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ORDERED ELECTRON-HOLE CONDENSATE AS A PERSPECTIVE LASER 2D ENVIRONMENT AT ROOM TEMPERATURES

A comparative analysis of the processes governing the formation and stability of the electron-hole (EH) exciton continuum in 3D and 2D semiconductors has been carried out on the basis of theoretical and experimental results obtained by the authors, as well as literature data. Using the phase diagrams, luminescence spectra, and lux-lux dependences, a substantial increase of the excitonic binding energy $E_{\rm ex}$ and the EH continuum stability as compared with those in the 3D case is demonstrated. The role of physical factors responsible for the growth of the excitonic binding energy $E_{\rm ex}$ and the EH continuum stability in the 2D case, namely, the exciton binding at shallow impurity centers, image forces, and the correlation factor, is analyzed. The peculiarities of the exciton-polariton and electron-hole continua are considered taking the Bose–Einstein and Fermi–Dirac statistics into account. Tasks for further theoretical studies are formulated, and the advantages of the application of macroresonator 2D ditches as optoelectronic devices of the new generation, which do not need the complicated and costly MBE technology, are indicated.

K e y w o r d s: electron-hole condensate, exciton continuum, capture centers, 2D excitons, macroditch resonator, induced radiation.

1. Introduction

Modern optoelectronics and nanophotonics have formed a demand for low-threshold and high-efficiency lasers that can further increase the processing and transmission rates of information signals by using photons instead of electrons, provide a higher localization of radiation both in space and time, and solve the problem of impedance matching for electrical circuits inherent in traditional electronic schemes. In this paper, by analyzing the experimental and theoretical results obtained earlier by us and other authors, we show that the electron-hole (EH) condensate is a promising 2D laser medium at room temperatures. High concentrations of EH plasma at its intensive excitation (in particular, using lasers) are a precondition for the formation of a stable exciton EH fluid. This fact was established relatively long ago as a result of studies of EH droplets. The latter, as the increasing exciton concentration reaches a certain critical limit, undergoes a gas-liquid phase transition, which in traditional 3D semiconductors, in most cases, takes place at cryogenic temperatures (see, e.g., works [1–3]). This fact is associated with low values of the exciton binding energy for most semiconductor materials, in particular, groups III-V with the predominantly covalent type of interatomic bond. The quantum gas-liquid transition in the EH system with the change of the distance between spatially separated electrons and holes in quantum wells and the criteria for the existence of a metastable phase were considered in work [4]. In our works [1, 5, 6], it was shown for a wide class of semiconductors (Si, GaAs, InP, GaN) that the exciton binding energy at the surface and, accordingly, the stability of 2D-surface exciton states are substantially higher than in the bulk.

2. Analysis of Basic Theoretical Relationships and Experimental Results for Ordered Electron-Hole Condensate

The parameters of the exciton continuum were analyzed on the basis of phase diagrams (Fig. 1), photoluminescence spectra (Fig. 2), and lux-lux characteristics (Fig. 3). The results obtained revealed an increase in the exciton binding energy due to the following factors:

• the number of particles required to screen the EH interaction in the 2D case is smaller than that in the 3D one,

• the quantum size effect,

• the influence of the surface centers of exciton capture,

• specular image forces,

• the influence of correlation forces at high exciton concentrations [7].

The coordinate dependences of the corresponding components of the binding energy in the electron-hole subsystem are illustrated in Fig. 4. In the case where the electron and the hole are located in the same plane, a two-dimensional exciton is described by the Schrödinger equation written in the 2D Cartesian coordinate system as follows:

$$\left[-\frac{\hbar^2}{2\mu^*}\frac{\partial^2}{\partial x^2}-\frac{\hbar^2}{2\mu^*}\frac{\partial^2}{\partial y^2}+\frac{q^2}{\varepsilon z}\right]\psi=E\psi,\qquad \frac{1}{\mu^*}=\frac{1}{m_n}+\frac{1}{m_p}, \tag{1}$$

where $(x,0)$ is the the electron coordinates, $(0,y)$ the hole coordinates, $z=\sqrt{x^2+y^2}$ is the distance between the electron and the hole, μ^* is the reduced effective mass, and m_n and m_p are the effective masses of the electron and hole, respectively. The eigenvalues

