5. Eletskiĭ AV *Usp. Fiz. Nauk* **167** 945 (1997) [*Phys. Usp.* **40** 899 (1997)]
6. Dresselhaus M S, Dresselhaus G, Eklund P C *Science of Fullerenes and Carbon Nanotubes* (San Diego: Acad. Press, 1996)
7. Tans S J et al. *Nature* (London) **386** 474 (1997)
8. Bockrath M et al. *Science* **275** 1992 (1997)
9. White C T, Todorov T N *Nature* (London) **393** 240 (1998)
10. Egger R, Gogolin A O, cond-mat/9803128; *Eur. Phys. J. B* **3** 281 (1998)
11. Bockrath M et al. *Nature* (London) **397** 598 (1999)
12. Haddon R C *Nature* (London) **388** 31 (1997)
13. Lin M F, Chuu D S *Phys. Rev. B* **57** 6731 (1998)
14. Odintsov A A, Smit W, Yoshioka H, cond-mat/9805164
15. Tsebro V I, Omel'yanovskiĭ O E, Moravskiĭ A P *Pis'ma Zh. Eksp. Teor. Fiz.* **70** 457 (1999) [*JETP Lett.* **70** 462 (1999)]
16. Mendelssohn K *Proc. R. Soc. London Ser. A* **152** 34 (1935)
17. Büttiker M, Imry Y, Landauer R *Phys. Lett. A* **96** 365 (1969)
18. Szopa M, Zipper E *Int. J. Mod. Phys. B* **9** 161 (1995)
19. Stebelski M, Lisovski M, Zipper E *Eur. Phys. J. B* **1** 215 (1998)
20. Lisovski M, Zipper E, Stebelski M *Phys. Rev. B* **59** 8305 (1998)
21. Haddon R C *Nature* (London) **378** 249 (1995)
22. Chauvet O et al. *Phys. Rev. B* **52** 6963 (1995)
23. Ovchinnikov A A *Phys. Lett. A* **155** 95 (1994)
24. De Heer W A et al. *Science* **268** 845 (1995)
25. Colbert D T et al. *Science* **266** 1215 (1994)
26. Kiselev N A et al. *Carbon* **37** 1093 (1999)
27. Eletskiĭ A V, Smirnov B M *Usp. Fiz. Nauk* **163** 33 (1993) [*Phys. Usp.* **36** 202 (1993)]
28. Eletskiĭ A V, Smirnov B M *Usp. Fiz. Nauk* **165** 977 (1995) [*Phys. Usp.* **38** 935 (1995)]
29. *Null-balance Magnetometer with Capacitance Sensor. Internal Specification.* International Laboratory of High Magnetic Fields and Low Temperatures (Wroclaw, 1998)
30. Campbell A M, Evetts J E *Critical Currents in Superconductors* (London: Taylor and Francis, 1972)
31. Brandt E H *Rep. Prog. Phys.* **58** 1465 (1995)
32. Kasumov A Yu et al. *Science* **284** 1508 (1999)
33. Frank S et al. *Science* **280** 1744 (1998)

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Two-dimensional excitonic polaritons and their interaction

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Considerable recent attention has been focused on semiconductor microcavities (MCs) with plane Bragg mirrors owing to the possibility of profoundly altering the exciton properties in quantum wells placed in the antinode of the electromagnetic field [1]. The system of strongly interacting excitons and photons in an MC is described in terms of excitonic MC polaritons. These polaritons are realized only in an MC with a relatively high Q factor, when the interaction between the photon mode (C) in the MC and the exciton mode (X) in the quantum well exceeds their broadenings. The excitonic polaritons in plane MCs are quasi-two-dimensional and are characterized by extremely small masses, $\sim 10^{-5} m_0$, and large dimensions, on the order of a micron [2]. Unlike volume excitonic polaritons, which are stable quasiparticles, they have a very short lifetime, on the order of several picoseconds. The qualitative difference derives from the fact that MC polaritons annihilate without conservation of the momentum in the direction perpendicular to the plane of the quantum well.

