Dependence of the four-wave mixing signal on the spectrum of excited states in GaAs/AlGaAs multiple quantum wells

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The selective excitation of different groups of states in GaAs/AlGaAs multiple quantum wells has been used to study the dynamics of the four-wave mixing signal in the heavy-exciton spectral band as a function of the spectrum of excited states. A notable shortening of the coherent state lifetime has been detected under conditions when coherent wave packets are excited. The main features of the recorded correlation curves can be interpreted in terms of a density-matrix formalism based on the model of multilevel noninteracting quantum oscillators. It follows from the comparison between experimental data and theoretical calculations that, in particular, when the spectral width of coherently excited states increases, the lifetime of coherent states is largely controlled by the destructive interference, rather than scattering. © *1996 American Institute of Physics.* [S1063-7761(96)02210-X]

1. INTRODUCTION

The recent progress in (sub)picosecond laser techniques has led to a further development of coherent laser spectroscopy, in which the dominant position is occupied by degenerate four-wave mixing spectroscopy.[1] Degenerate four-wave mixing is used as an efficient tool for studying various scattering mechanisms in solids, since they control the decay of coherent motion in optical excitations, hence the phase relaxation times of these states.[2]

The existence of many quantum interference effects, however, indicates that the spectrum of excited states may essentially determine the optical response of a solid. For example, the decay of the induced polarization **P** in real time t after excitation of a solid by an optical pulse is purely exponential (with a dephasing time constant T_2) only when a homogeneously broadened transition is excited.[3,4] In the presence of inhomogeneous broadening of the spectral line, the polarization decay may be faster owing to destructive interference between excited states with slightly different energies.[4,5] When two states with close energies are excited, the interference may manifest itself as an oscillation superposed on the polarization decay curve,[6] and if continuum states are excited, the lifetime of the coherent response may be shortened considerably. In the latter case, a wave packet, i.e., a coherent superposition of many excited states, is generated, and the fast decay of the macroscopic polarization in this packet may be due to the destructive interference between bound and dissociated exciton states. Previously the dynamics of the four-wave mixing signal due to the coherent excitation of wave packets was studied in Ge (Ref. 7) and in InGaAs/GaAs multiple quantum wells.[8] Whether the states included in the theoretical model of the latter work[8] are the same as those observed in the experiment seems somewhat dubious because the optical signal was recorded without spectral resolution. At the same time, in many experiments using femtosecond laser pulses the effect of wave packets, which probably takes place because the spectral width is larger than the exciton binding energy, was not taken into account in the analysis of experimental results.[9] The aim of the present work was to study both theoretically and experimentally the four-wave mixing signal in the spectral range around the heavy-exciton ground state in GaAs/AlGaAs multiple quantum wells as a function of the shape of the energy spectrum of excited states.

In our experiment, we have studied the dynamics of the four-wave mixing signal depending on the spectrum of excited states by selectively exciting different groups of states in the same sample containing GaAs/AlGaAs multiple quantum wells. The comparison of experimental data with theoretical calculations has yielded parameters of excited exciton states, namely the energy difference between the levels of light and heavy excitons, the phase relaxation time of quasiparticles, and the magnitude of the heavy-exciton inhomogeneous broadening. We have demonstrated that when the spectrum of excited states is sufficiently broad, the shortening of the lifetime of coherent states is controlled by the destructive interference. The calculation of the output signal due to excitation of continuum states based on the density-matrix formalism has revealed some features not detected in the analysis based on the semiconductor Bloch equations.[7]

2. EXPERIMENTAL TECHNIQUE AND RESULTS

A diagram of the experimental facility measuring the dynamics of induced polarization by the four-wave mixing technique is given in Fig. 1. The pumping beam E generated by a tunable titanium-sapphire laser (pulse width 150 fs, spectral width ~30 meV, repetition rate 70 MHz) is divided into two beams having an approximately equal intensity of 0.1 to 1 W by a beam splitter BS. These beams are focused on the sample surface into a spot with a diameter of less than