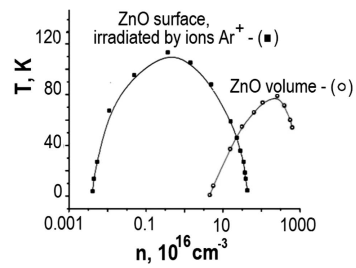

Fig. 1. Phase diagrams of 2- (left) and 3-dimensional (right) EH condensates [7]

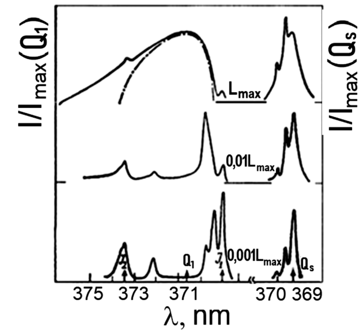

Fig. 2. Photoluminescence spectra of the initial (left) and anode-bombarded (right) ZnO at various excitation levels; the dash-dotted curve is the form of the EH condensate curve theoretically calculated for $n_0 = 5\times10^{18}$ cm^{-3} and $T = 4$ K [6]

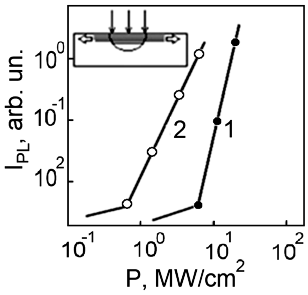

Fig. 3. Dependences of the intensity of stimulated radiation by 3D EH plasma in GaAs (*1*) and quasi-2D EH plasma in Si_3N_4–GaAs structures (*2*) on the excitation intensity. Inset: schematic representation of quasi-2D EH plasma expansion outside the excitation region [5]

of Eq. (1) for the energy spectrum E and the amplitudes A for the bound states ($E<0$) in the 2D case look like

$$E_{ns}=-\frac{R_y}{(n+1/2)^2},\quad A_s=\frac{A_0}{(n+1/2)^3}, \tag{2}$$

where $n=0,1,2...$, $R_y=\frac{q^4\mu^*}{2\varepsilon^2\hbar^2}$ is the ground state energy (Rydberg), and $A_0=\frac{\hbar^2\varepsilon}{4\mu^* q^2}$ the Bohr radius of two-dimensional exciton.

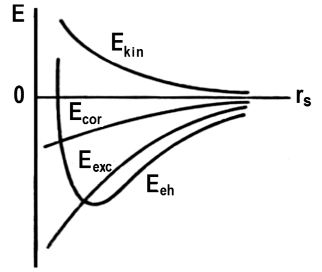

Fig. 4. r_s-dependences of the binding energy components in the electron-hole subsystem: $E_{\rm kin}$ is the kinetic energy, $E_{\rm cor}$ the correlation energy, $E_{\rm exc}$ the exchange Coulomb energy, $E_{\rm eh}$ the binding energy of electron-hole liquid [7]

For comparison, in the 3D case,

$$E_{nv} = \frac{R_y}{n^2}, \quad A_v = \frac{A_0}{n^3}, \quad n = 1, 2.... \tag{3}$$

For a free exciton ($E > 0$), the spectrum is continuous,

$$E_{\rm kin} = \frac{\hbar^2 k^2}{2\mu^*}. \tag{4}$$

By comparing Eqs. (2) and (3), one can see that if the dielectric constant ε is constant, the binding energy of the ground state in the 2D case is 4 times higher than in the 3D case. This is a result of the motion quantization along the coordinate perpendicular to the plane. Furthermore, an additional factor that increases the exciton binding energy is the growth of either of the effective masses, which reduces the non-binding contribution to the binding energy from the kinetic energy of motion $E_{\rm kin}$ (4) along the corresponding coordinate [7]. We should also consider the peculiarities of the lattice state in the two-dimensional case where μ^* can grow, and ε can decrease at the contact with vacuum or vice versa, the increase of ε at the contact with a metal and the decrease of μ^* due to the inter-valley redistribution or elastic lattice deformation.