The quasi-two-dimensional polaritons in MCs constitute a very interesting object of research. First, the light confinement in an MC is responsible for a strong multiplication of the electromagnetic field and should therefore result in a dramatic enhancement of nonlinear effects. Second, the MC polaritons are bosons. Owing to the smallness of their mass, a polariton system admits, unlike an exciton one, attainment of a significant filling of states for a relatively low overall density, whereby the effect of the internal fermion structure of the excitons can be neglected. In this case, the features predicted for bosons, like stimulated scattering and condensation in the momentum $\mathbf{k}$ space (Bose condensation) [3], would be expected to manifest themselves in the system of MC polaritons.

Among the manifestations of stimulated scattering is a superlinear increase of the polariton radiation at high excitation densities. This mode was discovered by Pau et al. [4] in studies of a GaAs/AlAs MC under intense light excitation with photon energies $\hbar\omega$ exceeding the width of the GaAs forbidden band E_g. The effect was interpreted as a sequel to the Bose condensation of polaritons and was termed 'boser.' More comprehensive studies [5] revealed that the stimulated emission arises in circumstances where the Coulomb interaction in the system is strongly screened owing to a high density of photoexcited carriers and the exciton–photon interaction is suppressed, i.e., is realized in the mode of weak interaction. Relying on their calculations, Kira et al. [5] arrived at the conclusion that there is no way of realizing a boser in MC structures in the mode of strong interaction. However, the observation of the superlinear polariton luminescence mode in the strong interaction mode was reported recently in II–VI semiconductor compound MC structures, where the exciton binding energy and, hence, the critical density is significantly higher than in GaAs [6]. However, it is pertinent to note that the excitation with $\hbar\omega > E_g$ is extremely inefficient for the realization of a high filling of the polariton states, because the time for the relaxation to the polariton states at the bottom of the lower polariton branch (LPB) is comparable with the polariton lifetime.

Here, in order to realize a high LPB filling, we resorted to the resonance excitation either to the bottom of the upper polariton branch (UPB) or directly to the LPB 1–3 meV above its minimum. Furthermore, unlike earlier papers, we excited the system using circularly polarized light. It turned out that the polariton spin relaxation time τ_s in this case was significantly shorter than their lifetime τ_l. We were therefore able to realize a spin-polarized polariton system with a relatively high degree of spin polarization and observe a variety of nonlinear effects both in the radiation intensity and in the degree of circular polarization.

We studied nonlinear effects in the emission of a GaAs/AlAs MC with a set of InGaAs quantum wells in the active layer under resonance of the photon and exciton modes at $k = 0$, when the polaritons with $k = 0$ are half exciton and half photon in nature. A tunable titanium–sapphire laser was used to excite the MC luminescence. The sample was placed in a cryostat at a temperature of 1.8–10 K. The experiments were carried out on structures with a Rabi splitting of 5–7 meV and an energy mismatch of the X and C modes $|\Delta| < 0.6$ meV. The polariton dispersion law measured for low excitation densities is shown in Fig. 1b.

Figure 1a gives the MC emission spectra recorded under nonresonance excitation above E_g. For low P, the emission

