Bose condensation of finite-lifetime particles with excitons as an example

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Bose-Einstein condensation (BEC) of a dilute nonequilibrium gas of finite-lifetime bosons (e.g., excitons in a semiconductor) generated by an incoherent external source and cooled in a phonon thermostat is investigated. Even at $T = 0$ (T is the thermostat temperature) BEC arises only if the gas density exceeds some threshold $n > n_c \approx V_0^{-1} \tau_{\text{phon}} / \tau$ (where V_0^{-1} is the wave vector of the generation region, τ is the boson lifetime and τ_{phon} the mean boson-phonon scattering time). In many cases the kinetic equation for $T = 0$ can be solved analytically. Some features of the phase transition with BEC in a nonequilibrium system are demonstrated in the case of simple exactly soluble models. It is shown that in the case of excitons or biexcitons the possibility of BEC is controlled by the slow relaxation of low-energy (subsonic) particles with $\tau_{\text{phon}} \approx 10^{-4} - 10^{-3}$ s and $V_0^{-1} \approx (mS/\hbar)^3 \approx 10^{15}$ cm^{-3} (m is the exciton mass, S the velocity of sound). For $\tau \approx 10^{-6}$ s the threshold density should be very high—$n_c \gtrsim 10^{17}$ cm^{-3}; at such densities the distance between excitons is of the same order of magnitude as their radius. Thus it is impossible to obtain BEC of a dilute (bi)exciton gas only by phonon cooling provided the lifetime is not very long, $\tau \gtrsim 10^{-3}$ s. This could explain why BEC has not been observed in a (bi)exciton system in the case of incoherent excitation.

INTRODUCTION

Since the time of Einstein's prediction in 1924[1] of the Bose-Einstein condensation effect in an ideal Bose gas, no real physical system has been found in which this phenomenon is experimentally observed. Liquid ^{4}He is a system with very strong interaction. Attempts to obtain Bose condensation in dilute, very cold atomic hydrogen did not succeed.[2] Great hopes were placed on semiconducting electronic excitations of the Bose type—excitons (bound hydrogenlike states of electrons and holes),[3,4] and also on biexcitons (excitonic molecules). A large number of experimental and theoretical studies (a survey of which can be found in Refs. 5, 6) have been devoted to the problem of Bose condensation of (bi)excitons. In a number of semiconductors a (bi)excitonic gas with a Bose-Einstein (non-Maxwellian) distribution has been successfully obtained[7,8] (see also Ref. 6) and the degeneracy limit has even been approached; however, a Bose condensate with $\mathbf{k} = 0$ has not been recorded—with the exception of experiments using two-photon excitation,[9] in which coherent biexcitons are directly excited.

Observation of Bose condensation in an excitonic system is favored by: small particle mass and, consequently, comparatively high transition temperatures; the possibility of easily varying the gas density by changing the level of optical excitation of the semiconductor; the presence of self-luminescence, the analysis of the spectrum of which permits observation of the particle energy distribution function and, therefore, allows one to fix the fact of Bose condensation. At the same time, luminescence is a consequence of the finite lifetime of (bi)excitons—a property of excitonic "material" which distinguishes it in a fundamental way from ordinary material and complicates Bose condensation. As far as we know, the effect of the finite exciton lifetime on Bose condensation has gone practically unstudied in the literature—in comparison, for example, with the role of many-body effects.[1]

The finite lifetime is a consequence of the fact that excitons, like all electron and hole states in general, are semiconductor excitations. Experiments on them require a constant, or initial, excitation, and any state of an excitonic system, even a stationary or constant excitation, is fundamentally nonequilibrium. This very circumstance is essential to the possibility of quantum generation of an excitonic system.

