

Nonlinear absorption of femtosecond light pulses in bulk crystals under interband multiphoton resonance conditions

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Abstract. A theory of the nonsteady-state absorption of supershort light pulses in bulk materials and heterostructures with quantum wells and wires when multiphoton resonance at interband transitions occurs is developed. The absorbed energy dependence on the detunings of the multiphoton resonance and on the pulse width is investigated. The interaction between two consecutive short pulses (pump-probe spectroscopy within the femtosecond regime) with bulk crystals and low-dimensional structures under the conditions of two-photon resonance at interband transitions and transitions between size-quantized sublevels is studied.

1. Introduction

Femtosecond light pulses are widely used in studying interaction of powerful laser radiation with transparent insulators and semiconductor nanostructures [1]. The carrier time τ_p (known as the relaxation time of momentum of carrier) is usually determined by scattering on longitudinal optical lattice vibrations and ordinarily is hundreds of femtoseconds in bulk crystals. In heterostructures τ_p is also determined by intrasubband transitions due to interaction of electrons with optical phonons. When the pulse width $\sqrt{\sigma} < \tau_p$, conventional methods of calculating the optical transition probabilities, relying on the concept of the number of transitions per unit time, are not adequate. Thus, the formulas obtained for the case of quasi-steady-state electromagnetic fields [2] cannot be used for a consistent interpretation of experimental data concerning the nonlinear response of materials to femtosecond light pulses [3], and other methods are required.

2. Transitions between discrete electron levels

Let a linearly polarized light with frequency ω propagate a length L in a medium along x direction. The absorbed power per unit area is then determined by [3]

$$J = -\omega L F_0(t) P_s(t) \quad (1)$$

where $F_0(t)$ is the electric field amplitude of the wave, while $P_s(t)$ is the medium polarization of the reactive nature. We assume that the field does not change significantly as it passes through



the length L of the medium. We take the light pulse in a Gaussian shape:

$$F_0(t) = f_0 \exp \left[-(t - t_0)^2 / \sigma \right] \quad (2)$$

Further to simplify the calculations, we considered the nonlinear polarization for the case of a three-level system with discrete levels (for example, quantum dots and impurities) when multiphoton resonance corresponds to a transition between levels 0 and 1, i.e., $E_1 - E_0 = n\hbar\omega$ (Fig.1a). For even values of n , the allowed transitions are $0 \rightarrow 2$ and $1 \rightarrow 2$, while, for odd n , the transitions $0 \rightarrow 1$ and $1 \rightarrow 2$ are allowed.

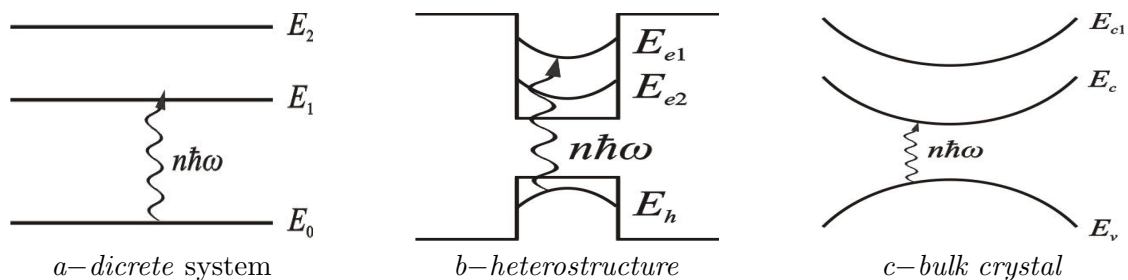


Fig.1. Models of the band materials

In what follows, we also consider absorption in a three-band model of a one-, two- or three-dimensional systems (Fig.1b-c). Using Eq.(1), for the energy absorbed in unit volume the following approximate formula is obtained

