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COHERENT ELECTRON–HOLE BCS STATE: STUDY OF DYNAMICS

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The first experimental study of the evolution of a coherent electron–hole (e–h) BCS-like state in bulk GaAs at room temperature is presented. We demonstrate explicitly that the total spontaneous emission from e–h pairs located within the conduction and valence bands approaches zero when the radiative recombination of the e–h BCS state occurs. This confirms that a vast majority of electrons and holes available is condensed at the very bottoms of the bands and forms the BCS state. An average lifetime of this state is measured to be around 300 fs. We also show that the coherence of electrons and holes of the BCS state preserves for a much longer time compared to the intraband relaxation time T_2.

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Electron–hole (e–h) systems in semiconductors and their interactions with a resonant electromagnetic emission have been among foremost topics in condensed matter physics for a long time. 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, the macroscopic quantum state can be either the excitonic Bose–Einstein condensate or a cooperative e–h state that is similar to the BCS state of the Cooper pairs in a superconductor [1]. Semiconductors, including quantum-well structures and microcavity structures, which are highly excited with an optical emission, are generally considered as the most promising candidates for observation of these phenomena [2–5]. The number of publications devoted to excitonic Bose condensates, their superfluidity, excitonic insulators, and a crossover from Bose condensation to BCS states rapidly increased [6–9].

In our recent papers [10, 11], we presented the first experimental results of the investigation of spectral characteristics of the radiative recombination of a transient BCS-like e–h state in bulk GaAs at room temperature. Instead of an optical pumping, we used a strong current injection in a p–i–n semiconductor structure for achieving very high e–h densities. The e–h concentrations attained in our experiments were so high that the average interparticle distance (34–58 Å) was about or even smaller than the de Broglie wavelength of an e–h pair in GaAs at room temperature. The latter is about 107 and 60 Å for the electron–light hole and electron–heavy hole pairs, respectively. The collective pairing of electrons and holes and their condensation were responsible for the spectral features of the observed emission, including a large spectral shift of the emission line peak towards the longer wavelengths. Fitting of the optical spectra with some theoretical curves allowed us to estimate the order parameter Δ of the e–h quantum state (the e–h pairing gap), which proved to be about 2 meV [10]. We have also demonstrated that the order parameter decreases from 2.1 to 1.2 meV with increasing the e–h density, whereas the Fermi energy of the quasiparticles lies in the range from 4 to 8 meV [11]. The latter values are much smaller than the Fermi energy of electrons in GaAs (60–170 meV) at room temperature and carrier concentrations $(2\text{–}6) \cdot 10^{18}$ cm^{-3}

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achieved experimentally.

It is obvious that in contrast to the normal BCS state of the Cooper pairs, any e–h BCS-like state is essentially unstable due to recombination of electrons and holes. The radiative decay of the e–h BCS state should exhibit basic features of the cooperative recombination or superradiance of an ensemble of quantum oscillators. Some characteristics of the cooperative emission of electrons and holes have been reported previously [12–14]. Because the e–h BCS-like state possesses a macroscopic polarization and occupies a sizeable portion of the active region of a semiconductor structure, it should explicitly display the coherent interaction of the recombination emission field with e–h pairs of the BCS state. By observing this coherent interaction, we can prove that e–h pairs located at different parts of the structure (or of the BCS state) have a common phase, i.e., that they are coherent.

In this paper, we present additional experimental results for properties of the transient e–h BCS-like state. The main goal is to investigate the temporal behavior and evolution of the cooperative state. The observations are based on (a) direct measurements of time-resolved spectra of the e–h BCS state recombination emission utilizing a streak camera and a monochromator and (b) the fringe-resolved second harmonic generation (SHG) autocorrelation. The former method allows us to find which part of the e–h ensemble occupies which energy places in the bands and at what time intervals. The latter technique allows measuring coherent properties of the recombination emission.

We use the same semiconductor structures and the pumping technique as in our previous work and their description can be found elsewhere [10, 11]. The output emission was monitored in time domain using a single-shot streak camera with a temporal resolution about 1.5 ps. Time-resolved optical spectra were also detected by the streak camera. In this case, the emission from the samples was initially collimated on a diffraction grating having 600 lines/mm. The beam deflected at the third diffraction order was then focused on the input slit of the streak camera. This allows us to observe emission features in both time and frequency domains. The technique is discussed in detail in the book [15].