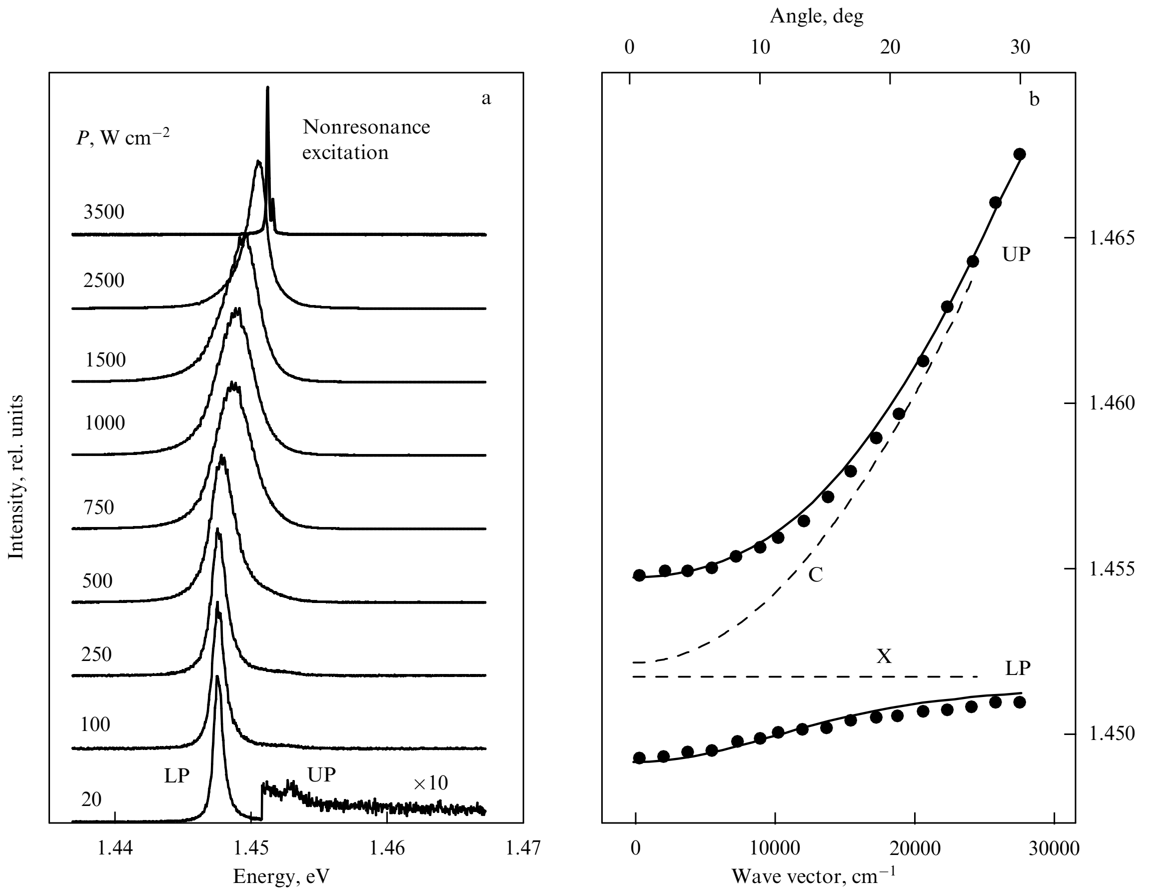

Figure 1. MC emission spectra under nonresonance excitation above E_g recorded for $T = 6$ K, $\Phi = 0°$, and different excitation densities P (a). Polariton dispersion law measured at low excitation densities (b).

spectrum shows two lines corresponding to the LPB (LP) and UPB (UP) $k = 0$ polariton emission. With increasing P, the LP line monotonically broadens and shifts by 3 meV toward higher energies, and its position for $P > 1500$ W cm^{-2} corresponds to the exciton mode energy. This implies that the exciton – photon system passes into the mode of weak interaction owing to screening of the Coulomb interaction in a dense photoexcited system. As the excitation density is increased further, a narrow line appears in the spectrum, which corresponds to oscillation with the photon mode energy. This behavior is identical to that observed previously [4].

The behavior of the MC emission changes qualitatively when passing to resonance excitation with circularly polarized σ^+ light to the bottom of the UPB. Figure 2 shows the MC emission spectra recorded with two circular polarizations, σ^+ and σ^- for two angles $\Phi = 0°$ and 25°. They correspond respectively to the emission of polaritons with wave vectors $k < 10^3$ cm^{-1} [strongly mixed exciton – photon states (LP)] and $k = 2.6 \times 10^4$ cm^{-1} (the admixture of the photon mode is insignificant, exciton-like states (X)). The σ^+-polarized radiation (the LP$^+$ and X$^+$ peaks) is significantly more intense than the σ^--polarized radiation (the LP$^-$ and X$^-$ peaks), allowing a conclusion that $\tau_s > \tau_l$. The behavior of the spectra for $\Phi = 0°$ and 25° is qualitatively different. With increasing excitation density, the LP$^+$ peak shifts toward higher energies by about 1.3 meV and exhibits, for $P > 600$ W cm^{-2}, superlinear growth and significant line narrowing. By contrast, the LP$^-$ peak exhibits a sublinear dependence, is hardly shifted in energy, and slightly broadens. The emission of exciton-like polaritons ($\Phi = 25°$, Fig. 2b) exhibits a sublinear dependence in both polarizations, and the ratio of integrated intensities of the lines in the σ^+ and σ^- polarizations I_{X^+}/I_{X^-} is hardly changed. Furthermore, it is evident from Fig. 2b that the X line experiences a strong broadening with increasing P, as would be expected due to the exciton – exciton collisions. Note, however, that we are dealing with an exciton – photon system in the strong interaction mode up to the maximum excitation density used. The LPB (LP$^+$) and UPB (UP$^+$) energies as functions of P are given in Fig. 2c. The LPB energy was determined from the emission spectra by the position of the LP$^+$ peak (Fig. 2a), while the UPB position was found from the spectral position of the peak in the LPB luminescence photoexcitation spectrum for $k = 0$ (the UP$^+$ peak). One can see from Fig. 2c that the LP$^+$ and UP$^+$ energy changes are equal in magnitude and opposite in sign, but the Rabi splitting does not vanish completely. The screening of the Coulomb interaction with increasing excitation density leads to a reduction of the splitting LP$^+$ – LP$^-$ from 5.5 meV to 3 meV for $P = 1300$ W cm^{-2}. Therefore, the nonlinear effects in the polariton radiation take place in the strong interaction mode, i.e., refer to mixed exciton – photon rather than purely photon modes, as with band-to-band excitation (Fig. 1a).

