Spatial and temporal characteristics of signals representing transient Raman scattering and photon echo under two-photon excitation conditions

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An analysis is made of the correlation characteristics of signals representing transient Raman scattering and Raman photon echo in the case of a two-photon resonance. It is shown that frequency conversion is possible in the case of an arbitrary time profile of a biharmonic pump pulse and that a correlation analysis can be carried out on slow envelopes of the fields which can be of the same or different frequencies. The feasibility of using Raman photon echo signals for the formation and transformation of transient optical images is considered. It is shown that photon echo signals at the anti-Stokes and Stokes frequencies make it possible to form simultaneously real and virtual images and time may be traversed in the forward and mirror-reversed directions.

INTRODUCTION

Nonlinear optical spectroscopy is a relatively young and rapidly developing branch of the physics of interaction of optical radiation with matter.[1] Transient coherent spectroscopy is gaining in popularity[2–4] particularly in studies of electron spectra of atoms and ions in gaseous and solid-state phases. The photon echo, first observed in ruby under one-photon resonance conditions almost 25 years ago,[5] occupies the central position among coherent processes.

The power of coherent light sources used in nonlinear optics has recently increased considerably, making it possible to observe multiphoton resonance effects. The ability to create coherent quantum states by a two-photon resonance has been discussed in the literature sometime ago.[6,7] Considerable progress has been made in theoretical understanding of nonlinear coherent transient processes occurring in the course of a two-photon resonance interaction.[8–19] For example, a lucid vector model, which provides a simple description of the processes of two-photon excitation of a resonant medium, was proposed in Ref. 11 and can be used to describe also the appearance of the photon echo.[12] The conditions for the observation of photon echo and the conditions for spatial phase matching were formulated in Refs. 10, 13, and 16. Coherent transient effects were considered in Refs. 17–19 under conditions of energy level degeneracy.

Undoubtedly the physical understanding has been improved by the results of experimental investigations of transient processes under two-photon excitation conditions.[20–25] The Raman echo in n-type CdS was discovered experimentally in 1976 (Ref. 20). Two years later a two-photon echo was observed in sodium vapor.[21] The Raman echo had subsequently been investigated in atomic thallium vapor[22,23] and also in molecular nitrogen.[24,25] This provided foundation of transient band scattering echo spectroscopy.

The photon echo is known to exhibit a great variety of time characteristics. They include reproduction of a complex temporal profile of a coding pulse along forward and reversed directions on the time scale,[26–32] pair correlations in time, time convolutions of signal envelopes,[33] etc. These characteristics of formation of a complex temporal structure of coherent optical echo signals are due to the Fourier-transforming properties of multilevel resonance systems,[34] which are of practical interest in optical data processing.

Equally important are linear properties of quantum systems capable of generating echo responses in spatial transformations of wavefronts,[35–39] when the amplitude of the response is directly proportional to one of the fields of the exciting pulses. However, up to now these properties have been considered in the literature only in the specific case of one-photon resonance conditions. When two-photon excitation takes place, a one-photon transition between resonant energy levels is usually electric-dipole-forbidden or even completely forbidden. This increases the radiative lifetime and facilitates the appearance of a long-term (slow) stimulated photon echo.

It would therefore be of interest to generalize the results for a one-photon resonance to two-photon resonant interaction processes (two-photon absorption and Raman scattering of light) and identify the conditions under which such nonlinear systems exhibit linear properties in the sense defined above.

The present paper is a theoretical analysis of the temporal and spatial characteristics of the Raman scattering signals and of the Raman photon echo associated with the Fourier-transforming properties of resonant systems in the case of two-photon transitions. We shall investigate in detail the transient radiation emitted at Raman frequencies under the influence of biharmonic pumping and also the photon echo. We shall show that frequency transformation of an arbitrary temporal profile of a coding pulse and its reproduction in the forward and time-reversed directions are possible, and that a correlation analysis of slow envelopes of the amplitudes of the excitation pulses can be carried out. We shall formulate the conditions for utilization of a resonant medium, in which two-photon transitions take place, as spectrally selective holograms for the reconstruction of transient optical images in the forward and time-reversed directions.

1. PRINCIPAL EQUATIONS

We shall consider a resonant medium subjected simultaneously to two ultrashort light pulses (for the sake of simplicity we shall assume that they are of the same duration $\delta_1 = \delta_2$) representing biharmonic pumping:

$$\mathbf{E}(\mathbf{r}, t) = \sum_{\alpha=1}^{2} \mathrm{Re}\{\mathbf{E}_\alpha(\mathbf{r}, t) \exp(i\mathbf{k}_\alpha \mathbf{r} - i\omega_\alpha t)\}. \qquad (1.1)$$

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Here, $\mathbf{E}_\alpha(\mathbf{r},t)$ is a slowly varying amplitude of the electric field of a light wave; ω_α and $\mathbf{k}_\alpha$ are the frequency and the wave vector. We shall consider the specific case when $\omega_0 = \omega_1 \pm \omega_2$ (the plus sign corresponds to two-photon absorption and the minus sign corresponds to Raman scattering) is close to the atomic frequency $\omega_{ba} = (E_b - E_a)/\hbar$ of a forbidden transition.

