FEMTOSECOND PULSES AND PROCESSES

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Properties of electrons and holes during femtosecond cooperative emission

P P Vasil'ev, H Kan, H Ohta, T Hiruma

In memory of academician Nikolai Gennadievich Basov

Abstract. **Spectral and energetic characteristics of the cooperative recombination of high density electrons and holes in bulk GaAs are experimentally studied at room temperature. It is shown that the properties and parameters of femtosecond superradiant pulses are conditioned by the collective properties of electrons and holes. Electron – hole pairing and the formation of a short-living coherent e – h BCS state distinguish strongly the regime of cooperative emission from all radiative e – h recombination regimes, which have been observed earlier. The dependences of the energy gap (the order parameter), the Fermi energy, and the band gap of the coherent e – h BCS state on the concentration of electron-hole pairs are obtained.**

Keywords: *electron – hole pairs, Cooper states, femtosecond pulses*

1. Introduction

One of the most interesting problems of physics of condensed state at present is a study of collective properties of electrons and holes in semiconductors, including their pairing and formation of Bose condensates [1 – 3]. Excitons are formed at low enough temperatures as a result of pairing of low density electrons and holes. In the high-density case, when the average distance between particles becomes smaller than the exciton radius, due to collective interactions in the medium of electron – hole pairs the properties of electron – hole pairs prove to be similar to those of an ensemble of Cooper pairs in a superconductor [4 – 6]. The presence of a resonance electromagnetic (optical) internal or external field assists often in the formation of such a state. This coherent cooperative state of an electron – hole ensemble is often referred to as an electron – hole BCS state.

It has been shown theoretically that, similarly to Cooper pairs, there is an energy gap in the spectrum of elementary excitations of quasiparticles in an e – h BCS state [4 – 9]. In addition, spectra of optical absorption (amplification) and luminescence of a semiconductor with this state differ strongly from normal spectra of the radiative recombination of electrons and holes [9 – 11]. For instance, there appears a gain region in the long wavelength range at the photon energies corresponding to excitonic levels and lower. This region is connected with the recombination of electrons and holes which are at the coherent BCS state.

On the other hand, we have already paid attention [12 – 14] to the fact that the regime of the cooperative spontaneous recombination in semiconductor laser structures, described in Refs [15 – 17], can be explained by the formation of a short-living coherent e – h BCS state. Due to a special geometry of the structure and the original method of pumping, we managed to obtain a very high density of electrons and holes [$(2-6)\times10^{18}$ cm^{-3}]. We found that the line center of the cooperative electron – hole recombination lies 15 – 20 and 35 – 40 meV lower than the center of lasing and normal spontaneous emission lines, respectively. The line shape of the cooperative recombination was originally approximated by a convolution of the densities of states of electrons and holes and the Fermi functions of their energy distributions. This is similar to a description of the recombination for an electron – hole liquid [18 – 20]. This approximation well describes the shape of the emission line but it gives for the temperature of the electron – hole plasmas and the Fermi energy the values of 1 – 3 and 3 – 8 meV, respectively [12], which is difficult to explain.

This contradiction can be readily overcome by assuming that paired electrons and holes form a coherent BCS state during the cooperative recombination. Then the emission line shape is well approximated by a convolution of the densities of states of Fermi quasiparticles obtained by the Bogolubov canonical transformation [4, 5] and their energy distribution functions [13]. In this case, the approximation parameters are the energy gap Δ (the order parameter), the Fermi energy (the chemical potential) of the quasiparticles, the renormalised band gap, and the effective width of the energy levels. This approach allowed us to explain adequately the experimental results [12 – 14].

In this paper, we describes the experiments that are a continuation of the studies published elsewhere [12 – 14]. The aim of the paper is to obtain additional experimental data that confirm the fact of the formation of a coherent electron – hole BCS state. Moreover, it was necessary to find out how parameters of the coherent electron – hole BCS state depend on the pumping intensity, the carrier concentration, parameters of the semiconductor structures, etc.

