Investigation of the optical gain spectra in two-dimensional quantum well heterostructures

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We have investigated the optical gain spectra in GaAs–$Al_xGa_{1-x}As$ two-dimensional quantum well heterostructures at 4.2 K over a wide range of excitation levels (L from 0.04 to 10 MW/cm^2). In this paper we show that when a three-dimensional electron-hole plasma (3D EHP) located in the bulk GaAs coexists with a 2D EHP at the GaAs–$Al_xGa_{1-x}As$ heterostructure, the stimulated emission is generated predominantly in the 2D EHP. Under these circumstances the threshold laser excitation at which optical gain first appears in the 2D EHP is anomalously low (two orders of magnitude smaller than that of the 3D EHP), and the corresponding value of the optical gain coefficient is 2.5 times higher than in bulk GaAs. We have analyzed the shape of the stimulated optical gain spectrum for the 2D EHP, taking into account outward streaming of the EHP, superheating of the plasma and variation of the electron-hole density in the latter; using this information we then determined the fundamental parameters of the strongly-nonequilibrium 2D EHP, including its density, temperature, and drift velocity v_D. Based on our determination of v_D, we estimate the ambipolar diffusion coefficient and mobility of the nonequilibrium charge carriers in the 2D EHP.

1. INTRODUCTION

The quasi-two-dimensional nonequilibrium electron-hole plasma (EHP) created by laser excitation near an insulator-semiconductor boundary is characterized by a number of important and distinctive features: a lower pump threshold for stimulated emission, a larger value of the optical gain coefficient compared to that of a three-dimensional (3D) EHP, etc.[1,2] These features are due to "compression" of the EHP against the hetero-interface, leading to an increase in the concentration of nonequilibrium charge carriers over that of a bulk semiconductor under the same photoexcitation conditions.

It is interesting to investigate the optical gain (as a necessary condition for laser excitation) in GaAs–$Al_xGa_{1-x}As$ heterostructures with a single two-dimensional (2D) quantum well containing one type of carrier (electrons or holes). To do so, we laser-excite the system at a power level sufficient to create a three-dimensional EHP (in the GaAs bulk) and a two-dimensional EHP (at the GaAs–$Al_xGa_{1-x}As$ heterostructure) at the same time; using the stimulated-emission spectrum we then can determine which transition is exhibiting the optical gain.

It should be noted that a number of papers have reported using the optical gain technique to determine the concentration and temperature dependence of carriers in these types of nonequilibrium EHP, in order to investigate scattering processes, renormalization of the bands, etc. These papers discuss both bulk semiconductors[3,4] and inhomogeneous systems, including multi-quantum-well structures[5–7] and single wells placed between wide-gap regions.[8] However, systematic investigations of optical gain spectra (including the case where a 3D EHP and a 2D EHP are excited at the same time) have not been carried out to date.

In this paper we investigate the spectra of spontaneous and stimulated emission in GaAs–$Al_xGa_{1-x}As$ heterostructures over a wide range of excitation intensities, and compare the thresholds at which optical gain first appears for the two-dimensional and three-dimensional EHP. Based on our analysis of the shape of the optical gain spectrum, we then determine the fundamental parameters of EHP created at extremely high excitation levels. By taking into account outward streaming effects we can determine the kinetic (drift) characteristics of the 2D plasma state and compare it with the 3D case.

2. METHODS OF INVESTIGATION AND EXPERIMENTAL RESULTS

We obtained the heterostructures used in this work by the method of molecular-beam epitaxy. We first deposited a layer of undoped GaAs (with an unintentional impurity concentration $N \leqslant 10^{15}$ cm^{-3}) on a semi-insulating GaAs substrate, followed by a thin (~ 70 Å) spacer layer of undoped $Al_{0.3}Ga_{0.7}As$, and then by a thicker (~ 700 Å) layer of doped $Al_{0.3}Ga_{0.7}As$ ($n \approx 10^{18}$ cm^{-3}). The mobility of the two-dimensional electron gas at the GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructure attained a value of roughly 10^5 cm^2/V sec at 77 K in these samples.