The interaction of light pulses with a medium will be allowed-for in the dipole approximation, and quantum-mechanical properties of a resonant medium of a system will be described by components of the density matrix. Then, in the interaction representation the main equations become

$$i\hbar\dot{\hat{\rho}} = [\hat{V}_{int}(t), \hat{\rho}], \tag{1.2}$$

where

$$V_{int}^{mn}(t) = \sum_{\omega_\alpha} V_{mn}(\omega_\alpha)\exp(-i\omega_\alpha t + i\omega_{mn}t) \tag{1.3}$$

(the summation is carried out over positive and negative values of ω_α), where

$$V_{mn}(\omega_\alpha) = -{}^1/_2\mathbf{d}_{mn}\mathbf{E}_\alpha(\mathbf{r},\ t)\exp(i\mathbf{k}_\alpha\mathbf{r}), \tag{1.4}$$

and the matrix elements V_{mn} satisfy the relationships $V_{mn}(-\omega_\alpha) = V_{nm}^*(\omega_\alpha)$.

Equation (1.2) is simplified by dropping the terms describing relaxation processes, which implies that $\delta_i < T_2$, where δ_i are the durations of the exciting light pulses and T_2 is the characteristic time governing the homogeneous width of a spectral line.

The initial system of equations (1.2) for the density matrix with the interaction Hamiltonian described by Eqs. (1.3) and (1.4) together with the wave equation for $\mathbf{E}(\mathbf{r},t)$, represents a complete and sufficiently general description of nonlinear dynamics of the parameters of matter (population of energy levels, macroscopic dipole moment) and of optical fields (phase and amplitude). Under some resonance conditions this system usually simplifies: for example, in the case of a one-photon resonance it simplifies to the reduced system of Maxwell-Bloch equations.[2]

A powerful method is available for simplifying the initial system in the case of various resonance interactions and an arbitrary degeneracy of the energy levels: this is the unitary transformation method.[8] This method was used in Ref. 17 in an investigation of the polarization properties of optical pulses interacting with a medium under two-photon resonance conditions and the relevant reduced Maxwell-Bloch equations were derived; these equations describe the interaction with degenerate atomic and molecular energy levels when the degeneracy is due to the different orientations of the total angular momentum of a quantum object. In the present study we shall concentrate entirely on the derivation of the temporal profiles of optical pulses. In this case the polarization properties of the pulses are of secondary importance so that we shall consider the case of a nondegenerate quantum system. This case is often encountered in crystals with paramagnetic impurities if a very specific polarization of optical pulses transmitted by a crystal is selected. It is then simpler and more convenient to apply the familiar averaging method developed by Bogolyubov and Mitropol'skiĭ and then used in Ref. 40 to deal with a resonant interaction of light with matter. According to this method, the steady-state "fast" parts of the density matrix are described by

$$\tilde{\rho}_{mn}(t) = \frac{1}{\hbar}\sum_{k,\omega_\alpha}\left\{\frac{V_{kn}(\omega_\alpha)\rho_{mk}}{(\omega_{kn}-\omega_\alpha)}\exp[i(\omega_{kn}-\omega_\alpha)t] - \frac{V_{mk}(\omega_\alpha)\rho_{kn}}{(\omega_{mk}-\omega_\alpha)}\exp[i(\omega_{mk}-\omega_\alpha)t]\right\}. \tag{1.5}$$

Substituting these expressions on the right-hand side of Eq. (1.2) and averaging in a time interval $\omega_\alpha^{-1} \ll T \ll \delta_i$, we find that the slowly varying parts of the density matrix are described by the equations

$$\dot{\rho}_{ba} = i\Delta\rho_{ba} - iv\exp(i\varphi)n, \tag{1.6}$$
$$\dot{n} = 4\,\mathrm{Im}\,[\rho_{ba}v^*\exp(-i\varphi)].$$

