Raman scattering of laser radiation by atoms participating in an oscillatory movement

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The process of scattering of intense coherent electromagnetic radiation by an ensemble of hydrogen-like atoms is considered theoretically. It is supposed that in this ensemble the standing sound wave is excited, and the external radiation is resonant to the transition between the two lower energy levels of these atoms. The oscillations of atoms about the equilibrium state are taken into account by the drift operator in the Hamiltonian of the nonlinear Schrödinger equation written for one of the atoms. It is obtained, that the scattered radiation should include combinations of frequencies of the incident radiation and the sound wave.

The phenomenon of light diffraction on standing sound waves in a medium is well known. First of all, one relates this phenomenon to the spatial oscillations of the density of scattering particles. Then one usually can calculate the energy distribution in the diffraction pattern sufficiently precise. However, such an approach to the description of light scattering is not comprehensive. In particular, it does not take into account the fact that the molecules and atoms form clusters and van der Waals molecules. Individual atoms in such formations have equilibrium positions, about which they harmonically oscillate. So one can expect that at scattering of light on an ensemble of such atoms Raman-shifted frequencies can appear in the spectrum of scattered radiation. This paper is devoted to theoretical consideration of such a Raman scattering of light by van der Waals molecules.

The initial suppositions are the following: light is presented as a plane monochromatic wave of high intensity (the density of energy of the wave is significantly greater than the energy which a molecule can accumulate in a unit volume)

$$\mathbf{E} = \mathbf{E}_0 \cos[\omega t - (\mathbf{K}\cdot\mathbf{R})], \tag{1}$$

its frequency is close to the frequency of transitions of the atoms from excited state to the ground one. In Eq. (1) $\mathbf{E}_0$ is the amplitude of the light wave, ω is the cyclic frequency, t is time, $\mathbf{K}$ is the wave vector, $\mathbf{R}$ is the coordinate of the point. One can consider the medium as an ensemble of hydrogen-like atoms, which initially are in the ground state and can transit to the second energy level under the effect of radiation. These atoms take part in the oscillations caused by excitation of a standing sound wave in the medium:

$$\mathbf{V} = 2\mathbf{V}_0 \cos(\mathbf{K}_0\cdot\mathbf{R})\cos(\omega_0 t), \tag{2}$$

where $\mathbf{V}_0$ is the amplitude of the velocity of the medium involved into the sound wave oscillations, ω_0 is the cyclic frequency of this wave, $\mathbf{K}_0$ is its wave vector.

As the atoms of the medium are involved into the joint motion, one can not consider them as isolated formations. The change of their states should cause the change of the states of neighbor particles, and, hence, cause the change of the state of these atoms themselves through the effect of the neighbors. This fact is taken into account in our paper by means of the nonlinear Schrödinger equation[1,2] which has the following form for the hydrogen-like atoms accurate to the terms of smaller order of magnitude.

$$i\hbar\frac{\partial\Psi}{\partial t} = \frac{1}{1+i\alpha}\frac{1}{2m}\hat{\mathbf{P}}^2\Psi + e\varphi\Psi + (\mathbf{E}\cdot\mathbf{d})\Psi + \frac{m}{2e}(\dot{\mathbf{V}}\cdot\mathbf{d})\Psi + \frac{i\alpha}{1+\alpha^2}\langle\Psi|\frac{1}{2m}\hat{\mathbf{P}}^2|\Psi\rangle\Psi. \tag{3}$$

Standard notations are introduced in Eq. (3): the parameter α (the value is greater than zero) takes into account the density of the surrounding medium in implicit form, this parameter is greater in a more dense medium.

One can found the time-regular solutions of Eq. (3) by solving the equation of the form

$$i\hbar\frac{\partial\psi}{\partial t} = \frac{1}{1+i\alpha}\frac{1}{2m}\hat{\mathbf{P}}^2\psi + e\varphi\psi + (\mathbf{E}\cdot\mathbf{d})\psi + \frac{m}{2e}(\dot{\mathbf{V}}\cdot\mathbf{d})\psi. \tag{4}$$

The functions Ψ and ψ are related by the following formula

$$\Psi = \psi/\langle\psi|\psi\rangle^{1/2}. \tag{5}$$

If one takes into account the selection rules valid for the hydrogen atom in the dipole approximation, one can represent the solution of Eq. (4) in the form

$$\psi = b_1(t)\exp\left(-\frac{i}{\hbar}E_1 t - \gamma_1 t\right)\psi_1(\mathbf{r}) + \exp\left(-\frac{i}{\hbar}E_2 t - \gamma_2 t\right)[b_2(t)\psi_2(\mathbf{r}) + b_3(t)\psi_3(\mathbf{r})]. \tag{6}$$

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Here E_i is the energy of the stationary state of the hydrogen-like atom, γ_i is the attenuation constant. The wave functions $\psi_i(\mathbf{r})$ are orthogonal to each other, and the matrix elements $\mathbf{d}_{12}$ and $\mathbf{d}_{23}$ and the complex conjugated values of the dipole moment are different from zero.

