})/2 + i\Omega - \varepsilon\gamma_{21}/4\Omega; \tag{6}$$
$$\beta_2 \approx (i\varepsilon - \gamma_{21})/2 - i\Omega + \varepsilon\gamma_{21}/4\Omega. \tag{7}$$

In Eqs. (6) and (7), ε is the frequency detuning

$$\varepsilon = \omega - (E_2 - E_1)/\hbar, \tag{8}$$

γ_{21} is the parameter equal to the difference

$$\gamma_{21} = \alpha\,(E_2 - U_{22})/\hbar - \alpha\,(E_1 - U_{11})/\hbar\,. \tag{9}$$

In what follows, we assume that no exact resonance occurs ($|\varepsilon| \gg |\gamma_{21}|$), but the frequency detuning is sufficiently small: $|\varepsilon| \ll \omega_{21}$), Ω is the Rabi frequency

$$\Omega = \sqrt{\frac{\varepsilon^2}{4} + \frac{e^2}{4\hbar^2}\,|(\mathbf{E}\,\mathbf{r}_{12})|^2}. \tag{10}$$

The substitution of the wave function $\psi(\mathbf{r}, t)$ into the expression for the average dipole moment of the considered atom

$$\langle\mathbf{d}(t)\rangle = \langle\psi(\mathbf{r}, t)\,|\,\mathbf{d}\,|\,\psi(\mathbf{r}, t)\rangle =$$
$$= \langle\tilde{\psi}(\mathbf{r}, t)\,|\,\mathbf{d}\,|\,\tilde{\psi}(\mathbf{r}, t)\rangle / \langle\tilde{\psi}(\mathbf{r}, t)\,\tilde{\psi}(\mathbf{r}, t)\rangle \tag{11}$$

under condition that α is considerably less than unity and the duration of the external light action essentially exceeds the value $\frac{1}{|\gamma_{21}|}$ leads to the following expression:

$$\langle\mathbf{d}(t)\rangle = [(e\,\mathbf{r}_{12}\,F_n^* \exp(-i\omega t + i\mathbf{K}\,\mathbf{R}) +$$
$$+ e\,\mathbf{r}_{21}\,F_n \exp(i\omega t - i\mathbf{K}\,\mathbf{R})) +$$
$$+ \frac{\alpha m e}{\hbar}(\mathbf{V}(\mathbf{rr})_{11} + (\mathbf{rr})_{12}\,F_n^* \exp(-i\omega t + i\mathbf{KR}) +$$
$$+ (\mathbf{rr})_{21}\,F_n \exp(i\omega t - i\mathbf{K}\,\mathbf{R}) +$$
$$+ (\mathbf{rr})_{22}|F_n|^2)]/(1 + |F_n|^2 +$$
$$+ \frac{\alpha m e}{\hbar}\,|(\mathbf{V}\,\mathbf{r}_{21})\,F_n|\cos(\omega t - \mathbf{K}\,\mathbf{R} + \varphi)). \tag{12}$$

When deriving formula (12) we assumed that the vectors $\mathbf{r}_{21}$ and $\mathbf{E}_0$ are parallel. The value F_n is defined by the relation

$$F_n = i2\hbar\,\beta_n / e(E_0\,\mathbf{r}_{12}), \tag{13}$$

where the parameter n takes the values

$$n = 1 \text{ for } \varepsilon > 0,\ \gamma_{21} < 0;\ \varepsilon < 0,\ \gamma_{21} > 0; \tag{14}$$
$$n = 2 \text{ for } \varepsilon > 0,\ \gamma_{21} > 0;\ \varepsilon < 0,\ \gamma_{21} < 0. \tag{15}$$

The additional phase φ introduced into the cosine argument, in the denominator, is determined by the ratios between $\mathbf{r}_{12}$ and F_n. Besides, the matrix elements $(\mathbf{rr})_{ij} = \langle\psi_i|\mathbf{rr}|\psi_j\rangle$ have been introduced into Eq. (12).