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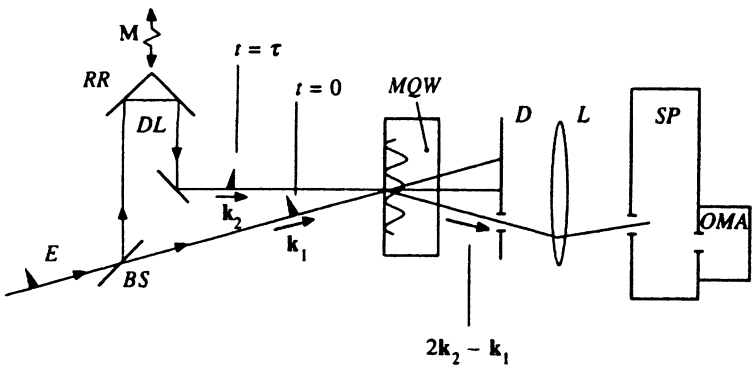

FIG. 1. Diagram of the facility for measuring induced polarization by the four-wave mixing technique.

100 μm, which corresponds to a carrier density of 10^8 to 10^{10} cm^{-2}. One of the pulses propagates along the wave vector $\mathbf{k}_1$; the other one, propagating along the vector $\mathbf{k}_2$, is delayed by a delay line DL containing a retroreflector RR and a micrometer screw M so that it should be delayed with respect to the first pulse by the time τ. The diffracted light propagating along the wave vector $2\mathbf{k}_2-\mathbf{k}_1$ is selected by a diaphragm D, focused by a lens L on the input slit of the spectrometer SP with a resolution of 0.1 meV, and detected by an OMA-1V multichannel optical analyzer in the spectral range around the heavy-exciton line.

In our experiments we used samples containing twenty GaAs quantum wells with a nominal thickness of 116 Å separated by $Al_{0.3}Ga_{0.7}As$ barriers with a thickness of 150 Å. The samples were grown on (100) GaAs substrates, which were subsequently eliminated by selective etching. The sample was placed inside an optical cryostat in helium vapor at a temperature of 5 K. The absorption spectrum of multiple quantum wells is shown by the solid line in Fig. 2. The two peaks centered at 1.5597 eV and 1.5687 eV are due to the $n=1$ states of heavy and light excitons, respectively. The step near 1.572 eV is due to the excited $n=2$ level of the heavy exciton overlapping with higher bound states of the exciton and with the continuum of dissociated states. The half-widths of the heavy and light exciton lines are 1.2 and 1.5 eV, respectively, mostly due to local fluctuations in the well width of one to two atomic layers around its nominal 116 Å width.[10]

The contributions of different bound and dissociated exciton states to the induced polarization were changed by tuning the laser spectral maximum $\hbar\omega_L$. For example, at $\hbar\omega_L=1.545$ eV (curve *1* in Fig. 2) only the lowest $1s$-state of the exciton with the heavy hole is excited, whereas at $\hbar\omega_L=1.555$ eV (curve *2* in Fig. 2) $1s$-states of excitons with both light and heavy holes are generated. If the spectral maximum is tuned to $\hbar\omega_L=1.590$ eV (curve *3* in Fig. 2), mostly continuum states are excited.

One can see in Fig. 3 (curve *1′*) that when the ground $1s$-state of the exciton containing a heavy hole is excited, the curve of the diffracted light intensity I versus delay time τ (correlation curve) has a maximum at $\tau=1$ ps and then drops exponentially with a time constant of about 40 ps. When $1s$-states of excitons containing light and heavy holes are generated simultaneously, the correlation curve is radically different: the intensity has a maximum at $\tau=0$ and falls exponentially with the delay with a time constant of about 4 ps and also has oscillations with a period of about 450 fs (curve *2′*). When states of the continuum are generated, the correlation curve sharply drops with a time constant of less than 0.2 ps (curve *3′*).

