

PACS numbers: 42.50.Fx, 71.35.Ee; 42.55.Px
DOI: 10.1070/QE2002v032n12ABEH002353

Coherent electron – hole state and femtosecond cooperative emission in bulk GaAs

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

***Abstract*. The conditions for obtaining a collective coherent electron – hole state in semiconductors are discussed. The results of the experimental study of the regime of cooperative recombination of high-density electrons and holes (more than 3×10^{18} cm^{-3}) in bulk GaAs at room temperature are presented. It is shown that the collective pairing of electrons and holes and their condensation cause the formation of a short-living coherent electron – hole BCS-like state, which exhibits radiative recombination in the form of high-power femtosecond optical pulses. It is experimentally demonstrated that almost all of the electrons and holes available are condensed at the very bottoms of the bands and are at the cooperative state. The average lifetime of this state is measured to be of about 300 fs. The dependences of the order parameter (the energy gap of the spectrum of electrons and holes) and the Fermi energy of the coherent BCS state on the electron – hole concentration are obtained.**

Keywords: *Bose condensate, coherent state, ultrashort light pulses, semiconductor laser*.

1. Introduction

Electron – hole systems in semiconductors and their interactions with resonant electromagnetic fields have been among foremost topics in condensed matter physics for a long time [1 – 3]. One of the most notable features of such systems is their ability to form macroscopic quantum states under appropriate conditions. For instance, depending on the $e-h$ density, a macroscopic quantum state can be either an excitonic Bose – Einstein condensate (the low-density regime) or a cooperative $e-h$ state, which is similar to the collective state of Cooper pairs in a superconductor (BCS state, the high-density regime) [3]. Quantum-well and microcavity structures, excited by an optical coherent field, have been generally considered as the most promising candidates for the observation of these macroscopic quantum states [4 – 7].

One of the key points of the formation of a Bose condensate is the process of pairing of electrons and holes and formation of either single bound states (excitons) or a cooperative state under the high density regime (collective pairing). Such a coherent cooperative state of an electron – hole ensemble is often called the electron – hole BCS state [3]. The presence of a resonant internal or external electromagnetic (light) field assists frequently to the pairing process.

However, unlike a Bose condensate obtained in atomic traps, and BCS states of Cooper pairs in superconductors, any cooperative state of electrons and holes in semiconductors must be intrinsically unstable due to their recombination. It is obvious that the collective decay of a coherent electron – hole state must possess properties of superradiance.

In recent papers [8 – 11], the author studied superradiance in a system of electrons and holes of high density, where the coherent interaction of an optical field with an active semiconductor was discovered for the first time. It was also demonstrated how a strong optical gain in the semiconductor could overcome a destructive role of the phase relaxation. Moreover, it was shown that properties of the cooperative emission could be explained by the condensation of electron – hole pairs and by the formation of an unstable coherent BCS-like electron – hole state at room temperature [12 – 14]. The main parameters of this state were determined, including the order parameter Δ.

In this paper, we summarise the experimental data available and published earlier and present a number of new results on the investigation of the coherent cooperative electron – hole state in bulk GaAs.

2. Conditions of the condensation of electron – hole pairs

It is well known that, unlike electrons and holes, which are Fermi particles, a bound electron and hole (an exciton) is a Bose particle (at the first approximation at least). Hence, an ensemble of excitons should experience Bose condensation under certain conditions. The exciton condensation has been really observed at very low temperatures and low densities upon optical pumping of Cu_2O [4] and excitation of GaAs/AlGaAs quantum-well structures by picosecond laser pulses [15].

Since an electron and a hole have opposite electric charges and the Coulomb attractive forces act between them, they can get paired spontaneously without any additional physical mechanism. This distinguishes electron – hole pairs from Cooper pairs where the attraction

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Received 28 June 2002
Kvantovaya Elektronika **32** (12) 1105 – 1112 (2002)
Translated by P.P. Vasil'ev

between the electrons is determined by the interaction with phonons. However, it is worth to point out that in our case a resonant optical field plays a great role in the electron–hole pairing and in establishing of coherency in the ensemble. This field is constantly present in the system due to the electron–hole recombination.

The process of pairing and formation of a coherent ensemble of electrons and holes is illustrated in Fig. 1. Let an unbound electron and a hole and a photon be at the beginning. The elctrons and holes are produced upon pumping the semiconductor by intense current pulses (see Section 3). Photons are constantly generated due to the radiative recombination of electrons and holes. A photon can cause the recombination of an electron–hole pair with the generation of another photon, which is absolutely identical to the original photon. Thus, at the intermediate stage, we have two coherent photons. There also exists an opposite process of the generation of an electron–hole pair from a photon. Therefore, at the final stage there exist the photon and the bound electron and hole, which are coherent with the photon field.

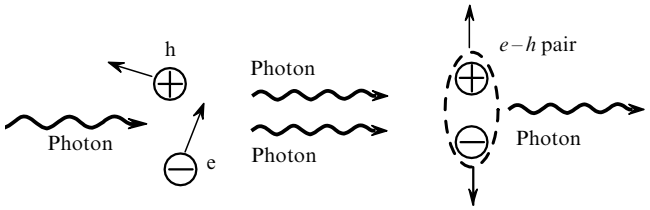

Figure 1. Photon-mediated paring of an electron and a hole.