In order to investigate the photoluminescence (PL) spectra we used a LGN-502 CW argon laser and a pulsed Nd^{3+}:YAG laser (the second harmonic of the latter). We used a MDR-23 monochromator to record the emission spectra, and found the spectra of the optical gain coefficients according to the method described in Refs. 2 and 9.

In Fig. 1 we show PL spectra in the region 1.50 to 1.52 eV for GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructures under CW excitation by the argon laser as a function of excitation energy. Near the fundamental absorption edge of GaAs we observe the well-studied emission structure due to excitons bound to donors and acceptors (for our experimental conditions these features were resolved at an illumination intensity $L \approx 1$ W/

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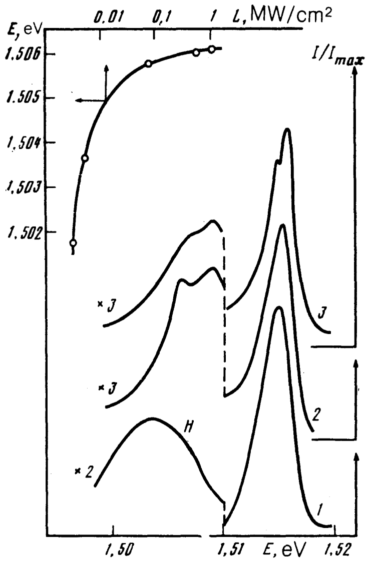

FIG. 1. PL spectra of GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructures for three excitation power densities L in MW/cm^2: *1*—0.05; *2*—0.5; *3*—1; the temperature was 4.2 K. Above and to the left is the dependence of the energy position of the H-band on the excitation power density.

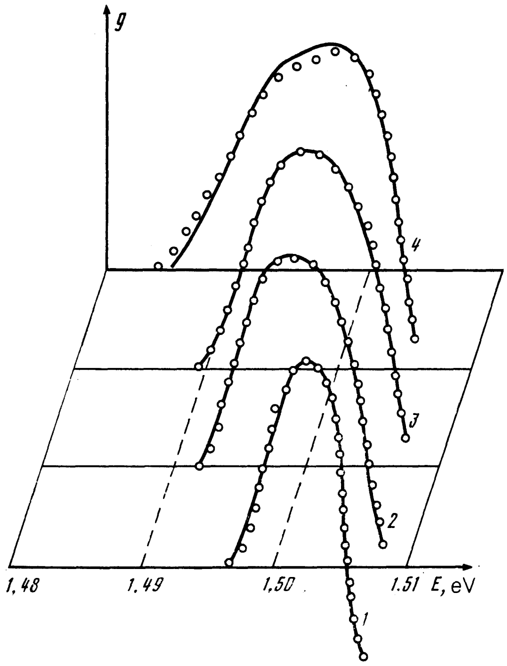

FIG. 3. Optical gain spectra of GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructures for various excitation power densities L; the points are experimental ($T = 4.2$ K), the solid traces are calculated; L in MW/cm^2 is: *1*—0.04; *2*—0.8; *3*—2; *4*—10.

cm^2). In the spectral region 1.50–1.51 eV a PL band appears (called the H-band in Ref. 10) which is characteristic of the GaAs–$Al_xGa_{1-x}As$ system. For the heterostructures we investigated the energy position of this band shifted toward the short-wavelength region from 1.502 eV to 1.506 eV as the excitation power density increased from $3 \cdot 10^{-3}$ W/cm^2 to 1 W/cm^2. In this case the "rate" of the shift falls off with increasing pump power; for $L \approx 0.5$ W/cm^2 the H-band splits in two, and the intensity of the short-wavelength component increases strongly as the excitation power density increases.

