Semiconductors, Vol. 37, No. 5, 2003, pp. 526–531. Translated from Fizika i Tekhnika Poluprovodnikov, Vol. 37, No. 5, 2003, pp. 546–551.

A Local Specific Feature of Variation in the Spectrum of Picosecond Superluminescence upon Adding Excited Carriers to a Non-Fermi Electron–Hole Plasma in GaAs

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Submitted June 10, 2002; accepted for publication September 9, 2002

Abstract—A dense hot electron–hole plasma and picosecond superluminescence appeared upon pumping of a GaAs layer by a picosecond (*ex*) optical pulse. The distribution of electrons over the conduction band was modulated with a period equal to the LO-phonon energy. The effect of additional pumping of GaAs (p_i) by an optical pulse with a photon energy of $\hbar\omega_p < \hbar\omega_{ex} - 0.1$ eV on the superluminescence was investigated. In the case of simultaneous pumping by *ex* and p_i pulses, a local maximum or minimum appeared in the spectrum of relative increase in the superluminescence energy at the photon energy, which corresponds to the peak in the superluminescence-energy spectrum of the active region of the GaAs layer. The local maximum appeared when the electrons that were excited by the p_1 pulse to the level with a depleted population emitted LO phonons (one phonon per each electron) and recombined. The local minimum appeared when electrons were excited by the p_2 pulse to the level with the Fermi population. The spectral width of the local maximum and minimum turned out to be narrower than the calculated width of the energy level from which the electrons recombine. *© 2003 MAIK "Nauka/Interperiodica".*

1. INTRODUCTION

The purpose of this study is to clarify the influence of additional excitation of charge carriers by a pulse (p_i) with a photon energy of $\hbar\omega_p < \hbar\omega_{ex}$ on the intense picosecond superluminescence that occurs upon the pumping of GaAs by a powerful picosecond (*ex*) optical pulse. The electrons were excited by p_i pulses to the region near the bottom of the conduction band, where the electron-energy distribution was strongly modulated, i.e., differed from the Fermi distribution. The experiments were carried out at room temperature.

Let us initially describe the situation that occurs in the case of pumping of GaAs by the *ex* pulse with the photon energy $\hbar\omega_{ex} = 1.558$ eV and a duration of 14 ps. Upon the interband absorption of a powerful *ex* pulse in a thin (~1-μm) GaAs layer, a dense hot electron–hole plasma (EHP) arises and picosecond superluminescence occurs [1–4]. The latter is considered to mean induced radiative recombination in an active GaAs medium without a resonator with a characteristic relaxation time of ~10 ps. The superluminescence relaxation is interrelated with the relaxation of the temperature and concentration of the EHP [5, 6]. As estimations show, the superluminescence intensity integrated over the spectral range exceeds 10^8 W/cm^2.

Under these conditions, the energy distribution of electrons in the conduction band is modulated by oscillations with a period $\hbar\omega_{LO}$ ($\hbar\omega_{LO}$ is the energy of the longitudinal optical (LO) phonon) [7]. Periodically located regions arise in which the populations of energy levels are depleted in comparison with the population characteristic of the Fermi distribution of electrons. The following physical mechanism was suggested to explain the modulation of the electron-energy distribution [7].

The superluminescence intensity B_ω increases with an increase in the superluminescence-recombination rate of charge carriers: $(dn/dt)_R \propto \int \alpha_\omega B_\omega d\omega$. Here, ω is the frequency of superluminescence radiation and α_ω is the absorption coefficient in the light-amplification region; the integral is taken over the spectral amplification band. An increase in $|(dn/dt)_R|$ prevents $|\alpha_\omega|$ from increasing in the amplification region. For high superluminescence intensities, this causes depletion of the inverse population of the energy levels near the bottom of the conduction band, from which the electrons are induced to recombine, compared with the population in the case of the Fermi distribution of EHP. Such a depletion is represented by the difference between two absorption spectra for the amplification region, one of which is experimental and the other is calculated under the assumption of Fermi distribution for EHP (Fig. 1). Let us call this difference the dip in the amplification range. The shape of the dip spectrum is similar to that of the part of the superluminescence-energy spectrum which is located above the energy W_s^t (the superluminescence energy at those two values of $\hbar\omega$ which determine the dip boundaries [8]) (Fig. 1).