P P Vasil'ev P N Lebedev Physics Institute, Russian Academy of Sciences, Leninsky prosp. 53, 119991 Moscow, Russia;
e-mail: peter@mail1.lebedev.ru
H Kan, H Ohta, T Hiruma Central Research Lab, Hamamatsu Photonics K.K., 5000 Hirakuchi, Hamakita City, 434 Japan

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2. Experiment

Semiconductor laser structures based on GaAs/AlGaAs heterostructures, which were similar to those described earlier [15, 16, 21], were used in the experiments. The laser structures were grown by the MOCVD technique on *n*-GaAs substrates. A 5–6 μm wide mesa structure was etched in the upper heavily doped *p*-GaAs layer of some samples. The other samples had tapered optical waveguides with a width increasing from 5 to 20–40 μm. The semiconductor structures were photolithographically divided into three sections along the structure axis. Two of them (amplifying sections), which were located at the crystal ends, had a single electrical contact and were pumped by current pulses with an amplitude of 0.4–1.5 A. The repetition rate of the pump current pulses was from 10 Hz to 14 MHz and duration was varied from 1 to 10 ns. The middle (absorbing) section of the semiconductor structure was connected to a dc voltage source. The reverse voltage up to −10 V was applied to this section. A photograph of one of the structures is presented in the inset of Fig. 1b. The total length of the structures was 250, 350, and 450 mm.

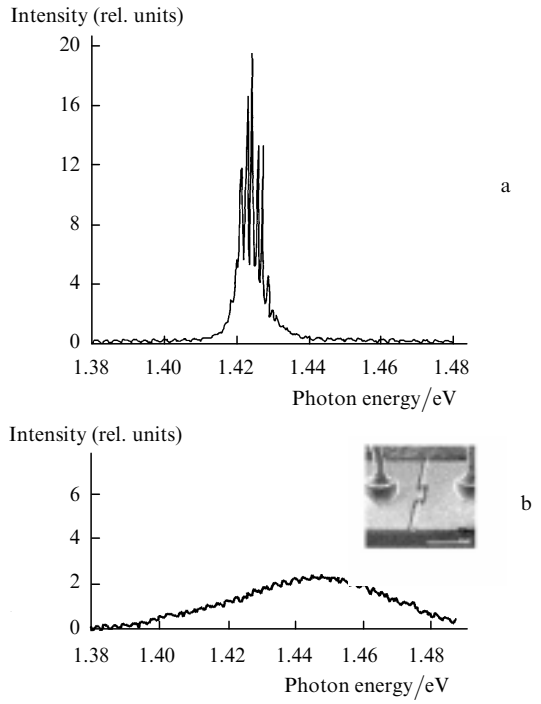

Figure 1. Typical spectra of lasing (a) and amplified spontaneous emission (b) observed from semiconductor structures. Inset: microscopic photograph of a three-section structure.

The lasing threshold under the uniform injection was less that 70–110 mA, depending on the length and width of the active region. Despite a large amplitude of the current pulses, the average current through the structures did not exceed 10–15 mA. This provided the active region temperature at the level of the temperature of the heat sink. All the measurements were carried out at room temperature. Other details of the experimental setup are described in Refs [15, 16].

Three emission regimes are possible in the semiconductor structures, depending on the pumping method and its parameters. They differ from each other both qualitatively and quantitatively. In the first case, lasing takes place under the uniform current injection into all the sections of a structure. A typical lasing spectrum is shown in Fig. 1a. The longitudinal cavity modes formed by the crystal facets are clearly distinguished in the spectrum. The center of the emission line is located at 1.424 eV or the wavelength of 872 nm. Relaxation oscillations are often observed in the time domain until the emission achieves the steady-state level. Passive *Q*-switching occurs when the central part of the structure is disconnected from the current source or when a small reverse voltage ($-1 \div -3$ V) is applied to it. This regime has been studied some long ago and described in detail in the literature (see, for instance, [21]). Output optical pulses under *Q*-switching have a typical duration of 10 ps and longer and a peak power of about 1 W.

In the second case, the amplified spontaneous emission (ASE) is observed when the amplitude of pumping pulses applied to the amplifying sections is not large enough or when the reverse voltage across the central section is very large. In this case, the output power is small (tens and hundreds of microwatts) and a long nanosecond optical pulse is emitted. A typical ASE spectrum is shown in Fig. 1 b. The spectrum is very broad, its centre being located at 1.45 eV (857 nm).

And, finally, the third and most interesting case is the cooperative recombination of electron–hole pairs, which has been described for the first time in Refs [15,16]. Recall that the output pulse of the collective recombination has a typical duration of 300–500 fs and a peak power of above 20 W. Coherent beatings of optical field with a frequency of around 1 THz and a doublet optical spectrum are observed for some samples [15]. The centre of the cooperative emission line in this regime is always strongly shifted to the red and corresponds to the recombination of electrons and holes which are condensed at the very bottom of the bands.

