

GENERATION OF COHERENT PHONON PULSES BY HOT ELECTRONS IN NON-CRYSTALLINE SEMICONDUCTORS

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We study the cooperative generation of phonon pulses in semiconductors by groups of inverted equidistant multilevel coupled excitations (electrons, excitons). Such equidistant systems take place in chalcogenide glasses with fractal structure. It is demonstrated that the cooperative generation of non-equilibrium localized phonons can be observed in experimental studying of the relaxation of groups of excited atoms after the passage of short laser pulse through the glass sample. The increasing of the absorption coefficient in the process of the excitation of glasses by short laser pulse with relatively small power is conditioned by the generation of the non-equilibrium coherent localized phonons in the process of the relaxation of non-equilibrium excitations. These coherent non-equilibrium localized phonons strongly change the topology of the random potential and, thus, open a new channel of the interband light absorption. These transitions take place with simultaneous participation of the photons and localized phonons.

Keywords: Nonlinear optics, Chalcogenide glasses, Fractals

1. Introduction

Recently a great attention was devoted to the studying of cooperative generation of the non-equilibrium phonons in semiconductors [1,2]. In the ref. [1] it was shown that superradiance takes place not only in the multi-atom inverted two-level systems. For example, superfluorescence and amplified spontaneous emission of 29-cm^{-1} phonons were observed following inversion of the $\bar{E}(^2E) - 2\bar{A}(^2E)$ acoustic transition of Cr^{3+} in ruby. In this work it was demonstrated that the conditions for both of these manifestations of phonon avalanches are selected via the Cr^{3+} concentration. The authors of this work describe the superfluorescence in terms of a pendulum equation of the acoustic Bloch vector. It should be noted, that rate equations of the level and phonon populations account for amplified spontaneous emission.

In another ref. [2] using the picosecond pump and probe technique the authors have detected oscillations of photoinduced transmission and reflection in thin films of $a\text{-As}_2\text{Te}_3$ and cis-polyacetylene. These oscillations are due to the generation and propagation of coherent acoustic phonons in the film. Recently the non-linear interband absorption in such chalcogenide glass semiconductors (CGS) as As_2Se_3 , AsSe , GeSe_2 and $\text{As}_{22}\text{Se}_{33}\text{Ge}_{45}$ has been studied in ref. [3]. Here the hysteresis dependence of the output light intensity as a function of the input light intensity was observed. This hysteresis takes place due to the generation of the non-equilibrium phonons, which change the absorption coefficient.

As the possibility of the coherent generation of the non-equilibrium phonons was observed in refs. [1,2] in our paper we propose a new cooperation mechanism, which account for the non-linear dependence of the absorption coefficient in CGS. That is why we study the interband excitation of the CGS by a short laser pulse in the same way as it was done in Andriesh – Chumash experiments [3]. It is clear that the relaxation of the excited electrons and holes (or certain number of excited atoms [4]) in the quantum wells of the random potential is accompanied by the generation of the non-equilibrium localized phonons [5,6]. The avalanche of these phonons with the wave-length of the order of the

magnitude modulation length of the random potential in the CGS drastically changes the form of this potential. We suppose that random potential modulates the bottom of the conductor band and the top of the valency band of the CGS. The non-equilibrium localized phonons with such wave-length also change this modulation and this effect opens a new channel of the interband light absorption. For this reason the absorption coefficient increases with the increasing of the light intensity and this phenomenon can be explained by means of photoinduced light absorption in CGS with the participation of the fractons. In fact, the coefficient of the interband light absorption can be represented as a sum of two parts

$$\alpha = \alpha_0 + \beta n, \quad (1)$$

where α_0 is the part of the absorption coefficient, which does not depend on the temperature, βn is the part of the absorption coefficient, which depends on the mean number of the localized phonons n .

Thus, in the process of the interband light absorption the transitions with the simultaneous participation of light quanta and localized phonons play an essential role. The second part of the absorption coefficient in eq. (1) essentially changes in the process of the relaxation of the excited electrons into the localized states in the optical gap of CGS. This relaxation is accompanied by the coherent generation of the non-equilibrium phonons $\Delta n = n - n_0$, where n_0 is the mean number of the equilibrium localized phonons.