$$J_n \approx -\omega \left| M_{if}^{(n)} \right|^2 n_0 \frac{2^{\frac{7n}{2}} - \frac{3}{2} \pi^{\frac{n}{2}} \sqrt{5} e^{2n}}{\varepsilon_\infty^{n/2} c^n \hbar^{2n-1} \sigma^{-1+n/2}} W^n \exp \left(-\frac{\omega_R^2}{n\sqrt{\pi}} \sigma \right) \lambda_n \quad (3)$$

where e is the electron charge, c is the speed of light, ε_∞ is the high-frequency permittivity of the medium, n_0 is the number of three-level systems (quantum dots or impurity atoms) in unit volume, $\omega_R = n\omega - \omega_{fi}$ is the detuning of the n -photon resonance under assumption that $\omega_R \ll \omega$, W is the total energy passing through a unit area during the laser pulse, $M_{if}^{(n)}$ are the n -order compound matrix elements of the electron coordinate operator between the initial i and final f states [4] and

$$\lambda_n = \sum_{p=1}^n \exp \left(-\Theta_p^{(n)} \gamma \sqrt{\sigma} \right) \quad (4)$$

The following approximations for the quantities entering Eq.(4) can be made

$$\Theta_1^{(n)} \approx \frac{1}{8}(n+9), \Theta_2^{(n)} \approx \frac{1}{3\sqrt{2\pi}} \left(1 + 3\sqrt{7n} \right), \Theta_3^{(n)} \approx \frac{6\sqrt{2\pi}}{13} \left(1 + \frac{2}{\sqrt{\pi n}} \right),$$

$$\Theta_4^{(n)} \approx \frac{1}{3\sqrt{\pi}} \left(1 - \frac{23\sqrt{3\pi}}{2n} \right), \Theta_5^{(n)} \approx \frac{1}{8} \sqrt{\frac{37\pi}{3}}.$$

Let us note, that the preexponential factor in dependence (3) is proportional to $\sigma^{1-n/2}$. In addition, the σ dependence is contained in the exponent and in the factors λ_n in the right-hand side of Eq. (3). While, in the case of long pulses ($\gamma\sqrt{\sigma} \gg 1$) the absorbed energy is proportional to $\sigma^{1-n/2}$ only. Thus, for systems with discrete levels, including quantum dots, impurity centers, etc., the nonlinear multiphoton resonance response to ultrashort and longer laser pulses, may be essentially different.

3. Nonlinear multiphoton resonance response of heterostructures with a continuous spectrum

In the case of interband (intersubband) transitions, the electron system of a crystal can be approximately represented as a collection of noninteracting three-level systems each of which is characterized by its own wave vector \mathbf{k} (three-, two-, or one-dimensional) and the corresponding energies $E_i(\mathbf{k})$.

Let us denote the dimensionality of the structure by D ($D = 3$ in the case of bulk crystals, $D = 2$ for quantum wells, and $D = 1$ for quantum wires). Then, the energy of the ultrashort pulse absorbed by the D -dimensional system under n -photon resonance conditions is determined by the formula

$$J_n^{(D)} = 2\Xi_D \int_0^\infty dk k^{D-1} J_n(k), \quad (5)$$

where $\Xi_1 = Ln_1/(2\pi)$, $\Xi_2 = Sn_2/(2\pi)$, $\Xi_3 = \Omega/(4\pi)$, L is the length of quantum wire, S is the area of quantum well, n_1 is the number of quantum wires per unit area normal to their direction, and n_2 is the number of quantum wells per unit length in the growth direction. Integration over k space is carried out by the saddle-point method. For the energy absorbed in unit volume from (5) we get

$$J_n^{(D)} = \eta_D \frac{2^{(7n-4)/2} \pi^{(2n-1)/4} 5^{1/2} n^{1/2} \omega \mu^{1/2} e^{2n}}{\varepsilon_\infty^{n/2} c^n \hbar^{(4n-1)/2} \sigma^{(n-1)/2}} W^n \lambda_n |M^{(n)}|^2. \quad (6)$$