The depletion of populations leads to non-compliance with the principle of detailed balancing. As a result, intense transitions of electrons to the levels with depleted inverse population occur; these transitions are accompanied by the emission of LO phonons. The transitions with the emission of LO phonons turn out to be so intense that they lead to the formation of another region of population depletion in the conduction band. The second depletion region is located above the first one; the energy spacing between these regions is equal to the energy of the LO phonon $\hbar\omega_{\mathrm{LO}}$. The second depletion region results in the formation of a shoulder in the experimental absorption spectrum at $\hbar\omega > 1.417$ eV (Fig. 1). The further extension of this process upward over the conduction band leads to the periodic modulation of the electron-energy distribution within the band.

We used the mechanism of modulation described in [7] to explain the effects we revealed:

(i) the modulation of the bleaching (increase in transparency) spectrum of the GaAs layer by *phonon* oscillations with a period

$$\Delta = \hbar\omega_{\mathrm{LO}}(1 + m_e/m_h),$$

where m_e and m_h are the effective masses of an electron and a heavy hole, respectively [7];

(ii) the energy transport of electrons which occurs via emission of LO phonons in the course of picosecond superluminescence in GaAs [9];

(iii) "LO-phonon" correlation between the picosecond-superluminescence spectrum and the specific features of the absorption spectrum of GaAs in the case of the non-Fermi distribution of carriers [8].

The effects reported in [7–9] allow one to assume that, under the above experimental conditions, the relaxation times $\tau_{e\text{-LO}}$ and $\tau_{c\text{-}c}$ become comparable. Here, $\tau_{e\text{-LO}}$ is the time of such relaxation of electrons to the bottom of the conduction band, under which the electrons, emitting LO phonons, undergo transitions to the level with depleted population [9]. The quantity $\tau_{c\text{-}c}$ is the time of intraband energy relaxation of electrons due to collisions between the carriers, leading to the Fermi distribution of electrons. The fact that the times $\tau_{e\text{-LO}}$ and $\tau_{c\text{-}c}$ become comparable has not been explained theoretically yet and continues to arouse interest. For comparison, according to the calculations [7–9], in the dense quasi-equilibrium EHP, $\nu_{e\text{-LO}}^{-1} \gg \tau_{c\text{-}c}$ ($\nu_{e\text{-LO}}$ is the rate of emission of LO phonons by an electron) for the electrons from the energy region in which the above effects were observed.

It is worth noting that the largest depth of the dip within the amplification region at the photon energy

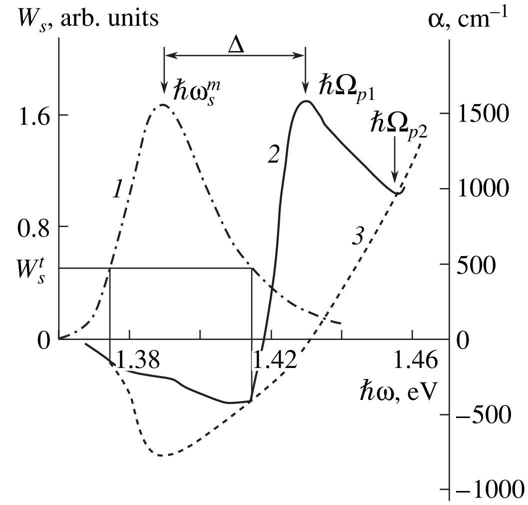

Fig. 1. (*1*) Spectrum of superluminescence radiation emerging from GaAs normally to the surface of the active region of the epilayer, (*2*) experimental light-absorption spectrum of photoexcited GaAs, and (*3*) calculated light-absorption spectrum for the case of Fermi distribution of the EHP in GaAs. The figure is taken from [8]. The arrows point to the photon energies $\hbar\Omega_{p_1}$ and $\hbar\Omega_{p_2}$ of the pulses p_1 and p_2, which were used in this study, as well as to the photon energy $\hbar\omega_s^m$.