The goal of the present study is to analyze the possibility of forming a Bose condensate in a system of nonequilibrium particles with a finite lifetime. We study the simplest model of an ideal boson gas, having a finite lifetime, generated by an incoherent external source, and thermalizing due to interaction with a phonon thermostat. It turns out that even for a $T = 0$ thermostat temperature, a Bose condensate is formed in such a system when the excitation exceeds some threshold value (that is, beginning with some critical value of the density of the boson gas, depending on the boson lifetime and the cooling rate). Thus, if in the case of a dilute gas of stable particles the basic experimental difficulty in observing Bose condensation is ultra-low cooling, in the case of a nonequilibrium system there also arises a requirement on the excitation intensity.

For example, for a dilute exciton gas[2] at $T = 0$ the critical density is of order $10^{15} \tau_{\text{phon}} / \tau$ (cm^{-3}), where τ is the exciton lifetime, and τ_{phon} is the characteristic scattering time of a slow exciton on phonons; the exciton speed is less than that of sound. Since the latter is very high, $\tau_{\text{phon}} \approx 10^{-3} - 10^{-4}$ s, from which it follows that phonon relaxation cannot ensure Bose condensation of a dilute exciton gas, unless the excitons have a very long lifetime—on the order of milliseconds and longer. [An excitonic gas can be considered dilute if its density is less than $(10^{-2} - 10^{-1}) a_B^{-3} \approx 10^{15}$ cm^{-3}, where a_B is the Bohr radius of the exciton. At higher densities it is impossible to neglect, as we have in the present study, exciton-exciton interaction and the composite nature of the exciton.]

An earlier analogous problem (for a biophysical model) was discussed by Fröhlich[13] and not long ago by Duffield[14]—in the simplest approximation, in which the matrix

element for interaction with photons is taken to be independent of the transmitted momentum. In contrast to these studies, we limit ourselves to the $T = 0$ case, which allows a clear solution in a series of simple models of the kinetic equation; this leaves the possibility open of taking into account the dependence of the matrix element on the transmitted momentum. As a result, in the exactly soluble models one can study the properties of a phase transition (at $T = 0$) with Bose condensate formation in a nonequilibrium system with relaxation, and also evaluate the threshold excitation intensity necessary to observe Bose condensation. Naturally, the values of threshold intensity obtained at $T = 0$ are lower bounds, since they grow as the temperature increases.

The structure of this study is as follows. In Sec. 1 we introduce the familiar kinetic equation (at $T = 0$) for a gas of bosons with a finite lifetime, generated by an incoherent external source, with cooling by phonon emission. The conditions for forming a Bose condensate are analyzed. In Sec. 2 we discuss the simplest model with a boson-phonon interaction matrix element independent of the transmitted momentum (the Fröhlich model). In this approximation we not only successfully obtain the Bose condensation conditions and calculate the stationary boson distribution function in the statistical limit, but also track how, in the limit of a transition to an infinite volume, a phase transition arises upon Bose condensate formation. Besides this, the condensate formation time (which, as in a system of stable particles,[15] diverges in the statistical limit) is evaluated. In Sec. 3, a model with a power-law dependence of the matrix element on the transmitted momentum is considered; this is responsible for the relaxation of slow $3D$-excitons due to an isotope-scattering type process. In the Appendix, τ_{phon} is evaluated for slow excitons.

1. BOSE CONDENSATION OF A WEAKLY NONIDEAL BOSON GAS WITH A FINITE LIFETIME

We will consider (at $T = 0$) a weakly nonideal boson gas with a finite lifetime, generated by pulses from an external source distributed in space and thermalized by weak interaction with a phonon thermostat. This is the simplest nonequilibrium model in which we can expect Bose condensation. We will begin with a system in a finite volume V. Bose condensation will be studied in the statistical limit $V, N \gg 1$, $n = N/V \propto$ const. (N is the total number of particles in the system.)