In the system (1.6) the quantity n is the population inversion equal to $\rho_{bb} - \rho_{aa}$; the quantity Δ is the difference between the Stark shifts of the resonant energy levels under the influence of all the fields:

$$\Delta(\mathbf{r},t) = \sum_{\alpha=1}^{2}\frac{\varkappa^{bb}(\omega_\alpha) - \varkappa^{aa}(\omega_\alpha)}{4\hbar}|E_\alpha(\mathbf{r},t)|^2; \tag{1.7}$$

the coefficients $\varkappa^{mn}(\omega_\alpha)$ are components of the polarizability of an atom or an ion which is in the n-th energy state, at a frequency ω_α:

$$\varkappa^{mn}(\omega_\alpha) = \frac{2}{\hbar}\sum_\mu \frac{|d_{m\mu}|^2\omega_{\mu m}}{(\omega_{\mu m}^2 - \omega_\alpha^2)}, \tag{1.8}$$

where v is a matrix element of the energy operator for the average motion:

$$v(\mathbf{r},t) = \frac{\varkappa^{ba}}{4\hbar}E_1(\mathbf{r},t)E_2(\mathbf{r},t), \tag{1.9}$$

where

$$\varkappa^{ba} = \frac{1}{\hbar}\sum_\mu d_{b\mu}d_{\mu a}\left(\frac{1}{\omega_{\mu b}+\omega_1} + \frac{1}{\omega_{\mu b}+\omega_2}\right) = \frac{1}{\hbar}\sum_\mu d_{b\mu}d_{\mu a}\left(\frac{1}{\omega_{\mu a}-\omega_1} + \frac{1}{\omega_{\mu a}-\omega_2}\right) \tag{1.10}$$

represents the second-order polarizability; the phase φ in the phase exponent $\exp(i\varphi)$ occurring in Eq. (1.6) is governed by the detuning ε of the frequency of a two-photon transition and by the wave vector of biharmonic pumping:

$$\varphi = \varepsilon t + \mathbf{K}\mathbf{r},\ \varepsilon = \omega_{ba} - \omega_0,\ \mathbf{K} = \mathbf{k}_1 + \mathbf{k}_2. \tag{1.11}$$

It should be noted that Eq. (1.10) is subject to the conditions

$$\omega_{\mu b} + \omega_1 = \omega_{\mu a} - \omega_{ba} + \omega_1 \approx \omega_{\mu a} - \omega_2,$$
$$\omega_{\mu b} + \omega_2 = \omega_{\mu a} - \omega_{ba} + \omega_2 \approx \omega_{\mu a} - \omega_1.$$

Moreover, Eqs. (1.9)–(1.11) apply to the case of two-photon absorption (in all the subsequent treatment an allowance for the excitation in the Raman scattering case has to be made by the substitutions $\omega_2 \to -\omega_2$, $E_2 \to E_2^*$).

Separating the rapidly oscillating factors of the spatial variables in the density matrix

$$\rho_{ba}(\mathbf{r},\ t)\exp(-i\varphi) = \sigma_{ba}(\mathbf{r},\ t) = \sigma,$$

we find that in the case of the slowly varying part of σ we obtain equations which are fundamental in the theory of a two-photon resonance:

$$\dot{\sigma} + i(\varepsilon - \Delta)\sigma = -ivn,\ \dot{n} = 4\,\mathrm{Im}\,(\sigma v^*). \tag{1.12}$$

The macroscopic dipole moment in a medium is governed by the average value of the dipole moment operator

$$\langle d(\mathbf{r}, t)\rangle = \mathrm{Sp}\,\hat{\rho}\hat{d}_{int}(t) = \sum_{k,n}\rho_{kn}d_{nk}\exp(-i\omega_{kn}t). \quad (1.13)$$

However, since in a two-photon resonance a one-photon transition can be completely forbidden, the best conditions for the observation of a coherent response are obtained when the resonant medium is subjected to a probe field. We shall now determine the macroscopic dipole moment of the medium in the presence of an additional illumination field

$$E(\mathbf{r}, t) = \mathrm{Re}[E_p(\mathbf{r}, t)\exp(i\mathbf{k}_p\mathbf{r} - i\omega_p t].$$

It should be noted that during the action of a biharmonic pump field on a resonant medium the role of the probe field is played directly by the field of frequencies ω_1 and ω_2. In the calculation of the macroscopic dipole moment we have to allow for nonresonance and resonance contributions. We shall give only the final result, omitting cumbersome intermediate steps:

$$P_{\text{н}}(\mathbf{r}, t) = N_0(\varkappa^{aa}(\omega_p)\langle\sigma_{aa}\rangle + \varkappa^{bb}(\omega_p)\langle\sigma_{bb}\rangle) \times \mathrm{Re}\,[E_p(\mathbf{r}, t)\exp(i\mathbf{k}_p\mathbf{r} - i\omega_p t)],$$

$$P(\mathbf{r}, t) = \mathrm{Re}\,\{N_0\varkappa^{ab}(\omega_p)\langle\sigma_{ba}(\mathbf{r}, t)\rangle E_p(\mathbf{r}, t) \quad (1.14)$$