The aim of the present work is to show in what manner the excited electrons in the quantum wells of the random potential can generate the correlated localized phonons. These coherent localized vibrational excitations decay into acoustic phonons which propagate through the sample of CGS.

2. The model of cooperative relaxation

Let us first consider the case in which such quantum wells appear in CGS due to the random potential that there are only two quasielectron states in the wells: excited and ground. Similar quantum wells can appear in the process of the excitation of certain number of atoms (or groups of atoms) by laser pulse [4,7]. In general, the energies of the two levels are more or less random quantities, depending on such factors as the particular configuration of atoms surrounding the two minima, on local strains, etc. [4]. In this case the new quasibonds between the atoms, belonging to the certain group can occur. Such a non-equilibrium cluster states of atomic group can relax into the ground states, generating the non-equilibrium localized phonons. Among randomly distributed quantum wells of quasielectrons one can always pick out the quasiequidistant subgroups of two-level states in a such randomly distributed states. In the other words, the spectrum of two-level states with random distance between the ground and excited states can be divided into the spectrum of quasiequidistant groups of two-level states. For example, if we consider a single localized electron with the transition energy $\hbar\omega_\alpha = E_{2\alpha} - E_{1\alpha}$ one can always find the large number of other localized electrons with the same transition energy between the ground and excited states. Therefore, let us introduce the subgroup distribution function ρ^α of the electron states in the quantum wells with the same energy distance $\hbar\omega_\alpha$. Every subgroup α of coupled electrons generates localized phonons with the proper frequency $\hbar\omega_\alpha$. The certain quasiequidistant subgroup can generate a coherent Dicke pulse [8] of the non-equilibrium localized phonons. The localized phonons decay into the acoustic phonons. The analogous two-level systems were proposed by Anderson et al. [4]. Such two-level systems occur when the certain number of excited atoms (or group of atoms) have two equilibrium positions. The mechanism of the excitation of the localized electron-hole pairs (or groups of atoms) by laser pulse and the generation of the localized non-equilibrium phonons can be described by the following system of the differential equations

$$\begin{aligned} \frac{1}{c} \frac{dI_s(t,s)}{dt} + \frac{dI_s(t,s)}{dz} &= -(\alpha_0 + \beta n) I_s(t,z), \\ \frac{dN_e^\alpha}{dt} &= -\frac{N_e^\alpha}{\tau_0^\alpha} + (\alpha_0 + \beta n) \frac{V_0 I_s(t,s)}{\hbar\omega_0} \rho^\alpha - \frac{N_e^\alpha N_g^\alpha}{\tau^\alpha} - \frac{n_\alpha}{\tau_0^\alpha} (N_e^\alpha - N_g^\alpha), \end{aligned} \quad (2)$$

$$\frac{dN_g^{\alpha'}}{dt} = \frac{N_e^{\alpha'}}{\tau_0^{\alpha'}} + \frac{N_e^{\alpha'} N_g^{\alpha'}}{\tau^{\alpha'}} + \frac{n_{\alpha'}}{\tau_0^{\alpha'}} (N_e^{\alpha'} - N_g^{\alpha'}),$$

$$\frac{dn}{dt} = -\frac{I}{\tau_f} (n - n_0) + \sum_{\alpha'} \frac{dN_g^{\alpha'}}{dt}$$

where $N_e^{\alpha'}$ and $N_g^{\alpha'}$ are the mean numbers of the excited electrons and the electrons in the ground state of α' subgroup, respectively, α' enumerates the number of the equidistant subgroups of the two-level systems, $\alpha' = 1, 2, 3, \dots, N_s$, V_0 is the excitation volume by laser pulse of the sample of CGS, I_s is the light intensity in the sample of CGS, c is the light velocity, $\rho^{\alpha'}$ is the subgroup distribution function, having the following property $\sum_{\alpha'=1}^{N_s} \rho^{\alpha'} = 1$, $n_{\alpha'} = n \rho^{\alpha'}$, $\tau^{\alpha'} / N^{\alpha'}$ is the cooperative decay time of subgroup α' , $N^{\alpha'} = N_g^{\alpha'} + N_e^{\alpha'}$, $\tau_0^{\alpha'}$ is the spontaneous decay time of a single electron from the excited to the ground state of subgroup α' , τ_f is the leaving time of the phonons from the active localized region of the acoustic phonon modes [9]. This system can be obtained from Maxwell-Bloch equations when the laser-pulse duration is more large than the relaxation time of electron polarization in CGS. The cooperation effect is possible in case when the wave-length of the localized phonons is of the same order as the distance between the quantum wells of α' subgroup.