The optical field generates $e-h$ pairs with a total wavevector equal to zero because the photon wavevector is small compared to that of electrons and holes. Hence, at the initial stages of evolution a coherent ensemble (the cooperative state) with zero total wavevector originates from all electrons and holes when the electromagnetic field induces correlations in the electron–hole system. Due to the exponential increment of the phasing coherent field towards the ends of the structure, the cooperative state is highly nonuniform in space. It is located preferably near the both facets of the crystal. It is obvious that collisions between particles prevent the establishment of coherency. We discuss this issue below.

Let us now find out under which conditions the condensation of $e-h$ pairs is possible. A system of electrons and holes with the density n transforms into the condensed state, when the so-called quantum-degeneracy criterion is satisfied [3]

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

where $\lambda_D = (2\pi\hbar^2/MkT)^{1/2}$ is the de Broglie wavelength and M is the mass of an $e-h$ pair. In bulk GaAs, the electron mass is $m_e = 0.07$, the heavy hole mass is $m_{hh} = 0.5$, and the light hole mass is $m_{lh} = 0.08$ (in units of the free electron mass). At room temperature, λ_D is equal to about 107 and 60 Å for a pair of electron-light hole and pair of electron-heavy hole, respectively. One can see from Eqn (1) that an increase in the concentration relaxes requirements imposed on the condensation temperature. Assuming that an electron–hole pair is an ideal boson, the exact value of the condensation temperature can be found from the relation [16]

$$n\left(\frac{2\pi\hbar^2}{MkT}\right)^{3/2} = 2.612. \tag{2}$$

The dependences of the condensation temperature on the electron–hole pair concentration are shown in Fig. 2 for different values of the mass of the pair. One can see that to achieve condensation at room temperature, a higher $e-h$ density is required when the mass of the pair increases. Fig. 2 shows that for achieving condensation in GaAs at room temperature in an ensemble of electron–light hole pairs, the concentration should be $n \approx 2.4 \times 10^{18}$ cm^{-3}. Due to a special geometry of the structure and the original method of pumping, we managed to achieve a very high density of electrons and holes [$(2-6) \times 10^{18}$ cm^{-3}] in our experiments [8, 9]. At such densities of electron–hole pairs, the average interparticle distance $r_s = (3/4\pi n)^{1/3}$ is from 49 to 34 Å, which is smaller than the de Broglie wavelength calculated above. Therefore, the de Broglie wavelengths of individual pairs overlap in space, and we may expect the onset of the condensation in the electron–hole ensemble.

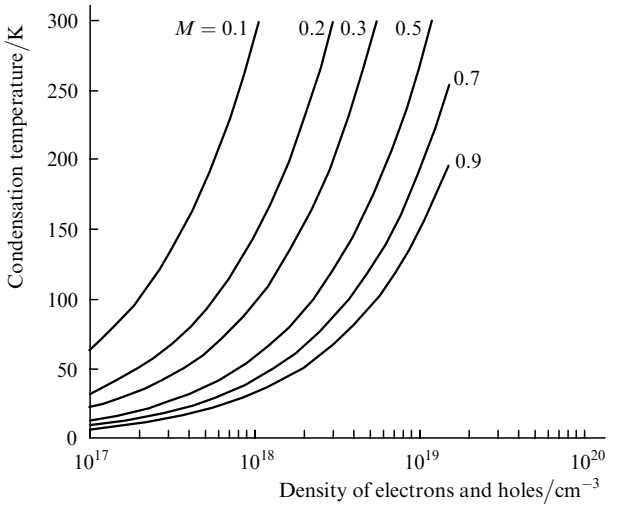

Figure 2. Dependences of the condensation temperature on the density of $e-h$ pairs for different values of the effective mass M of the pair (in units of the free electron mass).

In our case of high densities and room temperature ($kT \approx 25$ meV), one cannot consider excitons (the exciton binding energy in bulk GaAs is around 4 meV and the exciton radius a_0 is about 140 Å). Thus, the coherent state of paired electrons and holes should be similar to the state of an ensemble of electrons in a superconductor when the characteristic size of Cooper pairs is much larger than the average distance between the electrons (the pairs overlap strongly in space). It is the collective radiative decay of this coherent $e-h$ ensemble that is experimentally observed in the form of a high-power femtosecond pulse [12, 13]. Note that the transition of the system into the coherent state must be abrupt because, when the nearest $e-h$ pairs are being phased, they make the rest pairs to be polarised by their total Coulomb field and force them to make a transition into the coherent state. Therefore, if electron–light hole pairs underwent a transition into the cooperative state, they can

facilitate the transition of electron-heavy hole pairs into this state, because the critical temperature for the former pairs is much smaller than that for the latter at the same concentration.

3. Dynamics of the formation of the coherent state and its temporal characteristics

Three-section semiconductor laser structures based on p–i–n GaAs/AlGaAs heterostructures (see Fig. 3a), which are similar to those described earlier, were used in the experiments. All the details of the experimental setup can be found elsewhere [8, 9, 12, 13]. The structures were pumped by nanosecond current pulses with an amplitude of up to 1 A.