where

$$\eta_1 = n_1/\Delta_n^{1/2}, \eta_2 = (2\mu/\hbar)^{1/2} n_2, \eta_3 = 2\mu\Delta_n^{1/2}/(\pi\hbar)$$

and μ is the reduced mass of electron and hole and $\hbar\Delta_n = n\hbar\omega - \tilde{E}$, $\omega_R = \Delta_n - \frac{\hbar k^2}{2\mu}$. In the case of three-dimensional crystal, $\tilde{E} = E_g$, where E_g is the bandgap width. In the case of quantum wells or quantum wires, \tilde{E} stands for the energy gap between initial and final subbands of quantum confinement at $\mathbf{k} = 0$.

It should be noted that the dependence of the absorbed energy on the detuning of resonance from the fundamental band edge, Δ_n , leads to the same result as in the case of quasi-steady-state field:

$$J_n^{(3)} \propto \sqrt{\Delta_n} \quad (7)$$

This Eq. corresponds to the density of electron states for bulk crystal ($D = 3$). Similarly, it is found that the dependence of the absorbed energy on duration of field action also is the same as in the case of quasi-stationary.

Here, the situation differs from the case of a system with discrete levels. It is clear to understand if we take into account the following reasons. When we consider the continuous electron spectrum, all the spectral components that form the ultrashort light pulse, participate in the resonant multiphoton transitions on interband transitions, each Fourier component in the proper point of the Brillouin zone. For this reason, the situation becomes similar to the case of steady-state absorption of monochromatic light.

The dependences of the absorbed energy on the pulse width in the case of low-dimensional structures ($D = 1, 2$) have the same character as in bulk crystals. The explanation of this fact is the same as in the case of bulk materials.

The dependences of the absorbed energy on the detuning from resonance Δ_n in the case of two- and one-dimensional structures again coincides with their quasi-steady-state analogs. For example, in two-photon absorption process $J_2^{(1)} \propto \Delta_2^{-1/2}$ and $J_2^{(2)} \propto \text{const}$ for $D = 1, 2$, respectively.

4. Pump-Probe spectroscopy within the femtosecond two-photon resonance regime

In recent years, in experimental studies of multiphoton absorption by wide-band semiconductors and insulators the method of the femtosecond "pump-probe" spectroscopy is used widely [5]. Pump-probe spectroscopy within the femtosecond regime uses two consecutive laser pulses. The first pulse (known as pump) excites the sample, the second pulse (known as probe) probes the change in the optical properties, due to action of the first powerful pump pulse. Let the system interact with two consecutive ultrashort pulses with frequencies ω_1 and ω_2 , respectively. The time delay between the pulses is of the same order as the pulse durations, that is $\tau \leq 100$ fs. The lights are linearly polarized and they propagate in optically thin medium, i.e. single-photon transitions in this medium are absent. Two-photon resonance corresponds to a transition between levels 0 and 1, i.e., $E_1 - E_0 = \hbar(\omega_1 + \omega_2)$. In the dipole approximation the interaction operator is

$$\hat{V}_t = \sum_{i=1}^2 \hat{d}F_{\omega_i}(t)e^{-i(\omega_i t - k_{\omega_i} z)} + H.c. \quad (8)$$

where k_{ω_i} are the wave vectors of light pulses. So, using the formulas (1) we can write the final expression for absorbed energy from probe pulse in unit volume

$$J_{2,pp}^{(0)}(\omega_R, \tau, \sigma_1, \sigma_2, \gamma) \approx \sum_{i=1}^2 M_i f_i, \quad (9)$$

where

$$M_i = n_0 \omega_2 \frac{\sigma_1 \sigma_2}{\sigma_1 + \sigma_2} \sqrt{\pi} \frac{e^4 (x_{20} x_{21})^2}{\hbar^3} \left(\frac{32\pi W_1 W_2}{c^2 \varepsilon_\infty \sqrt{\sigma_1 \sigma_2}} \right) \left(\frac{1}{\omega_{20} - \omega_i} \right) \left(\frac{1}{\omega_{20} - \omega_2} + \frac{1}{\omega_{20} - \omega_1} \right),$$