$\hbar\omega_s^m$ is larger than the amplification factor α measured at $\hbar\omega_s^m$ by a factor of 2. Here, $\hbar\omega_s^m$ is the photon energy at which the superluminescence spectrum of the active region of GaAs is peaked (Fig. 1). It follows from this relation that the inverse frequency λ^{-1} of induced recombination transitions, which are accompanied by emission of photons with the energy $\hbar\omega_s^m$, is also comparable with the intraband relaxation time $\tau_{c\text{-}c}$.

In this study, in the spectrum of substantial relative increase in the energy of picosecond superluminescence, which is caused by the additional excitation of electrons by the p_i pulse, an abnormally narrow local minimum and maximum were detected. This phenomenon, observed in the case of simultaneous pumping of GaAs by picosecond *ex* and p_i pulses, is described and discussed in detail below.

2. EXPERIMENTAL

In this study, we analyzed a sample irradiated by an *ex* pulse. The thickness and composition of the epitaxial layers, as well as the pulse parameters, were identical to those used in [8]. Therefore, the results obtained in [8] (see Fig. 1) are completely applicable to this study.

The sample was an $Al_{0.22}Ga_{0.78}As$–GaAs–$Al_{0.4}Ga_{0.6}As$ heterostructure with layer thicknesses of 1.2, 1.6, and 1.2 μm, respectively. The heterostructure was grown by molecular-beam epitaxy on a GaAs(100) substrate.

Then, the heterostructure was separated from the substrate. This operation was not performed in [8]. The concentrations of donor and acceptor impurities in the heterostructure did not exceed 10^{15} cm^{-3}. The $Al_xGa_{1-x}As$ layers were designed to stabilize the surface recombination and mechanical strength. These layers are transparent to light of the frequency used in the experiment. An antireflection coating was deposited on the outer surfaces of the $Al_xGa_{1-x}As$ layers. As a result, the portion of light reflected normally to the layer surface was no more than 2%.

The heterostructure was irradiated by one (*ex*) or two ($ex + p_i$) pulses with an approximately Gaussian spatial distribution of intensity; the pulses were focused onto a single point. The duration of both p_i and *ex* pulses was equal to 14 ps. The diameters of the focal spots of the *ex* and p_i pulses were equal to 0.5 and 0.46 mm, respectively. In the case of pumping of GaAs by the p_i pulse only, the EHP concentration was just close to the threshold one, at which superluminescence appears.

The photoexcited active region of the GaAs layer, where superluminescence occurred upon pumping by the *ex* pulse and which was additionally enhanced upon pumping by the p_1 or p_2 pulses, was located at a distance of about 1.4 mm from the side face of the heterostructure. During the excitation pulse, this circumstance made it possible to exclude the following: (i) the effect of the positive feedback caused by the reflection of the superluminescence radiation from the side face of the heterostructure (the superluminescence radiation passed through the photoexcited region after the excitation pulse), and (ii) the effect of lattice defects near the side face of the heterostructure. The superluminescence radiation propagated mainly in the plane of the GaAs layer. After the emission from the side face of the heterostructure, the spectrum of the picosecond-superluminescence energy integrated over time, W_s, was measured in the solid angle of 4°.

Due to the absorption in the nonexcited region of the GaAs layer, the spectrum we measured differed from the superluminescence spectrum of the photoexcited active region of the GaAs layer. The superluminescence spectrum of the active region is shown in Fig. 1, which is taken from [8]. In [8], owing to the poor waveguiding properties of the sample, we measured the spectrum of radiation emerging from a similar sample normally to its surface. As was mentioned above, we used the *ex* pulse with the same parameters as those of the pumping pulse used in [8]. The thicknesses of the layers and the composition of the samples were also identical. Therefore, we can assume that Fig. 1 also shows the superluminescence spectrum measured in this study for the active region of the GaAs layer upon pumping by the *ex* pulse.