The superlinear growth of the LP$^+$ line intensity (I_{LP^+}) for $P > 700$ W cm^{-2} (Fig. 2a) is indicative of the onset of a new scattering mechanism. This mechanism may prove to be exciton – exciton scattering. However, the radical difference

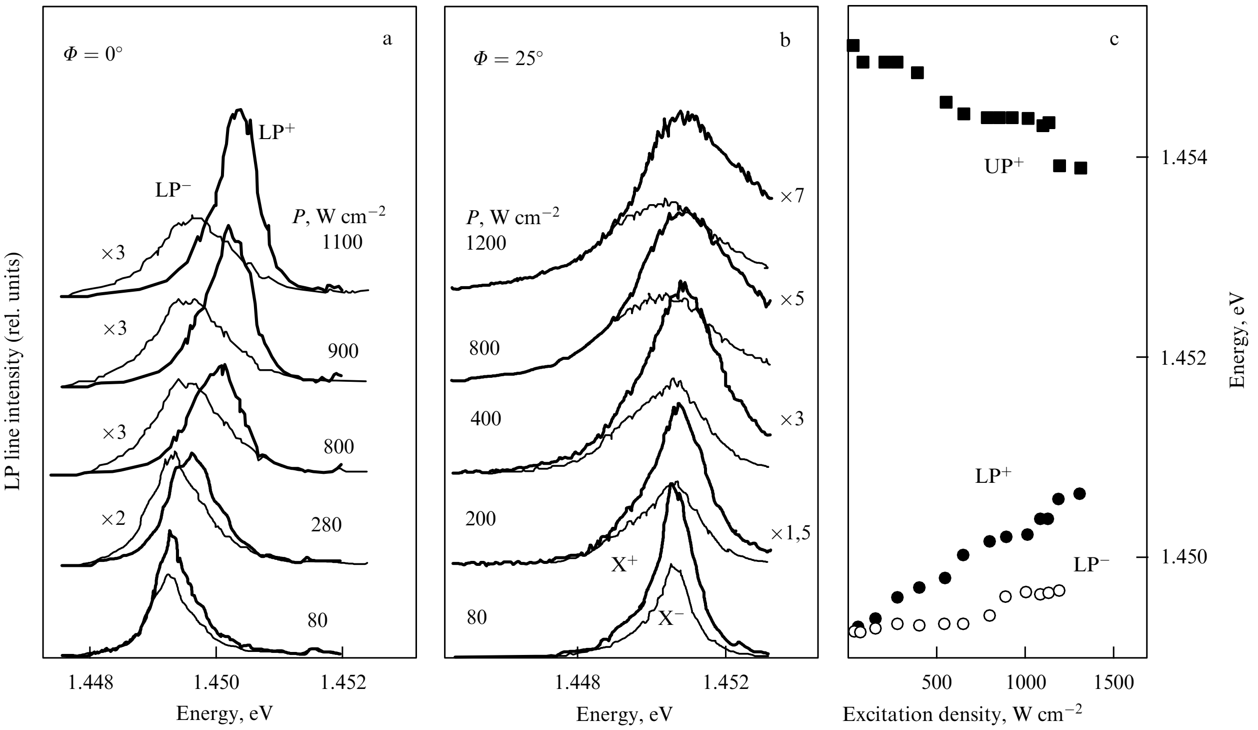

Figure 2. Polarized MC emission spectra, σ^+ and σ^-, for $\Phi = 0°$ (a) and 25° (b) recorded with an angular resolution of 0.5° under σ^+-light excitation to the bottom of the UPB. The intensity is normalized to the laser power. (c) LPB (LP^+) and UPB (UP^+) energies as functions of the excitation density.