Because a typical duration of superradiant pulses from semiconductor structures lies in the femtosecond range [13, 14], we also use the autocorrelation technique based on SHG for more precise pulsewidth measurements. This method has a femtosecond time resolution and permits measurements of both amplitude and phase relationships of the emission under test with femtosecond accuracy.

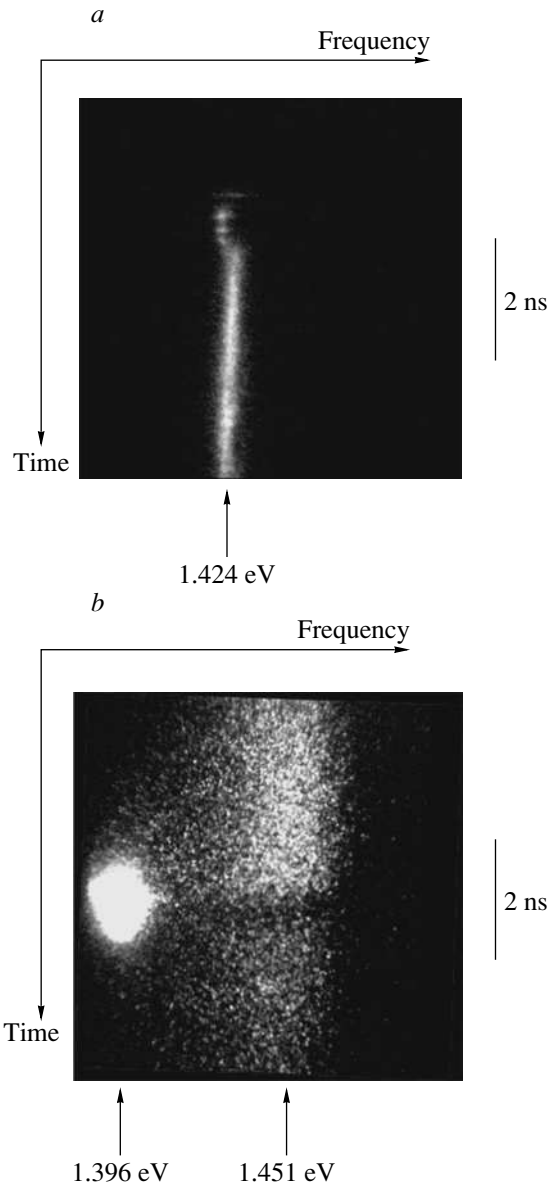

Fig. 1. Time-resolved spectra of (a) lasing and (b) spontaneous emission and superradiant pulse. The width of the spontaneous emission in Fig. 1b is so broad that its high-energy tail is not shown due to the limited input aperture of the streak camera

Figure 1 shows time-resolved optical spectra of a GaAs/AlGaAs p–i–n structure in two typical regimes. Laser emission can be generated from the structure when current pulses with a not large enough amplitude are applied on the amplifier sections and zero reverse bias is applied on the absorber section of the structure. A typical time-resolved optical spectrum of lasing is presented in Fig. 1a. Here, a photograph taken from

the streak camera screen is shown. The lasing starts from the relaxation oscillations (3–4 pulses in front of the trace), which are quite typical of semiconductor lasers. We note that the trace is quite narrow and its center is located at the same place on the frequency axis almost all the time. This means that the central wavelength of the emission varies in time only slightly. Individual modes of the spectrum are not resolved due to a relatively poor spectral resolution of the diffraction grating. The photon energy of the spectral peak is around 1.424 eV. This value was measured using a monochromator separately.

A completely different picture is observed when the e–h BCS state is formed and femtosecond superradiant pulses are generated as a result of its recombination. Figure 1*b* corresponds to this regime. The broad vertical stripe represents the ordinary spontaneous emission of nonpaired electrons and holes, whereas the bright spot on the left is the superradiant pulse due to the recombination of the e–h BCS state. Because the peak power of the superradiant pulse is typically over 10^4–10^5 times larger than the spontaneous background, its image on the picture is misshapen due to the overexposure. It is clearly seen in Fig. 1*b* how the femtosecond superradiant pulse develops from the spontaneous emission. As the carrier density increases in time, the spontaneous recombination of electrons and holes occurs at lower and lower frequencies (photon energies) because of the band gap shrinkage. When the carrier density is sufficiently high, the de Broglie wavelengths of individual e–h pairs start to overlap and the quantum-degeneracy criterion is fulfilled [10], the e–h BCS-like state develops. The phasing of wavefunctions of individual e–h pairs occurs via the common electromagnetic emission. The macroscopic polarization is building up at early stages of the leading edge of the superradiant pulse.