It is understandable that the shoulder at $\hbar\omega > 1.417$ eV in the light-absorption spectrum measured in this study, which is related to the depletion of the carrier population, also coincided with the shoulder in the spectrum measured in [8] and represented by curve *2* in Fig. 1.

Despite the partial absorption of superluminescence radiation in the passive region of the GaAs layer, we found some specific spectral features which allowed us to draw a number of substantial conclusions. In order to exclude, wherever possible, the effect of absorption of the superluminescence radiation in passive region of the GaAs layer, the spectrum of the relative increase in the superluminescence energy $W_s(ex + p_i)/W_s(ex) = f(\hbar\omega)$ was measured. This increase is due to the fact that the pumping by the p_1 pulse was added to the pumping of the sample by the *ex* pulse. Here, $W_s(ex + p_i)$ and $W_s(ex)$ are the energies of superluminescence for the cases of combined pumping by the *ex* and p_i pulses and pumping by the *ex* pulse solely, respectively.

In these experiments, p_i pulses with two values of the photon-energy, $\hbar\Omega_{p_1}$ and $\hbar\Omega_{p_2}$ (shown by arrows in Fig. 1), were used. The first value $\hbar\Omega_{p_1} = \hbar\omega_s^m + \Delta = 1.43$ eV coincided with the frequency corresponding to the shoulder peak in the absorption spectrum. Upon the absorption of the p_1 pulse, the electrons were excited to the energy level of the conduction band, which had the largest depletion of population in the second depletion region. The energy of this level exceeded by $\hbar\omega_{LO}$ the energy of the level which was maximally depleted in the first depletion region and from which the electrons recombined, emitting photons with the energy $\hbar\omega_s^m$. The variation in the superluminescence was investigated in detail for the spectral region close to the photon energy $\hbar\omega_s^m$. Upon simultaneous irradiation of the heterostructure with *ex* and p_1 pulses, the picosecond-superluminescence energy increased substantially compared with the case of superluminescence upon pumping of GaAs by the *ex* pulse solely. In this case, a local maximum at $\hbar\omega = \hbar\omega_s^m$ appeared in the spectrum of the relative increase in the superluminescence energy $W_s(ex + p_i)/W_s(ex) = f(\hbar\omega)$ (Fig. 2). The base width of the local maximum was 1.9 meV.

No local maximum was observed in the spectral dependences $W_s(ex + p_1)/W_s(ex) = f(\hbar\omega)$ measured for $\tau_d = -20$ ps and $\tau_d = 20$ ps, where the *ex* and p_1 pulses were not overlapped in time. Here, τ_d is the delay time of the p_1 pulse with respect to the *ex* pulse (Fig. 3). Thus, the local maximum appeared specifically under the conditions of simultaneous pumping by the *ex* and p_1 pulses.

The second energy value of the p_i pulse $\hbar\Omega_{p_2} = 1.455$ eV corresponds to the short-wavelength edge of the shoulder in the absorption spectrum (Fig. 1). Upon absorption of the p_2 pulse, the electrons were excited to that level in the conduction band, at which there was no population depletion. The energy of this level exceeds, by less than $\hbar\omega_{LO}$, the energy of the level to which elec-