3. DISCUSSION OF RESULTS IN TERMS OF MULTILEVEL NONINTERACTING SYSTEMS

Figure 3 clearly indicates that the shape of the correlation curve strongly depends on which bound and dissociated exciton states contribute to the induced macroscopic polar-

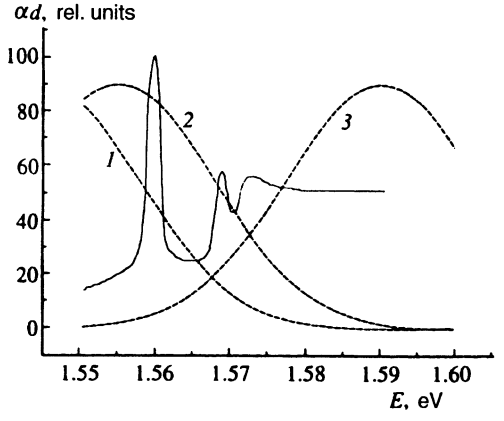

FIG. 2. Absorption spectrum of the sample (solid curve) and spectrum of laser pulses superposed on the exciton spectrum (dashed curves *1–3*).

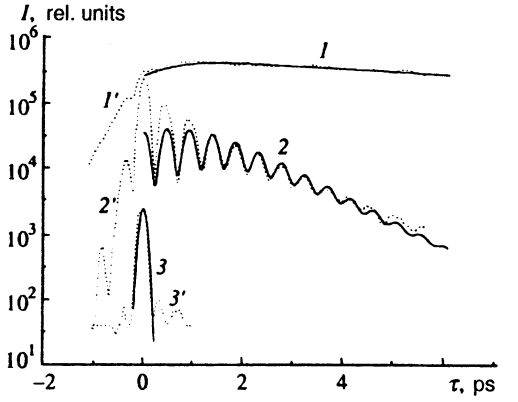

FIG. 3. (*1′–3′*) Experimental and (*1–3*) theoretical correlation curves for laser pulses whose spectra are given in Fig. 2.

ization. When a system with two homogeneously broadened levels is excited by laser pulses, the correlation curve is purely exponential with a time constant equal to $T_2/2$.[3,4] The curves given in Fig. 3, including curve *1'*, which corresponds to the generation of an exciton with a heavy hole, have features not observable on the curves for a two-level homogeneously broadened system. In particular, the maximum on curve *1'* is shifted in the direction of positive delays, curve *2'* has oscillations, and curve *3'*, which corresponds to the excitation of a wave packet, has a very narrow trailing edge.

As we will demonstrate below, the features of the curves of the diffracted light intensity versus delay time given in Fig. 3 can be interpreted in terms of the model of noninteracting multilevel quantum oscillators using the density-matrix formalism operator. The ground state will be denoted by the index a, the levels of the heavy and light excitons by the indices b and c, respectively, and the quasi-continuum states by the index s.

In our analysis of the correlation curve corresponding to the generation of the exciton with heavy hole, we have used the results of Yajima and Taira,[4] who considered excitation of a two-level system by optical δ-pulses. They demonstrated that in the case of inhomogeneously broadened levels the destructive interference between excited states with close energies leads to a faster decay of polarization than in the case of a two-level homogeneously broadened system, and the delay of the peak on the correlation curve is a consequence of the photon echo detected in time-resolved signals. At this point we should note that the exciton peak position in the sample is not constant owing to inevitable fluctuations in the width of the quantum well with an amplitude of one to two atomic layers. Consequently, an inhomogeneously broadened band, which can be described by a Lorentzian-Gaussian curve, is formed.[11] The shape of the correlation curve in this case is described by the function[4]

$$I(\tau)\propto\exp[-4\tau/T_2^{ba}]\mathrm{Erfc}[\sqrt{2}/(\Gamma_{hhx}T_2^{ba})-\Gamma_{hhx}\tau/\sqrt{2}], \tag{1}$$

where $\mathrm{Erfc}(X)=(2/\sqrt{\pi})\int_X^\infty\exp(-t^2)dt$, T_2^{ba} is the phase relaxation time of the excitons and Γ_{hhx} is the inhomogeneous broadening parameter of the heavy-exciton level.