Fig. 2 shows typical optical spectra of femtosecond pulses of the cooperative recombination from the semiconductor structure H2-33, which were obtained upon increasing the reverse voltage applied to the middle section of the structure. One can clearly see that the centre of the spectrum shifts to the red when the reverse voltage increases. The minimum photon energy at the centre of the spectrum is 1.398 eV and that at the long wavelength end of the spectrum is 1.392 eV. Note that the edge of the band gap of intrinsic GaAs at 300 K without taking into account the gap narrowing with increasing the carrier concentration corresponds to the photon energy of 1.424 eV. The shape of the cooperative recombination spectra is often asymmetric and is characterised by a steep long-wavelength wing and smooth short-wavelength wing. We measured the cooperative recombination spectra for many samples to obtain parameters of the coherent BCS state presented in the next section.

Some samples exhibited doublet spectra at the reverse voltage above 8 V (Fig. 3). In this case, the centre of the emission line lay even at a lower energy (1.392 eV). The doublet spectra were observed due to coherent beating of two spatial areas of the cooperative BCS state, which was observed earlier [15, 16].

It is obvious that due to intraband relaxation, only a part of all electrons and holes injected into the active region takes part in the coherent BCS state. The rest of them recombine spontaneously and independently from each other.

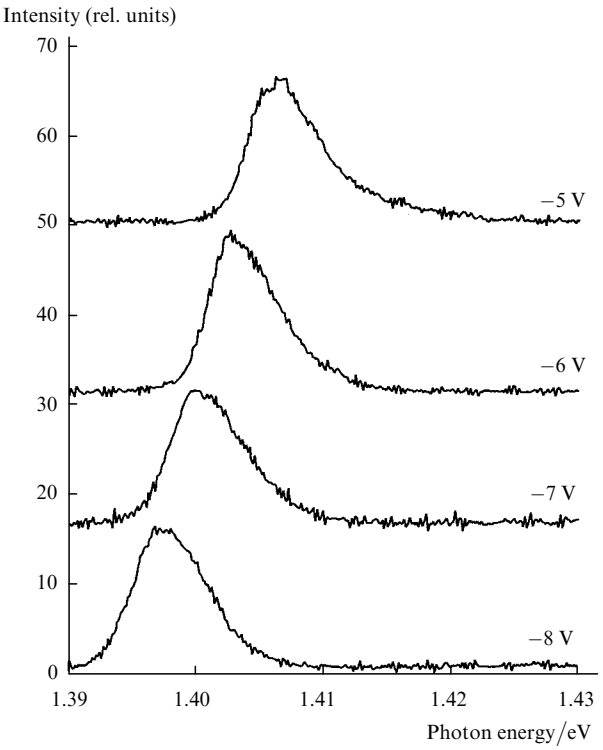

Figure 2. Cooperative emission spectra for different reverse voltages across the centre section of the H2-33 sample.

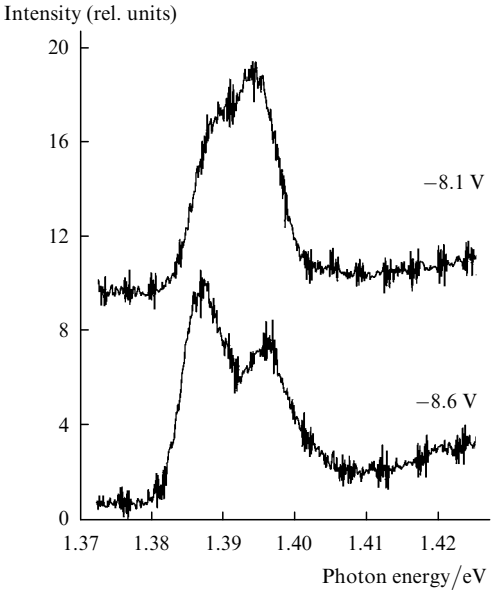

Figure 3. Cooperative emission spectra of the H2-32 sample with a doublet structure at a large reverse voltage.

We estimated the concentration of electrons and holes in the BCS state by measuring variations in the time delay of the generation of cooperative recombination pulses, the pulse energy, as well as the energy and power of the spontaneous background for different values of the reverse voltage and pumping current amplitudes. It turned out that the concentration of electron–hole pairs could be readily changed by varying the reverse voltage across the central section of the samples.