In thin films, which play the role of resonator, the induced laser generation of phonons is possible due to the reflection of phonon waves from the surfaces of the films. In this paper we discuss the problem of the generation of localized phonons, which don't have the possibility to propagate through the films. The last equation of the system (2) takes into account only the generation of such localized phonons in the decay process of the non-equilibrium localized electrons $\sum_{\alpha'} \frac{dN_g^{\alpha'}}{dt}$ and losses from the non-equilibrium phonon modes $-(n - n_0) / \tau_f$. In this equation we don't take into account the reflection of phonon waves from the surfaces of the films. This is valid in the case of large losses and thick films with thickness $\propto 5 \times 10^{-4} \text{ cm}$ [3]. That is why there are no oscillations of the light transmitted from the laser.

When the pulse duration τ_p has the same order as the relaxation times of the localized electrons and phonons subsystems one can find the stationary solution of the system (2). From the stationary conditions of variables N_e , n and I_s ($dI_s / dt = dn / dt = dN_e / dt = 0$) we obtain the expression for the non-linear absorption coefficient $\alpha(I_s) = -dI_s(z) / I_s dz$ in the following form

$$\alpha(I_s) = \alpha_0 + \beta \frac{n_0 + \tau_f V_0 \alpha_0 I_s(z) / \hbar \omega_0}{1 - \tau_f V_0 \beta I_s(z) / \hbar \omega_0}$$

One can observe that with the increasing of the light intensity the absorption coefficient increases. Such increasing was experimentally observed by the authors of refs. [3]. The nonstationary solution of the system of equations (2) leads to the hysteresis dependence of the output light intensity as a function of the input intensity.

When the time duration of the excitation pulse is shorter than the decay times of the localized electrons the pulse form can be approximated by δ -function ($I_s(z, t) = I_{s0} \tau_p \delta(t)$). In this case the number of excited electrons during the pulse duration is given by

$$N_e^{\alpha'} = \tau_p V_0 \frac{I_{s0}}{\hbar \omega_0} (\alpha_0 + \beta n_0) \rho^{\alpha'} = 2 j^{\alpha'}$$

where $2j^{\alpha'}$ is the number of the localized excited centers (electrons, holes) in the α' subgroup. From the second and third equations of the system (2) one obtains the decay equation for the inversion $\langle D_z^{\alpha'} \rangle = (N_e^{\alpha'} - N_g^{\alpha'}) / 2$

$$\frac{d \langle D_z^{\alpha'} \rangle}{dt} = -\frac{I}{\tau_0^{\alpha'}} (\langle D_z^{\alpha'} \rangle + j^{\alpha'}) - \frac{2}{\tau_0^{\alpha'}} n_0 \langle D_z^{\alpha'} \rangle + \frac{I}{\tau^{\alpha'}} (\langle D_z^{\alpha'} \rangle^2 - j^{\alpha'^2}). \quad (3)$$

Here \bar{n}_0 is the mean photon number of the external thermal field $\bar{n}_0 = [\exp(\hbar\omega_0 / K_B T) - 1]^{-1}$.