Perhaps the most interesting feature of Fig. 1*b* is a dark stripe that goes across the spontaneous emission at the time when the BCS state recombines. This implies that there are almost no electrons and holes within the bands that can recombine spontaneously. All the electrons and holes are condensed at the very bottoms of the bands and form the BCS state. During its radiative recombination, photons having the minimum possible energy are emitted. This energy is 1.396 eV in our case, while the peak of the spontaneous emission is at 1.451 eV. We recall that the nonrenormalized band gap in bulk GaAs at room temperature is 1.424 eV.

We calculated the intensity of total spontaneous emission from electrons and holes occupying energy levels inside the bands and plotted it against time. Figure 2 presents this plot. It is clearly seen how the spontaneous emission intensity almost approaches zero when the e–h BCS state is built and recombines. Its temporal position is shown by the arrow.

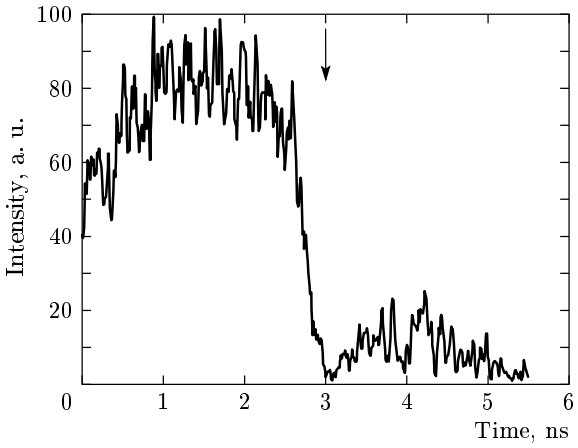

Fig. 2. Intensity of the spontaneous emission against time. The position of the superradiant pulse is shown by the arrow

Such a dynamic behavior is completely different from lasing or spontaneous emission. For example, when lasing starts in a semiconductor laser structure, the spontaneous emission power across the entire frequency range clamps at the threshold level. It is not possible to achieve any dip in the carrier distribution or to quench the spontaneous emission at neighbor frequencies due to ultrafast intraband relaxation processes. On the other hand, it is known [15] that in real semiconductor lasers, e–h pairs decohere rapidly with the typical time 10–100 fs. As a result, we find that for typical operating conditions, the ratio of the number of photons in the sample to the number of e–h pairs is $N_{photon}/N_{e-h} \approx 10^{-5}$–$10^{-4}$ [16]. But in the superradiant state, the photon field is linearly coupled to the order parameter and we have $N_{photon}/N_{e-h} \approx 1$. This implies that when the e–h BCS state recombines and photons are emitted from the structure, very few e–h pairs should be left in the structure. That is exactly what we experimentally see as the dark horizontal stripe in the middle of Fig. 1*b*.

Precise measurements of the pulsewidth of superradiant pulses allow us to estimate the typical lifetime of the e–h BCS state. Figure 3 presents an SHG intensity autocorrelation of superradiant pulses. Its full width at half maximum (FWHM) is around 460 fs. This value corresponds to the actual pulsewidth between 290 and 324 fs depending on the assumed pulse shape (Gaussian, sech, or asymmetric exponential shapes) [15]. It

