

Figure 6. Classification of the spectral lines of tin in the 2% interval about the 13.5-nm wavelength by their ionization stages. (Borrowed from Ref. [26].)

radiation source in the interval $\lambda = 13.5 \pm 0.135$ nm requires producing a plasma with a prevalence of the ions $\text{Sn}^{+10} - \text{Sn}^{+12}$.

Similar investigations of the spectra of indium ions with the nuclear charge $Z_C = 49$ performed in parallel confirmed the main results of tin spectrum analysis ($Z_C = 50$). It was noted that the 4–4 transition spectra for different ionization degrees gradually separate in wavelength as the nuclear charge decreases. This is favorable to a more reliable classification of transitions in each specific ion. Presently underway in the Department of Atomic Spectroscopy of ISAN is an investigation of the 4–4 transition spectra in Pd, Ag, and Cd ions ($Z_C = 46–48$) intended to verify the results of our analysis of tin ionic spectra. Nevertheless, even now the data obtained are advantageously used as the spectroscopic foundation for modeling, development, and optimization of 13.5 nm lithography radiation sources based on tin ions.

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References

1. Martin W C, Zalubas R, Hagan L, Natl. Stand. Ref. Data Ser. Natl. Bureau Stand. NSRDS-NBS 60 (Washington: National Bureau of Standards, 1978)
2. BIBL: Bibliography database on atomic spectra, <http://das101.isan.troitsk.ru/bibl.htm>
3. Biemont E, Palmeri P, Quinet P *Astrophys. Space Sci.* **269** 635 (1999); D.R.E.A.M. Database on Rare Earths at Mons University, <http://w3.umh.ac.be/~astro/dream.shtml>
4. Tolstikhina I Yu, Churilov S S, Ryabtsev A N, Koshelev K N, in *EUV Sources for Lithography* Vol. PM149 (Ed. V Bakshi) (Bellingham, Wash.: SPIE Press, 2006) p.113
5. Svendsen W, O'Sullivan G *Phys. Rev. A* **50** 3710 (1994)
6. Crosswhite H, Private communication (1976)
7. Aldenius M, Master Thesis (Lund: Depart. of Physics, Univ. of Lund, 2001)
8. Ryabchikova T, Ryabtsev A, Kochukhov O, Bagnulo S *Astron. Astrophys.* **456** 329 (2006)
9. Kochukov O, Ryabchikova T, Piskunov N *Astron. Astrophys.* **415** L13 (2004)

10. Cowan R D *The Theory of Atomic Structure and Spectra* (Berkeley: Univ. of California Press, 1981)
11. Wahlgren G M *Phys. Scripta* **T100** 22 (2002)
12. Biemont E, Quinet P *Phys. Scripta* **T105** 38 (2003)
13. Sugar J, Spector N J. *Opt. Soc. Am.* **64** 1484 (1974)
14. Den Hartog E A, Wickliffe M E, Lawler J E *Astrophys. J. Suppl.* **141** 255 (2002)
15. Ryabchikova T et al. *Astron. Astrophys.* **343** 229 (1999)
16. Wyart J F J. *Opt. Soc. Am.* **68** 197 (1978)
17. Wyart J F, Buache-Arnold C I *Phys. Scripta* **22** 583 (1980)
18. Mashonkina L, Ryabchikova T, Ryabtsev A *Astron. Astrophys.* **441** 309 (2005)
19. Wyart J-F, Tcham-Brillet W-Ü L, Churilov S S, Ryabtsev A N *Astron. Astrophys.* **483** 339 (2008)
20. Churilov S S, Kildiyarova R R, Ryabtsev A N, Kramida A E, Joshi Y N *Phys. Scripta* **50** 463 (1994)
21. Azarov V I, Joshi Y N, Churilov S S, Ryabtsev A N *Phys. Scripta* **50** 642 (1994)
22. Sugar J, Kaufman V, Rowan W L J. *Opt. Soc. Am. B* **9** 1959 (1992)
23. Azarov V I, Joshi Y N J. *Phys. B* **26** 3495 (1993)
24. Churilov S S, Ryabtsev A N *Opt. Spektrosk.* **100** 721 (2006) [*Opt. Spectrosc.* **100** 660 (2006)]
25. Churilov S S, Ryabtsev A N *Opt. Spektrosk.* **101** 181 (2006) [*Opt. Spectrosc.* **101** 169 (2006)]
26. Churilov S S, Ryabtsev A N *Phys. Scripta* **73** 614 (2006)

