Generation of laser radiation harmonics by an ensemble of three-level atoms in a shock wave

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One of possible mechanisms of generation of laser radiation harmonics in light scattered from a medium in the front of a shock wave is studied theoretically. Interference of upper quantum states can produce a pronounced effect on the spectrum of scattered radiation: combined frequencies can appear in the spectrum of scattered light along with the multiple frequencies, close to them.

It is well-known[1] that laser radiation harmonics can appear in the front of a shock wave arising at the laser-induced break-down of a medium. The nonlinear effects that can occur in the domain of a shock wave are the main cause leading to appearance, in the multipole moment of the ensemble of atoms that interacts with radiation, of summands containing harmonics of the incident exciting radiation. In this paper, we consider one of the ways to describe this phenomenon.

The situation is simulated in the following way. Assume that we have an ensemble of three-level atoms moving in the front of a shock wave. The upper two levels of atoms are assumed to be close in energy. It is assumed that movement of these atoms with respect to a medium whose atoms are not yet involved in the process occurs at a certain average velocity **V**. This velocity varies with acceleration **a** due to "friction" (interaction with motionless atoms). A plane monochromatic light wave whose frequency coincides with the frequency of atomic transition from the first level to the second is incident on this ensemble. The symmetry of atom states is assumed to be such that transition from the first level to the second one is possible in the dipole approximation. Adding parameters connected with the properties of the thermostat to the wave equation for the ψ-function, we phenomenologically take into account the effect of collisions of individual atoms with the particles of the surrounding medium which is considered as a thermostat.

Here we start from the nonlinear Schrödinger equation[2] obtained by the method of path integrals and describing the behavior of a subsystem of a large ensemble of particles:

$$i\hbar \frac{\partial \psi}{\partial t} = \frac{1}{1 + i\alpha} \hat{T} \psi + U\psi \frac{i\alpha}{1 + \alpha^2} \langle \psi | \hat{T} | \psi \rangle \psi, \quad (1)$$

where $\hat{T}$ is the efficient operator of kinetic energy (its form for the considered approximation is presented below); U is the efficient operator of potential energy; α is the parameter connected with the properties of the medium which involves the subsystem studied. Nonlinearity of this equation is connected with the reduction of the problem for an ensemble to a problem for a subsystem within it and implicitly reflects the reaction of the ignored part of the ensemble to changes in the subsystem isolated.[3]

The solution of this equation written for an optical electron of an individual atom interacting with the environment can be represented in the following form:

$$\psi = \frac{\psi(\mathbf{r}, t)}{[\langle \psi(\mathbf{r}, t) | \psi(\mathbf{r}, t) \rangle]^{1/2}}. \quad (2)$$

The value $\psi(\mathbf{r}, t)$ satisfies the Schrödinger equation simulating the behavior of an atom's electron in an anisotropic Markovian thermostat:

$$i\hbar \frac{\partial \psi}{\partial t} = \frac{1}{1 + i\alpha} \left[\frac{1}{2m} \left(\hat{\mathbf{P}} - \frac{e}{c} \mathbf{A} - (1 - i\alpha) \frac{m\mathbf{V}}{2} \right)^2 + \chi \right] \psi + e \left[\varphi - \frac{(\mathbf{A} \cdot \mathbf{V})}{2c} - \left(1 + i\alpha \frac{m\mathbf{V}^2}{8c} \right) \right] \psi, \quad (3)$$

where e and m are charge and mass of the electron; $\hat{\mathbf{P}}$ is the momentum operator; **A** and φ are vector and scalar potentials of the electromagnetic field containing the optical electron of an atom; α and χ are non-negative parameters connected with thermostat density (those are the larger the more frequent the shock perturbations of the atom are).

Omitting the summands of the second order of magnitude both in the equation and in its solution, one can represent the wave function satisfying Eq. (3) as the following product:

$$\psi(\mathbf{r}, t) = \exp[\alpha (m/(2\hbar)) (\mathbf{V} \cdot \mathbf{r})] \tilde{\psi}(\mathbf{r}, t), \quad (4)$$

where $\tilde{\psi}(\mathbf{r}, t)$ is a solution of the equation

$$i\hbar \frac{\partial \tilde{\psi}}{\partial t} = \frac{1}{1 + i\alpha} (\hat{H} - U) \tilde{\psi} + U \tilde{\psi} + (\mathbf{E} \cdot \mathbf{d}) \tilde{\psi} - \frac{m}{2e} (\mathbf{a} \cdot \mathbf{d}) \tilde{\psi}. \quad (5)$$

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The designations in Eq. (5) are standard: $\hat{H}$ is the Hamiltonian of a non-disturbed atom; U is the operator of its potential energy; $\mathbf{d}$ is the dipole moment operator; $\mathbf{E}$ is the strength of the electric component of the external electromagnetic field.

Using the eigenfunctions $\psi_k(\mathbf{r})$ of the non-disturbed Hamiltonian as a basis, one can represent the solution of Eq. (5) in the form

$$\tilde{\psi} = \sum_{k=1}^{3} C_k \exp\left(-\frac{i}{\hbar} E_k t - \gamma_k t\right) \psi_k(\mathbf{r}), \qquad (6)$$

$$\gamma_k = \alpha\,(E_k - U_{kk})/\hbar,$$

where U_{kk} are the diagonal elements of the matrix of the potential energy operator.