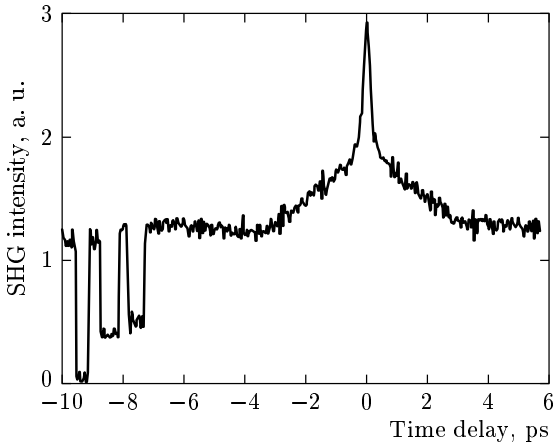

Fig. 3. Intensity autocorrelation of superradiant pulses

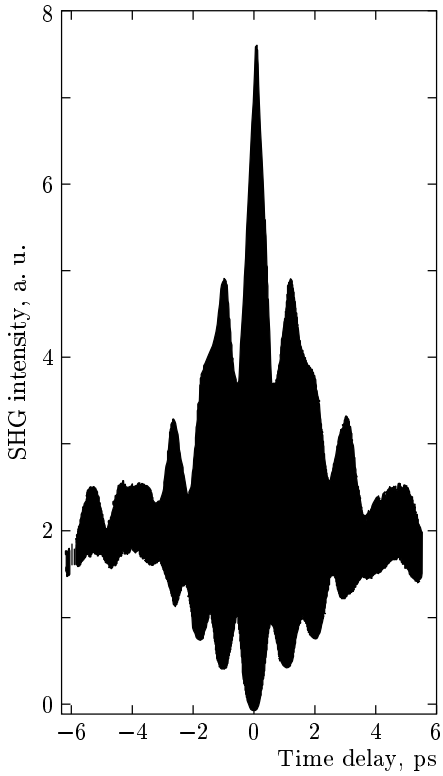

Fig. 4. Interferometric autocorrelation of superradiant pulses illustrating the coherent interaction of the photon field with e–h pairs of the BCS state

is worth to point out that the achieved pulsewidth of superradiant pulses is an absolute record among all ultrashort pulses generated by semiconductor lasers, including mode-locked, Q-switched, and gain-switched devices. The SHG trace exhibits a pedestal of 1.5–2 ps long, which originates from instabilities of the pulse shape and very large timing jitter. These are likely to be determined by intrinsic quantum-mechanical fluctuations of initial conditions of both the photon field and e–h system. Indeed, as noted in [7], the appearance of the large noise is a strong evidence for the presence of coherence in the e–h system. The noise amplitude is known to be inversely proportional to the number of statistically independent entities in an e–h system. Thus, large noise observed experimentally implies that only a few entities exist in the macroscopically large regions where the e–h BCS state is located.

Unlike intensity autocorrelations presented in Fig. 3, where all phase information is lost in averaging, fringe-resolved or interferometric autocorrelations can provide some information about phase relationships of the emission under study. This technique enables us to prove experimentally that different spatial regions of the e–h BCS state are coherent or have the same or coupled phases. First of all, we point out that all interferometric autocorrelations of ultrashort pulses generated by lasers have a single peak at zero time delay [15]. In the case of mode-locked pulses, there are additional peaks with fringes separated by the cavity round trip time.

Figure 4 shows the experimental interferometric autocorrelation of superradiant pulses from a semiconductor structure where two regions with a very high e–h density are formed. The separation between the nearest parts of the regions was less than 10 μm. The total length of the structure corresponds to the round trip time about 3.1 ps. The coherent beating of the photon field is clearly seen in Fig. 4. The shape of the trace resembles the beating of two oscillators and suggests that the coherency of interaction between the emission and different parts of the BCS state stays on for a few picoseconds while the pulse travels through the medium. Indeed, the very beginning of the recombination pulse induces a small macroscopic polarization while spreading in the medium. This polarization acts as a source to produce additional photon field that in turn creates more polarization. Dephasing processes prevent establishing the coherency of individual e–h pairs and the formation of the macroscopic polarization. But if the number of e–h pairs is sufficiently large, the optical gain in the medium can overcome the dephasing [14] and the phase transition of the e–h system into a coherent BCS-like state can occur. In this case, the BCS state occupies a sizeable portion of the sample and its deexcited parts can then be reexcited by a coherent emission from other regions of the cooperative state. This gives rise to the coherent ringing observed in the

output radiation in the form of multiple peaks of the interferometric autocorrelation as presented in Fig. 4.

We have pointed out earlier [10] that e–h interactions within the cooperative state do not affect the coherence of individual electrons and holes, which is quite similar to the Cooper pairs of a superconducting BCS state. This means that relaxation processes must be much slower compared to a system of noncorrelated e–h pairs. If we suppose that e–h pairs of the BCS state decohere at the same rate as in a normal bulk GaAs (more than 10^{13} s^{-1}), then we would not observe any beating of the photon field on a picosecond scale as shown in Fig. 4. The extremely long phase relaxation time of electrons and holes of the BCS state can be explained as follows.