At $T = 0$ the kinetic equation for the boson occupation number f_p has the form

$$\partial f_p/\partial t = g_p - f_p/\tau_p + (1 + f_p) V^{-1} \sum_{\varepsilon_p < \varepsilon_q} W(q, p) f_q - f_p V^{-1}$$

$$\times \sum_{\varepsilon_p > \varepsilon_q} W(p, q)(1 + f_q). \quad (1.1)$$

Here g_p, τ_p, and $\varepsilon_p = \varepsilon_0 + p^2/2m$ are respectively the generation rate, the lifetime and the frequency of the bosons; m is their mass; and $W(q,p)$ is the frequency of transition of states q into p with phonon emission.[3]

Equation (1.1) assumes that the phonon thermostat is in equilibrium at $T = 0$—that is, there are no phonons in the system. This equation is the limiting form (for $T = 0$) of the equation studied by Fröhlich[13] for the case of constant τ_p, $\tau_p = \tau =$ const, and uniform g_p inside a sphere of radius

p_{max}:

$$g_p = \begin{cases} g, & p < p_{max}, \\ 0, & p > p_{max}, \end{cases} \quad (1.2)$$

where $p \equiv |\mathbf{p}|$. In Ref. 13 it was assumed that

$$W(\mathbf{p}, \mathbf{q}) = \text{const} \cdot |\exp(\varepsilon_q/T) - \exp(\varepsilon_p/T)|. \quad (1.3)$$

Then the corresponding equation is greatly simplified, and the solution in the stationary case contains the Bose-Einstein factor:

$$f_p \propto \{\exp[(\varepsilon_p - \mu)/T] - 1\}^{-1},$$

where the value of μ, analogous to the chemical potential for the equilibrium case ($\tau = \infty$) tends from below toward the energy of the fundamental mode as the pumping intensity grows: $\mu \to \varepsilon_0 - 0$. The population of this mode grows without bound. In the statistical limit, examined in Ref. 14, a true phase transition takes place at some critical excitation $g = g_c$: μ becomes equal to ε_0 and forms a Bose condensate with $\mathbf{p} = 0$. We note that all the results of Ref. 14 are also obtained for a very special form of the function $W(\mathbf{p},\mathbf{q})$.

In this study, as in Refs. 13 and 14, the boson lifetime will be taken as constant, and the generation as uniform inside the region (1.2). In application to excitations in indirect semiconductors, the first condition is completely valid. The second condition, at first glance, seems inadequate. It is also not clear what the value of p_{max} might be. However, if the charge carriers in the semiconductor are excited sufficiently highly in the conduction band, then excitons arise only as a result of electron and hole cooling, initially due to emission of a cascade of optical phonons (the characteristic time for this process is 10^{-12} s), then a cascade of acoustic phonons (here the characteristic time is 10^{-10}–10^{-9} s), and finally due to binding into an exciton (10^{-9} s). The cooling process of both current carriers and excitons is sharply slowed in the region of small momenta $p < p_s = mS$ (where S is the speed of sound in the semiconductor). For $T < mS^2 \approx 1$ K (we are interested in just this low-temperature behavior), a "bottleneck" arises in this region of small pulses which controls the exciton distribution near $\mathbf{k} = 0$. Furthermore, in evaluation we will consider that the fast cooling process of electrons and holes and their binding in excitons causes this region to be uniformly populated by excitons; that is, we will assume $p_{max} = p$ in Eq. (1.2). Uniformly of population is ensured not only by the presence of different, practically equally probable, cascade cooling processes, but also by the fact that in the initial stages of cooling the excitations have Fermi statistics and do not undergo exchange attraction to the strongly and nonuniformly populated region of momenta near $\mathbf{k} = 0$.[4]