The coefficients C_k satisfy the following systems of equations which are valid in the approximation of rotating waves:

$$i\hbar \frac{\partial C_1}{\partial t} = C_2 \frac{(\mathbf{E}_0 \cdot \mathbf{d}_{12})}{2} \exp[-i\,(\mathbf{K}\cdot\mathbf{R}) - (\gamma_2 - \gamma_1)t] +$$

$$+ bC_3 \frac{(\mathbf{E}_0 \cdot \mathbf{d}_{13})}{2} \exp[-i\omega_{32}t - i(\mathbf{K}\cdot\mathbf{R}) - (\gamma_3 - \gamma_1)t]; \quad (7)$$

$$i\hbar \frac{\partial C_2}{\partial t} = C_1 \frac{(\mathbf{E}_0 \cdot \mathbf{d}_{21})}{2} \exp[i\,(\mathbf{K}\cdot\mathbf{R}) +$$

$$+ (\gamma_2 - \gamma_1)t] - (1-b)C_3 \frac{m}{2e}(\mathbf{a}\cdot\mathbf{d}_{23}) \times$$

$$\times \exp[-i\omega_{32}t - i(\mathbf{K}\cdot\mathbf{R}) - (\gamma_2 - \gamma_1)t]; \qquad (8)$$

$$i\hbar \frac{\partial C_3}{\partial t} = bC_1 \frac{(\mathbf{E}_0 \cdot \mathbf{d}_{31})}{2} \exp[i\omega_{32}t + i\,(\mathbf{K}\cdot\mathbf{R}) +$$

$$+ (\gamma_3 - \gamma_1)t] - (1-b)C_2 \frac{m}{2e}(\mathbf{a}\cdot\mathbf{d}_{32}) \times$$

$$\times \exp[i\omega_{32}t - (\gamma_3 - \gamma_1)t]. \qquad (9)$$

In Eqs. (7)–(9), $\mathbf{E}_0$ is the amplitude of the external electromagnetic wave which is resonant to the atomic transition from the second level to the first one; $\mathbf{R}$ is the radius vector of an atom; $\mathbf{d}_{ij}$ are the matrix elements of the dipole moment; b is the parameter which equals 1 if transitions between the first and third levels are allowed in the dipole approximation; otherwise, it equals zero (in this case, transitions between the second and third levels are supposed to be allowed). The system of equations was constructed under the assumption that attenuation constants are considerably less than the frequency ω_{32}, and the latter frequency, in its turn, is considerably lower than the frequency of incident electromagnetic radiation.

The solution of the system (7)–(9), without the regard of the parameter b, up to the terms of higher orders of smallness, has the form

$$C_1 = \left\{A_1 \exp\left[\frac{(\gamma_1 - \gamma_2)}{2} t + i\Omega t\right] + \right.$$

$$+ A_2 \exp\left[\frac{(\gamma_1 - \gamma_2)}{2} t - i\Omega t\right] +$$

$$\left. + A_3 \exp\,[(\gamma_1 - \gamma_3 - i\omega_{32})t]\right\} \exp\,[i(\mathbf{K}\cdot\mathbf{R})], \quad (10)$$

$$C_2 = B_1 \exp\left[\frac{(\gamma_2 - \gamma_1)}{2} t + i\Omega t\right] +$$

$$+ B_2 \exp\left[\frac{(\gamma_2 - \gamma_1)}{2} t - i\Omega t\right] +$$

$$+ B_3 \exp\,[(\gamma_2 - \gamma_3 - i\omega_{32})t], \qquad (11)$$

$$C_3 = D_1 \exp\left[\left(\gamma_3 - \frac{(\gamma_1 + \gamma_2)}{2} + i\Omega + i\omega_{32}\right) t\right] +$$

$$+ D_2 \exp\left[\left(\gamma_3 - \frac{(\gamma_1 + \gamma_2)}{2} - i\Omega - i\omega_{32}\right) t\right] + D_3. \quad (12)$$

Here Ω is the Rabi frequency:

$$\Omega = \sqrt{\frac{(\gamma_1 - \gamma_2)^2}{4} - \frac{|(\mathbf{E}_0 \cdot \mathbf{d}_{12})|^2}{\hbar^2}}\,. \qquad (13)$$

The constants A_i, B_i, and D_i are defined by the initial conditions.

If the expressions (10)–(12) contain summands describing the interference of quantum states, that leads, in calculating the dipole moment of an individual atom using $\tilde{\psi}(\mathbf{r}, t)$,

$$\langle \mathbf{d}(t) \rangle \approx \tilde{\mathbf{d}}\,[1 - \alpha m/(e\hbar)\,(\mathbf{V}\cdot\tilde{\mathbf{d}}) + \ldots], \qquad (14)$$

where

$$\tilde{\mathbf{d}} = \frac{\langle C_1\psi_1(\mathbf{r},t) + C_2\psi_2(\mathbf{r},t) + C_3\psi_3(\mathbf{r},t)\,|\,\mathbf{d}\,|\,C_1\psi_1(\mathbf{r},t) +}{\langle C_1\psi_1(\mathbf{r},t) + C_2\psi_2(\mathbf{r},t) + C_3\psi_3(\mathbf{r},t)\,|\,C_1\psi_1(\mathbf{r},t) +}$$

$$\frac{+ C_2\psi_2(\mathbf{r},t) + C_3\psi_3(\mathbf{r},t)\rangle}{+ C_2\psi_2(\mathbf{r},t) + C_3\psi_3(\mathbf{r},t)\rangle}\,; \qquad (15)$$

$$\psi_k(\mathbf{r}, t) = \exp\left(-i\frac{E_k}{\hbar} t - \gamma_k t\right) \psi_k(\mathbf{r}),$$

to appearance of summands oscillating not only at the frequency of external radiation and its harmonics but also at combination frequencies. These oscillations must evidently induce electromagnetic waves propagating mostly along the direction of the external laser radiation.

References

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