in the behavior of polariton emission in the σ^+ and σ^- polarizations cannot be attributed to the exciton–exciton scattering. In fact, for the polaritons photoexcited at the bottom of the UPB, a direct single-phonon scattering to the states at the bottom of the LPB is impossible. The polaritons are first scattered to the exciton states with large k ($> 10^6$ cm^{-1}) or to localized exciton states and only then relax to the polariton states with small k. For low P, the degree of polarization of the LP line, ρ_{LP}, is therefore somewhat lower than that of the X line, ρ_X. However, referring to Fig. 2, ρ_{LP} rises steeply while ρ_X hardly changes with increasing excitation density. In this case, the LP-to-X line intensity ratio remains invariable in σ^- and increases by nearly an order of magnitude in the σ^+ polarization. So the preferred scattering of the excitons with $k \sim 10^6$ cm^{-1} to the σ^+ states with $k \sim 10^3$ cm^{-1} (the LP line) in comparison with $k \sim 2 \times 10^4$ cm^{-1} (the X line) cannot be explained in the context of the exciton–exciton and exciton–phonon scattering without the assumption that there operates a mechanism of stimulated scattering to the σ^+ states at the bottom of the LPB. The stimulated scattering may originate when the filling ν of the state with $k = 0$ is higher than unity because the polaritons are bosons. Clearly, in circumstances where the excitation is effected by σ^+-polarized light and the spin relaxation time is high, the critical filling $\nu = 1$ is first attained for the σ^+ state. As this takes place, there emerge prerequisites to the stimulated scattering to exclusively this state irrespective of the scattering mechanism (exciton–phonon, exciton–exciton, or direct two-photon scattering of the exciting light). The latter requires simultaneous fulfillment of the energy and momentum conservation laws. One can see from the dispersion law in Fig. 1b that such a mechanism, too, can prove to be efficient for high photoexcitation densities only due to a strong broadening of the excitonic states. Additional calculations are required to determine the specific scattering mechanism. However, the above experimental data alone suggest that the use of resonance excitation near the bottom of the UPB, unlike the band-to-band excitation, allows the preparation of a polariton system wherein nonlinear effects reveal themselves in the form of a strong exciton–photon interaction. The relative fraction of low-k ($< 10^4$ cm^{-1}) polaritons remains not high enough, with the result that the nonlinear effects arise not far from the threshold of passage to the weak-coupling mode.

One can see from the polariton energy dispersion given in Fig. 1b that the excitation in the 'bottleneck' region, below the energy of free excitons E_X, is most attractive. In this case, there is reason to hope to avoid the filling of high-k excitonic states and, hence, heighten the fraction of the polariton states. However, in the experimental realization of this idea, a problem emerges stemming from the excitation of, primarily, localized excitons, for which the time of scattering to the polariton states with $k < 10^4$ cm^{-1} (hundreds of picoseconds) is far longer than their lifetime. Since the radiative decay time for the polaritons at the bottom of the LPB is extremely short (several picoseconds), their concentration proves, as a rule, to be low and the distribution over k is far from that of thermodynamic equilibrium. Only realizing the conditions whereby the scattering of the exciting radiation to the polariton states bypasses the states of localized excitons can qualitatively change the situation. Referring to the LPB dispersion law (see Fig. 1), this situation may arise in the resonance excitation near the point of inflection ($k_{ex} \sim 1.6 \times 10^4$ cm^{-1}, or $\Phi \sim 16°$). In this case, direct two-photon scattering to the polariton states with $\mathbf{k} = \mathbf{k}_{ex} - \mathbf{k}$ and $\mathbf{k} = \mathbf{k}_{ex} + \mathbf{k}$ is possible, wherein the energy and momentum conservation laws are obeyed simultaneously: $2\hbar\omega(\mathbf{k}_{ex}) = \hbar\omega(\mathbf{k}_{ex} - \mathbf{k}) + \hbar\omega(\mathbf{k}_{ex} + \mathbf{k})$, and therefore one would expect