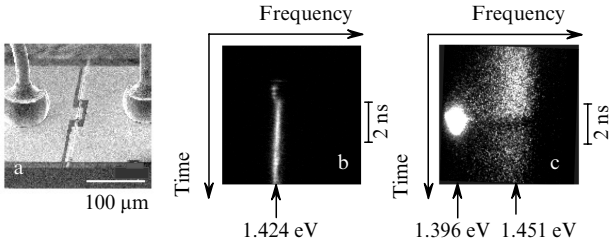

Figure 3. Microscopic photographs of a three-section laser structure (a) and time-resolved spectra of lasing (b) and cooperative-emission pulses (c) taken from the screen of the streak camera. The arrows show the positions of the specral lines of lasing, the cooperative, and spontaneous emission.

There exist three kinds of the generation in semiconductor structures depending on the pumping technique and its parameters. They differ both quantitatively and qualitatively from each other. First, lasing is observed under the uniform current injection into all sections of the structure. Passive Q-switching is observed when the central section is disconnected from the current source or when a reverse bias of a small amplitude (1–3 V) is applied to it. This regime has been studied for a long time and described in detail in the literature (see, for instance [17]). In this case, the output optical pulses upon Q-switching have a typical duration of 10 and more picoseconds and the peak power of the order of 1 W.

Second, the regime of amplified spontaneous emission is observed when the amplitude of the pulses pumping the amplifying sections of the structure is not large enough or when the reverse voltage across the central section is very large. In this case, the power of the output emission is low (tens and hundreds of microwatts), and a long nanosecond optical pulse is generated.

And, finally, the third and the most interesting case is the regime of the cooperative recombination of electron–hole pairs, which was described for the first time in [8, 9]. We investigated the spectral dynamics of the formation of the electron–hole BCS state using two-dimensional frequency–time diagrams detected by a single-shot streak camera with a temporal resolution of 1.5 ps. Typical frequency–time diagrams of standard lasing emission and cooperative recombination pulses are presented in Figs 3b, c, respectively. The intensity relaxation oscillations are observed at the beginning of the laser generation (Fig. 3b, 3–4 pulses at the front of the trace). This is quite typical for semiconductor lasers. Note that the trace is narrow enough, which implies a narrow spectrum. The trace centre is located virtually at the same place on the frequency axis, i.e., the central frequency varies insignificantly during lasing. The spectral peak is located approximately at 1.424 eV, as measured separately using a diffraction spectrometer.

A completely different picture is observed in the regime when the $e-h$ BCS state is formed and femtosecond pulses of the cooperative emission are generated (Fig. 3c). The broad vertical stripe represents the ordinary spontaneous emission of non-paired electrons and holes, whereas the bright spot on the left at the center of the photograph is the powerful cooperative emission pulse due to the recombination of the $e-h$ BCS state. Because the peak power of the superradiant pulse is typically $10^4 - 10^5$ times larger than the spontaneous background, its image on the picture is distorted due to the overexposure. It is clearly seen in Fig. 3c how the cooperative pulse develops from the spontaneous emission. As the carrier density increases in time under the action of a nanosecond current pulse, the band gap narrows down and the spontaneous recombination of electrons and holes occurs at lower and lower photon energies. When the density of $e-h$ pairs becomes high enough, the de Broglie wavelengths of individual $e-h$ pairs start to overlap and the quantum-degeneracy criterion Eqn (1) is fulfilled. It is at this moment that the $e-h$ BCS-like state is formed. The phasing of wavefunctions of individual $e-h$ pairs occurs via the common optical field. At the leading edge of the cooperative pulse, the macroscopic polarisation appears, which is typical for superradiance (see figures in paper [10]).

Perhaps the most interesting feature of Fig. 3c is a dark stripe, which goes across the spontaneous emission at the instance of time when the $e-h$ BCS state is formed and recombines. It means the absence of spontaneous emission at this moment, i.e., that there are almost no electrons and holes in the bands, which can recombine spontaneously. In other words, all the electrons and holes are condensed at the very bottoms of the bands and form the BCS state at this time. Upon the radiative recombination of this state, photons with a minimum possible energy are emitted. In our case, the line of cooperative emission is located at 1.396 eV, while the spontaneous emission line is located at 1.451 eV. Recall that the non-renormalised band gap in bulk GaAs at room temperature is 1.424 eV.

To prove this important fact, we calculated the intensity of total spontaneous emission upon recombination of electrons and holes, which occupy energy levels inside the bands, and plotted its dependence on time (Fig. 4). One can see that spontaneous emission almost vanishes when the $e-h$ BCS state recombines (as shown by the arrow).