$$\times \exp[i(\mathbf{k}_p + \mathbf{K})\mathbf{r} - i(\omega_p + \omega_0)t] + N_0\varkappa^{ba}(\omega_p)\langle\sigma_{ab}(\mathbf{r}, t)\rangle \times E_p(\mathbf{r}, t)\exp[i(\mathbf{k}_p - \mathbf{K})\mathbf{r} - i(\omega_p - \omega_0)t]\}, \quad (1.15)$$

where the angular brackets on the right-hand side of the expression denote, as usual, averaging over the scatter of the frequencies of the corresponding transitions with a characteristic distribution function $g(\varepsilon)$ describing the profile of an inhomogeneously broadened line $\langle\ldots\rangle = \int(\ldots)g(\varepsilon)d\varepsilon$ and the quantities $\varkappa^{ab}$ represent the second-order polarizability

$$\varkappa^{ab}(\omega) = \frac{1}{\hbar}\sum_{\mu} d_{a\mu}d_{\mu b}\left(\frac{1}{\omega_{\mu a} + \omega} + \frac{1}{\omega_{\mu b} - \omega}\right),$$

which satisfy the following symmetry relationships: $\varkappa^{ba}(-\omega) = \varkappa^{ab}(\omega)^*$. We note that when the frequency ω is equal to ω_1 or ω_2, the polarizability coefficients become identical and equal to the value given by Eq. (1.10): $\varkappa^{ba}(\omega_1) = \varkappa^{ba}(\omega_2) = \varkappa^{ba}$.

It follows from Eqs. (1.14) and (1.15) that the macroscopic dipole moment of nonresonance type appears always at the frequency of the additional illumination field ω_p. The macroscopic dipole moment of resonance type, including that creating a photon echo signal, appears at the Raman frequencies: the Stokes frequency $\omega_s = \omega_p - \omega_0$ and the anti-Stokes frequency $\omega_\alpha = \omega_p + \omega_0$.

We shall solve the system (1.12) by the method of successive approximations, which is equivalent to the case of "small areas" in the theory of a one-photon resonance.[26,32,24] We can easily show[2] that in the case of a two-ptoton resonance the approximation of small areas implies the inequality

$$\left|\int v(\mathbf{r}, t')\,dt'\right| < {}^1/_2, \quad (1.16)$$

where integration is carried out during a biharmonic pump pulse. When the inequality of Eq. (1.16) is satisfied, the first equation in the system (1.6) becomes bilinear:

$$\dot{\sigma} + i(\varepsilon - \Delta)\sigma = iv. \quad (1.17)$$

Since the coefficients that determine the Stark shift of resonant energy levels and the second-order polarizability of Eq. (1.10) are of the same order of magnitude, it follows that when the inequality of Eq. (1.16) is obeyed, we can also ignore the quantity Δ compared with the spectral width of a pulse, i.e., we can assume that also the following inequality is obeyed:

$$\int \Delta(t')\,dt' < 1. \quad (1.18)$$

Under these conditions, when the inequalities of Eqs. (1.16) and (1.18) are obeyed, Eq. (1.17) becomes a linear analog of the corresponding equations from the theory of a one-photon resonance Therefore, many results of this theory can be applied to a two-photon resonance provided we make the substitution $d_{ba}E/2\hbar \rightleftarrows v$ in the formulas. It should be noted that this substitution is valid only in the case of partial lifting of the forbiddeness of a quantum transition dipole approximation, such as that observed in a ruby crystal. Then, instead of one of the biharmonic pump pulses we can use a conventional excitation pulse at a frequency close to a resonance. We shall consider in detail the various special cases in the theory of a two-photon resonance using the approximation corresponding to Eqs. (1.16) and (1.18).

2. TEMPORAL CHARACTERISTICS OF RAMAN SCATTERING SIGNALS UNDER BIHARMONIC PUMPING CONDITIONS

As pointed out already, a biharmonic pump pulse creates quantum incoherence in a resonant medium (near-diagonal elements $\sigma_{ba} = \sigma$ of the density matrix become nonzero). In accordance with Eqs. (1.14) and (1.15), a polarization is excited in the medium and its components at the Stokes frequencies $\omega_{1,2} - \omega_0$ contribute to the field at the frequencies of the excitation fields ω_1 and ω_2 (we shall discuss the specific case of two-photon absorption).