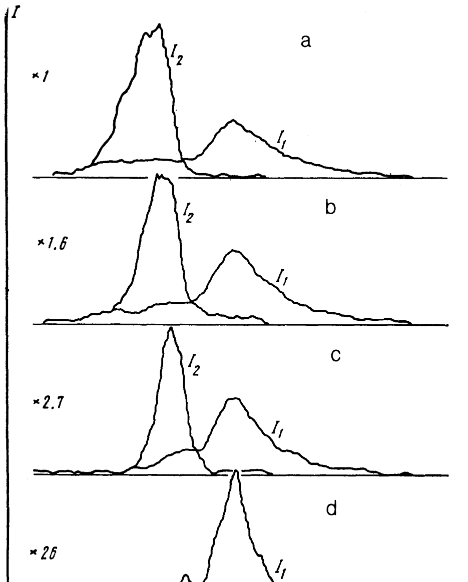

FIG. 2. Spontaneous (I_1) and stimulated (I_2) emission spectra of GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructures at 4.2 K; L in MW/cm^2 equals 10 (a), 2 (b), 0.8 (c) and 0.04 (d).

Figure 2 shows the spontaneous-emission and stimulated-emission spectra of the GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructures under pulsed laser excitation at various power densities. For the excitation levels used we observed two bands in the spontaneous-emission spectrum, one due to radiative recombination of electrons and holes in the nonequilibrium EHP located in the bulk GaAs (the short-wavelength part of the spectrum) and one due to recombination in the two-dimensional EHP localized at the GaAs–$Al_{0.3}Ga_{0.7}As$ boundary (the long-wavelength part of the spectrum). It is noteworthy that stimulated emission was observed in these structures only for the two-dimensional EHP.

The spectra of the optical gain coefficient g were found from the relation between the stimulated (I_2) and spontaneous (I_1) emission intensities measured at identical pump power densities:

$$\frac{I_2}{I_1} = \frac{\exp(gl) - 1}{gl}; \qquad (1)$$

here l is the length of that part of the laser beam producing gain (in our case $l = 130\,\mu$). Figure 3 shows the calculated spectra of g at four excitation power densities (the points are from experiment). Note the considerable broadening of the optical gain spectrum and its shift toward longer wavelengths as the excitation power density increases.

3. DISCUSSION OF RESULTS

The long-wavelength shift mentioned above in the energy position of the H-band as the excitation power density L increases from 3×10^{-3} W/cm^2 to 10^{-1} W/cm^2 is characteristic of donor-acceptor recombination and in this case could indicate either tunneling recombination of donor-acceptor pairs localized at the heteroboundary[11] or recombination in the two-dimensional system consisting of spatially separated electron and hole layers.[12] However, for $L > 10^{-1}$ W/cm^2 the shift becomes weak, while the H-band splits in two (see Fig. 1). The subsequent behavior of the doublet as a function of excitation intensity and temperature (i.e., the growth in intensity of the short-wavelength component with increasing L and T) indicates the presence of two electronic subbands in the two-dimensional quantum well formed at the GaAs–$Al_{0.3}Ga_{0.7}As$ heteroboundary. Under conditions of high optical excitation density ($L = 0.04$ to 10 MW/cm^2) the band caused by recombination of the two-dimensional electrons with photoexcited holes (i.e., the 2D EHP shifts toward the long-wavelength region because of many-body collective interactions (see Fig. 2).

Let us investigate the optical gain spectrum further. Figure 2 shows that intense stimulated emission is observed only in the spectral region of spontaneous luminescence of the 2D EHP. These results indicate that under conditions that give rise to coexisting 3D and 2D EHP the optical gain effects take place primarily in the 2D EHP. It is interesting to compare the threshold for appearance of optical gain and the values of g_{max} for the 3D and 2D EHP. In Fig. 4 we show the dependence of g_{max} on the excitation power density for the GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructure used in this paper along with the corresponding dependences taken from Ref. 2 for a Si_3N_4–GaAs structure (in which a quasi-two-dimensional EHP can be created) and GaAs with a free surface (with a 3D EHP). The excitation threshold at which optical gain appears in the Si_3N_4–GaAs structure is roughly a factor of 3 smaller than that of bulk GaAs; for the GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructure the excitation threshold is smaller by a factor of 60. At the same time, the maximum value of the optical gain coefficient for the Si_3N_4–GaAs is roughly 1.5 times larger than in bulk GaAs, while the value for the GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructure is 2.5 times larger. Thus, the transition from 3D EHP to 2D EHP significantly decreases the threshold for the appearance of optical gain while simultaneously increasing g. The main physical reason for this is the concentration of the plasma along one coordinate in the 2D case.[13]