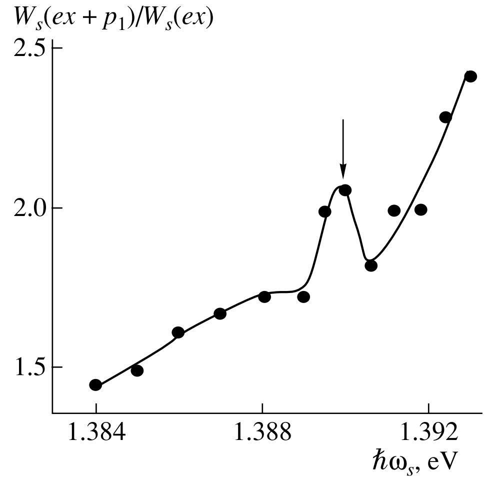

Fig. 2. Spectrum of relative increase in the superluminescence energy, caused by the addition of synchronous pumping of the sample by the p_1 pulse to pumping by the *ex* pulse. $\hbar\omega_{ex}$ = 1.558 eV, $\hbar\Omega_{p_1}$ = 1.43 eV. The arrow points to the photon energy $\hbar\omega_s \approx \hbar\omega_s^m \approx \hbar\Omega_{p_1} - \Delta$.

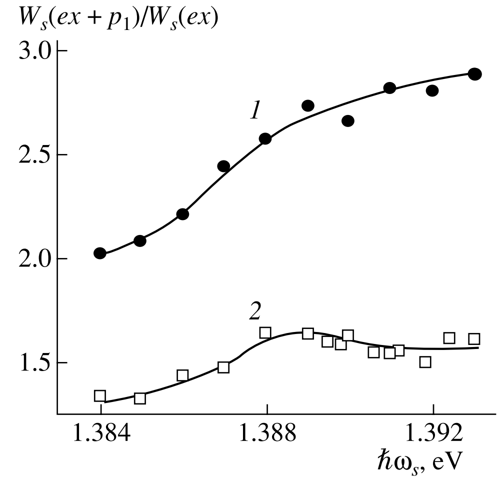

Fig. 3. Spectrum of relative increase in the superluminescence energy, caused by the addition of pumping of the sample by the p_1 pulse to the pumping by the *ex* pulse; the pulses were not overlapped in time. $\hbar\omega_{ex}$ = 1.558 eV, $\hbar\Omega_{p_1}$ = 1.43 eV; (*1*) τ_d = –20 ps, (*2*) τ_d = 20 ps (τ_d is the delay time of the p_1 pulse with respect to the *ex* pulse).

trons were excited due to the absorption of the p_1 pulse. Upon simultaneous pumping by the *ex* and p_2 pulses, a local minimum appeared in the spectrum of the relative increase in the superluminescence energy $W_s(ex + p_2)/W_s(ex) = f(\hbar\omega)$; however, this also occurred at the photon energy $\hbar\omega = \hbar\omega_s^m$ (Fig. 4). The base width of the local minimum was 1.7 meV.

3. RESULTS AND DISCUSSION

As was explained above, upon generation of EHP by the *ex* pulse, regions with a lowered population of the energy levels are formed in the conduction band, giving rise to corresponding depletion regions in the electron-energy distribution. These regions are located with the period $\hbar\omega_{LO}$. The electrons were excited to the level with the largest depletion in the second depletion region in the conduction band by the p_1 pulse with the photon energy $\hbar\Omega_{p_1}$, which resulted in a local enhancement of radiation with the photon energy $\hbar\omega_s \approx \hbar\omega_s^m \approx \hbar\Omega_{p_1} - \Delta$ (Fig. 2). The base width of the local maximum in the experimental spectrum $W_s(ex + p_1)/W_s(ex) = f(\hbar\omega)$ was equal to 1.9 meV. The appearance of a local maximum in the $W_s(ex + p_1)/W_s(ex) = f(\hbar\omega)$ spectrum points to the following. After the excitation of electrons by the p_1 pulse, some of them emit LO phonons (one phonon per electron) and undergo transitions to the levels with the largest depletion in the first depletion region at the bottom of the conduction band. At these levels, the electrons, having experienced no energy relaxation due to the interactions with other electrons of the conduction band, are forced to recombine, thus enhancing the superluminescence at $\hbar\omega_s \approx \hbar\omega_s^m$. This confirms the fact that $\tau_{e\text{-LO}}$ and $\tau_{c\text{-}c}$ are comparable. Hence, the effective lifetime of an electron at such a level, $\tau_{SR} = \lambda^{-1}$, and the quantity $\tau_{c\text{-}c}$ are also comparable. Here, λ is the rate of forced recombination transitions at $\tau_d \approx 0$, during which photons with an energy $\hbar\omega_s^m$ are emitted. We should note that the same assumption, based on the value of the dip depth in the amplification region, was formulated in the Introduction.