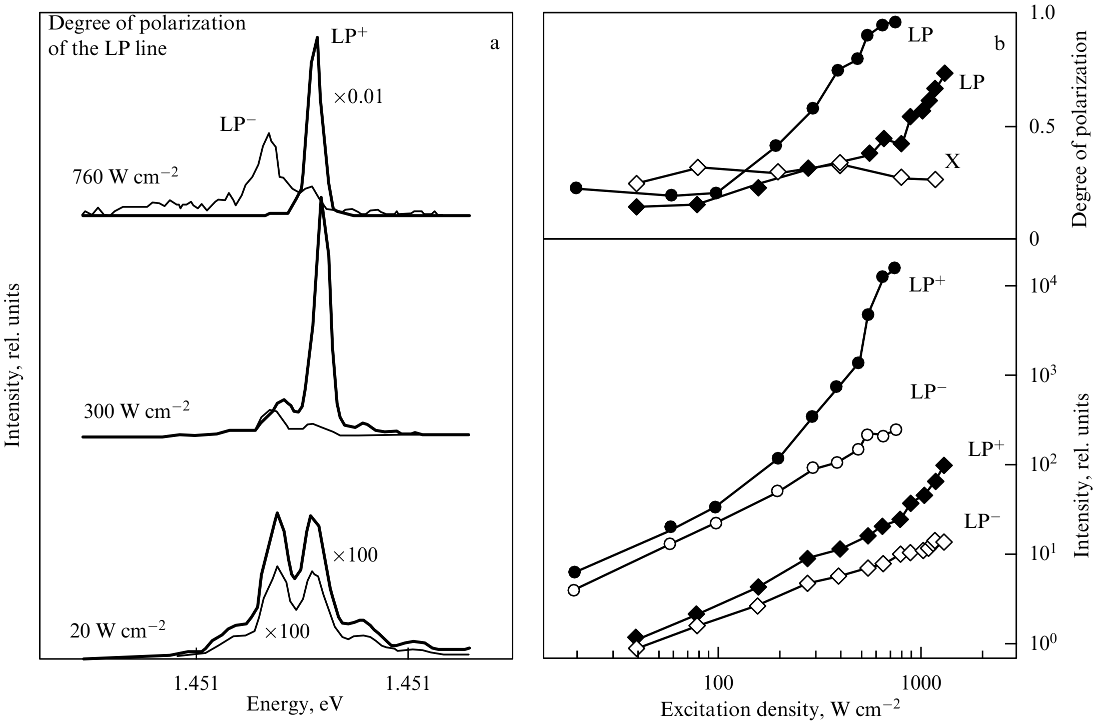

Figure 3. (a) Spectra of polariton radiation at 2 K in two polarizations for $\Phi = 0°$ recorded under σ^+-light excitation to the LPB for $\Phi = 16°$. (b) LP line intensity and degree of its circular polarization as functions of the excitation density with σ^+-light resonance excitation to the bottom of the UPB (UP) (rhombi) and to the LPB for $\Phi = 16°$ (LP) (circles).

a sharp increase in the polariton excitation efficiency at high excitation densities.