Fig. 4 shows the dependences of the time delay of the generation of femtosecond pulses of the cooperative recombination on the reverse voltage for samples H2-101 and H2-33 at two pumping currents. The femtosecond pulses appear on the leading edge or near the top of the nanosecond pulse of the spontaneous recombination.

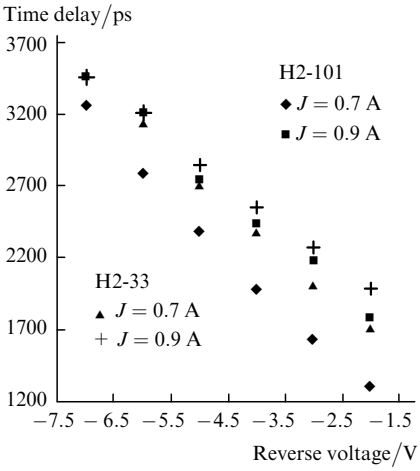

Figure 4. Time delay of the onset of a cooperative emission pulse as a function of the reverse voltage for H2-101 and H2-33 samples at different currents.

The delay appears because a cooperative BCS state can be formed only when a large enough number of electron–hole pairs is accumulated. One can clearly see from Fig. 4 that the generation of a cooperative pulse is delayed when the reverse voltage is increased. The increase in the reverse voltage results in the enhancement of absorption at the central part of the semiconductor structure and in the red shift of the absorption edge. In other words, higher concentrations of electrons and holes are required. Note that the observed dependences are linear.

We calculated the concentration of electron–hole pairs in the BCS state using the following technique. A charge accumulated in the active region before the emission of a cooperative recombination pulse was calculated using the known values of the amplitude and duration of the current pulses and the pumping efficiency in these structures [16]. The period of the optical pulses was also measured. Then, the number of electrons and holes in the active region was calculated. Their concentration was calculated using the dimensions of the active region (length, width, and thickness). A portion of the electrons and holes that were in the coherent BCS state and a portion of the electrons and holes that recombined spontaneously were determined using the experimental data (the optical pulse energy and the energy of the spontaneous background between the pulses). The concentration of the former was in a range of $(2-6)\times 10^{18}$ cm^{-3}, depending on the pumping conditions.

An increase in the carrier concentration with increasing reverse voltage is also confirmed by the dependence of the spontaneous emission intensity on the reverse voltage. The intensity of spontaneous emission from the samples H2-32 and H2-33 as a function of the reverse voltage for different pumping currents is presented in Fig. 5. One can see that the spontaneous emission intensity increases with increasing absolute value of the voltage.

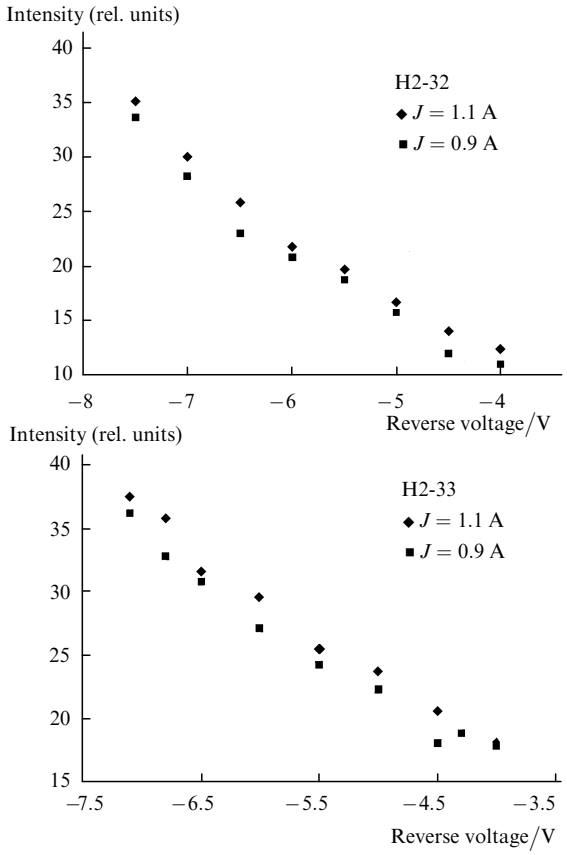

Figure 5. Intensity of the spontaneous background for H2-32 and H2-33 samples at the pumping currents of 1.1 and 0.9 A. (V),

