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The influence of collisions on the temporary shape of stimulated echo hologram in gas

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Abstract. In this paper, we investigate the influence of collisions with the change of particle velocity direction in a gas on the reproduction of the temporary shape of the object laser pulse in the stimulated echo hologram response. Due to such collisions, the frequency shifts of the radiation of atoms in the gas randomly vary (spectral diffusion within the heterogeneously broadened line). It is shown, that such diffusion leads to the not correlated heterogeneous broadening in the gas at the different time intervals and the partial loss of system phase memory, which results in a partial loss of retrieved information encoded in the temporal form of the object laser pulse.

1. Introduction

In gas media, the formation of stimulated echo-holograms (SEH) is gualitatively different from their analogs in a solid object. The differences are associated with the movement of "working" particles, which leads to the need to take into account the collisions and random reorientations of particles in the formation of optical coherent responses of the medium. In a solid object, each "working" particle has its own transition frequency (heterogeneous broadening of the resonance line). During the formation of photon echo signal in a solid object, at each point within the sample is a phasing of the induced dipole moments during the echo response appearance and this phasing does not depend on the excitation direction. In gases, however, velocities of the individual particles are vector quantities, so each point within the sample is associated with its own direction in the space (the directed nature of the Doppler effect). Therefore, the amplitude-phase information contained in the object wave turns out to be "encoded" in the streams of particles moving with certain velocities. In addition, the phasing of individual dipole moments in the gas at the time of echo formation does not necessarily occur at each point of the sample, but phasing of the different velocity "packs" of the particles moving with various velocities [1-6]. Elastic collisions lead to random changes in the angle between the direction of the observation and the velocity of the particle, which in turn leads to random shifts in the emission frequency of each particle (spectral diffusion). This can have an effect on the formation of the SEH, even in case of the parallel wave vectors of the exciting laser pulses. Therefore, it is in the interest to consider the effect of the spectral diffusion on the temporal form of the SEH response [2-6].

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2. The basic equations

The description of the formation of the echo-holograms in gases requires simultaneous consideration of the Doppler shifts in the frequencies of the particles emission, changes in their position and the orientation in space, collisions of particles, and also degeneracy of their energy levels over some quantum number. In case of the impact mechanism of the broadening of the resonance transition line, the equation for the density matrix in the laboratory coordinate system has the form [2]:

$$\left(\frac{\partial}{\partial t} + \frac{\partial\Delta\vec{r}}{\partial t}\nabla\right)\rho_{\gamma\alpha}^{J_{1}J_{2}}\left(1 - \frac{\vec{n}}{c}\frac{\partial\Delta\vec{r}}{\partial t}\right)^{-1} = \frac{i}{\hbar}\left(H_{J_{1}}^{0} - H_{J_{2}}^{0}\right)\rho_{\gamma\alpha}^{J_{1}J_{2}} + \frac{i}{\hbar}\sum_{\beta}\left(V_{\gamma\beta}^{J_{1}J}\rho_{\beta\alpha}^{J_{2}} - V_{\beta\alpha}^{J_{2}}\rho_{\gamma\beta}^{J_{1}J}\right) - N\left\langle\upsilon\left(\sigma_{\gamma\alpha}' - i\sigma_{\gamma\alpha}''\right)\right\rangle\rho_{\gamma\alpha}^{J_{1}J_{2}} \tag{1}$$

where J_i is the total moment of *i*-th level, indexes α , β , γ denote sublevels degenerate levels, $\Delta \vec{r}$ is the displacement of the particle due to its movement on the considered time interval, \vec{n} is the direction of propagation of the laser excitation pulse, $V_{\beta\beta'}^{JJ'}$ are the matrix elements of the interaction operator of the atom with electromagnetic radiation, H^0 is Hamiltonian of the unperturbed atom, N is the concentration of the perturbing particles, υ is the relative velocity of the atom and the perturbing particle, σ and σ'' are cross-sections for the broadening and shifting of the corresponding spectral line, c is the velocity of propagation of the excitation laser pulse in the medium.