A more important effect is the appearance of a microscopic dipole moment and of an associated field at the anti-Stokes frequencies $\omega_{1,2} + \omega_0$:

$$P(\mathbf{r}, t) = \mathrm{Re}\,\{N_0\varkappa^{ab}(\omega_1)\langle\sigma_{ba}(\mathbf{r}, t)\rangle E_1(\mathbf{r}, t)\exp[i(2\mathbf{k}_1 + \mathbf{k}_2)\mathbf{r} - i(2\omega_1 + \omega_2)t] + N_0\varkappa^{ab}(\omega_2)\langle\sigma_{ba}(\mathbf{r}, t)\rangle E_2(\mathbf{r}, t) \times \exp[i(\mathbf{k}_1 + 2\mathbf{k}_2)\mathbf{r} - i(\omega_1 + 2\omega_2)t]\}. \quad (2.1)$$

In fact, Eq. (1.12) considered in the approximation of Eqs. (1.16) and (1.18) admits a solution for an arbitrary time dependence of the slow amplitudes of the fields at frequencies ω_1 and ω_2:

$$\sigma(t) = i\int_{-\infty}^{\infty}\theta(t - t')\exp[-i(t - t')\varepsilon]v(t')\,dt'. \quad (2.2)$$

and also

$$\langle\sigma(t)\rangle = 2\pi i g(0)v(t), \quad (2.3)$$

where $\theta(t)$ is a unit function. In Eq. (2.3) it is assumed that the duration of a biharmonic pulse exceeds the time T_2^* characterizing an inhomogeneous broadening of a resonance line.

It therefore follows that a resonant medium performs a nonlinear functional transformation of slow envelopes of the fields $E_1(t)$ and $E_2(t)$ characterized by different frequencies

ω_1 and ω_2 into a field at the Raman frequencies $2\omega_1+\omega_2$ and $\omega_1+2\omega_2$ with an envelope which is a product of the type $E_1^2(t)E_2(t)$ or $E_1(t)E_2^2(t)$. In the specific case when the duration of one of the pulsed fields of biharmonic pumping exceeds the duration of the other, for example, if $\delta_1>\delta_2$, the temporal profile of the Raman scattering signal at the frequency $2\omega_1+\omega_2$ repeats the slow envelope of the field amplitude at the frequency ω_2. Consequently, the process of nonlinear resonant interaction of light with matter has some features of linear effect. Similarly, when the opposite inequality $\delta_1<\delta_2$ is obeyed, the temporal profile of the function $E_1(t)$ is manifested in the slow envelope at the Raman scattering frequency $\omega_1+2\omega_2$.

3. TEMPORAL AND CORRELATION CHARACTERISTICS OF RAMAN PHOTON ECHO SIGNALS

We shall now consider the excitation of photon echo signals by a sequence of biharmonic pump pulses. Using the analogy of Eqs. (1.17) with the equations from the theory of a one-Photon resonance,[34] we find that in the case of near-diagonal elements of the density matrix σ at the instants of appearance of a two-pulse photon echo signal we have

$$\langle\sigma(t)\rangle=-i\int g(\varepsilon)V_2^2(\varepsilon)V_1^*(\varepsilon)\exp[-i\varepsilon(t-t_e)]\,d\varepsilon, \qquad (3.1)$$

where τ_e is the instant of appearance of a photon echo, defined by $2\tau+\delta_1+\mathbf{Kr}/\omega_0$; $\mathbf{K}$ is the wave vector defined by a sequence of biharmonic pump pulses: $\mathbf{K}=2\mathbf{K}^{(2)}-\mathbf{K}^{(1)}$, $\mathbf{K}^{(i)}=\mathbf{k}_1^{(i)}\pm\mathbf{k}_2^{(i)}$; $V_i(\varepsilon)$ is the Fourier transform of the time dependence $v_i(t)$ which describes the ith biharmonic pulse

$$V_i(\varepsilon)=\int v_i(t)\exp(i\varepsilon t)\,dt. \qquad (3.2)$$

As in the case of a one-photon resonance, the resonating atoms transform the time dependence of the product of slow envelopes at frequencies ω_1 and ω_2 into a Fourier tramsform, and the Fourier spectrum of a sequence of two biharmonic pulses in a photon echo signal is a product of the Fourier transforms of the individual pulses.

It follows from Eq. (3.1) that in the case of spectrally wide biharmonic excitation pulses, where we can ignore the dependence on the detuning ε in the functions $V_1(\varepsilon)$ and $V_2(\varepsilon)$ (or assume that the inequalities $\delta_i\ll T_2^*$ are satisfied, where $i=1$ or 2), the average value of the near-diagonal element of the density matrix simplifies to

$$\langle\sigma(t)\rangle=-2\pi i V_2^2(0)V_1^*(0)F(t), \qquad (3.3)$$

where

$$F(t)=\int\frac{d\varepsilon}{2\pi}g(\varepsilon)\exp[-i\varepsilon(t-t')]=G(t-t'), \qquad (3.4)$$

$$t'=2\tau+\delta_1+\delta_2+\mathbf{Kr}/\omega_0,$$

and the function $G(t)$ is the Fourier transform of a function $g(\varepsilon)$ representing the distribution of elementary emitters in respect of the detuning within the limits of an inhomogeneously broadened line.