By multiplying $\langle\mathbf{d}(t)\rangle$ by the concentration of atoms nearby the point $\mathbf{R}$, one can obtain the polarization of a unit volume of the medium.

The summand standing in the numerator and proportional to α is responsible, first, for the effects as rotation of the polarization plane of scattered radiation[4] and it is unimportant for this study. So we do not take it into account in what follows. This makes it possible to write Eq. (12) in the form

$$\langle\mathbf{d}(t)\rangle =$$
$$= \frac{[e\mathbf{r}_{12}\,F_n^* \exp(-i\omega t + i\mathbf{KR}) + e\mathbf{r}_{21}\,F_n \exp(i\omega t - i\mathbf{KR})]}{[1 + |F_n|^2 + \frac{2\alpha m}{\hbar}|(\mathbf{V}\mathbf{r}_{21})\,F_n|\cos(\omega t - \mathbf{KR} + \varphi)]}. \tag{16}$$

The numerator in the Eq. (16) describes the dipole moment induced by the resonant radiation in a separate atom,[5] accurate to a factor close to unity. The denominator involves an additional summand that appeared here due to the assumption that the atom is in an anisotropic thermostat. This summand depends not only on the average drift velocity of the considered system with respect to the thermostat but also on the parameters of external radiation. By expanding $<\mathbf{d}(t)>$ into a series over the powers of the parameter α

$$<\mathbf{d}(t)> = \alpha^0 <\mathbf{d}(t)>_0 + \alpha^1 <\mathbf{d}(t)>_1 + \\ + \alpha^2 <\mathbf{d}(t)>_2 + \ldots, \quad (17)$$

one can easily see that the series terms contain harmonics of the external radiation. The most significant term is $\alpha<\mathbf{d}(t)>_1$:

$$\alpha <\mathbf{d}(t)>_1 = \alpha\, (e\mathbf{r}_{12}\, F_n^* \exp(-\,i\omega t + i\mathbf{KR}) + \\ + e\mathbf{r}_{21}\, F_n \exp(i\omega t - i\mathbf{KR}))\, 2m\,|(\mathbf{Vr}_{21})\, F_n| \times \\ \times \cos(\omega t - \mathbf{KR} + \varphi)/[\hbar\,(1 + |F_n|^2)^2]. \quad (18)$$

It is easy to see that the value (18) differs from zero if the angle between the vectors $\mathbf{V}$ and $\mathbf{E}_0$ is not equal to $\pi/2$, the value $\alpha<\mathbf{d}(t)>_1$ being maximal when the vectors are parallel. This means that the scattered radiation should contain the component with the doubled frequency of the incident radiation. This frequency doubled wave must propagate along the same direction as the incident one. Its amplitude depends on atom concentration, parameter α (i.e., on the thermostat density), and on the velocity $\mathbf{V}$. Since the induced dipole moment of the atom at the doubled frequency differs from that at the fundamental frequency of $\alpha<\mathbf{d}(t)>_1$ by the factor

$$\mu = \alpha\, 2m\,|(\mathbf{Vr}_{21})\, F_n|/[\hbar\,(1 + |F_n|^2)], \quad (19)$$

it can be easily determined.

The substitution, into Eq. (19), of the parameters that are close in their values to real ones shows that the following estimate of μ:

$$\mu \approx 10^2\, \alpha\, |\mathbf{V}|/c \quad (20)$$

is valid.

According to Eq. (20), in the case when the thermostat is sufficiently dense ($\alpha \approx 10^{-3}$–10^{-2}) and the velocity of the directed motion of the ensemble of particles disturbing the atom is commensurable with the speed of light, the dipole moment of an atom at the doubled frequency is close to the "resonant" value. This points to the fact that the effect of frequency doubling may be sufficiently strong to be detectable in experimental observations.

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