On substituting the expression (6) in Eq. (4) it is easy to obtain the system of differential equations

$$i\hbar\frac{\partial b_1}{\partial t} = b_2(\mathbf{E}\cdot\mathbf{d}_{12})\exp\left(-i\frac{E_2 - E_1}{\hbar}t - (\gamma_2 - \gamma_1)t\right),$$

$$i\hbar\frac{\partial b_2}{\partial t} = b_1(\mathbf{E}\cdot\mathbf{d}_{21})\times$$

$$\times\exp\left(-i\frac{E_1 - E_{12}}{\hbar}t - (\gamma_1 - \gamma_2)t\right) + b_3\frac{m}{2e}(\dot{\mathbf{V}}\cdot\mathbf{d}_{23}),$$

$$i\hbar\frac{\partial b_3}{\partial t} = b_2\frac{m}{2e}(\dot{\mathbf{V}}\cdot\mathbf{d}_{32}). \qquad (7)$$

The system (7) is written under assumption that the frequency of laser radiation is some orders of magnitude greater than the frequency of sound.

If one supposes that the duration of the laser pulse in significantly greater than the lifetime of the excited state, the following relationships are valid accurate to a constant factor for all coefficients $b_i(t)$ in adiabatic approximation of sound oscillations

$$b_1(t) = \exp\left\{\left[-i\left(\Omega + \frac{\varepsilon}{2}\right) - \frac{(\gamma_2 - \gamma_1)}{2}\left(1 - \frac{\varepsilon}{2\Omega}\right)\right]t\right\}, \qquad (8)$$

$$b_2(t) = \exp\left\{\left[-i\left(\Omega - \frac{\varepsilon}{2}\right) + \frac{(\gamma_2 - \gamma_1)}{2}\left(1 + \frac{\varepsilon}{2\Omega}\right)t\right]\right\}\times$$

$$\times\exp[i(\mathbf{K}\cdot\mathbf{R})]\left[1 - \left(\frac{m\omega_0}{e\hbar\Omega}\right)^2\left|(\mathbf{V}_0\cdot\mathbf{d}_{23})\right|^2\times\right.$$

$$\left.\times\cos^2(\mathbf{K}_0\cdot\mathbf{R})\sin^2(\omega_0 t)\right], \qquad (9)$$

$$b_3(t) = \exp\left\{\left[-i\left(\Omega - \frac{\varepsilon}{2}\right) + \frac{(\gamma_2 - \gamma_1)}{2}\left(1 + \frac{\varepsilon}{2\Omega}\right)t\right]\right\}\times$$

$$\times\frac{m\omega_0}{\hbar\Omega}(\mathbf{V}_0\cdot\mathbf{d}_{32})\cos(\mathbf{K}_0\cdot\mathbf{R})\sin(\omega_0 t). \qquad (10)$$

In formulas (8)–(10) ε is the deviation of the frequency of laser radiation from the natural frequency of the atom:

$$\varepsilon = \frac{E_2 - E_1}{\hbar} - \omega, \qquad (11)$$

Ω is the Stark shift:

$$\Omega = \frac{\left|(\mathbf{E}_0\cdot\mathbf{d}_{12})\right|}{2\hbar}. \qquad (12)$$

Substitution of the obtained relationships into the wave function leads to the following formula in calculating the induced dipole moment:

$$\langle\mathbf{d}\rangle \approx \langle\Psi|\mathbf{d}|\Psi\rangle = \{\mathbf{d}_{12}\exp[(-i\omega t) + i(\mathbf{K}\cdot\mathbf{R})]\times$$

$$\times\left[1 - \left(\frac{m\omega_0}{e\hbar\Omega}\right)^2\left|(\mathbf{V}_0\cdot\mathbf{d}_{23})\right|^2\cos^2(\mathbf{K}_0\cdot\mathbf{R})\sin^2(\omega_0 t)\right] +$$

$$+\mathbf{d}_{23}\left(\frac{m\omega_0}{e\hbar\Omega}\right)(\mathbf{V}_0\cdot\mathbf{d}_{32})\cos(\mathbf{K}_0\cdot\mathbf{R})\sin(\omega_0 t) +$$

$$+\text{c.c.}\}\Big/\left\{2 - \left(\frac{m\omega_0}{e\hbar\Omega}\right)^2\left|(\mathbf{V}_0\cdot\mathbf{d}_{23})\right|^2\cos^2(\mathbf{K}_0\cdot\mathbf{R})\sin^2(\omega_0 t)\right\}. \qquad (13)$$

The terms describing in Eq. (13) the oscillations with the sound frequency show that, in addition to the main frequency, both low-frequency electromagnetic waves (odd harmonics of the sound frequency) and the waves of the frequency $\omega \pm 2n\omega_0$ (n is an integer number) should be present in the spectrum of scattered radiation, i.e., the Raman scattering of light should take place. Spatial modulation of the dipole moment value of an individual atom shows that the formalism developed in this paper is applicable to description of the phenomenon of diffraction of an intense laser radiation on the standing sound waves.

References

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