The base width of the local maximum in the experimental spectrum $W_s(ex + p_1)/W_s(ex) = f(\hbar\omega)$, as was mentioned above, was equal to 1.9 meV. In this case, the width ΔE of the level from which an electron recombines emitting a photon with an energy $\hbar\omega_s^m$ should be equal to $\Delta E \le 1.9$ meV. According to the uncertainty principle, the effective lifetime of an electron at this level $\tau_{SR} \ge 3.5 \times 10^{-13}$ s and the rate $\lambda \le 2.9 \times 10^{12}$ s^{-1}. The evaluations by formulas (6.2.35) and (4.3.19) from [10] for quasi-equilibrium carrier distribution yield the following times (i) of relaxation of the perturbation of the Fermi gas at an energy of 25 meV due to the electron–electron scattering, $\tau_{e\text{-}e} = 4 \times 10^{-15}$ s, and (ii) of absorption of an LO phonon by an electron with an energy of 25 meV, $\tau_{e\text{-}ph} = 9 \times 10^{-14}$ s. For calculations, we used 5×10^{18} cm^{-3} as the value of the concentration of electron–hole pairs and T = 52 meV for the temperature. These values correspond to the calculated absorption spectrum with the Fermi distribution of the EHP, which is shown in Fig. 1. The times $\tau_{e\text{-}e}$ =

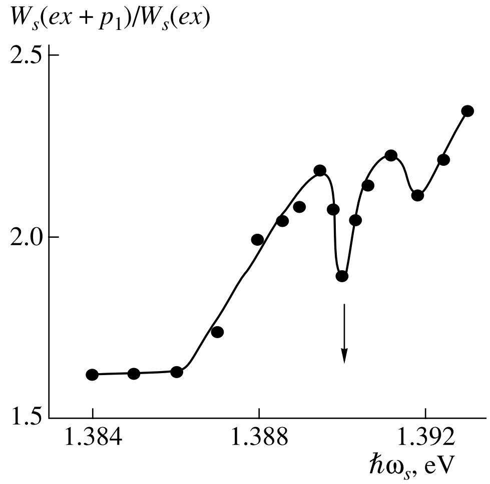

Fig. 4. Spectrum of relative increase in the superluminescence energy, caused by the addition of synchronous pumping of the sample by the p_2 pulse to the pumping by the *ex* pulse. $\hbar\omega_{ex}$ = 1.558 eV, $\hbar\Omega_{p_2}$ = 1.455 eV. The arrow point to the photon energy $\hbar\omega_s \approx \hbar\omega_s^m$.

4×10^{-15} s and $\tau_{e\text{-}ph} = 9 \times 10^{-14}$ s turned out to be substantially smaller than the time $\tau_{SR} \geq 3.5 \times 10^{-13}$ s. Therefore, doubt arises whether it is possible to use the estimates obtained under the assumption that the true modulated carrier distribution is the approximated Fermi distribution in the situation under investigation. Apparently, there are either some unknown reasons for increasing the times $\tau_{e\text{-}e}$ and $\tau_{e\text{-}ph}$ or some unknown specific features of the physical mechanism of formation of the local maximum in the spectrum of the relative increase in the picosecond superluminescence.