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Strong correlations and new phases in a system of excitons and polaritons. A polariton laser

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One of the most beautiful phenomena in many-particle physics — Bose–Einstein condensation (BEC) in a system of particles with nonzero mass m obeying the Bose statistics — was predicted by Einstein already in 1925 soon after the publication of Bose's paper on the thermodynamically equilibrium distribution of photons. Einstein showed that at a temperature T below the critical value

$$T_c = \frac{3.31 \hbar^2}{m} n^{2/3} \quad (1)$$

(where n is the concentration of particles), the integral of the distribution function over all momenta decreases to below the total number of particles; in order to resolve this paradox, he assumed that all 'missing' particles lie in the one and only state with the lowest energy (and zero momentum). If $T \leq T_c$, the thermal de Broglie wavelength is of the order of the average distance between particles or is larger than it, such that the BEC occurs only in the quantum mode for a system of particles. If $T \rightarrow 0$, all particles in a system of noninteracting particles form the condensate.

After P Kapitza discovered the superfluidity of ^4He , F London suggested that superfluidity may stem from BEC. This was a brave hypothesis because BEC was then predicted only for noninteracting particles, while the interaction in condensed ^4He is strong. The BEC theory was generalized to the case of interacting particles only later (see [1–3] and the references therein). It was found that the interaction 'exhausts' the Bose condensate such that not more than 9% of particles in ^4He stay in the condensate even at $T \rightarrow 0$. This was shown by first-principle quantum Monte Carlo calcula-

tions and by evaluations based on experiments with neutron scattering in liquid ^4He at temperatures lower than the temperature of transition to the superfluid state. In reality, the number of atoms in the condensate was calculated in these experiments as a result of subtraction of two large numbers, i.e., the total number of particles and the number of above-condensate particles. This procedure is equivalent to determining a captain's weight by weighing a ship with the captain on board and subtracting the weight of the ship and is hardly conclusive in view of the accuracy achieved in the experiment. Consequently, to experimentally observe Bose condensation, it was necessary to pass to a system of weakly interacting atoms in which condensate exhaustion due to interaction is negligible, that is, to start using Bose gases at a low concentration of particles and hence, as follows from formula (1), at ultralow temperatures (in the range of nanokelvins). This dictated the need to develop a spectacular technology of laser cooling and building traps for neutral atoms (see, e.g., [4] and the references therein). This is why it took 70 years after Einstein's brilliant prediction for the experiments of Ketterle, Cornell, and Wieman and their coworkers (those among the most beautiful experiments of the 20th century) to observe Bose–Einstein condensation of cold atoms in a trap.¹

In fact, because the BEC temperature is proportional to the particle mass, BEC could be realized at substantially higher temperatures, for example, with a positronium, or quasiparticles of Bose excitations in semiconductors—Wannier–Mott excitons (see [3, 6, 7] and the references therein) or polaritons in optical microcavities (see [8–12] and the references therein).

Estimates show that Bose condensation of excitons could be already obtained at temperatures of the order of several kelvins and the Bose condensation of polaritons at even higher temperatures. There is a difficulty here, however: quasiparticles have finite lifetimes due to their short recombination times (for excitons) and still shorter lifetimes (of the order of only ten picoseconds) of photons in an optical microcavity owing to its moderate quality (for polaritons).

For the Bose condensation to be produced, it is necessary that the time for a system of quasiparticles to reach thermal equilibrium and form the condensate be substantially shorter than the quasiparticle lifetime. This was one of the reasons why it was suggested rather long ago to use a system of excitons with spatially separated electrons (e) and holes (h) (see [13–15] and the references therein). Spatial separation in coupled quantum wells or in a single quantum well in a strong electric field reduces the recombination rate by several orders of magnitude. Furthermore, if the exciton concentration is not very high, repulsion between parallel electric dipoles occurring in response to spatial separation of electrons and holes prevents the standard coalescence of electrons and holes into metallic e–h-drops [16] and stabilizes the exciton phase in an isotropic (or nearly isotropic) electron–hole system. In the range of sufficiently low temperatures, we can expect to observe, as the concentration increases, a quantum crossover from the mode of BEC of dipole excitons [14] (or a local e–h pair in an equilibrium system) to the Bardeen–Cooper–Schrieffer (BCS) regime, i.e., to pairing of spatially separated electrons and holes (first discussed in the case of three-dimensional insulators by Keldysh and Kopayev [17] (see also [18])) such that the size of the Cooper pairs much

exceeds the distance between them.² The spatial separation of electrons and holes plays another important role in the entire range of controlling parameters in which the coherent phase exists. First of all, in principle, it allows the observation of nondecaying *electric* currents associated with the superfluid motion of neutral excitons or Cooper e–h pairs as a whole. Second, the spatial separation suppresses tunneling and also phase fixation in an *equilibrium* e–h system, thus allowing superfluidity [15] (a three-dimensional equilibrium exciton dielectric has no superproperties). And finally, it permits observation of the Josephson effect between separately nonsuperconducting e- and h-layers [15].