The best fit of the theoretical formula (1) to experimental data is achieved with $\Gamma_{hhx}=0.73$ meV and $T_2^{ab}=40$ ps (curve *1* in Fig. 3). These values are in a good agreement with the half-width γ of the heavy-exciton line (1.2 meV) in the linear absorption spectrum ($\gamma=1.665\Gamma_{hhx}$).

It is known that when two oscillators with slightly different energies are excited in a system, the effect of interference between them may appear as an oscillation superposed on either polarization decay[6] or correlation curves.[6,12] The oscillations on the correlation curve *2'* in Fig. 3, corresponding to the case of a three-level system, are true quantum beats. A theoretical analysis of the four-wave mixing signal in the case of simultaneous coherent excitation of two oscillators with due account of the inhomogeneous broadening was undertaken by Cundiff[13] and Erland *et al.*[14] A general approach to the correlation between resonance shifts due to the inhomogeneous broadening was proposed by Cundiff,[13] but the agreement with experimental data was better when the case of full correlation was considered. The one-to-one correspondence between the shifts of the resonances in the case of full correlation[14] was described in the linear approximation:

$$\omega_{ca}-\omega_{ca}^0=\theta(\omega_{ba}-\omega_{ba}^0).$$

Here $\hbar\omega_{ca}$ and $\hbar\omega_{ba}$ are the local transition energies to the light and heavy exciton state, respectively, at a site in a quantum well with a given thickness, $\hbar\omega_{ca}^0$ and $\hbar\omega_{ba}^0$ are the same transition energies corresponding to the nominal quantum well thickness, θ is a constant. One can easily prove that this approach implies the light-exciton level broadening with the parameter $\theta\Gamma_{hhx}$. In the approximation that the matrix elements of the dipole moment for exciton transitions are equal, we have obtained the following formula for the correlation curve in this case:

$$I(\tau)\propto A^2(\tau)+B^2(\tau)+2A(\tau)B(\tau)\cos(\Delta\omega\tau), \tag{2}$$

$$A(\tau)=\exp\left(-\frac{2\tau}{T_2^{ba}}\right)\mathrm{Erfc}\left(\frac{1}{T_2^{ba}\Gamma_{hhx}}-\frac{\Gamma_{hhx}\tau}{2}\right),$$

$$B(\tau)=0.5\exp\left[-\left(\frac{\theta}{T_2^{ba}}+\frac{1}{T_2^{ca}}\right)\tau\right]\mathrm{Erfc}\left(\frac{1}{T_2^{ba}\Gamma_{hhx}}-\frac{\Gamma_{hhx}\tau\theta}{2}\right).$$

Here T_2^{ba} is the phase relaxation time of light excitons, and $\hbar\Delta\omega$ is the energy difference between the central components of inhomogeneously broadened exciton levels. If we take the parameters of the exciton level broadening derived from the half-widths of the exciton absorption lines, namely 1.2 and 1.5 meV (in this case $\theta=1.25$), the best fit of the theoretical curve *2* in Fig. 3 to the experimental data (curve *2'* in Fig. 3) is achieved for $T_2^{ba}=4.7$ ps, $T_2^{ca}=1.7$ ps, and $\Delta\omega=13.5\cdot10^{12}$ s^{-1}. The notable discrepancy between the experimental and theoretical curves around $\tau=0$ is caused by the overlap of optical pulses, which was not taken into account in deriving Eq. (2) for the correlation curve. The larger damping parameter $1/T_2^{ba}$ when the peak of the laser spectrum is shifted from the position 1 to 2 (Fig. 2) is caused by the effect of the exciton density, which is considerably higher in the latter case. The energy difference $\hbar\Delta\omega=8.95$ meV derived from the quantum beat period $T_{\mathrm{beat}}=465$ fs agrees with the energy difference of 9 meV between the $1s$-states of the light and heavy excitons detected in linear absorption spectra.