If only the second biharmonic pulse is spectrally wide, the temporal profile $\langle\sigma(t)\rangle$ and the photon echo signal are described by expressions of the type

$$F(t)=\int d\xi G(\xi)v_1^*(\xi-t+t'+\delta_1). \qquad (3.5)$$

However if the characteristic duration of the first coding biharmonic pulse exceeds the characteristic relaxation time T_2^* of the polarization, but is less than the duration of the probe pulse, a mirror-like reproduction of the time profile of the first recording pulse appears in the Raman photon echo signal:

$$\langle\sigma(t)\rangle=-2\pi i g(0)V_2^2(0)v_1^*(t'+\delta_1-t), \qquad (3.6)$$

where the time profile of the coding pulse [one of the factors $E_1(t)$ or $E_2(t)$ occurring in $v(t)$ and representing slow envelopes of the fields at different frequencies ω_1 and ω_2] can be reconstructed at the Stokes and anti-Stokes Raman scattering frequencies.

We can similarly consider the case when a second biharmonic pulse plays the role of the coding pulse. As in the theory of a one-photon resonance,[34] the time profile of the coding pulse is not reproduced in the signal of a two-pulse photon echo, but is governed by an expression which is of the convolution integral type:

$$\langle\sigma(t)\rangle=-2\pi i g(0)V_1^*(0)F(t),$$
$$F(t)=\int v_2(t-t'+\delta_2-\xi)v_2(\xi)\,d\xi. \qquad (3.7)$$

The opportunities for using the photon echo effect in time processing of optical signals can be greatly extended by the use of a three-pulse regime for the excitation of a resonant medium. In calculating the macroscopic dipole moment responsible for the formation of a stimulated photon echo signal in a medium we must bear in mind that the second biharmonic pulse transfers the "coherence" created by the first biharmonic pump pulse to the diagonal elements of the density matrix, i.e., to the population. The quantity $\langle\sigma(t)\rangle$ governing the Raman stimulated photon echo signal at the Stokes and anti-Stokes frequencies can then be described by

$$\langle\sigma(t)\rangle=-2i\int g(\varepsilon)V_3(\varepsilon)V_2(\varepsilon)V_1^*(\varepsilon)\exp[-i\varepsilon(t-t_e)]\,d\varepsilon, \qquad (3.8)$$

where

$$t_e=2\tau_1+\tau_2+2\delta_1+\delta_2+\mathbf{Kr}/\omega_0,$$
$$\mathbf{K}=\mathbf{K}^{(3)}+\mathbf{K}^{(2)}-\mathbf{K}^{(1)},\quad \mathbf{K}^{(i)}=\mathbf{k}_1^{(i)}\pm\mathbf{k}_2^{(i)}.$$

We shall now consider various special cases of the general relationship (3.8). If durations of all three biharmonic pulses are less than the relaxation time T_2^*, then

$$\langle\sigma(t)\rangle=-4\pi i V_1^*(0)V_2(0)V_3(0)F(t),$$
$$F(t)=G(t-t''),\quad t''=\tau_e-\delta_1+\delta_2+\delta_3. \qquad (3.9)$$

However, if in a sequence of three biharmonic pulses, two are of considerable spectral width and the duration of the coding pulse exceeds the time T_2^*, then $\langle\sigma(t)\rangle$ is described by the expression

$$\langle\sigma(t)\rangle=-4\pi i g(0)V_i^*(0)V_k(0)F_l(t), \qquad (3.10)$$

where the indices i, k, and l satisfies the cyclic transposition conditions, and the function $F_l(t)$ represents possible temporal profiles stimulated photon echo signals:

$$F_i(t)=v_3(t-t''+\delta_3),$$
$$F_l(t)=v_2(t-t''), \qquad (3.11)$$
$$F_l(t)=v_1^*(t''-\delta_1-t).$$

Finally, when the spectral width of just one of the biharmonic pump pulses exceeds the spectral width of a line and the inequality $\delta_i > T_2^*$ is obeyed by the two biharmonic pulses, a stimulated photon echo signal can be subjected to a correlation analysis and the time profile of the photon echo envelope is described by one of the functions

$$\begin{aligned} F(t) &= \int d\xi v_2(\xi) v_3(t-t''+\delta_3-\xi), \\ F(t) &= \int d\xi v_3(\xi) v_1^*(\xi+t''-\delta_1-t), \\ F(t) &= \int d\xi v_2(\xi) v_1^*(\xi+t''-\delta_1-t). \end{aligned} \tag{3.12}$$

Therefore, in the case of stimulated Raman photon echo signals it is found, as in a one-photon resonance, that a correlation analysis is possible in the case of envelope fields of the same frequency and of different frequencies when these fields satisfy the condition for a two-photon resonance. In this sense the potential applications of the stimulated photon echo in temporal processing of optical signals are greater in the case of two-photon excitation. In contrast to three-level systems, the correlation function due to different-frequency envelope fields does not contain a characteristic scale factor.[34]