As regards the form of the function W, in actuality the matrix element for phonon interaction usually decreases as the phonon momentum decreases. Therefore $W(\mathbf{p},\mathbf{q})$ does not have the form of Eq. (1.3), and the steady-state distribution is not Bose-Einstein. However, for $T = 0$ and a power-law form of $W(\mathbf{p},\mathbf{q})$, the steady-state equation (1.1) in the statistical limit turns out in many cases to be exactly soluble. The simplest example, when $W(\mathbf{p},\mathbf{q})$ is just constant (the Fröhlich model), is examined in Sec. 2. Another example (Sec. 3) corresponds to phonon cooling of $3D$ excitons in the region $p < p_S$ due to isotope scattering. A dependence of the

form

$$W(\mathbf{p},\ \mathbf{q})=w|\varepsilon_\mathbf{p}-\varepsilon_\mathbf{q}|^\nu, \qquad (1.4)$$

arises (see the Appendix), with w determined by (A8), and $\nu = 3$.

In the statistical limit, accounting for the possibility of forming a Bose condensate with density

$$n_0 = \lim_{V \to \infty} f_0/V,$$

Eq. (1.1) can be written in the form

$$\partial n_0/\partial t = n_0 \Big[\int_{0<q<p_{max}} d\mathbf{q} W(\mathbf{q},0)f_\mathbf{q} - \tau^{-1} \Big], \qquad (1.5)$$

$$\partial f_\mathbf{p}/\partial t = g + [1+f_p] \int_{p<q<p_{max}} d\mathbf{q} W(\mathbf{q},\mathbf{p})f_\mathbf{q}$$

$$-f_\mathbf{p} \Big[\int_{0<q<p} W(\mathbf{p},\mathbf{q})(1+f_\mathbf{q})d\mathbf{q} + \tau^{-1} + n_0 W(\mathbf{p},0) \Big]. \qquad (1.6)$$

Here $d\mathbf{q} \equiv (2\pi)^{-d} dq_1 dq_2 \dots dq_d$, and d is the spatial dimensionality of the system.

Before going on to an analysis of specific models, we will make several observations. First, the solution (1.5), (1.6) satisfies the obvious law of conservation of particle number

$$(\partial/\partial t + 1/\tau)n = \int_{0<q<p_{max}} d\mathbf{q} g, \qquad (1.7)$$

where

$$n = n_0 + \int_{0<q<p_{max}} d\mathbf{q} f_\mathbf{q}$$

is the total boson concentration.

Second, the right-hand part of Eq. (1.5) for the Bose condensate density is proportional to n_0. Therefore, the solution of this equation with the initial condition $n_0|_{t=0} = 0$ is $n_0(t) = 0$. A Bose system also has this property for $\tau = \infty$. In conditions when Bose condensation is possible, this solution is unstable. However, in order to evaluate the time for development of this instability, it is necessary to consider a finite system. It turns out that the time diverges for $V \to \infty$. This result is known for systems of stable bosons.[15] In Sec. 2B we will turn to a discussion of this result, using the example of the exactly soluble Fröhlich model.

Finally, we will examine under what conditions a stationary solution of the system of equations (1.5), (1.6), can contain a Bose condensate. It follows from Eq. (1.5) for $n_0 > 0$ that the condition

$$\int_{0<q<p_{max}} d\mathbf{q} W(\mathbf{q},0)f_\mathbf{q} = \tau^{-1} \qquad (1.8)$$

should be fulfilled; this has an obvious physical meaning: the departure of particles from the Bose condensate should, under steady-state conditions, be completely balanced by their arrival from other modes. The intensity of boson generation clearly does not enter into this condition of balance in the statistical limit, since

$$\lim_{V \to \infty} g\tau/V = 0,$$

that is, the Bose condensate is not generated directly by the external source.