Such a spectral dynamics is completely different from the dynamics of lasing or spontaneous emission. Indeed, when lasing appears in a semiconductor, the spontaneous emission intensity becomes fixed over the entire spectral range at the threshold level. Because of ultrafast intraband relaxation in the semiconductor, it is impossible to burn a spectral hole in the energy distribution of electrons and holes and to reduce spontaneous emission near the lasing line and all the more at remote energy levels. It is known that the coherence of individual $e-h$ pairs in semiconductor lasers is destroyed for 10–100 fs. As a result, under typical operating conditions, the ratio of the number of photons in the sample to

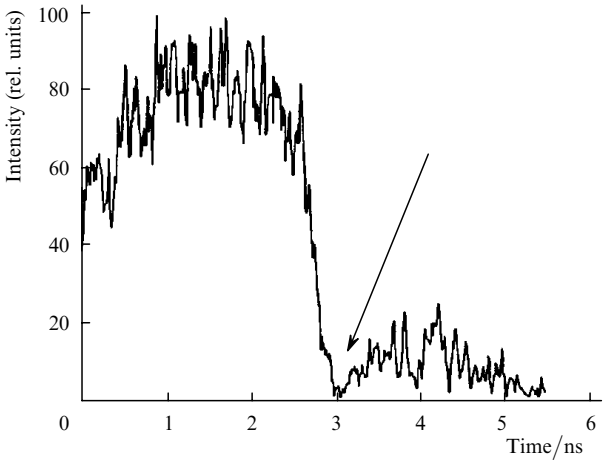

Figure 4. Time dependence of the spontaneous emission intensity. The arrow shows the position of the superradiant pulse.

the number of $e-h$ pairs is $N_{ph}/N_{e-h} \simeq 10^{-4} - 10^{-5}$ [18]. On the other hand, in the superradiant state, the photon field is linearly coupled to the order parameter, and we have $N_{ph}/N_{e-h} \simeq 1$. This means that the number of $e-h$ pairs remained in the structure immediately after the recombination of the electron–hole BCS state should be rather small. This is exactly what we see as the dark horizontal stripe in the middle in Fig. 3c. Then, the intensity of spontaneous emission begins to rise again due to injection of new electrons and holes by the pump current pulse.

Precise measurements of the duration of cooperative pulses were performed using both a streak camera and the SHG autocorrelation technique. Two typical envelopes of cooperative recombination pulses detected by the streak camera are presented in Fig. 5. The pulse duration on the screen is very close to the temporal resolution of the camera (1.5 ps), which corresponds to the actual pulse duration of less than 1 ps. One can see that the main femtosecond pulse lies on a background of duration ~ 10 ps. The nature of this background requires a separate study.

To measure the duration of cooperative emission pulses more accurately, we used the technique based on intensity autocorrelation functions having a femtosecond time resolution. Fig. 6 presents the typical SHG intensity autocorrelation function for superradiant pulses (i.e., the dependence of the SHG intensity on the delay between pulses in a scanning Michelson interferometer). It has a full-width at half maximum (FWHM) of about 460 fs. This value corresponds to an actual pulse duration from 290 (for an asymmetric exponential shape of the envelope) to 320 fs (Gaussian shape) [17]. According to the theory of superradiance [19, 20], the shape of pulses is far from a Gaussian and is closer to the asymmetric exponential shape, i.e., the actual pulse duration is shorter than 300 fs. Note that the duration of superradiant pulses achieved in our experiments is an absolute record among all ultrashort pulses generated by semiconductor lasers without additional pulse compression by external optical elements.

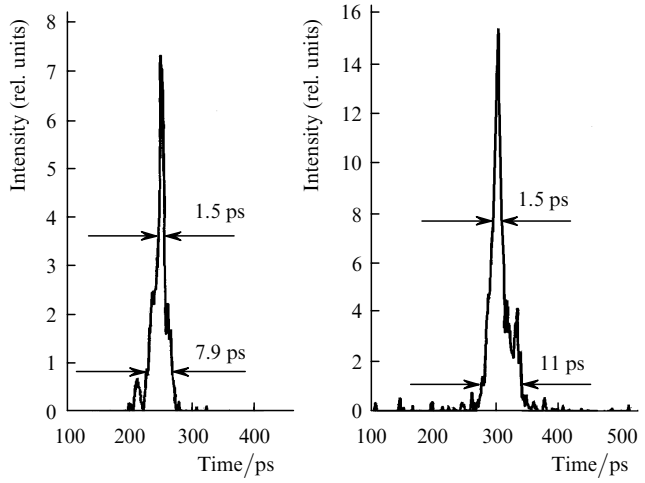

Figure 5. Typical envelopes of cooperative emission pulses recorded with the streak camera.

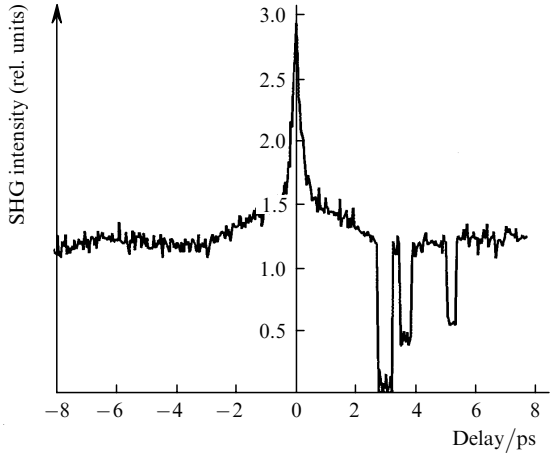

Figure 6. Intensity autocorrelation function of cooperative-emission pulses.