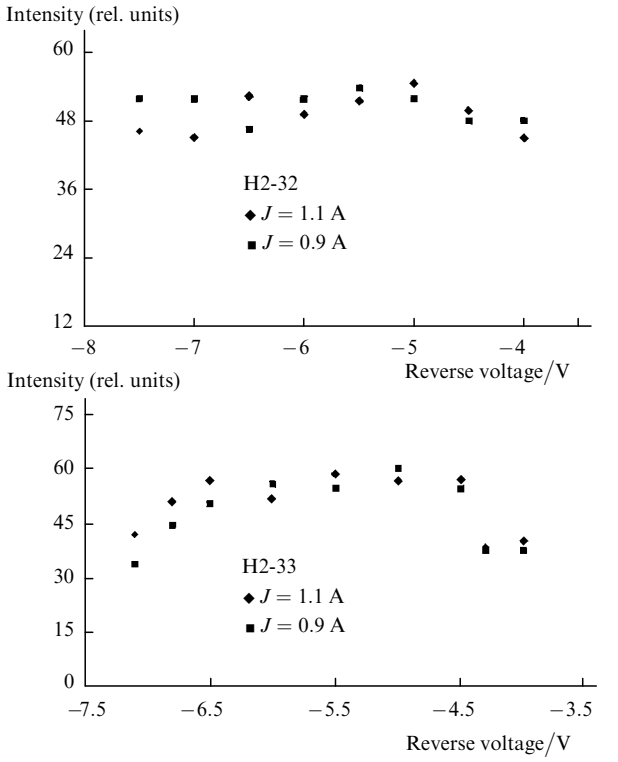

Figure 6. Energy of cooperative emission pulses for H2-32 and H2-33 samples at the pumping currents of 1.1 and 0.9 A.

A completely different picture is observed for the dependences of the energy of cooperative emission pulses on the reverse voltage. Fig. 6 shows these dependences for samples H2-32 and H2-33 for different pumping currents. One can see that in the first case (the upper graph), the pulse energy does not virtually depend on the reverse voltage, i.e., on the concentration of electron–hole pairs. In the second case, it is difficult to make a decision about the existence of any dependence. However, it is likely that the amplitude of the pumping current also does not affect the energy of the generated pulses.

3. Discussion

To understand better the dynamics of accumulation of electrons and holes in the laser active region and the dynamics of spontaneous emission, we calculated the carrier concentration using the rate equations. The calculations confirmed an increase in the concentration upon the delay of cooperative recombination pulses caused by the increase in the reverse voltage. The concentration increases almost linearly in the time delay range of about 3–4 ns, which corresponds to the leading edge of the spontaneous emission pulse.

To obtain the information about parameters of the coherent electron–hole BCS state, we used the method of approximation of cooperative recombination spectra discussed in detail in Refs [13, 14]. The cooperative recombination spectra $I(\hbar\omega)$ are well described by a simple convolution of the density of states $\rho(E)$ of quasiparticles and distribution functions $v^2(E)$ of quasiparticles as

$$I(\hbar\omega) = I_0 \int_{E_g'}^{\hbar\omega} \rho(E)\rho(\hbar\omega - E_g' - E) \times v^2(E)v^2(\hbar\omega - E_g' - E)\mathrm{d}E, \quad (1)$$

where $\hbar\omega$ is the quantum energy; E_g' is the renormalised band gap; I_0 is a constant. The energy spectrum of quasiparticles has a gap Δ, which is bounded above by the binding energy of an e–h pair (an exciton), which is about 4 meV for GaAs. The density of states $\rho(E)$ of quasiparticles has a singularity at $E = \Delta$. To eliminate the divergence, it is necessary to take into account the width of energy levels by introducing the effective density of states [13].

As with Cooper pairs, the distribution function $v^2(E)$ of the quasiparticles is very close to the Fermi distribution with $kT = \Delta/1.76$ and can be determined by numerical methods [6–8]. We approximated it by the function

$$v^2(E) = \frac{1}{2}\left\{1 - \frac{E-\mu}{\left[(E-\mu)^2 + \Delta^2(E)\right]^{1/2}}\right\}, \quad (2)$$

where μ is the Fermi energy, and the gap Δ depends, generally speaking, on the quasi-momentum (energy) and should be calculated self-consistently [6–8].