A number of interesting optical effects, such as the stimulated light backscattering and other nonlinear optical effects, can be observed in a system of Bose-condensed dipole excitons [21].

However, Bose condensation is impossible in an extended two-dimensional system (of excitons) owing to the divergence of phase fluctuations in the Bose condensate even though superfluidity may arise in this system at a temperature below the Kosterlitz–Thouless transition point. BEC becomes possible in two-dimensional exciton traps created by nonuniform deformation [22], lithography, the nonuniform electric field of the tip of the scanning probe microscope or special-profile control electrode [23], or a ‘natural’ trap involving localization of excitons in a random potential due to impurities, boundary roughness, etc. [24].

We have calculated the properties of such traps and the distribution profile of excitons in them. Furthermore, we constructed a quasilocal generalization of the Kosterlitz–Thouless theory [25], which describes the transition to the superfluid state in an extended trap (e.g., produced by nonuniform strain).

As the concentration or the dipole moment of excitons increases, the role of correlation effects grows; in particular, they allow the formation of a liquid superfluid phase of excitons [14] and also of a new phase, the exciton crystal [26]. The results of our quantum Monte Carlo calculations [27] show that a crystal is formed in a system of dipole excitons at the Lindemann parameter equal to 0.23, which corresponds to a quantum phase transition at the dimensionless density of excitons $nr_0^2 = 290$, where n is the concentration of excitons, r_0 is a parameter of the dimension of length, $r_0 = md^2/(4\pi\hbar^2)$, and d is the exciton dipole moment. (Another scenario of quantum crystallization in a two-layer e–h system, stemming from the large difference between the effective masses of e and h, was studied in [28].)

In fact, fairly large deviations from the behavior typical of a weakly interacting electron gas are already detected for substantially lower parameters nr_0^2 : the condensate is found to be significantly exhausted, a peak appears on the static structural factor that signifies the formation of a short-range order in the system of dipole excitons, the excitation spectrum deviates substantially from the Bogolyubov profile, and a roton minimum appears on the dispersion curve [29].

At present, serious progress is taking place in the experimental realization of a specific form of excitons [23, 24, 30]. It is interesting that the exciton systems currently being studied by experimenters are, as follows from our analysis [27, 29], rather strongly correlated.

² No such crossover occurs in two-layer graphene in the absence of magnetic fields because excitons do not form in a band spectrum with a zero gap [19]. The regime produced in two-layer graphene in strong magnetic fields is the Bose condensation of dipole magnetoexcitons [20].

¹ The spectacular success in this field was described, for instance, in [1, 2].

Strong magnetic fields offer an interesting possibility of controlling the effects of strong correlation of excitons and of generating a crystalline excitonic phase. The effective (magnetic) mass of excitons greatly increases in a strong magnetic field [31]; this increases the characteristic length parameter r_0 , which in turn determines the controlling parameter of the quantum phase transition to the crystalline phase, i.e., the dimensionless exciton density nr_0^2 . The result is the magnetic-field-induced quantum crystallization of excitons.

At the moment, the results of our quantum Monte Carlo simulation leave open the question of whether a supersolid phase in an extended dipole system is possible at all (crystal ordering and superfluidity are expected to coexist in the supersolid phase [32]). However, by using quantum Monte Carlo simulation, we were able to establish that supersolids can exist in mesoscopic systems of dipoles in traps [33]. It was simultaneously found that the concentration of the superconducting component decreases as the number of particles increases. If the phase of the exciton crystal is taken into account, the phase diagram of a two-layer system of electrons and holes with the interlayer distance d much greater than the effective Bohr radius a_0 must take the form shown in the figure (as d decreases, the existence range of the crystalline phase contracts and then disappears, and a liquid exciton phase appears at small $d \leq a_0$).