Note that there exists an additional pedestal of duration 1.5–2 ps, on which the main femtosecond pulse is located. This pedestal probably appears due to fundamental quantum-mechanical fluctuations in the system of electron–hole pairs – photons and the uncertainty in the initial phase of the decay of the BCS state. These circumstances result in large fluctuations of the shape of superradiant pulses.

Indeed, as was shown in [5], the appearance of an intense noise is a convincing evidence of the condensation and the establishment of coherency in the system of $e-h$ pairs. It is well-known from statistical physics that the noise amplitude is inversely proportional to the number of statistically independent parameters of the system. Therefore, the experimentally observed intense noise (fluctuations of the shape, duration, and timing jitter) is the obvious manifestation of the fact that the number of independent variables strongly decreases when a macroscopic region of coherency appears, where the electron–hole BCS state is located. Let us point out that optical pulses without the pedestal shown in Fig. 6 are generated at all pulsed operating regimes of the laser structure (mode locking, Q-switching, etc.) when fundamental quantum-mechanical fluctuations do not appear explicitly.

Let us discuss now the coherent properties of the cooperative state and emission. It is known that interactions between Cooper pairs do not lead to loss of coherence in the ensemble [21]. In our case of the cooperative $e-h$ state, we have a similar situation. It was discovered experimentally

that the coherence of the interaction of the optical field with the $e-h$ system was preserved for anomalous long times, much longer than the transverse relaxation time T_2 (less than 100 fs) [8, 9]. Moreover, the classical superradiance should be observed on a time scale shorter than 100 fs [19, 20]. The experimentally observed coherent beating with a frequency of over 1 THz, which lasted a few picoseconds [8], suggests that the coherence in the cooperative state is preserved for a few hundreds of femtoseconds, although the time between collisions of the particles in the ensemble is shorter than 10 fs. Unlike intensity autocorrelation functions (Fig. 6), where all phase information is averaged out, interferometric autocorrelations, which were recorded with the resolution of individual fringes and femtosecond accuracy, allow us to retrace the phase relations of emission.

Fig. 7 shows the experimental interferometric autocorrelation of cooperative-emission pulses from a semiconductor structure, where two regions with a very high $e-h$ density are located close to each other (at a distance of less than 10 μm). The total length of the structure corresponds to the round-trip time of about 3.1 ps. The coherent beating of the photon field is clearly seen. The shape of the trace resembles the beating of two coherent oscillators and suggests the coherent interaction of emission with different parts of the cooperative state upon pulse propagation between the facets of the crystal.

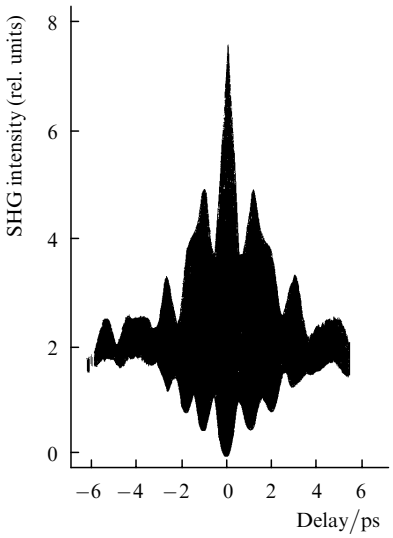

Figure 7. Fringle-resolved autocorrelation of cooperative-emission pulses illustrating the coherence of the interaction of the optical field with $e-h$ pairs of the cooperative state.

This can be explained by the fact that the $e-h$ interactions within the cooperative state do not affect the coherence of individual electrons and holes. The coherence of the ensemble is also preserved upon collisions with external electrons and holes, which do not belong to the cooperative state, because the ensemble probably requires a hit as a whole, i.e., like a large 'molecule'. Since the number of $e-h$ pairs in the cooperative state is huge (about 10^8 [9]) and its total mass is much larger than the electron (hole) mass, the coherence of the ensemble is almost not changed during the interaction. In other words, the phase relaxation time of the coherent ensemble should be longer than T_2 and should increase with increasing number of the particles.

4. Spectral characteristics of emission

As noted in Section 3, lasing and amplified spontaneous emission were observed in addition to the cooperative emission depending on the pumping of the laser sections. Typical spectra of lasing and amplified spontaneous emission are shown in Fig. 8. The longitudinal modes of the cavity formed by the chip facets are clearly distinguished in Fig. 8a. The emission line is located at 1.424 eV ($\lambda = 872$ nm). Relaxation intensity oscillations are observed, which precede the establishment of the steady-state level of emission (Fig. 3b). The spectrum of amplified spontaneous emission is shown in Fig. 8b. It is very broad, and its maximum is located at 1.45 eV ($\lambda = 857$ nm), coinciding with the maximum of the spontaneous emission line obtained using the streak camera (Fig. 3c).

