

Influence of sound and low-frequency electromagnetic waves propagating through a substance on spectral composition of scattered laser radiation

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The nonlinear wave equation derived by the method of path integrals is applied to describe scattering of laser radiation. The equations for the field strength at Raman frequencies are obtained in the dipole approximation.

Correct description of laser radiation scattering by different media is one of the key problems in optics since the advent of lasers and formation of nonlinear optics as an independent research area in physics. A wide variety of different factors and physical phenomena that should be taken into account simultaneously when solving the problem makes it very intricate to analyze and solve. Therefore, in spite of a great bulk of experimental data, no unified theory has been proposed that is capable of giving comprehensive answers to all questions.

In this paper we develop an approach allowing one to consider all problems associated with the radiation scattering from a unified point of view. The approach enables one to avoid unnecessary complication of the problem and to obtain results that agree well with the observed experimental data. The proposed method is applied to the study of the influence of sonic and low-frequency electromagnetic waves propagating through a substance on the spectral composition of scattered laser radiation. Such a model is chosen because collective phenomena in media can play a significant part in the mechanism of Raman scattering and stimulated Raman scattering, as well as in dynamics of transition from one type of scattering to another.

Actually, the methods for solution of this problem are based on two approaches: (1) quantum equations describing the response of a medium to the external electromagnetic field can be derived from the Liouville–von Neumann equation^{1,2} or (2) quantum equation can be obtained from phenomenological usage of the main properties of a thermostat (in particular, assumption of the Markovian properties of the processes occurring). The former approach fits neatly into the classic schemes of quantum mechanics thus being attractive for the rigorous and logically sequential actions resulting in solution of the problem. However, this method can give mathematically correct and useful results only in limiting cases and, as a rule, under some additional assumptions.

This work is based on the latter approach. The method of path integrals³ allows one to take into account the influence of surroundings on the behavior of a selected molecule already at the stage of constructing the wave equation. Assuming the processes

occurring in the medium to be Markovian (what is rather an evident physical assumption) and introducing the additional weighting function discriminating trajectories by their probabilities, we can derive⁴ the wave equation, that includes the velocity of the system motion in a thermostat as one of the parameters, in the following form:

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

where α and χ are the parameters of the thermostat.

The joint effect of vibrations of atoms and of the external field leads to appearance of dipole moments in atoms (due to the interaction with the inner electrons). The dipole moments oscillate at the frequencies of the external field, frequencies of the atomic oscillations, and their combinations.

The vibrations of atoms near the ground state can be considered harmonic to a good approximation. Consequently, in Eq. (1) we can introduce

$$\mathbf{V} = \mathbf{V}_0 \cos(\omega t + f), \quad (2)$$

where ω is the frequency of atomic vibrations. Strictly speaking, we should take into account vibrations at multiple frequencies too, but such a substitution only complicates the mathematics not changing the task qualitatively. The value of V_0 can be estimated starting from the virial theorem

$$V_0 = \sqrt{2\hbar\omega/M} \quad (3)$$

or from the theorem on energy distribution over the degrees of freedom

$$V_0 = \sqrt{3kT/M}. \quad (4)$$

The estimates obtained agree by the order of magnitude.

Let us introduce two assumptions more, that in our opinion are physically well justified: (1) molecules have no constant dipole moment and (2) the external field induces the dipole moment in one of atoms, which has the natural frequency close to that of the external field.

Let us substitute

$$\psi(\mathbf{r}, t) = \psi_1(\mathbf{r}, t) \exp \left[-\frac{i\chi t}{\hbar(1+i\alpha)} + \frac{ie}{\hbar c} \times \left(\left(\mathbf{A} + \frac{e}{c}(1+i\alpha) \frac{m\mathbf{V}}{2} \right) \cdot \mathbf{r} \right) + \frac{im}{8\hbar}(1+i\alpha) \int \mathbf{V}^2 dt \right] \quad (5)$$

into the Eq. (1) that involves the velocity in the form (2).

For the new wave function $\psi_1(\mathbf{r}, t)$, the nonlinear equation

$$i\hbar \frac{\partial \psi_1}{\partial t} = \frac{1}{1+i\alpha} (H - U) \psi_1 + \left(U + \frac{e}{2} \mathbf{V} \cdot \int \mathbf{E} dt \right) \psi_1 + (\mathbf{E} \cdot \mathbf{d}) \psi_1 - \frac{m}{2e} (1-i\alpha) \left(\frac{\partial \mathbf{V}}{\partial t} \cdot \mathbf{d} \right) \psi_1 - \frac{m}{8} (1+i\alpha) \cdot \mathbf{V}^2 \psi_1 \quad (6)$$

is valid, where \mathbf{E} is the strength of the external light field; \mathbf{d} is the dipole moment. Since quantum states split due to interaction with the thermostat, the equation for the wave function keeping its normalization in time has the form

$$\psi = \psi / \sqrt{\langle \psi | \psi \rangle}. \quad (7)$$

Hereinafter the subscript of the wave function is omitted for brevity.