Thus, we can estimate the parameters of the electron–hole BCS state by approximating the observed cooperative recombination spectra by expressions (1) and (2). One of such spectra is shown in Fig. 7. The experimental data are shown by the asterisks, and the approximation is shown by the solid line. The parameters of the curve are the values of Δ, μ, E_g', and the effective width Γ of energy levels. One can see that the calculated curve fits very well to the experimental data. Such an agreement was achieved for all cooperative emission spectra without any exceptions.

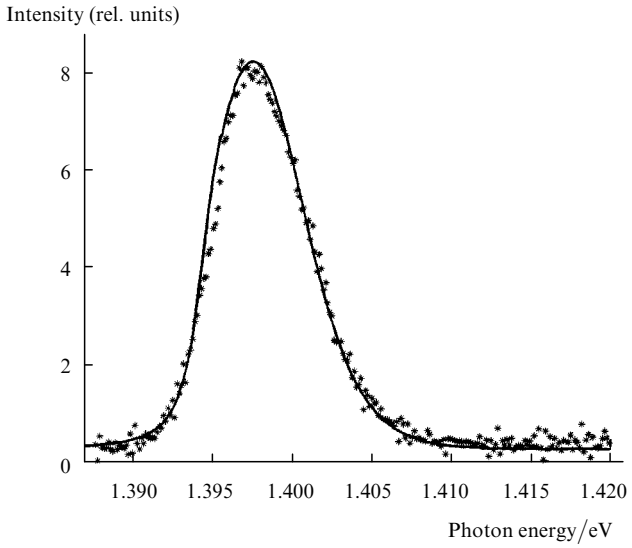

Figure 7. Typical approximation of the cooperative emission spectrum according to Eqns (1) and (2) at $E'_g = 1.392$ eV, $\mu = 7.1$ meV, $\Delta = 2.26$ meV, $\Gamma = 1.2$ meV.

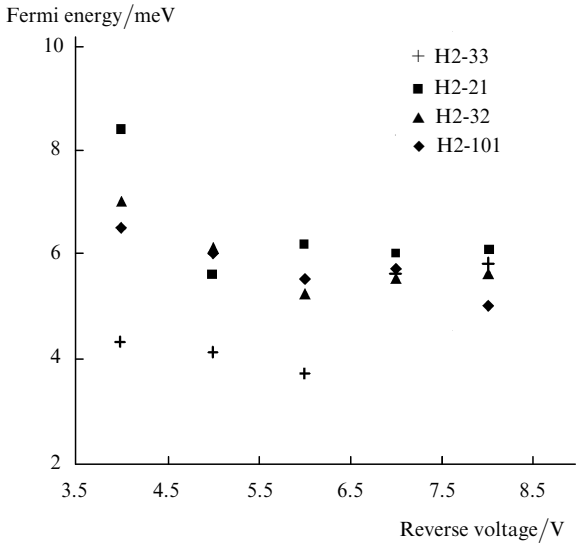

Figure 9. The Fermi energy of quasiparticles as a function of the reverse voltage (the concentration of electron–hole pairs) for the same samples as in Figure 8.

Dependences of the parameters of the coherent BCS state on the reverse voltage (the electron–hole concentration) for four samples are presented in Figs. 8–10. One can see that, in accordance with theoretical predictions, the energy gap Δ for all the samples decreases with increasing reverse voltage. The value of Δ is more than two times smaller than the exciton binding energy, being the same for all the samples within an accuracy of 10 %. The plots of the Fermi energy are shown in Fig. 9. Note that the Fermi energy of the quasiparticles in the coherent BCS state at the achieved concentrations is much smaller than that of electrons (over 100 meV) in GaAs.