It is clear from the optical gain spectra shown in Fig. 3 for the GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructure that as the level

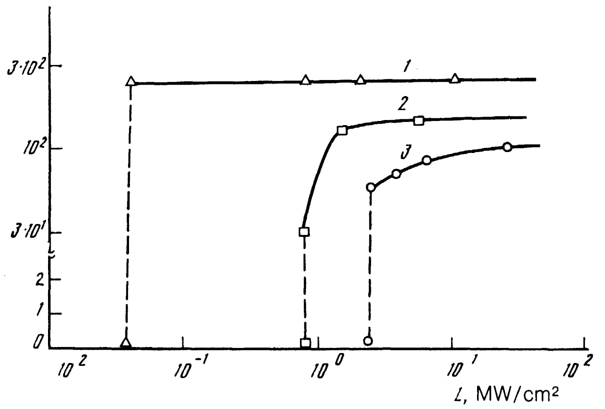

FIG. 4. Dependence of the optical gain coefficient (at its maximum) on the excitation power density for an GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructure (1), the insulator-semiconductor structure Si_3N_4–GaAs (2) and GaAs (3) at 4.2 K.

of excitation increases the spectra broadens, predominantly because of pulling toward the long-wavelength region. We can model this type of behavior quite well by taking into account the so-called "outward streaming effect" in the dynamics of the nonequilibrium carriers; this effect is a consequence of forces acting on the carriers associated with the gradient of the Fermi pressure $\mathbf{F} = \nabla p/n$, where $p = n\, df/dn - f$ (f is the free energy per unit volume). The outward streaming effect also leads to the appearance of an aureole of radiation beyond the edge of the excited region of the sample.[14,15] and to additional broadening due to reradiation.[16]

The experimental optical gain spectra obtained here, whose shapes are determined by the product of the density of states and a combination of the occupation functions of electronic states f_e and hole states f_n,

$$g(E) \sim \rho(E)\,(f_e + f_h - 1), \tag{2}$$

can be analyzed by using shifted distribution functions for the electrons and holes[16]:

$$f_{e,h} = \left(\exp\left\{\left[\frac{\hbar^2}{2m_{e,h}}(\mathbf{k}+\mathbf{k}_{D_{e,h}})^2 - F_{e,h}\right] \Big/ k_B T\right\} + 1\right)^{-1}, \tag{3}$$

where $\mathbf{k}_{D_{e,h}} = m_{e,h}\mathbf{v}_0/\hbar$, v_0 is the average drift velocity for outward streaming of the carriers, $F_{e,h}$ is the quasi-Fermi level, and T is the effective temperature of the electron-hole plasma. In our calculations we first averaged Eq. (2) over the angle between $\mathbf{k}$ and $\mathbf{k}_{D_{e,h}}$; by comparing the spectral densities $g(E)$ obtained in this way with the experimental data, we can determine the fundamental parameters of the plasma for each specific case: the carrier density n, the temperature T, the drift velocity v_D, and the ambipolar diffusion coefficient D.

Studies of electron-hole plasmas in CdS and CdSe under conditions of three-dimensional outward streaming based on the spectral dependence of the optical gain were carried out previously in Ref. 16. It should be noted, however, that the calculations of Ref. 16 did not include the temperature dependence $F_{e,h}(T)$ of the Fermi levels; in practice this effect must be taken into account because of the considerble superheating of the plasma. In this paper we also have calculated the optical gain coefficient of the two-dimensional EHP excited in GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructures with two-dimensional quantum channels, including $F_{e,h}(T)$, and have compared our results with the experimental spectra. In our experiments a certain pulling of the spectrum $g(E)$ toward the long-wavelength region is observed in these structures even for relatively small excitation levels. Therefore, in order to improve the agreement, we have replaced the stepwise distribution function for an ideal two-dimensional plasma $\rho(E) = \theta(E - E_g)$ by a function with an exponentially washed-out long-wavelength edge; by comparing the corresponding calculated line shape for $g(E)$ with the experimental shape for $v_D = 0$ (i.e., weak excitation) we found the value ΔE of thei "natural" washing-out of the long-wavelength edge. In previous calculations this quantity was taken to be an invariant parameter even at high pump levels.[17]