Figure 3a gives the experimental polariton emission spectra recorded for 2 K in two polarizations for $\Phi = 0°$ and under σ^+-light excitation for $\Phi = 16°$. A comparison of Figs 2a and 3a shows that the behavior of emission spectra with resonance excitation to the bottom of the UPB and to the point of inflection of the LPB are qualitatively similar. However, in the latter case the total density of photoexcited excitons and, hence, the screening of the Coulomb interaction in the system is significantly lower and it is possible to attain substantially stronger nonlinearities before the polariton states begin to collapse owing to the screening of the Coulomb interaction. In this case, the LP line in the σ^+ polarization narrows down to 0.07 meV and its degree of circular polarization exceeds 95%. The dependences of the LP line intensity and the degree of circular polarization ρ_{LP} on the excitation density are given in Fig. 3b. Reference to Fig. 3b shows that four excitation density regions with qualitatively different behavior can be distinguished. In the first region, $P < 150$ W cm^{-2}, the dependences of I_{LP^+} and I_{LP^-} on P are close to linear while $\rho_{\mathrm{LP}}(P)$ varies only slightly and lies between 30 and 40 %. This is a region of linear absorption, in which primarily localized excitons are excited, the scattering to the polariton states at the bottom of the LPB is extremely weak, and the quantum yield does not exceed a tenth of one percent. Further, in the $P = 150-450$ W cm^{-2} region, a quadratic $I_{\mathrm{LP}^+}(P)$ dependence is observed, the $I_{\mathrm{LP}^-}(P)$ dependence remains close to linear, and ρ_{LP} begins to rise steeply. This is the region in which the two-photon scattering $2\hbar\omega(\mathbf{k}_{\mathrm{ex}}) = \hbar\omega(k=0) + \hbar\omega(\mathbf{k} = 2\mathbf{k}_{\mathrm{ex}})$ comes to be the most efficient mechanism of filling the polariton states, and the quantum yield of the LP radiation begins to grow strongly. A direct confirmation of this process is the occurrence, in this excitation density domain, of a narrow line in the MC emission spectrum above the energy of the excitation light photon. This line is located at the energy $\hbar\omega \sim 2\hbar\omega(\mathbf{k}_{\mathrm{ex}}) - \hbar\omega(k=0)$, is observable at recording angles $\Phi = 30° - 34°$ that correspond precisely to the quantity $\mathbf{k} = 2\mathbf{k}_{\mathrm{ex}}$, and is strongly ($\rho > 50\%$) polarized, like the LP line for $k = 0$. As P increases further, the quadratic dependence of $I_{\mathrm{LP}^+}(k=0)$ is first replaced with a steeper, nearly exponential one, and then, for $P > 650$ W cm^{-2}, begins to saturate. In this case, the $I_{\mathrm{LP}^-}(P)$ dependence for $k = 0$ is distinctly sublinear, the degree of circular polarization of the LP line for $k = 0$ and for $\mathbf{k} = 2\mathbf{k}_{\mathrm{ex}}$ is close to unity, and the quantum radiation yield in the $\mathrm{LP}^+(k=0)$ mode for $P = 1000$ W cm^{-2} approaches 10%. The transition from the quadratic to a nearly exponential $I_{\mathrm{LP}^+}(P)$ dependence is indicative of the onset of stimulated two-photon scattering. As already noted, this process develops, owing to the bosonity of polaritons, when $\nu_{\mathrm{LP}}(k=0)$ exceeds unity. An additional confirmation of the stimulated nature is a nearly 100% polarization of the $\mathrm{LP}(k=0)$ line in this region. We also note the absence of exponential growth of the polariton radiation intensity in the region $0 < k < k_{\mathrm{ex}}$. For them, the energy and momentum conservation laws are also obeyed in the process of two-photon scattering, but $\nu_{\mathrm{LP}}(k) \ll \nu_{\mathrm{LP}}(0)$.

In the region $P = 450-650$ W cm^{-2}, the exponential growth of $I_{\mathrm{LP}^+}(k=0)$ is afforded both due to the efficiency increase of direct two-photon scattering and to the stimulated scattering from the photoexcited states of localized excitons and polaritons with $k > 0$ with the emission of acoustic phonons. The latter process becomes efficient for $\nu_{\mathrm{LP}}(E) > \nu_{\mathrm{ph}}(E)$, where the energy E is reckoned from the bottom of the LPB and $\nu_{\mathrm{ph}}(E)$ is the occupation number of the phonon mode. For $T = 2-5$ K, ν_{ph} declines rapidly with increasing E, and the inequality $\nu_{\mathrm{X}}(E) > \nu_{\mathrm{ph}}(E)$ is easily attained even for $E \sim 0.5$ meV. The exponential growth of the coefficient of conversion of the exciting radiation with energy $\hbar\omega_{ex}$ to the radiation with energy $\hbar\omega(k = 0)$ saturates on reaching a value of the conversion coefficient of about 10%. This is a relatively high performance criterion among the known types of optical parametric converters. A comparison of the dependences of I_{LP^+}, I_{LP^-}, and ρ_{LP} on the excitation density plotted in Fig. 3b for the case of resonance excitation to the bottom of the UPB and to the LPB for $\Phi = 16°$ shows that their behavior is qualitatively similar. However, in the latter case the threshold of the onset of nonlinearities is significantly lower owing to a more efficient two-photon scattering, with the effect that it is possible to attain higher polariton densities at the bottom of the LPB.