In contrast to the Bose condensation of excitons, which occurs spontaneously [6–9], the condensation of e–h pairs and formation of the BCS state in our case is caused by the resonant electromagnetic emission. The presence of the absorber section in the semiconductor structure results in the absorption of the emission travelling through the structure at all wavelengts except a narrow region at the very bottom of the bands. In this region of the longest possible wavelengths, we have net gain. The radiative recombination of e–h pairs leads to the generation of long-wavelength photons that are amplified and in turn create e–h pairs. These bound pairs are bosons and remain coherent with each other and the optical field for some time. Because of the very fast intraband relaxation, electrons and holes from upper energy levels in the bands occupy the levels at the bottom that happed to be free almost immediately. At very high carrier concentrations (larger than $(3\text{–}4) \cdot 10^{18}$ cm^{-3}), all energy levels within the band 30–60 meV from the bottom are occupied by electrons. This implies that there is no place within this band for an electron of a bound pair if it becomes free and becomes a fermion again instead of being a part of a boson. That is why the bound pairs are stable at room temperature. The number of such bound e–h pairs increases in time as the optical field travels back and forth between the facets of the crystal. Because the bosons originate from the electrons and holes occuping the lowest possible energy levels, their kinetic energy is very low. This explains the very small value of the Fermi energy of the quasiparticles mentioned above (less than 8 meV). The detailed explanation of this phenomenon is the task of our forthcoming paper.

In conclusion, we present the first direct measurement of dynamics of a coherent e–h BCS state in a semiconductor heterostructure at room temperature. We demonstrate experimentally that almost all electrons and holes are condensed at the very bottoms of the bands when the BCS state is formed. A typical lifetime of the e–h BCS state is measured to be about 300 fs. The macroscopic size of the BCS state results in the coherent interaction of the recombination photon field with e–h pairs of the cooperative state on the picosecond time scale, the coherence of individual electrons and holes being unaffected by their collisions.

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REFERENCES

1. *Bose–Einstein Condensation*, ed. by A. Griffin, D. W. Snoke, and S. Stringari, Cambridge University Press, Cambridge (1995).
2. L. V. Keldysh and Yu. V. Kopaev, Fizika Tverd. Tela **6**, 2791 (1964).
3. C. Comte and G. Mahler, Phys. Rev. B **34**, 7164 (1986).
4. T. J. Inagaki, T. Iida, and M. Aihara, Phys. Rev. B **62**, 10852 (2000).
5. P. R. Eastham and P. B. Littlewood, Phys. Rev. B **64**, 235101 (2001).
6. D. Snoke, J. P. Wolfe, and A. Mysyrowicz, Phys. Rev. Lett. **64**, 2643 (1990).
7. L. V. Butov, A. Zrenner, G. Abstreiter, G. Bohm, and G. Weimann, Phys. Rev. Lett. **73**, 304 (1994).
8. L. V. Butov, A. L. Ivanov, A. Imamoglu, P. B. Littlewood, A. A. Shashkin, V. T. Dolgopolov, K. L. Campman, and A. C. Gossard, Phys. Rev. Lett. **86**, 5608 (2001).
9. A. V. Larionov, V. B. Timofeev, J. Hvam, and K. Soerensen, Pis'ma v Zh. Eksp. Teor. Fiz. **75**, 233 (2002) [JETP Lett. **75**, 200 (2002)].
10. P. P. Vasil'ev, H. Kan, H. Ohta, and T. Hiruma, Phys. Rev. B **64**, 195209 (2001).
11. P. P. Vasil'ev, H. Kan, H. Ohta, and T. Hiruma, Quant. Electr. **31**, 870 (2001).
12. P. P. Vasil'ev, Quant. Electr. **24**, 540 (1994).
13. P. P. Vasil'ev, Quant. Electr. **27**, 860 (1997).
14. P. P. Vasil'ev, Quant. Electr. **29**, 842 (1999).
15. P. Vasil'ev, *Ultrafast Diode Lasers: Fundamentals and Applications*, Artech House, Norwood, MA (1995).
16. P. B. Littlewood and X. Zhu, Phys. Scripta **T68**, 56 (1996).