NONLINEAR INTERACTION OF OPTICAL BIHARMONICS IN A GASEOUS MEDIUM WITHOUT INVERSION CENTERS

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This study investigates observed interactions between second-order optical biharmonics with difference frequencies characteristic of intra-molecular spin motion. The simultaneous solutions of equations of motion for electron shells and nuclear and generalized Maxwell equations for electromagnetic waves are found. It is shown that a complete orthogonal series of solutions for the observed condition are comprised of Bessel \( J_n(z) \) functions. Leading from the properties of these functions it can be proved that bimodal light at a distance \( L \) gives rise, for example, to red and blue side lobes \( \pm L \) of comparable intensities. The results obtained may be useful in finding and developing low energy methods for destroying inversion symmetry in gaseous media.

INTRODUCTION

It is known that the application of optical biharmonics to coherent active Raman spectroscopic (CARS) methods for exciting molecular vibrations on difference frequencies allows for several orders of increase in the level of detected signal relative to spontaneous scattering. In the last few years, this method has also been applied to the study of polaritons, quasiparticles which arise from the interaction of optical phonons with photons having the frequency \( \omega = E_{ph}/\hbar \), where \( E_{ph} \) is the photon energy. Polariton CARS is based on the build up of polaritons pumped by the field created by two relatively high intensity laser beams with frequencies \( \omega_1 \) and \( \omega_2 \) whose differences is equal to the polariton frequency \( \omega_p \). Scattering of the probe wave with frequency \( \omega \) (\( \omega_1 \) or \( \omega_2 \) can be used) results from polaritons that are in phase throughout the volume at the frequency \( \omega_{as} = \omega_1 + \omega_p = 2\omega_1 - \omega_2 \). Since the polaritons exhibit significant dispersion, the efficiency of build up, and consequently the intensity of the CARS signal depends not only on the difference frequency of the excited fields (\( \omega_1 - \omega_2 \)), but also on the difference between the wave vectors of these fields \( \kappa_1 - \kappa_2 \). Observation of the dependence of the CARS signal intensity on the magnitude of phase detuning \( \Delta \kappa_p = \kappa_1 - \kappa_2 - \kappa_p \) (spectroscopy in \( \kappa \)-space) permits the characteristics of polariton quenching in the medium to be measured. A more detailed look at the technology of detecting polariton signals using CARS is carried out in [2]. In particular, that study shows that the efficiency of the observed signal is due to avalanche three-photon processes, characterized by second order nonlinear susceptibility \( \chi^{(2)} \). Since gaseous media exhibit centrosymmetry, the second order processes are forbidden by symmetry rules.

The study of the interaction mechanisms for optical biharmonics (using similar frequencies) with intrinsic particle vibrations in a gaseous medium, introduced in this paper is, in our opinion, extremely useful in determining the possibilities of circumventing this forbiddance.

1. FUNDAMENTAL EQUATIONS

For simplicity, lets consider the case of a diatomic molecule like nitrogen or oxygen. We will consider, as is normally the case, that the vibrational motions of the molecule associated with a displacement of the nucleus from equilibrium by a value \( \rho \), can be considered as vibrations of a particle with reduced mass \( M \) in a potential field \( U(\rho, r) \) which arises from the...
interaction of nuclei with each other and with the surrounding electrons. We will also assume that the nuclear displacement \( \rho \) and the electron shell radius \( r \) are parallel to the external field \( E \) (reduces to vector and tensor indices in the equations), which is composed of two spectral components \( \omega_1 \) and \( \omega_2 \) where the difference between them is a few inverse centimeters or less, i.e. characteristic of spin frequencies.

To account for electron–phonon coupling in the equations of motion for electron shells and nuclei, the molecular potential energy is assumed to take the form of a series of terms which deviate by degrees from the equilibrium values for \( r \) and \( \rho \):

\[
U = U(r, \rho) = U(0, 0) + \cdots + U'_r(0, 0) r^2 + \frac{1}{2} \omega_{srs} r^2 + \cdots,
\]

where the right hand side contains only linear and quadratic terms in \( r \).

Taking (1) into account, equations are proposed for polarization and molecular oscillations of the form [1, 3]

\[
\begin{align*}
\dot{r} + 2 \gamma r + \omega_r^2 r &= \frac{2BM}{Nm} \rho r + \frac{e}{m} E(x, t), \\
\dot{\rho} + 2\Gamma \dot{\rho} + \Omega \rho &= \beta r^2,
\end{align*}
\]

where \( e, m \) are the electron charge and mass; \( M \) is the reduced mass of the molecule; \( \beta = -u_{RR}''(0, 0)/M; \omega_0, \Omega \) are the intrinsic oscillation frequencies of the electron and nucleus, respectively; \( N \) is the electron density; \( \gamma, \Gamma \) are quenching parameters, \( E(x, t) \) is the applied electric field.

We are interested in the case of resonant interaction of bimodal light with intrinsic molecular vibrations induced by polarization oscillations. As demonstrated in [3], if the incident light contains two harmonics \( (\omega_1, \kappa_1) \) and \( (\omega_2, \kappa_2) \), then as a result of nonlinear interaction, polarization is manifested not only on frequencies \( \omega_1 \) and \( \omega_2 \), but spectral components of polarization are also manifested on combined frequencies: low multiples \( \Omega = \omega_1 - \omega_2, \kappa = \kappa_1 - \kappa_2 \), as well as harmonics \( \omega_n = \omega_{1,2} = n\Omega, \kappa_n = \kappa_{1,2} \pm n\kappa \) (\( n \) is an arbitrary integer).

As a result, we assume that the field of the interacting light waves takes the form of superimposed combined harmonics

\[
E(x, t) = \sum_n e_n a_n(x) e^{-i\omega_n t + i\kappa_n x},
\]

where \( e_n, a_n \) are the polarization and amplitude vectors for the light waves; \( x \) is the direction of propagation of the light; \( n \) are indices which can take on either positive or negative integer values.

Naturally, in order to obtain a closed system of equations, it is necessary to supplement the equations of motion (2) and (3) with equations that describe the propagation dynamics of the light. For a complex dielectric medium this takes the well known form [4, 5]

\[
\frac{1}{c^2} \frac{\partial^2 D}{\partial t^2} + \Delta E = 0.
\]

Here \( D \) is the electric induction vector associated with the polarization vector \( E \) and \( P \) through the linear relation

\[
D = \varepsilon E = (1 + 4\pi\alpha(\omega)) E = E + 4\pi P,
\]

where \( \alpha(\omega) \) is the polarizability of the medium.

If we assume that \( \omega_1 = \omega_2 = \omega \) in (6) and we substitute the average interacting wave packet \( \omega \) into the expression for polarizability, after the standard computations we get the form

\[
\alpha(\omega, \rho) = \alpha(\omega) \left[ 1 + \frac{\beta M \rho}{m (\omega^2 - \omega^2)} \right].
\]

Putting (6) and (7) into (5), we get an equation for light waves interacting with optical oscillations in the form

\[
\frac{\partial^2}{\partial t^2} \left[ 1 + 4\pi\alpha(\omega) \left( 1 + \alpha(\omega) \frac{2BM}{Nm \rho} \right) \right] E + e^{i\omega} \Delta E = 0,
\]

1002