We consider a system of exciton polaritons, the one that is currently popular (see [8–11])—an optical microcavity placed between two Bragg mirrors with one or several quantum wells placed in the cavity. In an optical cavity, photons with small longitudinal momenta (significant at low temperatures) have the dispersion law

$$\varepsilon_{\text{ph}}(k_{\parallel}) = c\sqrt{k_{\parallel}^2 + \left(\frac{\pi N}{L}\right)^2} \approx E_0 + \frac{k_{\parallel}^2}{2m_{\text{ph}}}, \quad (2)$$

where $m_{\text{ph}} = (\pi N)^2/(cL^2)$ is a quantity that can be logically described as the effective longitudinal photon mass, k_{\parallel} is the longitudinal momentum, L is the transverse width of the microcavity, and E_0 is the ground-state energy; in what follows, we consider transverse quantization with $N = 1$. For the parameters used in the experiment, m_{ph} is of the order of $10^{-4} - 10^{-5}$ of the electron mass m_e . The geometry of the system is selected such that at small momenta, the

resulting dispersion curve of the photon in the cavity intersects the dispersion curve for quasi-two-dimensional excitons in the quantum well placed in an optical microcavity. As a result of the interaction between two types of bosons—cavity photons and excitons—Rabi splitting is produced in the intersecting dispersion curves and a new quasiparticle, the excitonic polariton, is generated, which is a superposition of the cavity photon and exciton and has two dispersion branches (the upper and the lower polaritons).

Because m_{eff} on the lower polariton branch must coincide with $m_{\text{ph}} \sim (10^{-4} - 10^{-5})m_e$, it follows that according to estimate (1), the Bose condensate temperature of polaritons should be very high at a sufficiently high level of pumping—up to room temperature!

It could be possible to observe the transition of polaritons (and of excitons) into a coherent state by observing the abrupt narrowing of the width of the photons emitted from the cavity and their statistics. The statistics of photons leaving the cavity would be those of the coherent phase of polaritons. If this were so, they would have the statistics of laser radiation, with no population inversion in the system. In this sense, the system is a laser without inversion.

Of course, Bose condensation is impossible in an extended two-dimensional system, but superfluidity of polaritons at temperatures below the Kosterlitz–Thouless temperature is possible, while this last must be fairly high owing to the polariton’s small mass. However, the following interesting questions arise at this point:

1. How is polariton superfluidity to be observed?

2. How can the Kosterlitz–Thouless temperature in a system of polaritons be found?

Superfluidity could be established by observing unusual features in the behavior of the diffusion coefficient of polaritons and/or in the dependence of the effect of polariton entrainment by the two-dimensional gas of electrons located in the vicinity of a quantum well with excitons on polariton pumping. The electron current should entrain the exciton component of the polariton due to the interaction between the electron charge and the dipole moment induced by this charge in the excitons located in quantum wells (see also [34]), while the exciton component in turn entrains the photon component of the polariton. The result should be a change in the angular distribution of photons emitted from the cavity. The entrainment coefficient would manifest a singularity at the Kosterlitz–Thouless transition point.

As regards the calculation of the Kosterlitz–Thouless transition temperature in a system of polaritons, the following difficulty is obvious here, in contrast to a similar calculation for a system of excitons [14]: the polariton dispersion law resulting from the anti-intersection of two quadratic laws is highly nonquadratic. Furthermore, the effective interaction between polaritons also depends substantially on the momentum, as a result of ‘entanglement’ of excitons and photons in polaritons, which depends on the longitudinal momenta. Consequently, it becomes impossible to use the standard Landau recipe for calculating the superfluid density based on applying the Galilei transformation to the reference frame comoving with the superfluid component. The superfluid density thus has to be found using a more general formalism of linear response and applying sum rules [35]. This procedure is shown to be formally equivalent to calculating the flux by introducing a fictitious charge for the polariton placed in a gauge field. It was demonstrated in [35] that due to phase fixation, only one Kosterlitz–Thouless

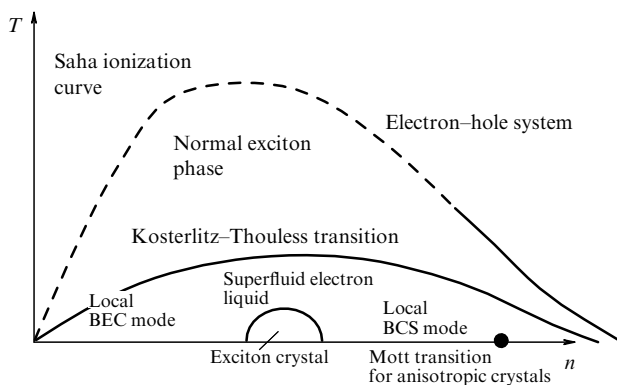


Figure. Phase diagram of a system of electrons and holes spatially separated in e- and h-layers at the interlayer separation d essentially larger than the effective Bohr radius a_0 (or in sufficiently strong magnetic fields H). As d (or H) decreases, the crystalline phase vanishes; liquid exciton phase emerges at $d \leq a_0$.

phase transition occurs between (local) condensates of excitons and cavity photons in the system; the transition temperature depends on the control parameters, the cavity geometry and the polariton splitting (Rabi splitting).