The very narrow trailing edge of the correlation curve *3'* in Fig. 3 detected when wave packets are generated in a GaAs/AlGaAs multiple quantum well indicates a sharp drop in the lifetime of coherent states, which is in an agreement with experimental data on Ge (Ref. 7) and InGaAs/GaAs multiple quantum wells.[8] In our analysis of the correlation curve in the case of packet generation we have used Eq. (A2) derived in the Appendix, which takes into account the overlap of laser pulses. All the results concerning this case have been obtained by numerical integration.

Equation (A1) indicates that the signal amplitude may be affected by the phase relaxation times of the heavy exciton

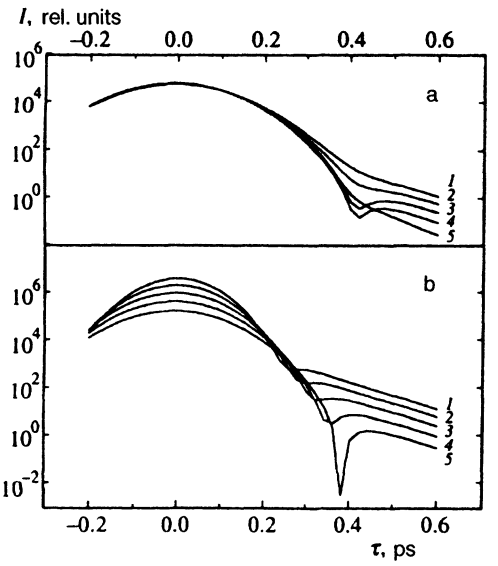

FIG. 4. (a) Shapes of correlation curves calculated for different values of the parameter $\Delta\omega_{\text{cont}}$: (*1*) 30; (*2*) 33.3; (*3*) 36.6; (*4*) 39.9; (*5*) 43.2 meV ($t_L=0.11$ ps); (b) for different laser pulse durations t_L: (*1*) 0.10; (*2*) 0.11; (*3*) 0.12; (*4*) 0.13; (*5*) 0.14 ps ($\Delta\omega_{\text{cont}}=40$ meV). The rest of the parameters used in the calculations are $T_2^{ba}=4$ ps, $\Gamma_{hhx}=0.7$ meV, $T=0.4$ ps, $\omega_L-\omega_{ba}=-32$ meV.

and continuum states, the inhomogeneous broadening parameter for heavy excitons, the position ω_L of the laser spectrum peak in the continuum, and the "effective width" $\Delta\omega_{\text{cont}}$ of the continuum states. The latter parameter was intended to take account of the drop in the dephasing time with the energy in the continuum, which leads to a decrease in the contribution of higher states to coherent radiation. From the formal viewpoint, this means that the summation in Eq. (A1) is performed only over the quasi-continuum states within the interval $\Delta\omega_{\text{cont}}$.

Typical shapes of correlation curves calculated for different effective widths of the continuum spectrum are shown in Fig. 4a. The graph indicates that at a constant laser pulse duration the shapes of the curves around the maximum weakly depend on the parameter $\Delta\omega_{\text{cont}}$, whereas the decay rate of the polarization slightly increases with this parameter. Curve *4* indicates that there is a rather narrow interval (with a width of about 2 meV) of the parameter $\Delta\omega_{\text{cont}}$ in which an additional maximum should exist. Our calculations show that this interval should shift with the peak laser frequency, so that it could be detected in experiment only when the laser is tuned to a certain band within the continuum. This effect could be used, in principle, to determine the parameter $\Delta\omega_{\text{cont}}$. The amplitude of the second maximum, however, is several orders of magnitude lower than that of the main peak, which makes its detection quite difficult. In particular, in our measurements of the correlation curves the second peak amplitude would be considerably lower than the noise level.