Equation (6) can be solved by the method of successive approximations. For further simplification let us assume the external field to be a monochromatic wave with the frequency ω_0 close to the frequency ω_{21} of the transition from the first level to the second. This choice allows us to restrict ourselves to the two-level approximation. Besides, we believe that the frequency detuning $\varepsilon = \omega_0 - \omega_{21}$ far exceeds the width of the spectral line and the matrix elements of the quadrupole moment are nonzero.

The external field can be presented in the form

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_0 \cos(\omega t - \mathbf{k}\mathbf{r}) + \delta\mathbf{E}(\mathbf{r}, t), \quad (8)$$

where \mathbf{k}_0 is the wave vector of the incident wave; $\delta\mathbf{E}$ is the strength of the electric field generated by the system which is related to the polarization of the unit volume in the dipole approximation by the well known relation

$$\delta\mathbf{E}(\mathbf{r}, t) = \int_V \frac{1}{c^2 \mathbf{r}} \left[\mathbf{n} \left(\mathbf{n} \cdot \frac{\partial^2 \mathbf{P}}{\partial t^2} \right) - \frac{\partial^2 \mathbf{P}}{\partial t^2} \right] d\mathbf{V}. \quad (9)$$

The solution of Eq. (6) for the polarization vector splits into three terms.^{5,6} First two terms are responsible for radiation at the frequency of the external field and at the frequency of natural oscillations, respectively. The last term is of greatest interest; it is responsible for radiation at Raman frequencies. In the case of correlation between vibrations of atoms in the medium, this term can be presented, accurate to a constant, as

$$P = C \cos(\omega_1 t - \mathbf{k}_1 \mathbf{r}), \quad (10)$$

where $\omega_1 = \omega_0 \pm 2\omega$; $\mathbf{k}_1 = \mathbf{k}_0 \pm 2\mathbf{k}$. The "+" sign corresponds to the anti-Stokes component, while the "-" sign corresponds to the Stokes component; ω_0 is the frequency of the external field; ω is the frequency of vibration of atoms in the medium; \mathbf{k} is the wave vector of the wave that induces synchronization, which in the general case can be induced by light and sound waves propagating through the medium. Correspondingly, the following equation

$$\mathbf{k} = \mathbf{k}_s + \mathbf{k}_l, \quad (11)$$

is valid in the general case for the wave vector. In Eq. (11) \mathbf{k}_l is the wave vector of the synchronizing light wave; \mathbf{k}_s is the wave vector of the synchronizing sound wave.

The next our assumption simplifies the mathematics, while not decreasing the usefulness of the results to be obtained. The external field is considered propagating along the Z axis. Assume that the distance to the observation point is far longer than the system dimensions [this assumption allows us to ignore amplitude attenuation with distance and consider \mathbf{r} to be constant in integrating in Eq. (9)]. The system is chosen to be a parallelepiped with the coordinates $-l, +l$ along the Z axis and $-b, +b$ along the X and Y axes. To bring theory in correspondence with the experiment, we assume $l \gg b$.

In integration we take into account the lags due to the finite speed of electromagnetic wave propagation and symmetry of the integration limits. Thus, for the strength of the field at Raman frequencies we have δE accurate to a constant and a time-dependent factor:

$$\delta E = \omega_1^2 \frac{\sin(\xi_x b)}{\xi_x} \frac{\sin(\xi_y b)}{\xi_y} \frac{\sin(\xi_z l)}{\xi_z}, \quad (12)$$

where

$$\begin{aligned} \xi_x &= \frac{\omega_1 \cos\alpha}{c} - k_1 \cos\alpha_1, \\ \xi_y &= \frac{\omega_1 \cos\beta}{c} - k_1 \cos\beta_1, \\ \xi_z &= \frac{\omega_1 \cos\gamma}{c} - k_1 \cos\gamma, \end{aligned} \quad (13)$$

$\cos\alpha$, $\cos\beta$, and $\cos\gamma$ are the direction cosines of the point at which the field is to be determined; $\cos\alpha_1$, $\cos\beta_1$, and $\cos\gamma_1$ are the direction cosines of the vector \mathbf{k} .

One can see from Eqs. (12) and (13) that the field emitted by the system can have peaks or dips depending on the direction of the wave vector \mathbf{k} , thus providing spatial quantization of the radiation.

The method of computer simulation was used to study the behavior of the intensities of Stokes and anti-Stokes components of radiation depending on the orientation of the wave vectors of light and sound synchronizing waves. In the simulation we used the following values of the parameters: speed of light of $3 \cdot 10^8$, sonic speed of 2500, ω_0 of 10^{15} , ω of 10^{13} , l of 0.1, b of 10^{-5} (what corresponds to the dimensions of a laser beam). Below we present the directional patterns of the

scattering intensity as functions of the direction along which the synchronizing waves propagate through the substance.

Figure 1 illustrates the case with a sound wave as a synchronizing wave. It should be considered separately, because it is most that frequent in practice. Actually, at the exposure of a substance to laser radiation, temperature gradient inevitably arises in the substance, what results in the appearance of sound waves.