Bose condensation of polaritons is possible in two-dimensional traps for polaritons. To localize the polariton gas, it suffices to localize just one of its components (photonic or excitonic). The following two methods are thus possible for implementing a polariton trap. The first is based on building photon confinement using a microcavity of a variable width or creating a dielectric permittivity inside the microcavity dependent on the longitudinal coordinates. The second method consists in building exciton confinement by applying an external potential to the quantum well with excitons or by nonuniformly deforming the system. This method was experimentally realized by Snoke and his group (see [22] and the references therein).

In view of this, we have theoretically investigated the properties of the Bose-condensed gas of polaritons in a two-dimensional trap [11, 36, 37]. A two-component analog of the Gross–Pitaevskii equation was studied in one of the approaches to describing Bose condensates of weakly interacting polaritons [36]. In this approach, the Bose condensate of polaritons is described as two connected condensates convertible into each other — a Bose condensate of excitons and another of microcavity photons.

The wave function of the polariton condensate has two components: the respective wave functions $\psi(r)$ and $\chi(r)$ of the photon and exciton condensate. It was assumed that practically all cavity photons and excitons in the quantum well are in the condensate state at $T=0$ and it would therefore be possible to obtain the two-component analog of the Gross–Pitaevskii equation.

The energy functional of a coupled system of exciton and cavity photon condensates has the form

$$E[\psi, \chi] = \int \left\{ -\frac{1}{4\pi} \frac{\varepsilon}{\alpha} \psi^* \left(L(r) \frac{\partial^2}{\partial r^2} + \frac{\partial L(r)}{\partial r} \frac{\partial}{\partial r} + \frac{L(r)}{r} \frac{\partial}{\partial r} \right) \psi + \left(2\pi \frac{\varepsilon}{\alpha} \frac{1}{L(r)} - \mu \right) |\psi|^2 - \frac{1}{2} \chi^* \left(\frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} \right) \chi + (V(r) - \mu) |\chi|^2 + \frac{g}{2} |\chi|^4 + \frac{\Omega}{2} (\psi^* \chi + \chi^* \psi) \right\} 2\pi r dr, \quad (3)$$

where V is the confining potential for excitons, Ω is the energy of polariton splitting, $\alpha = e^2/(\hbar c) = 1/137$, ε is the dielectric permittivity of the medium in the cavity, μ is the chemical potential of the system, common for both subsystems, g is the exciton–exciton coupling constant, and L is the width of the optical microcavity, which depends on the cavity radius in general.

After varying the energy functional with respect to ψ^* and χ^* , we arrive at a system of coupled equations for the two-component condensate, which takes the following form in polar coordinates:

$$\begin{aligned} & -\frac{1}{4\pi} \frac{\varepsilon}{\alpha} \left(L(r) \frac{\partial^2}{\partial r^2} + \frac{\partial L(r)}{\partial r} \frac{\partial}{\partial r} + \frac{L(r)}{r} \frac{\partial}{\partial r} \right) \psi(r) + \left(\frac{2\pi}{L(r)} \frac{\varepsilon}{\alpha} - \mu \right) \psi(r) + \frac{\Omega}{2} \chi(r) = 0, \\ & -\frac{1}{2} \left(\frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} \right) \chi(r) + (V(r) - \mu) \chi(r) + g |\chi(r)|^2 \chi(r) + \frac{\Omega}{2} \psi(r) = 0. \end{aligned} \quad (4)$$

The range of values of the chemical potential of the system was determined in the Thomas–Fermi approximation, which allows the localization of the polariton gas. For the ‘exciton’ trap created by applying an external potential, this region is $\pi\varepsilon/(L\alpha) - \Omega/2 < \mu < \pi\varepsilon/(L\alpha)$. For a photon trap in which localization is produced due to the symmetry of the microcavity, the chemical potential must fall in the interval $\pi\varepsilon/(L(0)\alpha) - \Omega/2 < \mu < \pi\varepsilon/(L(\infty)\alpha) - \Omega/2$. As the chemical potential increases, not only the total number of particles but also the relative fraction of photons in the condensate increases.