The above-indicated factors affecting the energy E_{ns} include the exciton binding at surface centers, similar to the "dusty plasma" effect [8], when the giant forces of the oscillators of bound excitons reveal themselves [9],

$$f_t = 8\left(\frac{\mu^*}{m_0}\frac{E_e}{E_t}\right)^{3/2}\frac{\pi A_s^3}{V} f_{\rm ex} \gg f_{\rm ex}, \tag{5}$$

where E_e is the exciton binding energy, E_t the exciton binding energy at the defect, V the unit cell volume, $f_{\rm ex}$ and A_s, are the oscillator strength and radius, respectively, and m_0 is the mass of a free electron. The key issue, which has not been fully clarified and needs to be resolved when considering this phenomenon, is the finding of the relation between the exciton binding energy and the oscillator strength, the parameters of surface centers (in particular, their concentration), which is important from the application viewpoint.

Among the external factors affecting the exciton binding energy in the 2D case, we should distinguish the influence of the adjacent medium by means of specular image forces [10, 11],

$$V_{fm} \cong \frac{q^2}{\varepsilon_1}\frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2}\frac{1}{4d}, \tag{6}$$

where V_{fm} is the potential energy of an electric charge in the electrostatic field at the point located at the distance d from the interface, and $\varepsilon_{1,2}$ are the dielectric constants of two adjacent media (semiconductor and insulator or metal). If $\varepsilon_2 < \varepsilon_1$ (ε_2 is the dielectric permittivity of insulator), excitons are repelled from the surface, otherwise, if $\varepsilon_2 > \varepsilon_1$ (ε_2 is the dielectric permittivity of a metal), they are attracted to the surface at frequencies ω much lower than the plasma resonance frequency ω_p, due to the known relation [10]

$$\varepsilon_2(\omega) \cong 1 - \frac{\varpi_p^2}{\varpi^2}. \tag{7}$$

In work [10], it was shown that, provided $\varepsilon_2 > \varepsilon_1$ (ε_2 describes a metal) and in a vicinity of the interface ($d/A_s \sim 0.1$, where A_s is the exciton radius), the exciton binding energy $E_{\rm ex} > 10R_y$. This factor leads to a significant increase of $E_{\rm ex}$ and the lifetime growth of the exciton state with respect to electron-hole pairs, which is important while developing 2D low-threshold emitting macroditch systems.

Let us consider another important factor that stabilizes the electron-hole continuum located outside the EH condensate region in the phase diagram (Fig. 1). This is a correlation interaction (or, in terms of quantum chemistry, a crystal field), which reflects the influence of the field regularity associated with a multiparticle interaction through the Coulomb potential.

In the low-temperature interval, excitons are almost ideal bosons with the spins equal to zero or one, which makes the phenomenon of Bose–Einstein condensation (BEC) possible. At higher temperatures, the deviation from this ideal behavior is associated with the presence of a term in the commutator of the exciton creation and annihilation operators related to the exciton interference (see, e.g., work [12]). However, at high concentrations, the behavior of excitons can differ significantly from that of bosons due to their interference. There is the classic work [13], where excitons, owing to the indicated factor, are described as fermions whose degeneracy temperature, by order of magnitude, is equal to the energy of interaction between neighbor magnetic moments.

In what follows, we also consider an electron-hole Fermi system under conditions of rather high excitation intensity, which makes the interference effects between the excitons substantial. For such a system, a phenomenon similar to the Wigner crystallization of an electron gas [14] becomes feasible. The growth of the ground state energy of such a system in the 2D case, if compared with the 3D one, takes place due to the growth of the correlation correction (with the negative sign corresponding to the attraction) as a result of the reduction of the average distance r_s between the particles in the 2D case as compared with that, r_v, in the 3D case: $\left|-q^2/r_s\right| > \left|q^2/r_v\right|$. From the physical point of view, the correlation interaction is responsible for a higher ordering degree in the system, in which the distance between the charged particles, $r_v(n) \sim 1/n^{1/3}$, always remains larger than the effective exciton radius, $r_v(n) > A_v$, or, in the two-dimensional case, $r_s(n) > A_s$, where $r_s(n) \sim 1/n^{1/2}$.

Nevertheless, in real solid-state radiating exciton structures at excitation levels close to and higher than the population inversion (i.e. at sufficiently high excitation intensities), the concentration of nonequilibrium charge carriers (NCCs) may exceed the limits of exciton existence on the phase diagram (Fig. 1) in both the 2D and 3D cases. As a result, if the excitation increases, the exciton subsystem decays into electron-hole pairs, at which, provided the corresponding amplification, the forced generation of radiation with a certain low-energy shift takes place (see, e.g., work [15]).