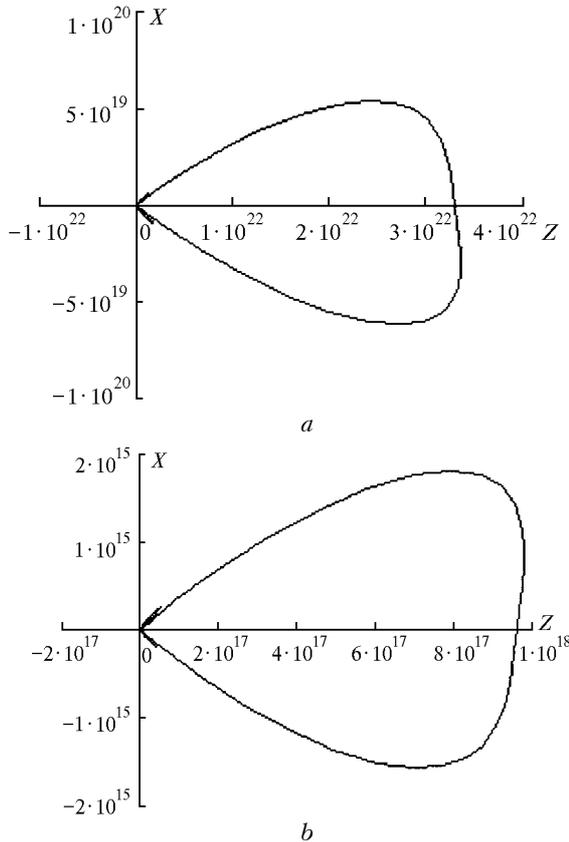


Fig. 1. Directional pattern of the scattering intensity of the Stokes (a) and anti-Stokes (b) components in the case of sonic synchronization.

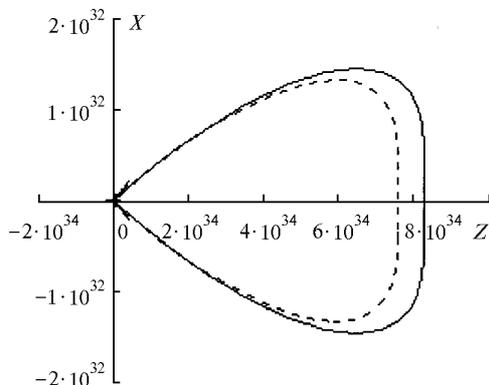


Fig. 2. Directional pattern of the scattering intensity of the Stokes (solid curve) and anti-Stokes (dashed curve) components in the case of light synchronization.

Analysis of Eq. (12) shows that the maximum value of the intensity should be expected in the case when a sound wave propagates at an angle to the direction of

the wave vector of the external field with the direction cosine approximately equal

$$\cos \gamma_{ak} = v/c, \tag{14}$$

where v is the speed of sound wave propagation.

In the case considered the Stokes component exceeds the anti-Stokes one by several orders of magnitude, what agrees well with the experiment.

Figure 2 shows the directional pattern of the scattering intensity for the case when vibrations are synchronized by a light wave propagating in parallel to the external field.

Figure 3 shows the directional pattern of the scattering intensity in the case of no correlation between vibrations of atoms. Besides the slight spatial quantization of the Stokes component, we can also explain the behavior of the anti-Stokes component. As seen from the figure, the radiation of the anti-Stokes component must be cone-shaped, what closely agrees with the experimental data on stimulated Raman scattering.

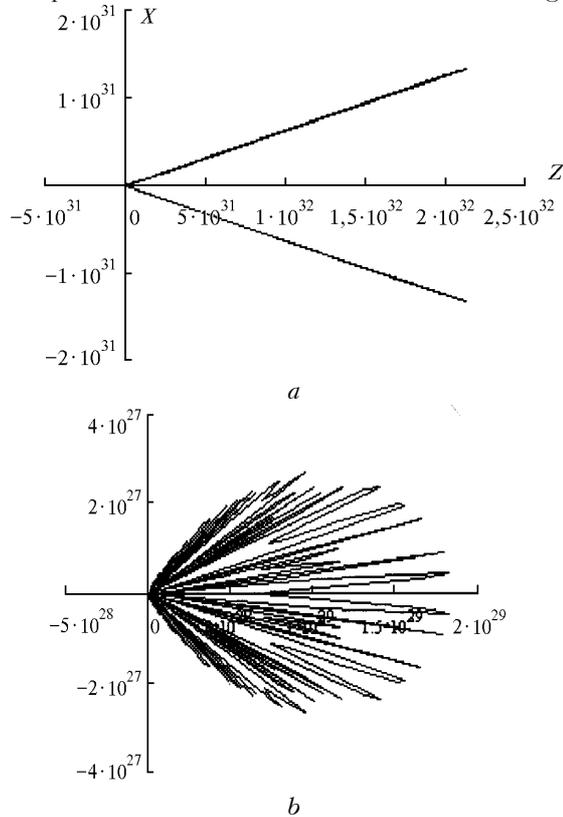


Fig. 3. Directional pattern of the scattering intensity of the anti-Stokes (a) and Stokes (b) components in the case of no synchronizing waves.

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