We emphasize that we here assumed that the polariton system is in thermal equilibrium (this is also true for our papers [11, 37], in which a different approach was used for describing the polariton condensate in a trap). With real systems currently under study, the polariton lifetime in a cavity is so far too short to achieve thermal equilibrium, and hence the quality of the optical cavity needs essential improvement. Nevertheless, appreciable line narrowing is observed as pumping increases, plus the second-order time correlation function also changes greatly, which points to coherence emerging in the system. In view of this, it is very important to conduct a detailed and consistent analysis of the kinetics of the polariton condensate formation.

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References

1. Pitaevskii L, Stringari S *Bose-Einstein Condensation* (Oxford: Clarendon Press, 2003); Pitaevskii L P *Usp. Fiz. Nauk* **176** 345 (2006); **168** 641 (1998) [*Phys. Usp.* **49** 333 (2006); **41** 569 (1998)]
2. Griffin A *Excitations in a Bose-condensed Liquid* (Cambridge: Cambridge Univ. Press, 2005); Griffin A, Nikuni T, Zaremba E *Bose-Condensed Gases at Finite Temperatures* (Cambridge: Cambridge Univ. Press, 2005)
3. Griffin A, Snoke D W, Stringari S (Eds) *Bose-Einstein Condensation* (Cambridge: Cambridge Univ. Press, 1995)
4. Metcalf H J, van der Straten P *Laser Cooling and Trapping* (New York: Springer, 1999); Minogin V G, Letokhov V S *Davlenie Lazernogo Izlucheniya na Atomy* (Laser Light Pressure on Atoms) (Moscow: Nauka, 1986) [Translated into English (New York: Gordon and Breach Sci. Publ., 1987)]
5. Ketterle W *Rev. Mod. Phys.* **74** 1131 (2002)
6. Keldysh L V, in *Bose-Einstein Condensation* (Eds A Griffin, D W Snoke, S Stringari) (Cambridge: Cambridge Univ. Press, 1995) p. 246
7. Moskalenko S A, Snoke D W *Bose-Einstein Condensation of Excitons and Biexcitons* (Cambridge: Cambridge Univ. Press, 2000)
8. Weisbuch C et al. *Phys. Rev. Lett.* **69** 3314 (1992); Imamoğlu A, Ram R J *Phys. Lett. A* **214** 193 (1996)
9. Kavokin A, Malpuech G *Cavity Polaritons* Vol. 32 *Thin Films and Nanostructures* (San Diego: Elsevier, 2003)
10. Gippius N A et al. *Usp. Fiz. Nauk* **175** 327 (2005) [*Phys. Usp.* **48** 306 (2005)]; Kylakovskii V D et al. *Usp. Fiz. Nauk* **173** 995 (2003) [*Phys. Usp.* **46** 967 (2003)]
11. Snoke D *Nature Phys.* **4** 673 (2008); *Science* **298** 1368 (2002); Balili R et al. *Science* **316** 1007 (2007)
12. Marchetti F M et al. *Phys. Rev. B* **77** 235313 (2008)
13. Keldysh L V, Yudson V I *Pis'ma Zh. Eksp. Teor. Fiz.* **22** 556 (1975) [*JETP Lett.* **22** 274 (1975)]; *Zh. Eksp. Teor. Fiz.* **71** 738 (1976) [*Sov. Phys. JETP* **44** 389 (1976)]; *Solid. State Commun.* **19** 391 (1976); Lozovik Yu E, in *Vsesoyuz. Soveshchanie po Dielektricheskoi Elektronike, Tashkent, 1973. Tezisy Dokladov* (Collected Papers of the USSR Conf. on Dielectric Electronics, Abstracts of Contributions) (Tashkent: FAN, 1973)