So far we have considered the behavior of a polariton system under resonance excitation by circularly polarized light. The efficiency of two-photon scattering depends on how far the doubled frequency is offset from the energy of optically operative transitions. In our case, the energy $2\hbar\omega(k_{ex})$ is close (only $1-2$ meV lower) to the energy of the ground biexcitonic state. This state in InGaAs quantum wells is a spin singlet and is optically operative under excitation by linearly polarized light. When excited by elliptically polarized light, its luminous activity monotonically falls to zero on increasing the degree of circular polarization of the exciting radiation ρ_{ex} from zero to unity. One would therefore expect a sharp increase in the two-photon scattering in going over to linearly polarized light.

Figure 4 shows how ρ_{ex} and the LP line intensity depend on ρ_{LP}. For a low excitation density, when the two-photon scattering is weak, I_{LP} increases only slightly as ρ_{ex} decreases from unity to zero, while ρ_{LP} falls off smoothly from 0.3 to zero. For $P = 540$ W cm^{-2}, the behavior of I_{LP} and ρ_{LP} changes substantially: I_{LP} increases severalfold, primarily for $\rho_{ex} < 0.6$. In this case, ρ_{LP} also rises significantly with decreasing ρ_{ex} down to $\rho_{ex} = 0.6$. For a higher excitation density, in the saturation region, the degree of circular polarization remains close to 100% over a broad range of $\rho_{ex} > 0.4$, and only then drops to zero. This behavior of the intensity and the degree of polarization of the LP line under excitation by elliptically polarized light is additional evidence in favor of the facts that, for high P, we are dealing with a two-photon process and this process is a stimulated one.

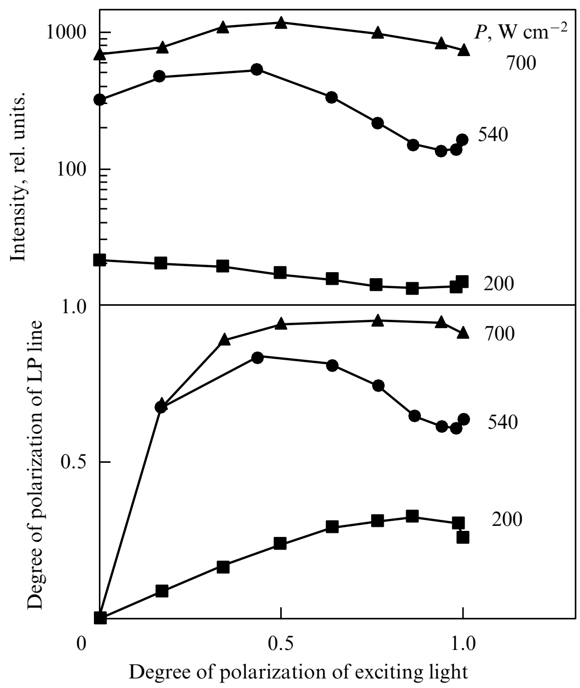

Figure 4. Degree of circular polarization ρ_{LP} and LP line intensity, $I_{LP} = I_{LP^+} + I_{LP^-}$, as functions of the degree of circular polarization of the exciting light for three fixed excitation densities.

Thus, we have studied the emission and luminescence photoexcitation spectra of MCs with quantum wells over a broad range of resonance photoexcitation densities, which were recorded with a high angular resolution. Relying on these studies, we determined the conditions whereby the photoexcitation of a polariton system can be achieved with a high occupancy at the bottom of the LPB. Under these conditions, strong nonlinearities in the intensity and the degree of polarization of the polariton radiation were revealed and investigated. These nonlinearities were shown to be attributable to the bosonity of polaritons.

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References

1. Skolnick M S, Fisher T A, Whittaker D M *Semicond. Sci. Technol.* **13** 645 (1998)
2. Houdre R et al. *Phys. Rev. Lett.* **73** 2043 (1994)
3. Imamoglu A et al. *Phys. Rev. A* **53** 4250 (1996)
4. Pau S et al. *Phys. Rev. A* **54** 1789 (1996)
5. Kira M et al. *Phys. Rev. Lett.* **79** 5170 (1997)
6. Le Si Dang et al. *Phys. Rev. Lett.* **81** 3920 (1998)