4. DYNAMIC FORMATION AND TRANSFORMATION OF OPTICAL IMAGES, PERFORMED BY RAMAN PHOTON ECHO SIGNALS

Manifestation of linear properties in the case of nonlinear interactions of biharmonic light pulses with a resonant medium can be used in the formation and transformation of dynamic optical images by Raman photon echo signals under two-photon excitation conditions. A resonant medium then acts as a characteristic phase hologram, which has selective properties in respect of the Fourier spectrum of coding light pulses. However, the situation is significantly different from conventional holography because there is no need for a time overlap of optical pulses in order to create an interference pattern, in view of the coherent nature of the interaction of light pulses with a resonant medium and due to the phase memory properties.

We shall consider in greater detail the formation and transformation of transient optical images which can be performed by Raman photon echo signals. Let us assume that a plane optical wave with an electric field of amplitude E_0 illuminates a plane transparency T with an amplitude transmission $T(\mathbf{r},t)$, located in the $z = 0$ plane ($\mathbf{r}$ is the radius vector at right-angles to the z axis). A resonant medium of short length l is located at a distance L from the transparency and it acts as a dynamic hologram The field in the region of the hologram can be determined using the parabolic diffraction theory equation,[41] the solution of which can be represented conveniently by a Green function

$$E(\mathbf{r}, z, \tau) = E_0 \int G(\mathbf{r}-\mathbf{r}', z) T(\mathbf{r}', \tau) d\mathbf{r}', \tag{4.1}$$

where

$$G(\mathbf{r}-\mathbf{r}', z) = -\frac{ik\theta(z)}{2\pi z} \exp\left[\frac{ik(\mathbf{r}-\mathbf{r}')^2}{2z}\right]. \tag{4.2}$$

In the last expression the symbol $\theta(z)$ denotes a unit function, k is the wave number of a coding light pulse, and the time $\tau = t - L/c$ (c is the velocity of light) demonstrates that it is possible to consider transient object scenes.

We shall find it convenient to apply to the Green function, the "impulse representation," which gives

$$E(\mathbf{r}, L, \tau) = E_0 \int \frac{d\mathbf{q}}{(2\pi)^2} T_F(\mathbf{q}, \tau) \exp\left(i\mathbf{q}\mathbf{r} - i\frac{\mathbf{q}^2 L}{2k}\right), \tag{4.3}$$

$$T_F(\mathbf{q}, \tau) = \int d\mathbf{r}' T(\mathbf{r}', \tau) \exp(-i\mathbf{q}\mathbf{r}'). \tag{4.4}$$

Similarly the amplitude of the electric field of the signal representing the Raman photon echo with an arbitrary coordinate z can be described by the expression

$$E^e(\mathbf{r}, z, \tau) = \int \frac{d\mathbf{q}}{(2\pi)^2} E_F^e(\mathbf{q}, \tau) \exp\left(i\mathbf{q}\mathbf{r} - i\frac{(z-L)\mathbf{q}^2}{2k_\alpha}\right), \tag{4.5}$$

where $E_F^e(\mathbf{q},\tau)$ is the Fourier transform of the photon echo signal at $z = L$ the exit from a resonant medium (we shall assume that the thickness of the resonance layer of the medium is considerably less than the characteristic distance L) and the wave number k_α determines the photon echo signal at the Stokes and anti-Stokes frequencies.

We shall ignore the reaction of the medium on the biharmonic pump field and assume that the inequality $\alpha_2(E_1,E_2) < 1$ is obeyed, where $\alpha_2(E_1,E_2)$ is the two-photon absorption coefficient dependent on the amplitudes of the fields E_1 and E_2. In the case of small thicknesses of the resonant medium we shall also ignore the diffraction of light inside the resonant medium on the assumption that $(\max k)a_r^2/l < 1$, where a_r are the characteristic dimensions of the resonant medium and $(\max k)$ is the maximum value of all the possible wave numbers, including that of an additional illumination wave. Under these conditions the amplitude of the electric field of an echo signal at the exit from the resonant medium can be represented by

$$E_{s,a}(\mathbf{r}, l, \tau) = i\frac{2\pi\omega_{s,a}}{cn} \langle P_{s,a}(\mathbf{r}, \tau)\rangle l, \tag{4.6}$$

where n is the refractive index of the nonresonance type and $\langle P_{s,a}\rangle$ are the slow amplitudes of the polarization in a medium at the Stokes and anti-Stokes frequencies:

$$\begin{aligned} \langle P_s\rangle &= N_0 \varkappa^{ba}(\omega_p) \langle \sigma_{ab}(\mathbf{r}, \tau)\rangle E_p, \\ \langle P_a\rangle &= N_0 \varkappa^{ab}(\omega_p) \langle \sigma_{ba}(\mathbf{r}, \tau)\rangle E_p, \end{aligned} \tag{4.7}$$

and E_p is the amplitude of the electric field of the additional illumination wave (it is assumed that the duration of this wave exceeds the duration of the photon echo response).