Our calculations indicate that variations in the phase relaxation time T of the continuum and that of heavy exciton states, T_2^{ab}, as well as the inhomogeneous broadening parameter Γ_{hhx} within reasonable limits (namely, T is varied between 0.2 and 0.8 ps, T_2^{ba} from 1 to 5 ps, and Γ_{hhx} between 0.6 and 0.9 meV, which corresponds to changing the exciton line half-width from 1.0 to 1.5 meV) do not significantly change the shape of the correlation curve *4* given in Fig. 4a.

At a fixed parameter $\Delta\omega_{\text{cont}}$, the calculations of the peaks on the correlation curves and of the polarization drop rates increase with the laser pulse duration (Fig. 4b). A comparison between Figs. 4a and 4b demonstrates that most changes in the shapes of correlation curves are determined by the continuum states within the spectral band of the laser light, whose width is inversely proportional to its duration. Figure 4b also demonstrates that the decay rate of the polarization component due to the heavy-exciton state increases with the spectral width of laser light, i.e., the lifetime of appropriate coherent states drops with the spectral width of coherently excited continuum states. In our opinion, the insensitivity of the shapes of calculated correlation curves to changes in the dephasing parameter T_2^{ba} and the drop in the lifetime of coherent states with the width of the spectrum of coherently excited continuum states are clear indications of the dominant role of destructive interference between the polarizations of the bound $1s$-state and dissociated exciton states in the observed shortening of the lifetimes of coherent states. Our conclusion concerning the effect of the destructive interference on the lifetime of coherent states generated under conditions when wave packets are excited is confirmed by the results of the theoretical study of a time-integrated four-wave mixing signal in InGaAs/GaAs multiple quantum wells[8] and by experimental and theoretical investigations of the signal from Ge recorded with spectral resolution.[7]

The comparison between the experimental curve *3'* in Fig. 3 and the theoretical curve *3* yields $t_L=110$ fs corresponding to the laser pulse FWHM $t_L^*=190$ fs ($t_L^*=1.665t_L$). This result is in satisfactory agreement with the pulse duration of 150 fs derived from autocorrelation measurements.

4. CONCLUSION

We have concluded from our experimental data and confirmed by calculations based on the Bloch optical equations that the dynamics of the four-wave mixing signal essentially depends on the nature of the spectrum of excited states. The spectral broadening changes the contributions of the effects responsible for shortening of lifetimes of coherent states, namely the processes of scattering and destructive interference. Our experimental data can be satisfactorily interpreted in terms of the model of noninteracting multilevel systems, and the exciton parameters obtained by fitting theoretical calculations to measurements are in agreement with those derived from absorption spectra of multiple quantum wells. Thus our results indicate that time-resolved four-wave mixing measurements with (sub)picosecond laser pulses should be interpreted very cautiously when partial excitation of continuum states is unavoidable.

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APPENDIX

The response to the laser excitation of continuum states has been calculated using the following system of equations for the density operator components:

$$\dot{\rho}_{aa}=-\frac{i}{\hbar}\left[\rho_{ba}V_{ab}-\rho_{ab}V_{ba}+\rho_{ca}V_{ac}-\rho_{ac}V_{ca}+\sum_s(\rho_{sa}V_{as}-\rho_{as}V_{sa})\right],$$

$$\dot{\rho}_{bb}=-\frac{i}{\hbar}(\rho_{ab}V_{ba}-\rho_{ba}V_{ab}),$$

$$\dot{\rho}_{cc}=-\frac{i}{\hbar}(\rho_{ac}V_{ca}-\rho_{ca}V_{ac}),$$

$$\dot{\rho}_{ss}=-\frac{i}{\hbar}(\rho_{as}V_{sa}-\rho_{sa}V_{as}),$$

$$\dot{\rho}_{ba}=-\left(i\omega_{ba}+\frac{1}{T_2^{ba}}\right)\rho_{ba}-\frac{i}{\hbar}(\rho_{aa}-\rho_{bb})V_{ba},$$

$$\dot{\rho}_{ca}=-\left(i\omega_{ca}+\frac{1}{T_2^{ca}}\right)\rho_{ca}-\frac{i}{\hbar}(\rho_{aa}-\rho_{cc})V_{ca},$$

$$\dot{\rho}_{sa}=-\left(i\omega_{sa}+\frac{1}{T}\right)\rho_{sa}-\frac{i}{\hbar}(\rho_{aa}-\rho_{ss})V_{sa}.$$

Here $V_{\alpha\beta}$ is the matrix element of the interaction Hamiltonian with an electromagnetic field between the states α and β, and T is the phase relaxation time in the continuum.