$$f_i = \frac{71}{40} \exp \left[-\frac{2(\tau + a_i \sigma_1 \gamma / 4)^2}{\sigma_1 + \sigma_2} \right] \exp \left[\frac{2(a_i \sigma_1 \gamma)^2}{\sigma_1 + \sigma_2} - b_i \gamma \sqrt{\sigma} \right] \exp \left[-\frac{\sigma \omega_R^2}{2} \right]$$

and $\omega_{ij} = (E_j - E_i)/\hbar$ are the frequencies of the transitions between the j th and i th states in the electron system, ω_R is the detuning of two-photon resonance, $\sqrt{\sigma_i}$ are the pulse widths. $a_1 = 47/16, b_1 = 5/46, a_2 = 251/86, b_2 = 21/328$. These coefficients are obtained by numerical evaluation.

Using Eq.(9) we get the second pulse energy absorbed by D -dimensional system

$$J_{2,pp}^{(D)} = \Delta_2^{(D-2)/2} \frac{\alpha^{(D)} \mu^{D/2} e^4 \left(x_{20}^{(D)} x_{21}^{(D)} \right)^2 W_1 W_2}{\hbar^{3+D/2} c^2 \varepsilon_\infty^{(D)}} \frac{\omega_2}{\sqrt{\sigma_1 + \sigma_2}} \Omega_{ij} S_{ji}, \quad (10)$$

where we introduced the following notations

$$S_{22} = f_1, S_{11} = f_2, S_{12} = S_{21} = (f_1 + f_2)/2 \quad (11)$$

and f_i is the same as in formula (9) and

$$(\omega_{20} - \omega_i)^{-1} (\omega_{20} - \omega_j)^{-1} = \Omega_{ij}. \quad (12)$$

In Eq.(10) the summation over dummy indices $i, j = 1, 2$ is implied. $\alpha^{(D)}$ are the numerical coefficients for the D -dimension structures: $\alpha^{(1)} = 568/5, \alpha^{(2)} = 284\sqrt{2}\pi/5, \alpha^{(3)} = 568\pi/5$. For one- and two- dimensional systems the right hand side of formula (10) must be multiplied by the number of quantum wires per unit area normal to their direction, n_1 or by the number of quantum wells per unit length in the growth direction, n_2 , respectively. It should be noted

that the terms $a_i\sigma_1\gamma/4$ in both exponents in Eqs.(9) and (10) are closely related to polarization induced by the high-powered pump pulse.

The expressions obtained above show that the dependence of the absorbed energy on detuning of two-photon resonance from probe pulse has Gaussian form, i. e. $J_{2,pp}^{(0)} \propto \exp(-\sigma\omega_R^2/2)$.

The absorbed energy dependence on the delay between two pulses also has Gaussian shape.

5. Conclusion

We analyzed the nonlinear response of bulk materials and heterostructures with quantum wells, wires, and dots to supershort light pulses with widths less than the intraband (intrasubband) electron or hole relaxation time. The dependence of the absorbed energy on the detunings of multiphoton resonances is obtained for nanostructures of different dimensions. Similarly, the dependence of the absorbed light energy on the width of a supershort pulse with fixed energy per pulse is investigated. It is shown that these dependences for zero-dimensional objects (quantum dots, impurity centers) substantially differ from those that occur in the case of relatively long pulses. This circumstance can be important in interpretation of experimental data on the nonlinear response of heterostructures with quantum wells to supershort light pulses. The dependence of the absorbed energy on the detuning of the two-photon resonance and on the delay between the pump and probe pulses is also studied. It is shown that in this case the main role in absorption of supershort pulses plays the polarization induced by pump pulse.

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