In the case most interesting, from the practical point of view, the dependence $\langle \sigma_{ba}(\tau)\rangle$ reproduces in the forward or time-reversed direction the time structure of the coding pulse. It follows from the results in §3 that physically this situation occurs in a resonant medium when it is excited with two spectrally wide biharmonic light pulses and in this case the spectral width of the coding pulse is less than the inhomogeneous line width. Then, in the amplitude of the field of the Stokes and anti-Stokes wave of an echo response at the exit from a resonant medium is described by

$$E_s = \gamma_s E^*(\mathbf{r}, \tau-\tau_e), \quad E_a = \gamma_a E(\mathbf{r}, \tau-\tau_e), \tag{4.8}$$

where $\gamma_{s,a}$ are the coefficients dependent on the parameters of the resonant medium and of the interacting light pulses:

$$\gamma_{s,a} = (2\pi^2\omega_{s,a} N_0 \varkappa_{s,a}^{ba}(\omega_p) \varkappa^{ba} g(0) l E_p E'/cn\hbar) \Theta_1^* \Theta_3. \tag{4.9}$$

In the last expression the quantity Θ_i represents

the "areas" under biharmonic excitation pulses [$\Theta_i = \int v_i(t)dt$], which are plane waves, whereas E_p and E' are the amplitudes of the probe field and of a field additional to the coding pulse. It is assumed that the temporal profile of the coding pulse is "built into" the second biharmonic pulse.

It should be pointed out that the relationships described by Eq. (4.8) apply to the case of two-photon absorption. Under Raman scattering conditions the complex-conjugate amplitude of the coding pulse will determine the field of the photon echo signal at the anti-Stokes frequency.

Using Eqs. (4.3)–(4.5) and (4.8), we obtain the Fourier component of the response field at the exit from the resonant medium:

$$E_F{}^s(\mathbf{q}, \tau) = \gamma_s T_F{}^*(-\mathbf{q}, \tau-\tau_e) \exp(i\mathbf{q}^2 L/2k) E_0{}^*,$$
$$E_F{}^a(\mathbf{q}, \tau) = \gamma_a T_F(\mathbf{q}, \tau-\tau_e) \exp(-i\mathbf{q}^2 L/2k) E_0. \quad (4.10)$$

We can now readily obtain the conditions for the appearance of clear images:

$$-\frac{(z-L)}{k_s} + \frac{L}{k} = 0, \quad -\frac{(z-L)}{k_a} - \frac{L}{k} = 0,$$

the corresponding condition for the coordinates $z_{s,a}$

$$z_s = L(1+k_s/k), \quad z_a = L(1-k_a/k). \quad (4.11)$$

The distribution of the field in the $z_{s,a}$ plane is then

$$E_s(\mathbf{r}, z_s) = \gamma_s E_0{}^* T^*(\mathbf{r}, \tau-\tau_e),$$
$$E_a(\mathbf{r}, z_a) = \gamma_a E_0 T(\mathbf{r}, \tau-\tau_e). \quad (4.12)$$

It follows thus that at the instant of appearance of the photon echo signal in the case of two-photon excitation we can expect formation of optical images at wavelengths corresponding to the Stokes and anti-Stokes Raman scattering signals. We can see from the relationships in Eq. (4.12) that the planes of the real and virtual images formed by the Stokes and anti-Stokes waves are located on opposite sides of the resonant layer at $z = L$. When a resonant medium is excited so as to cause Raman scattering, the Raman photon echo signals at the Stokes and anti-Stokes frequencies are interchanged and a real image appears at the anti-Stokes frequency. If the time profile of a coding pulse is included in the first biharmonic pulse, the Raman photon echo signals at the anti-Stokes and Stokes frequencies form real and virtual images, and the time profile of each of the photon echo signals is mirror-reversed.

Therefore, Raman photon echo signals can be used to form simultaneously transient images one of which is real and the other virtual, and time can be traversed in the forward and mirror-reversed directions.

The various temporal and spatial characteristics of the Raman photon echo signals considered above are related, as in the case of a one-photon resonance, to the Fourier-transforming properties of resonant systems. However, in the case of two-photon excitation the possibility of their manifestation is higher, as indicated by the conditions for spatial phase matching and various transformations in the time domain. Consequently, depending on the relationships between the amplitudes of biharmonic pump pulses, their durations, and the time representing the processes of reversible and irreversible relaxation of the polarization, the Raman photon echo signals can exhibit various time dependences. Moreover, the linear Fourier-transforming properties of resonant systems manifested in the spatial frequency range may have various applications in dynamic holography.

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