14. Lozovik Yu E, Berman O L *Pis'ma Zh. Eksp. Teor. Fiz.* **64** 526 (1996) [*JETP Lett.* **64** 573 (1996)]; *Zh. Eksp. Teor. Fiz.* **111** 1879 (1997) [*JETP* **84** 1027 (1997)]; Lozovik Yu E, Berman O L, Ruvinskii A M *Pis'ma Zh. Eksp. Teor. Fiz.* **69** 573 (1999) [*JETP Lett.* **69** 616 (1999)]
15. Lozovik Yu E, Yudson V I *Solid State Commun.* **22** 117 (1977); Klyuchnik A V, Lozovik Yu E *J. Phys. C: Solid State Phys.* **11** L483 (1978); Lozovik Yu E, Klyuchnik A V *J. Phys. Low Temp.* **38** 761 (1980); Shevchenko S I *Phys. Rev. Lett.* **72** 3242 (1994); Lozovik Yu E, Poushnov A V *Phys. Lett. A* **228** 399 (1997)
16. Keldysh L V, in *Electron-Hole Liquid* (Amsterdam: North-Holland, 1986)
17. Keldysh L V, Kopaev Yu V *Fiz. Tver. Tela* **6** 2791 (1964) [*Sov. Phys. Solid State* **6** 2219 (1965)]
18. Kozlov A N, Maksimov L A *Zh. Eksp. Teor. Fiz.* **48** 1184 (1965) [*Sov. Phys. JETP* **21** 790 (1965)]; Halperin B I, Rice T M *Solid State Phys.* **21** 115 (1968); Guseinov R R, Keldysh L V *Zh. Eksp. Teor. Fiz.* **63** 2255 (1972) [*Sov. Phys. JETP* **36** 1193 (1973)]
19. Lozovik Yu E, Merkulova S P, Sokolik A A *Usp. Fiz. Nauk* **178** 757 (2008) [*Phys. Usp.* **51** 727 (2008)]; Lozovik Yu E, Sokolik A A *Pis'ma Zh. Eksp. Teor. Fiz.* **87** 61 (2008) [*JETP Lett.* **87** 55 (2008)]
20. Berman O L, Lozovik Yu E, Gumbs G *Phys. Rev. B* **77** 155433 (2008)
21. Lozovik Yu E, Poushnov A V *Phys. Rev. B* **58** 6608 (1998); Lozovik Yu E, Pushnov A V *Zh. Eksp. Teor. Fiz.* **115** 1353 (1999) [*JETP* **88** 747 (1999)]; Lozovik Yu E, Ovchinnikov I V *Phys. Rev. B* **66** 075124 (2002); Lozovik Yu E, Kurbakov I L, Ovchinnikov I V *Solid State Commun.* **126** 269 (2003); Lozovik Yu E, Ovchinnikov I V, Sharapov V A *Zh. Eksp. Teor. Fiz.* **125** 659 (2004) [*JETP* **98** 582 (2004)]
22. Balili R B et al. *Appl. Phys. Lett.* **88** 031110 (2006); Berman O L, Lozovik Yu E, Snoko D W *Phys. Status Solidi C* **3** 3373 (2006)
23. Gorbunov A V, Timofeev V B *Usp. Fiz. Nauk* **176** 651 (2006) [*Phys. Usp.* **49** 629 (2006)]; Timofeev V B, Gorbunov A V *Pis'ma Zh. Eksp. Teor. Fiz.* **83** 178 (2006) [*JETP Lett.* **83** 146 (2006)]
24. Butov L V *J. Phys. Condens. Matter* **19** 295202 (2007)
25. Lozovik Yu E, Kurbakov I L, Willander M *Phys. Lett. A* **366** 487 (2007)
26. Lozovik Yu E, Berman O L *Phys. Scripta* **58** 86 (1998); Kulakovskii D V, Lozovik Yu E, Chaplik A V *Zh. Eksp. Teor. Fiz.* **126** 979 (2004) [*JETP* **99** 850 (2004)]
27. Astrakharchik G E, Boronat J, Kurbakov I L, Lozovik Yu E *Phys. Rev. Lett.* **98** 060405 (2007); Ludwig P, Filinov A, Lozovik Yu E, Stolz H, Bonitz M *Contrib. Plasma Phys.* **47** 335 (2007)
28. Lozovik Yu E et al. *Zh. Eksp. Teor. Fiz.* **133** 348 (2008) [*JETP* **106** 296 (2008)]; Astrakharchik G E et al. *Phys. Rev. A* **75** 063630 (2007)
29. Vörös Z et al. *Phys. Rev. Lett.* **97** 016803 (2006)
30. Lozovik Yu E, Ruvinsky A M *Phys. Lett. A* **227** 271 (1997); Lozovik Yu E et al. *Phys. Rev. B* **65** 235304 (2002); Butov L V et al. *Phys. Rev. Lett.* **87** 216804 (2001)
31. Andreev A F, Lifshits I M *Zh. Eksp. Teor. Fiz.* **56** 2057 (1969) [*Sov. Phys. JETP* **29** 1107 (1969)]; Chester G V *Phys. Rev. A* **2** 256 (1970)
32. Lozovik Yu E, Volkov S Y, Willander M *Pis'ma Zh. Eksp. Teor. Fiz.* **79** 585 (2004) [*JETP Lett.* **79** 473 (2004)]
33. Lozovik Yu E, Nikitkov M V *Zh. Eksp. Teor. Fiz.* **111** 1107 (1997); **116** 1440 (1999) [*JETP* **84** 612 (1997); **89** 775 (1999)]
34. Lozovik Yu E, Semenov A G *Pis'ma Zh. Eksp. Teor. Fiz.* **86** 30 (2007); *Theor. Mat. Fiz.* **154** 372 (2008) [*JETP Lett.* **86** 28 (2007)]; *Theor. Math. Phys.* **154** 319 (2008)]; see Lozovik Yu E, Semenov A G, Willander M *Pis'ma Zh. Eksp. Teor. Fiz.* **84** 176 (2006) [*JETP Lett.* **84** 146 (2006)]
35. Voronova N S, Lozovik Yu E *Fiz. Tver. Tela* **50** 1496 (2008) [*Phys. Solid State* **50** 1555 (2008)]
36. Berman O L, Lozovik Yu E, Snoko D W *Phys. Rev. B* **77** 155317 (2008)
37. Deng H et al. *Science* **298** 199 (2002)