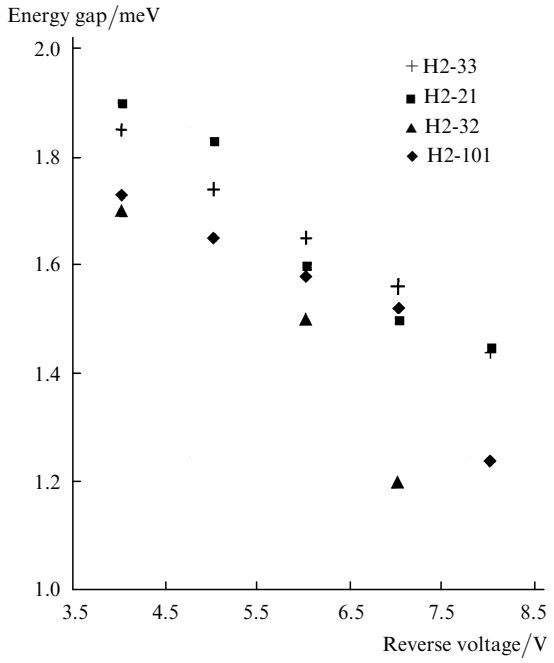

Figure 8. Dependence of the energy gap on the reverse voltage for four samples.

Fig. 10 shows the dependences of the renormalised band gap E'_g for the approximation according to expressions (1) and (2) for four samples (Fig. 10a) and the calculated dependence for GaAs [22, 23] (Fig. 10b). One can see that the band gap narrows down in fact linearly with increasing reverse voltage (the electron–hole concentration). The values of E'_g presented in Fig. 10b that were calculated for concentrations of $(2-7)\times 10^{18}$ cm^{-3} coincide well with the values in Fig. 10a. This confirms indirectly that we deal with very large carrier densities, which greatly exceed the values typical for the laser generation.

The red shift of the cooperative emission line with increasing reverse voltage can be caused by the following reason. The absorption edge of the central (absorbing) section of a sample shifts to the red when the voltage across this section is increased (the Franz–Keldysh effect). For the electron–hole paring and the appearance of a coherent BCS state in the amplifying sections of the structure, an inceptive coherent phasing field is required, capable of travelling freely through these regions [12,13]. This can be achieved by increasing the carrier concentration when the band gap of the amplifying sections of the sample decreases due to the filling of the bands by carriers. In this case, the emission, which comes from the very edge of the band, goes out of the long-wavelength absorption edge of the central section of the structure.

Let us now estimate the critical density of e–h pairs, which is required for their condensation. It is known [1] that a system of electrons and holes with the concentration n passes the condensed state when the so-called quantum-degeneracy criterion

$$n\lambda_D^3 > 1, \tag{3}$$

is valid, where $\lambda_D = (2\pi\hbar^2/MkT)^{1/2}$ is the de Broglie wavelength; M is the mass of an electron–hole pair. In GaAs, the electron mass is $m_e = 0.066$, the heavy hole mass is $m_{hh} = 0.45$, and the light hole mass is $m_{lh} = 0.084$. According to (3), the critical density slightly exceeds 5×10^{18} cm^{-3} at room temperature if the pairs are formed by heavy holes.

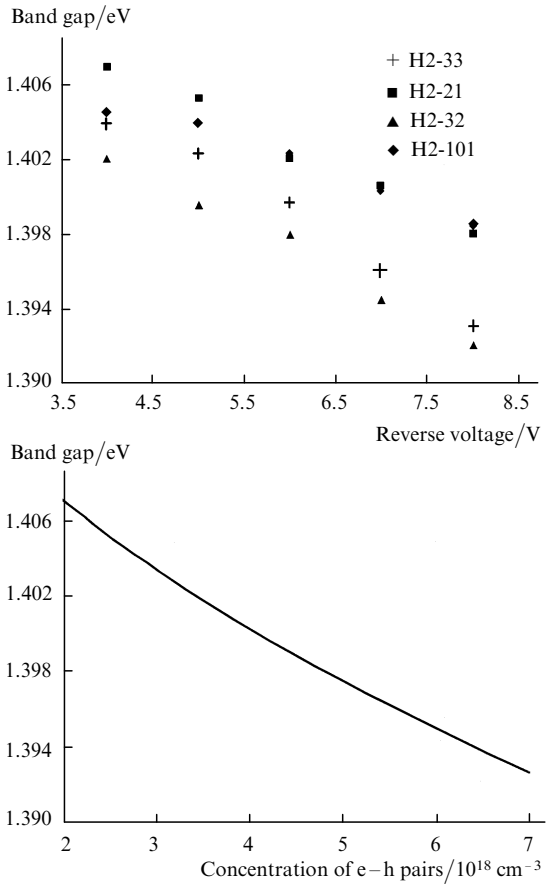

Figure 10. Dependence of the renormalised band gap E'_g on the reverse voltage (a) and the calculated dependence of E'_g on the concentration of electron–hole pairs for GaAs at room temperature (b).