The spectral heterogeneity in the gas is due to the molecular velocity spread and the dependence of the frequency shift Δ_j of the j-th molecule on the velocity (the Doppler effect). In the absence of collisions, the heterogeneous broadening during the different time moments and at the different energy transitions is fully correlated, since the frequency shifts are proportional to each other at different transitions and at different time intervals due to particle motion:

$$\Delta_j = \Omega_{0ik} \frac{\vec{v}_j \vec{n}}{c} = \Omega_{0ik} \frac{v_j \cos \varphi_j}{c}, \qquad (2)$$

where Δ_j is the frequency shift due to the motion of the *j*-th particle with a velocity of \vec{v}_j (in the absence of collisions), Ω_{0ik} is the central frequency of the *i*-*k* transition, and φ_j is the angle between the molecular velocity and the direction of observation

Let us consider the situation where the observations of a gas emission are made at different time moments separated by an interval τ at which elastic collisions of gas molecules can occur, leading to a change in the direction of the particles' velocity. These collisions would result in random changes in the projection of the velocity on the direction of the observation and, accordingly, random changes in frequency shifts (2) at each individual collision:

$$\Delta_{j}(\varphi_{j},\delta\varphi_{\eta}(\Delta)) = \Omega_{0ik} \frac{v_{j}\cos(\varphi_{j} + \delta\varphi_{\eta}(\Delta))}{c}, \qquad (3)$$

where $\delta \varphi_{\eta}(\Delta)$ is the change in the magnitude of the angle between the direction of the velocity of the molecule and the direction of observation due to a random change in the direction of the velocity of the molecule as a result of an individual η -th collision. Thus, each isochrome of the Doppler-broadened line breaks down over the time as a result of the spectral diffusion. This leads to non-correlation of the heterogeneous broadening in the gas at different moments of time and partial loss of the phase memory of the system.

Let us find the average number of collisions at the time interval τ between the moments of observation of the system and their effect on the radiation frequency of the molecule. Over a second

the molecule will, on average, pass a path equal to the average speed. Collisions with the change in velocity "distort" its path. We denote the effective diameter of the molecule by d and represent the molecule as a sphere. Then, the number of collisions of N_0 molecule with other molecules per second will be equal to the number of the molecules whose centers are in a cylinder of length equal to the mean velocity and the diameter of 2d [7]:

$$N_0 = \pi d^2 \bar{v} \rho$$
,

where ρ is the number of molecules per unit volume. But we need to make a correction to the fact that this molecule collides with not immobile molecules, but with moving molecules [1]. This circumstance will be taken into account if the average relative velocity is used instead of the mean absolute velocity. Consequently, $v_r \approx \sqrt{2}\overline{v}$. Therefore:

$$N_0 = \sqrt{2\pi d^2 \overline{\nu} \rho}.$$

Over the time interval τ between the moments of the observation time of the system, an average of $N(\tau) = N_0 \tau = \sqrt{2}\pi d^2 \overline{\nu} \rho \tau$ collisions will occur. This will lead to a change in the angle between the velocity of the molecule and the direction of observation in (3):

$$\Delta_{j}(\varphi_{j},\delta\varphi_{\eta}(\Delta)) = \Omega_{0ik} \frac{v_{j}\cos(\varphi_{j} + \sum_{\eta=1}^{N(\tau)}\delta\varphi_{\eta}(\Delta))}{c}, \qquad (4)$$

where the number of collisions is

$$N(\tau) = \frac{\overline{\nu}}{\overline{1}} \tau, \ \overline{\nu} = \sqrt{\frac{8kT}{\pi m}}.$$

3. The impact of collisions on the temporal shape of the stimulated echo-hologram in a gas

The described above spectral diffusion within the heterogeneously broadened line influences the temporal form of the SEH response [6]. Let us consider the case, where the information is included into the temporal form of the second laser pulse when writing SEH. In this case, the pulse sequence for excitation of a stimulated echo hologram is shown in figure 1.