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Inverted optical phonons in ion-covalent crystals

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1. Introduction

In this report, we discuss additional optical phonons, considered excess from the standpoint of selection rules, discovered in the majority of crystals with ion-covalent interatomic bonds, including their solid solutions. These ‘excess’ phonons are located inside the transverse-longitudinal splitting of the main phonons, where the real part of the crystal permittivity is negative, and are split by the crystal field into transverse optical (TO) and longitudinal optical (LO) phonons, the frequencies of ‘excess’ LO phonons turning out to be lower than those of ‘excess’ TO phonons.

Solid solution systems like $\text{Zn}_{1-x}\text{Cd}_x\text{S}$, $\text{Zn}_{1-x}\text{Cd}_x\text{Se}$, and $\text{ZnSe}_x\text{S}_{1-x}$ hold much promise for practical applications, in particular in optoelectronics, due to their unusual physical properties. Structures with quantum wells [1] and quantum dots [2] based on thin layers of these materials, which are candidates for light sources in the blue spectral region, were formed and investigated. Chromium-doped crystals of these materials have proven to show promise for making femtosecond lasers in the near-infrared (IR) region ($\lambda \approx 2.5\text{--}3.5\text{ }\mu\text{m}$) [3, 4].

The compositional disorder of a solid solution modifies the structural, vibrational, and optical properties of polar crystals. These changes give rise to special features in the lattice dynamics of ternary solid solutions of the substitution type (single-mode, two-mode, or intermediate behavior of the transverse ω_{TO} and longitudinal ω_{LO} vibration frequencies of the system [5]) as well as to the emergence of new modes (local, gap, or resonance (quasiresonance) excitations) and to the defect-induced density of phonon states [6].

In the rigid-ion model [7], for a diatomic crystal of the ZnS type, the phonon frequencies ω_{TO} and ω_{LO} are given by [8]

$$\omega_{\text{TO}}^2 = \omega_0^2 - \frac{4\pi}{3} \frac{e_B^2(m_1 + m_2)}{\sigma m_1 m_2}, \quad (1)$$

$$\omega_{\text{LO}}^2 = \omega_0^2 + \frac{8\pi}{3} \frac{e_B^2(m_1 + m_2)}{\sigma m_1 m_2}, \quad (2)$$

$$\omega_0^2 = -\frac{m_1 + m_2}{m_1 m_2} \sum_l \Phi^N \left(\begin{matrix} l \\ + - \end{matrix} \right), \quad (3)$$

where m_1 and m_2 are the positive- and negative-ion masses, l is the cell number, σ is the elementary cell volume, ω_0 is the frequency of triply degenerate atomic vibrations, neglecting long-range Coulomb forces, Φ^N are short-range force constants, which are independent of the ion position relative to the crystal surface, and e_B is the Born effective charge of an ion. The long-range Coulomb ion field in single crystals partly removes the triple degeneracy of a vibration with the frequency ω_0 , splitting it into a doubly degenerate vibration (ω_{TO}) and a nondegenerate one (ω_{LO}); the symmetry of atomic vibrations remains unaltered in this case.