We should note that the picosecond superluminescence relaxes after the pumping with a characteristic time of ~10 ps. The superluminescence energy was measured, i.e., the superluminescence intensity integrated over time. It is demonstrated in this study that, in the spectrum $W_s(ex + p_1)/W_s(ex) = f(\hbar\omega)$, a local maximum appeared during the synchronous pumping of the sample by the *ex* and p_1 pulses with $\tau_d \approx 0$ (Fig. 2). When the intensity of light in the *ex* pulse approximately passes through the maximum, the depletion of the electron populations, which is distributed with a period $\hbar\omega_{LO}$ in the conduction band, is strongest [7, 11]. The strongest superluminescence recombination, which is necessary to create the depletion, should also occur under these conditions. The density of nonequilibrium LO phonons is also close to the largest value during photoexcitation [12]. After the *ex* pulse, the depletion of populations and superluminescence recombination are substantially weaker [5–7, 11] and the density of nonequilibrium LO phonons is lower too. When the excitation by a p_1 pulse was carried out after the *ex* pulse with the delay time τ_d = +20 ps, no local maximum appeared in the spectrum of the relative increase in the superluminescence (Fig. 3). Consequently, the local maximum appears under conditions of strong depletion of populations, strong superluminescence recombination, and high density of nonequilibrium LO phonons.

The reason for the absence of a local maximum at τ_d = –20 ps, when the sample was initially irradiated with a p_1 pulse and then with a *ex* pulse (Fig. 3), is evident, and we will not discuss it.

At the present time, it is difficult to conclusively establish the physical mechanism of formation of the local minimum in the spectrum of relative increase in the superluminescence energy $W_s(ex + p_2)/W_s(ex) = f(\hbar\omega)$ (Fig. 4).

We may assume that the spectrum of energy states is renormalized due to the coherent interaction of the superluminescence radiation with electrons and a gap arises in this spectrum [13]. The spectral position of the gap coincides with the maximum of the superluminescence spectra. The gap width $2\hbar\lambda$ should be equal to the base width of the local minimum (1.7 meV) in the spectrum of relative increase in the superluminescence energy (Fig. 4). In this case, the frequency of interband transitions $\lambda = 1.3 \times 10^{12}$ s^{-1}. For GaAs,

$$\hbar\lambda = 0.017 J^{1/2} (\text{eV}).$$

Here, J is the intensity of superluminescence radiation measured in GW/cm^2. The superluminescence radiation should be monochromatic and, consequently, occupy a very narrow region in the vicinity of the photon energy $\hbar\omega_s^m$ in the superluminescence spectrum. Accordingly, we obtain the evaluation of $J = 2.5 \times 10^6$ W/cm^2. The interaction of electrons with the electromagnetic field is coherent if the frequency of interband transitions λ exceeds the frequency of intraband electron scattering $1/\tau$, i.e., $\lambda\tau > 1$. In order to satisfy this relation, τ should exceed 7.7×10^{-13} s. In the above analysis we found that the effective lifetime of electrons at the isolated level $\tau_{SR} \geq 3.5 \times 10^{-13}$ s. The estimations of the lifetimes τ and τ_{SR} made by us do not exclude the explanation of the local minimum considered here; however, they are obviously insufficient to confirm it.

There is another question, specifically, why the spectral widths of the local minimum and maximum turn out to be approximately equal? A possible reason is that the energy levels that lie within these extrema should have approximately the same energy. The electrons excited by the p_1 pulse have the same energy, and these electrons, each emitting one LO phonon, should undergo transitions to approximately the same level. If we assume that the local minimum represents a gap in the spectrum of the energy states of electrons, it is worth noting that monochromatic radiation is required to form the gap. For the spectrum of picosecond superluminescence, monochromatic radiation, in the extreme case, is the radiation caused by the recombination of electrons from the same level.

In our opinion, the issues analyzed in Section 3 are of interest for subsequent investigations both from a physical and practical points of view. The practical interest is due to the need to understand the physical processes that may occur in ultra-high-speed semiconductor optoelectronic devices, whose operation is based on the use of powerful ultrashort pulses of induced radiation and pumping (more than one pulse simultaneously).

ACKNOWLEDGMENTS

This study was supported by the Russian Foundation for Basic Research, project no. 01-02-16694.

We thank V.I. Perel', Yu.D. Kalifati, and G.N. Shkerdin for their helpful participation in discussions and useful advice.

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Translated by N. Korovin