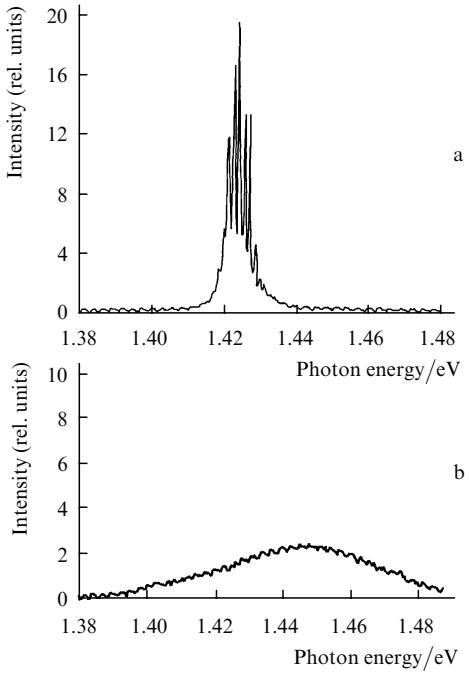

Figure 8. Typical spectra of lasing (a) and amplified spontaneous emission (b) observed in the structures under study.

The typical spectra of superradiant pulses obtained at different pumping levels in the laser structure are presented in Fig. 9. The absolute magnitude of the reverse voltage on the absorber section increases from 4.7 up to 7.0 V. Note that at voltages below −4.2 V, superradiance is less pronounced and is accompanied by lasing. It vanishes completely when the voltage approaches zero, and only lasing occurs in this case. The spectrum of cooperative emission is continuos. One can see from Fig. 9 that the spectrum shifts to the red and its width decreases with increasing the magnitude of the reverse voltage. The longer-wavelength emission band in Fig. 9 is located at 1.397 eV and its long-wavelength tail extends to 1.380 eV.

The red shift of the cooperative emission band with increasing the absolute magnitude of the reverse voltage can be caused by the following reason. The absorption edge of the central section of the sample shifts to the red when the voltage on this section is increased (the Frantz–Keldysh effect). An initial phasing coherent field, which can travel

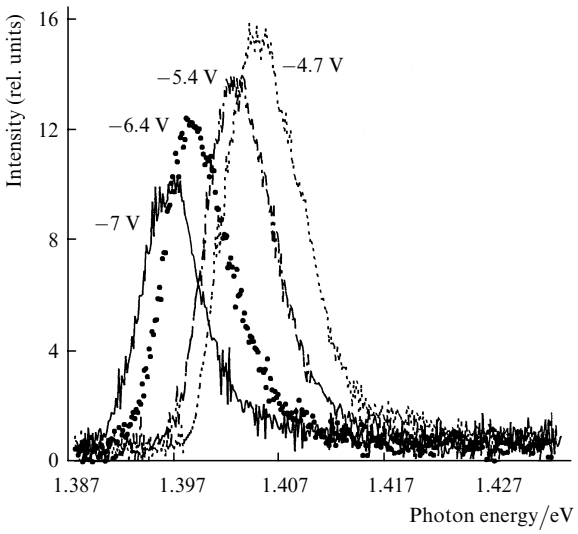

Figure 9. Spectra of cooperative emission for different values of the reverse voltage across the absorbing section of the structure.

freely through the amplifying sections of the structure, is required for electron–hole paring and the emergence of a coherent BCS state in these sections. This is possible only when the carrier concentration is increased and the band gap of the amplifying sections of the sample decreases due to the filling of the band. In this case, the emission, coming from the very edge of the band, lies beyond the long-wavelength absorption edge of the central section of the structure.

5. Determination of parameters of the cooperative state

To obtain information on the parameters of a coherent electron–hole BCS state, we used the method for approximation of spectra of the cooperative recombination, which was described in detail in [12, 13]. The cooperative recombination spectra $I(\hbar\omega)$ are well described by a simple convolution of the density of states $\rho(E)$ of Fermi quasiparticles, obtained by the Bogolubov canonical transformation [1, 2], and the energy distribution function $v^2(E)$ of the quasiparticles:

$$I(\hbar\omega) = I_0 \int_{E_g'}^{\hbar\omega} \rho(E)\rho(\hbar\omega - E_g' - E) \times v^2(E)v^2(\hbar\omega - E_g' - E)\mathrm{d}E, \quad (3)$$

where $\hbar\omega$ is the quantum energy; E_g' is the renormalised band gap; and I_0 is a constant. The validity of expression (3) in our case is conditioned by the following.

It is known that the Fermi distributions in the $e-h$ system are established very rapidly, in times that are only several times longer than the time between electron–hole collisions. In our case of a very large density, the characteristic time between collisions is far shorter than 10 fs. As is shown by the calculations [10] and experiments [8, 9], the characteristic times of cooperative emission are hundreds of femtoseconds, which is much larger than the time of establishing of the Fermi distributions. In addition, moments of the recombination of individual electron–hole pairs are correlated during the cooperative decay of the system, and all the $e-h$ pairs in the ensemble recombine almost simultaneously. This means that the dependence $I(\hbar\omega)$ will be determined by the energy distribution of electrons and holes according to expression (3) and will not be distorted by the reabsorption or amplification in other regions of the structure. And finally, the ultrashort lifetime of the cooperative state implies the broadening of the energy levels and, correspondingly, the integration over the energy. Indeed, according to the uncertainty principle, the shorter the lifetime of an electon or a hole, the broader the energy-level width. Because the cooperative state exists a few hundreds of femtoseconds only, the width of individual energy levels in the bands can be quite large (over 1 meV), which can be taken into account in the approximation of the spectra by expression (3).