Figure 1. The sequence of pulses at SEH excitation. Δt_i is the duration of the *i*-th resonant laser pulse, t_e is the time of appearance of the SEH response, $\tau = t_2 - t_1$ and $\tau_1 = t_3 - t_1$ are the time intervals between the exciting pulses, and P_1 , P_2 , P_3 are the resonant laser pulses.

The electric field strength of the SEH response with allowance for (1-4) in case of linear polarization of the exciting laser pulses will be determined by an expression similar to that obtained in [8-9]:

$$E \sim \int_{-\infty}^{\infty} \frac{\sin(\theta_{1}(\Delta))\sin(\theta_{2}(\Delta,\delta\varphi_{\tau}(\Delta)))\sin(\theta_{3}(\Delta,\delta\varphi_{\tau_{1}}(\Delta)))}{\theta_{1}'(\Delta)\theta_{2}'(\Delta,\delta\varphi_{\tau}(\Delta))\theta_{3}'(\Delta,\delta\varphi_{\tau_{1}}(\Delta))} \times \widetilde{S}_{1}^{*}(\Delta)\widetilde{S}_{2}(\Delta)\widetilde{S}_{3}(\Delta)\exp\left\{i\Delta[t\cdot\cos(\delta\varphi_{\tau}(\Delta))-\tau\cdot\cos(\delta\varphi_{\tau}(\Delta))-\tau_{1}\cdot\cos(\delta\varphi_{\tau_{1}}(\Delta))]\right\}g(\Delta)d\Delta,(5)$$

where

 $g(\Delta) = (1/\Delta_D \sqrt{2\pi}) \exp\{-(\Delta/\Delta_D)^2\} \theta_1(\Delta) = \theta_1'(\Delta) \delta_{\tau_1}, \theta_2(\Delta, \delta\varphi_\tau(\Delta)) = \theta_2'(\Delta, \delta\varphi_\tau(\Delta)) \delta_{\tau_2}, \theta_3(\Delta, \delta\varphi_{\tau_1}(\Delta)) = \theta_3'(\Delta, \delta\varphi_{\tau_1}(\Delta)) \delta_{\tau_3}$ are areas of laser pulses, $\tilde{S}_1(\Delta), \tilde{S}_2(\Delta), \tilde{S}_3(\Delta)$ are Fourier spectra of pulses, $\delta\varphi_\tau(\Delta), \delta\varphi_{\tau_1}(\Delta), \delta\varphi_\tau(\Delta)$ is the average random increase in the angle between the velocity of the molecule and the direction of observation during corresponding time interval and the corresponding isochrome of the Doppler-broadened line, Δ_D is the width of the heterogeneously (Doppler) broadened line, τ is the time interval between the first and the second pulses, τ_1 is the time interval between the 1st and 3rd pulses.

Numerical calculation of expression (5) in case of information encoding into temporal form of laser pulse for the SEH response is shown in figure 2 (a, b) in cases where the time intervals have been changed between the 2nd and 3rd excitation laser pulses at different vapor pressures of 174 Yb.



Figure 2. The temporal form of the stimulated photon echo response in the gas: $\tau = 5$ ns, $\tau_1 = 100$ ns, $T_2 = 1000$ ns, $T = 800 \circ \text{K}$, $d = 0.388 \cdot 10^{-7}$ cm, a) P = 1.5 Torr, b) P = 5.25 Torr.

4. Conclusions

Collisions with the change in velocity direction of gas particles lead to the destruction of the temporal form of the SEH response. Therefore, the reproducibility of the information in the SEH response due to collisions with the change in speed will deteriorate, which must be taken into account when creating an optical memory and information processing systems in gas media.

Acknowledgments

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