The third-order time-dependent macroscopic polarization derived from these equations by the common iteration technique contains several resonant terms. But in accordance with our experimental conditions, we retained in our calculations only those corresponding to the spectral band around the heavy exciton state:

$$\hat{P}^{(3)}(\omega_{ba},t,\tau)\propto\exp\left[-\left(i\Delta\omega_1+\frac{1}{T_2^{ba}}\right)t\right]\times\exp\left(\frac{\tau}{T_2^{ba}}\right)\int_{-\infty}^{t}dt'\int_{-\infty}^{t'}dt''\int_{-\infty}^{t''}dt'''\times\left\{E_2(t')\exp(i\Delta\omega_1 t')\left[E_1(t'')\times\exp\left(-\frac{t''}{T}\right)E_2(t''')\exp\left(\frac{t'''}{T}\right)\times\sum_s\exp(i\Delta\omega_s(t'''-t''))+E_2(t'')\times\exp\left(-\frac{t''}{T}\right)E_1(t''')\exp\left(\frac{t'''}{T}\right)\times\sum_s\exp(i\Delta\omega_s(t''-t'''))\right]\right\}.$$

Here $\hat{P}^{(3)}(\omega_{ba},t,\tau)$ is the macroscopic polarization complex amplitude, and we have written $\Delta\omega_1=\omega_{ba}-\omega_L$, $\Delta\omega_s=\omega_s-\omega_L$; $E_1(t)$ and $E_2(t)$ are the amplitudes of the exciting electromagnetic fields of the laser pulses 1 and 2, defined by the equations

$$E_1(t)\propto\exp\left(-\frac{t^2}{t_L^2}\right),\quad E_2(t)\propto\exp\left(-\frac{(t-\tau)^2}{t_L^2}\right).$$

The inhomogeneous broadening is taken into account through integration with respect to frequencies randomly distributed according to the Gaussian law with the parameter Γ_{hhx}. Thus we obtain the following expression for the full complex third-order polarization:

$$\hat{P}(t,\tau)^{(3)}\propto\exp\left(-\frac{1}{4}\Gamma_{hhx}^2t^2-\frac{t}{T_2^{ba}}-i\omega_{ba}^0t\right)\exp\left(\frac{\tau}{T_2^{ba}}\right)\int_{-\infty}^{t}dt'\int_{-\infty}^{t'}dt''\int_{-\infty}^{t''}dt'''\times\left\{E_2(t')\exp(i\Delta\omega_{ba}^0t')\left[E_1(t'')\times\exp\left(-\frac{t''}{T}\right)E_2(t''')\exp\left(\frac{t'''}{T}\right)\times\sum_s\exp(i\Delta\omega_s(t'''-t''))+E_2(t'')\times\exp\left(-\frac{t''}{T}\right)E_1(t''')\exp\left(\frac{t'''}{T}\right)\times\sum_s\exp(i\Delta\omega_s(t''-t'''))\right]\right\}=[J_1(\tau,t)+iJ_2(\tau,t)]\exp(-i\omega_{ba}^0t).\qquad(A1)$$

The correlation curve corresponding to excitation of the continuous spectrum in this case is the squared absolute value of the Fourier transformed real part of $P(t,\tau)^{(3)}$ at the point $\omega=\omega_{ba}^0$:

$$I(\tau)\propto\left[\int_{-\infty}^{+\infty}J_1(\tau,t)dt\right]^2+\left[\int_{-\infty}^{+\infty}J_2(\tau,t)dt\right]^2.\qquad(A2)$$

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