Figure 3 shows the spectra $g(E)$ calculated in this way, along with a comparison with experiment. In Fig. 5 we show n, T and v_D as functions of the excitation intensity L, which we obtained by fitting the line shape $g(E)$ for various L values. It is clear from Fig. 5 that as L increases a gradual in-

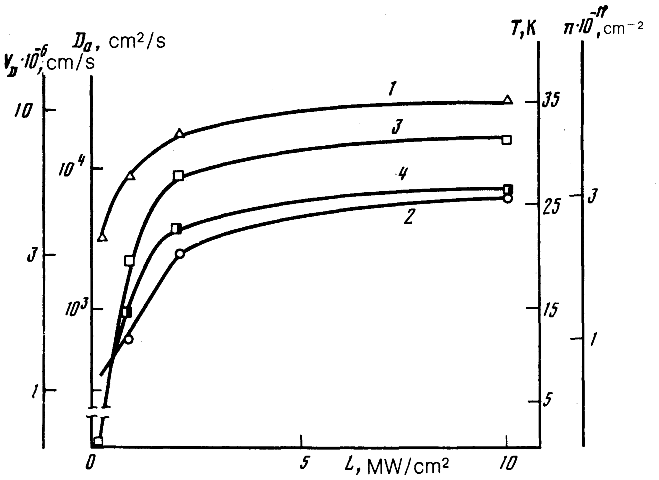

FIG. 5. Dependence of the fitting parameters (electron-hole pair density n (1), temperature T (2), and drift velocity v_D (3) used to bring the experimental and theoretical curves for $g(E)$ into agreement (Fig. 3); curve 4 gives estimated values for the ambipolar diffusion coefficient D_a.

crease in v_D is observed up to values which are the same order of magnitude as the saturated drift velocities caused by intense phonon generation.[18] At the same time, the superheating of the plasma increases markedly (up to ~ 20 K and the electron-hole pair density stabilizes. The saturation of the growth of $n(L)$ is caused by intense acceleration of the recombination processes, in particular due to Auger processes.[18]

Based on our data we can estimate the value of the ambipolar diffusion coefficient $D_a = v_D^2 \tau$ at large excitation levels, where τ is the carrier lifetime in the excited region (in our structures, $\gamma = 10^{-10}$ sec): $D_a \approx 6400$ cm^2/sec for $L = 10$ MW/cm^2 and $D_a \approx 3600$ cm^2/sec for $L = 2$ MW/cm^2 (Fig. 5). If we take into account the fact that the electron diffusion coefficient D_a is considerably larger than the hole diffusion coefficient D_h, the hole coefficient is estimated to be $D_h \approx D_a/2$. Estimates of the hole mobility based on the use of the Einstein relation for the two-dimensional case, i.e., $\mu_h = eD_h/F_h$, give very large values for the latter (from $3 \cdot 10^5$ cm^2/V·sec for $L = 0.8$ MW/cm^2 up to $\sim 10^6$ cm^2/V·sec for $L \geqslant 2$ MW/cm^2), which is within an order of magnitude of the experimental values of the hole mobility measured in specially designed GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructures with low background impurity concentrations[19] and of theoretically calculated values of μ_h for quantized channels.[20]

Thus, intense laser excitation of GaAs–$Al_{0.3}Ga_{0.7}As$ heterostructures simultaneously gives rise to both a three-dimensional electron-hole plasma (in the bulk GaAs) and a two-dimensional electron-hole plasma (at the heterostructure). Under these conditions we have observed substantial planar outward streaming of the plasma at a drift velocity of $v_D \approx 10^7$ cm/sec, which is close (for large L) to the limiting velocities for electron and hole transport in solids. The stimulated emission is created primarily in the 2D EHP. The threshold excitation at which optical gain appears in the 2D EHP is roughly two orders of magnitude smaller than in the 3D EHP, while the magnitude of the optical gain coefficient for GaAs–$Al_{0.3}Ga_{0.7}As$ is 2.5 times larger than it is in GaAs under the same photoexcitation conditions. These characteristics are very important in connection with the possible use of a nonequilibrium 2D EHP as a source of stimulated emission of light.

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