The joint density of states $\rho(E)$ of the quasiparticles has a singularity at $E = \Delta$ [22]:

$$\rho \propto \frac{E}{(E-\Delta)^{1/2}}. \quad (4)$$

To eliminate the divergence, the width of the energy levels should be taken into account. This can be done by introducing the effective densities of states. For a simple Lorentzian line shape, the effective density of states can be written as [13]

$$\rho_{\mathrm{eff}}(E) = \frac{1}{\pi}\int \frac{\rho(E)\mathrm{d}E_1}{\pi(E-E_1)^2 + \Gamma^2}, \quad (5)$$

where Γ is the effective width of the levels, which is independent of energy in a simplest case. Fig. 10 illustrates the dependence of the effective density of states of the quasiparticles on the energy [13].

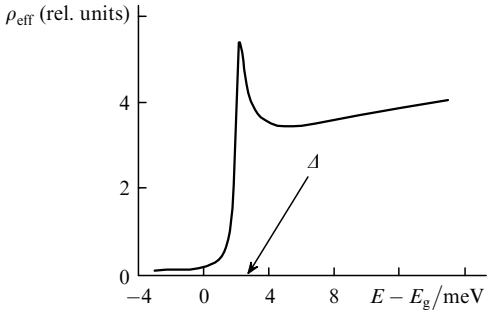

Figure 10. Effective density of states of Fermi quasiparticles as a function of energy. The arrow shows the position of the peak at $E = \Delta$.

As for 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 calculated numerically [22–24]. We approximated it by the function

$$v^2(E) = \frac{1}{2}\left\{1 - \frac{E-\mu}{[(E-\mu)^2 + \Delta^2(E)]^{1/2}}\right\}, \quad (6)$$

where μ is the Fermi energy, and generally speaking, the band gap Δ depends on the wavevector (energy) and should be calculated self-consistently [22–24].

Thus, we can estimate parameters of the electron–hole BCS state by approximating the observed spectra of cooperative recombination using Eqns (3)–(6). One of

such spectra is shown in Fig. 11, where the experimental data are shown by asterisk and the approximation is shown by the solid line. The parameters of the curve are the band gap Δ (the order parameter), the Fermi energy μ, the renormalised band gap E'_g, and the effective width Γ of energy levels. One can see that the calculated curve very well describes the experimental data. Such a good agreement was achieved for all the spectra of cooperative emission without an exception.

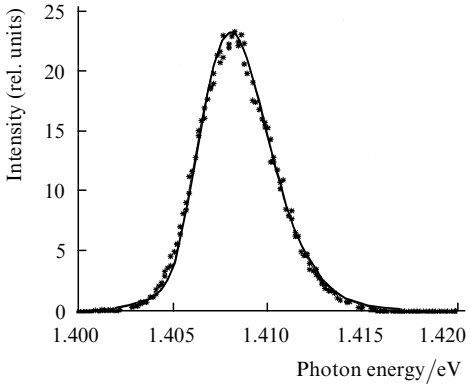

Figure 11. Typical approximation of the cooperative-emission spectrum according to Eqns (3)–(6) at $E'_g = 1.4008$ eV, $\mu = 7.3$ meV, $\Delta = 2.53$ meV, $\Gamma = 1.7$ meV.

Dependences of the order parameter Δ of the coherent BCS state on the reverse voltage (the electron–hole concentration) for four samples are presented in Fig. 12. As shown in [14], the carrier concentration rises almost linearly with increasing the reverse voltage. According to the theory [22–24], the band gap Δ of all the samples decreases with increasing density of $e-h$ pairs, as is clearly seen in Fig. 12. The value of Δ is more than two times smaller than the exciton binding energy. Note that the values of the parameter Δ are the same for all the samples within an accuracy of 10 %. Dependences of the Fermi energy of quasiparticles for the same samples are shown in Fig. 13. It should be pointed out that the Fermi energy of the quasiparticles in the coherent BCS state is much smaller than that of electrons (over 100 meV) in GaAs at room temperature and concentrations achieved.

6. Distinction of our experimental conditions on conditions of Bose condensation of excitons

Let us demonstrate now a qualitative distinction of our experimental conditions from conditions of Bose condensation of excitons at low temperatures described by other authors [4–7]. First, as follows from Eqns (1) and (2), the critical temperature of condensation rises with increasing the concentration of bosons. The densities of electrons and holes in our experiments exceed by many orders of magnitude those used in experiments [4–7]. This is one of conditions for observing the Bose condensation at room temperature.