For light holes and a combination of heavy and light holes, this value is a few times smaller and the condensation may start from pairs formed by electrons and light holes. According to the formulae presented in [4], the critical concentration of the condensation of light hole–electron pairs at room temperature is 2.3×10^{18} cm^{-3} only. The experimentally obtained densities of electron–hole pairs in a coherent state exceed this value.

At the obtained high densities and room temperature, excitons do not exist. The exciton binding energy in GaAs is about 4 meV, the exciton radius a_0 is 140 Å. The mean interparticle distance $r_s = (3/4\pi n)^{1/3}$ is in the range of 34–53 Å at the carrier densities of $(2-5) \times 10^{18}$ cm^{-3}. Thus, the coherent state of paired electrons and holes in our case should be similar to the state of electrons in a superconductor when the size of a Cooper pair is much larger than the distance between the electrons.

In our case, the crucial role in electron–hole pairing and the establishment of coherency in the ensemble belongs to an electromagnetic field, which always exists in the semiconductor. Because the photon momentum is small compared to the momenta of electrons and holes, the field creates and destroys pairs with zero total momentum. Thus, a coherent ensemble of particles (a coherent state) with zero total momentum arises from all the electrons and holes when the electromagnetic field induces correlations in the e–h system at the early stage of the development of cooperative emission. Due to the exponential rise of the phasing coherent field towards the structure ends, the cooperative state is highly spatially nonuniform and is located mainly at both facets of the crystal. The collective radiative decay of this coherent ensemble is observed as a high-power femtosecond pulse.

All the electrons and holes of the collective state are paired, the dimension of the pairs being much larger than the mean interparticle distance [4–9]. Similarly to Cooper pairs, the total momentum of each pair is equal to zero. That is why such a collective state is called an electron–hole BCS-like state. By analogy, the coherent electron–hole BCS state should have the minimum energy, and photon energy should also be minimal during its recombination.

It is known that the dissipation of Cooper pairs on each other does not lead to the loss of coherence in the ensemble. In our case of the cooperative e–h state, we have a similar situation. It has been experimentally found [15, 16] and confirmed theoretically [17] that coherent properties of the interaction of the optical field with the e–h system persisted for anomalously long time, much longer that the transverse relaxation time $T_2 < 100$ fs. Moreover, a classical case of superradiance should be observed at times knowingly shorter than 100 fs.

The experimentally observed coherent beatings, which lasted for a few picoseconds [15] suggest that the coherence in the cooperative state is preserved for a few hundreds of femtoseconds although the time between collisions in the ensemble is less than 10 fs. This can be explained by the fact that no loss of coherence occurs upon scattering of electrons and holes within the ensemble. The coherence of the ensemble is also preserved in collisions with external electrons and holes, which do not belong to the cooperative state because the ensemble seems to be involved in collisions as a whole, like a large molecule. Since the number of electrons and holes in the cooperative state is very large (about 10^8) and its total mass greatly exceeds that of a single electron (hole), the coherence of the ensemble does not virtually change upon the interaction. In other words, the phase relaxation time of the coherent ensemble should be larger than the time T_2 and should increase with increasing number of particles.

4. Conclusions

The experimental data presented above suggest that the emission of high-power femtosecond pulses from semiconductor structures is caused by radiative recombination of a cooperative state of electrons and holes. This unstable coherent state with a lifetime of a few hundred femtoseconds resembles an ensemble of Cooper pairs in a superconductor. It has been shown that the experimentally achieved concentrations of electrons and holes are larger than the critical values required for the condensation of e–h pairs at room temperature.

Experimental data have been obtained for the estimates of parameters of the coherent electron–hole BCS state, including the dependences of the order parameter Δ (the gap in the energy spectrum of quasiparticles), the Fermi energy, and the band gap on the density of injected electron–hole pairs. Note that the BCS-like gap of a degenerate electron–hole system, being smaller than the exciton binding energy (4 meV in our case), decreases with increasing concentration. In turn, the widths of the observed cooperative recombination spectra are quite broad (4–8 meV) due to the femtosecond lifetime of the collective state of electrons and

holes. The large width of the energy levels makes a direct observation of the energy gap in the spectrum of electrons and holes very problematic.

In conclusion, note that the problem of the dynamics of formation of the coherent electron – hole BCS state remains unsolved. Its solution will apparently take great experimental and theoretical efforts.

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