The influence of the correlation factor can be estimated on the basis of the Wigner crystallization criterion through the Mott electron localization constant C^*_{M} (see, e.g., work [7]),

$$n^{1/3}a^* = C^*_{\mathrm{M}} \cong 5 \times 10^{-2}, \tag{8}$$

where n is the NCC concentration, and a^* the effective Bohr radius of an electron.

Condition (8) is based on the requirement that the potential energy of attraction ΔE_{pot} should exceed the kinetic "loosening" energy E_{kin} ($\Delta E_{\mathrm{pot}} > E_{\mathrm{kin}}$), i.e.

$$n^{1/3} < \frac{m_n q^2}{\varepsilon \hbar^2} n^{1/3} \text{ and } n^{1/3} < \frac{2q^2}{3\varepsilon kT} \tag{9}$$

in the degenerate and non-degenerate cases, respectively. Estimates show that criterion (8) is not obeyed in the degenerate case at the NCC concentration of about 10^{18} cm^{-3}. In the non-degenerate case, it is obeyed only at sufficiently low temperatures ($T \leq 32$ K). Thus, the energy of correlation interaction is an additional factor. At sufficiently low temperatures but sufficiently high excitation levels, at which the exciton interference can no longer be neglected, it is aimed at ordering the structure of the EH continuum (Fig. 4).

3. Discussion and Conclusions

It should be noted that the role of various indicated factors remains unclear at length and is a subject of intense researches (see, e.g., works [16–18] and references therein). In any case, a reduction of the dielectric screening and an enhancement of the Coulomb interaction lead to intense exciton and multiparticle effects in 2D systems.

From whence, it follows that the excitation of the exciton 2D condensate as an active material for producing the stimulated radiation has considerable and principal advantages over the bulk case. Moreover, the 2D dimensionality conditions are satisfied even in a rather thick layer with nano-sized crystallites, which forms a nanocomposite film in a macroresonant ditch (see, e.g., work [19]). Figure 5 illustrates an example of such a structure with a resonator of the vertical type.

An essential feature of the condensed exciton EH state at temperatures below the critical phase transition temperature, when the interference factor is low and excitons are bosons, is the possibility of BEC [3, 20]. Under this condition, the particles collapse

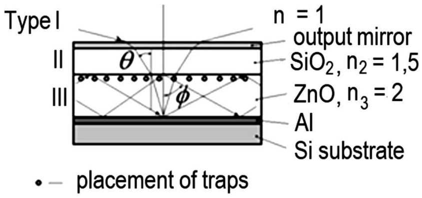

Fig. 5. Schematic diagram of a 2D-like device of the vertical type as a source of stimulated radiation: $T_{\rm ip}$ III is the mode supported by the active ZnO layer

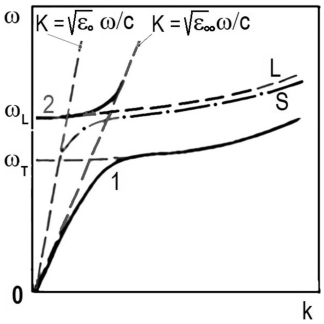

Fig. 6. Dispersion of surface exciton-polaritons: 0 corresponds to the dielectric constant of the environment, ε the dielectric constant at high frequencies, L the longitudinal wave dispersion, S the dispersion of the surface exciton-polariton wave [21]

into a macroscopic single-phase condensate, which reveals, in contrast to phonon polaritons, the spatial dispersion [21]

$$\varepsilon(\varpi,k)=\varepsilon_\infty\left(1+\frac{\varpi_L^2-\varpi_T^2}{\varpi_T^2-\varpi^2+\beta k^2-i\varpi\Gamma}\right),\qquad(10)$$