Second, unlike the Bose condensation of excitons, which occurs spontaneously, without any external force, in our case, the resonant optical field plays a decisive role in the condensation of $e-h$ pairs and the formation of the BCS-like state. It is well known that both the gain and absorption

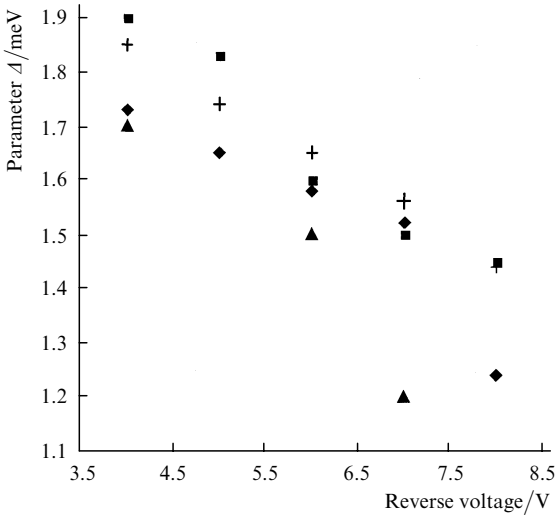

Figure 12. Dependences of the of the energy gap on the reverse voltage (the density of $e-h$ pairs) for four samples (shown by different symbols).

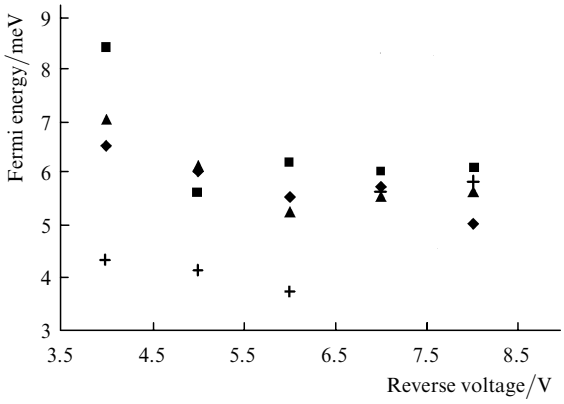

Figure 13. Fermi energy of quasiparticles as a function of the reverse voltage (the density of $e-h$ pairs) for four samples (see Fig. 12).

in a two-component medium (amplifier + absorber) can be readily controlled by applying the forward or reverse bias to different parts of a semiconductor structure. In this case, the absorption edge of a reverse-biased part shifts to the red due to the Frantz–Keldysh effect. As a result, it is possible to quench the generation at all frequencies and achieve purely spontaneous emission in the amplifying sections by increasing the reverse bias. Then, by increasing the pumping of the amplifier, it is possible to attain such a high carrier density that the gain overcomes absorption at the longest wavelengths due to the red shift of the band edge. The recombination of $e-h$ pairs located at the very bottom of the bands results in the generation of photons, which can be amplified. These photons, when being absorbed, generate bound $e-h$ pairs (bosons), which are coherent with the optical field and with each other for some time. Because of the very fast intraband relaxation, electrons and holes from the upper levels fill free spaces at the bottom virtually instantly. One can show that at very high densities [above $(3-4)\times 10^{18}$ cm^{-3}], all the sites in the conduction band within 30–60 meV from the bottom will be occupied by electrons. This means that, when a boson is decomposed into the electron and hole, the former has no free place where to go but to occupy a free energy level, which lies

30–60 meV above the bottom of the conduction band. Because the probability of such transition is very low, the bound pairs, generated by the optical field and coherent with it, will be stable at room temperature. The kinetic energy of these pairs is very low, which explains a small value of the Fermi energy of electrons and holes of the BCS state estimated earlier. A detailed mechanism and conditions of the condensation of $e-h$ pairs at room temperature will be considered in our next paper.

7. Conclusions

Therefore, the totality of experimental data suggests that an unstable coherent cooperative state of electrons and holes can exist in GaAs under appropriate conditions. The radiative recombination of this state is observed in the form of high-power femtosecond pulses. This unstable coherent state with a lifetime of a few hundreds femtoseconds is formed at room temperature. Some of its properties are similar to these of an ensemble of Cooper pairs in a superconductor. It has been shown that due to a strong pumping and a special geometry of p–i–n GaAs/AlGaAs structures electron–hole density achieved in experiments exceeds critical concentrations that are required for the $e-h$ condensation at room temperature. The resonant optical field plays a decisive role in the condensation.

Experimental data have been obtained for the estimate of the parameters of the coherent electron–hole BCS state. In particular, dependences of the order parameter Δ (the gap in the energy spectrum of quasiparticles) and the Fermi energy on the injected $e-h$ pair density have been obtained. Note that the BCS-like gap in a degenerate electron–hole system is smaller than the binding energy of excitons (4 meV in our case) and decreases with increasing concentration. The widths of the observed spectra of cooperative recombination are rather large (4–8 meV), which is determined by the femtosecond lifetime of the cooperative state of electrons and holes. In our opinion, this circumstance makes the direct observation of the energy gap in the electron–hole spectrum problematic when the energy levels are strongly broadened.

We have shown that the effective mass and concentration of $e-h$ pairs are the key parameters determining the $e-h$ condensation and the formation of the coherent BCS-like state at room temperature. To observe these effects in other semiconductor systems, it is necessary to find a semiconductor with minimum values of heavy and light holes. Strained-layer quantum-well structures are among the most promising devices. In these structures, the effective mass of heavy holes can be decreased a few times due to the reconstruction of the band structure with the introduction of strain.

In conclusion, we would like to note that academician N.G. Basov was the first who drew our attention to possible changes in the statistical properties of an electron–hole system in the case of the superradiance emission. It is his interest in our work that helped us to obtain these results.

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