Let us proceed to study the behaviour of the proposed system in the quasistationary case, when $dn/dt \ll (n - n_0) / \tau_f$. In this situation from the fourth equation of the system (2) it follows that

$$n = n_0 - \tau_f \sum_{\alpha} \int_0^t \exp(-(t-\tau) / \tau_0) \frac{d \langle D_z^{\alpha}(\tau) \rangle}{d\tau},$$

where one can find $\langle D_z^{\alpha} \rangle$ by solving eq. (3)

$$\langle D_z^{\alpha} \rangle = \frac{\tau^{\alpha}}{2\tau_0^{\alpha}} (1 + 2\bar{n}_0) - d^{\alpha} \tanh\left(\frac{d^{\alpha}}{\tau^{\alpha}} (t - t_0^{\alpha})\right), \quad (4)$$

where

$$d^{\alpha} = \sqrt{\left(\frac{\tau^{\alpha}}{2\tau_0^{\alpha}}\right)^2 (1 + 2\bar{n}_0)^2 + \left(\frac{j^{\alpha}}{\tau^{\alpha}} + \frac{1}{\tau_0^{\alpha}}\right) j^{\alpha} \tau^{\alpha}},$$

$$t_0^{\alpha} = \frac{\tau^{\alpha}}{d^{\alpha}} \ln \sqrt{\frac{d^{\alpha} - \frac{\tau^{\alpha}}{2\tau_0^{\alpha}} (1 + 2\bar{n}_0) + j^{\alpha}}{d^{\alpha} + \frac{\tau^{\alpha}}{2\tau_0^{\alpha}} (1 + 2\bar{n}_0) - j^{\alpha}}}$$

is the delay time of the subgroup α . In this case the stationary state solution for the non-linear absorption coefficient α takes the form

$$\alpha = \alpha_0 + \beta \left\{ n_0 - \tau_f \sum_{\alpha} \frac{d \langle D_z^{\alpha} \rangle}{dt} \right\} = \alpha_0 + \beta \left\{ n_0 + \tau_f \sum_{\alpha} \frac{d^{\alpha 2}}{\tau^{\alpha}} \operatorname{sech}^2\left(\frac{d^{\alpha}}{\tau^{\alpha}} (t - t_0^{\alpha})\right) \right\}. \quad (5)$$

The eq. (5) describes the relaxation of the non-linear absorption coefficient after the passage of the δ - pulse through the sample of CGS. It should be noted that these relaxations were also considered in refs. [3]. As follows from eq. (5) the relaxation depends on the cooperation law between the localized electrons. Every function sech^2 in eq. (5) has the maximum, when $t = t_0^{\alpha}$. The sum on α takes into account all subgroups of the equidistant two-level states, which have different delay times. In this situation the relaxation law of the absorption coefficient can be more broadened than that in case of single group of the equidistant two-level states. This broadening depends on the explicit form of the subgroup distribution function ρ^{α} .

3. Conclusions

Returning to our subgroups we observed that the low of coherent time excitation each phonon modes strongly depend: on the frequency of this phonons ω_{α} , on the number of excited electrons N_e^{α} , which can pass in the regime of cooperative generation. In the Dicke theory the time of coherent swing of phonon modes in the cooperative relaxation theory is named the delay time of phonon pulses. It is not difficult to observe that the group of excited electrons in the deep quantum wells generates more higher frequency phonon pulse with more short delay time. This law follows from the dependence of the delay time on the spontaneous relaxation time and number of excited electrons $t_0^{\alpha} = \tau_0^{\alpha} / N \ln 2N$. As the relaxation time τ_{α} is proportional with the third order of transition frequency ω_{α} , the delay time for the process of collective generation of higher frequency phonon is more less then the same time for phonon generation at lower frequency.

This effect can be experimental observed in the processes of propagation through the thin films of laser pulses with different duration and the same energy. If the time duration of pulses is comparable with the delay time of high energy phonons in the process of propagation of laser pulse through the films in absorption process the main contribution give only the higher frequency non-equilibrium phonons. For pulses with more large duration the low frequency coherent modes can be generated in time and make substantial contribution in the absorption processes of pulse. In the other

words, by changing the duration of laser pulses we can test the time and spectral behaviour of the process of coherent phonon generation.

As one can see from the eqs. (5) the non-linear induced absorption coefficient α' strongly depends on the relaxation law of hot electrons in the random potential. This law is determined by the form of the random potential and by the dynamic symmetry of the relaxation in the system. From eq. (5) it follows that for shallow quantum wells and for the SU(2) symmetry in case of deep quantum wells the non-linear absorption coefficient $\alpha \propto \text{sech}^2(t)$ and is similar to the Dicke superradiance pulse.

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