where $\beta=\hbar\omega_T/M$ is the spatial dispersion coefficient, M is the sum of the effective electron and hole masses, Γ the phenomenological damping factor, and ω_T and ω_L are the transverse and longitudinal exciton frequencies, respectively. The condensate in the state with unlimited lifetime exhibits the collective quantum behavior like a superfluid liquid [22]. At the beginning of researches, the BEC was observed only in rarefied atomic gases at micro-Kelvin temperatures. At present, encouraging results of the laser generation in micro-resonator ditches have been obtained. They showed a high efficiency of laser generation at room temperatures due to the dominance of exciton-polariton oscillations [23]. The deep dispersion of exciton-polariton particles, in particular, in the region with the negative values of dielectric constant – in the frequency interval from ω_{TO}(min) to ω_{LO}(max), Fig. 6 – leads to extremely small effective masses, which are $10^4\div10^5$ times smaller than the free electron mass. This occurs owing to the corpuscular-wave origin ("half-matter–half-light") of exciton-polaritons [24].

The low density of states ensures their high occupancy even at extremely low excitation levels. This circumstance gives a prospect of obtaining the extremely low-threshold and effective exciton-polariton microsources of induced radiation.

From the aforesaid, a conclusion can be drawn that, when the pump intensity increases, the condensed exciton state limited by the phase diagram (Fig. 1) goes beyond those limits to the state of heated EH gas, where the stimulated radiation generation takes place. Hence, the role of the exciton condensate is similar to that of a metastable level with a relatively long lifetime in laser systems and consists in the pre-accumulation of the NCC concentration in order to achieve the population inversion, which is required for the induced radiation emission to start.

On the basis of a comparison between the macro- and microditch resonators from the viewpoint of the feasibility and efficiency of their application as induced-radiation sources, the conditions and the factors giving rise to the exciton binding energy growth and, accordingly, the stability of the condensed EH state in the near-surface layers, which were considered above, allow the following advantages of the 2D EH continuum confined by a macroditch resonator to be distinguished:

• a possibility to enhance the specular image force factor by introducing a boundary layer with a higher refractive index, $\varepsilon_2>\varepsilon_1$, into the active material (for example, if ε_2 corresponds to a metal);

• the application of a metal layer to increase $E_{\rm ex}$; the layer is used as an element of the end mirror of the resonator ditch and, at the same time, as an adjacent medium for the specular reflection;

• the creation of additional surface centers of exciton capture, which enhance the stability of the condensed state, similarly to the "dusty plasma" effect;

• the action of correlation forces; under certain specific conditions, it can be an additional factor in maintaining the EH subsystem in the ordered state.

Those advantages, together with the simplicity of the resonator macroditch formation technology, make promising the development of 2D sources of induced radiation as more technologically attractive in comparison with 2D systems based on microresonant ditches. This is another demonstration of the prospects of using 2D semiconductor systems as a basis for creating a new generation of optoelectronic devices [25].

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ЕЛЕКТРОННО-ДІРКОВИЙ УПОРЯДКОВАНИЙ КОНДЕНСАТ ЯК ПЕРСПЕКТИВНЕ ЛАЗЕРНЕ 2D СЕРЕДОВИЩЕ ДЛЯ КІМНАТНИХ ТЕМПЕРАТУР

На основі теоретичних і експериментальних результатів, отриманих авторами, та детального літературного огляду зроблено порівняльний аналіз процесів формування і утримування електронно-діркового (ЕД) екситонного континууму у 3D та 2D напівпровідниках. За діаграмами фазового стану, спектрами фотолюмінесценції і люкс-люксовими залежностями продемонстровано суттєве збільшення енергії екситонного зв'язку E_{ex} та стабільності ЕД континууму в 2D порівняно з 3D випадком. Розглянуто вплив визначальних фізичних чинників, відповідальних за підвищення E_{ex} та стабільність ЕД континууму у випадку 2D: зв'язування екситонів на мілких домішкових центрах, сил дзеркального відображення та кореляційного чинника. Розглянуто особливості екситонно-поляритонного та електронно-діркового континуумів з урахуванням статистик Бозе–Ейнштейна та Фермі–Дірака з урахуванням можливості формування e-h кристалу. Сформульовані завдання для подальшого теоретичного розгляду, а також названо переваги і перспективи використання макрорезонаторних 2D кювет, що не потребують складної та витратної МПЕ технології, для створення оптоелектронних пристроїв нового покоління.

Ключові слова: електронно-дірковий конденсат, екстиноний континуум, центри захоплення, 2D екситони, макрокюветний резонатор, вимушене випромінювання.