Condition (1.8) and Eq. (1.6) allow determination of the asymptote of the stationary distribution function for $\mathbf{p} \to 0$:

$$f_\mathbf{p}|_{\mathbf{p}\to 0} = \begin{cases} \text{const}, & n_0 = 0, \\ \text{const} \cdot \Big[n_0 W(\mathbf{p},0) \\ \quad -\Big(\mathbf{p}\dfrac{\partial \varepsilon_\mathbf{p}}{\partial \mathbf{p}}\Big) \int d\mathbf{q} f_\mathbf{q} \dfrac{\partial W(\mathbf{p},\mathbf{q})}{\partial \varepsilon_\mathbf{p}}\Big|_{\mathbf{p}=0} \Big]^{-1}, & n_0 > 0. \end{cases} \qquad (1.9)$$

If W has the form (1.4) and $\nu > 0$ holds, and the boson spectrum is quadratic, then for $n_0 > 0$ we have $f_\mathbf{p} \propto 1/p^2$. Therefore, in the given dissipative system the Bogolyubov theorem on singularities of the $1/q^2$ type[16] is fulfilled. Since the function $f_\mathbf{p}$ should be integrable for $\mathbf{p} \to 0$, we can conclude that in one-dimensional and two-dimensional systems Bose condensation in a system of particles with a finite lifetime is impossible also for $T = 0$.

If $\nu = 0$, then $f|_{p\to 0} \propto$ const even for $n_0 > 0$—so that Bose condensation at $T = 0$ is possible both in one-dimensional and two-dimensional systems. Analysis of equations (1.1) for $T \neq 0$ shows that at finite temperatures the Bose condensate is destroyed.

The relation (1.9) allows us to analyze the question of the Bose condensate in a system with a finite lifetime and with other forms of the function W. For example, in the case $W(\mathbf{p},\mathbf{q}) = \varkappa|p^\xi - q^\xi|$, $\xi \geq 0$, a Bose condensate is possible only for $\xi < d$. Therefore, for specific forms of interaction a Bose condensate is not always possible even in the three-dimensional case. An example of such a situation ($\xi = 1$, $d = 1$), when with growth of the excitation intensity the steady-state distribution becomes more and more narrow, but a Bose condensate, strictly speaking, does not arise, is presented in Ref. 17.

2. THE FRÖHLICH MODEL

Let $W(\mathbf{p},\mathbf{q}) = $ const. Since a similar approximation was considered by Fröhlich in Ref. 13 [not for W, it is true, but for the value (1.3)], we call this the Fröhlich model. We have already said above that it does not, apparently, have a direct relation to the real exciton systems in semiconductors. However, many of the rules for Bose condensation in a system of particles with a finite lifetime have become clear from this simplest model.

We reduce Eqs. (1.5) and (1.6) to a more convenient form, transforming to dimensionless variables $\varepsilon = \varepsilon_\mathbf{p}/\varepsilon_s$, $\eta = \varepsilon_\mathbf{q}/\varepsilon_s$ and a dimensionless generation rate, condensate density and phonon relaxation time $G = g\tau$, $N_0 = n_0 V_0$ and $D = \tau_{phon}/\tau$. Here

$$\varepsilon_s = p_s^2/2m, \quad V_0 = {}^1\!/_2 d\Gamma({}^1\!/_2 d) (m\varepsilon_s/2\pi)^{-d/2} \propto (\lambda_s)^d$$

(Γ is the gamma function, λ_s is the deBroglie wavelength of bosons with $\varepsilon \approx \varepsilon_s$), and the value

$$\tau_{phon} = V_0/(w\varepsilon_s^\nu) \qquad (2.1)$$

is the characteristic scattering time on phonons for $\varepsilon \lesssim \varepsilon_s$ [for a dependence $W(\mathbf{p},\mathbf{q})$ of the form (1.4)].

Having set $\nu = 0$, we will turn to the Fröhlich model.
A. Stationary distribution. Let $\varphi(y) = f(y^{2/d})$ and

$$F(x) = \int_x^1 \varphi(y)dy, \quad 0 < x \equiv \varepsilon^{d/2} < 1. \tag{2.2}$$

In the stationary case the equations (1.5) and (1.6) take the form

$$N_0[F(0) - D] = 0, \tag{2.3}$$

$$F'[2F - G - D - x] - F - GD = 0. \tag{2.4}$$

The condensate density N_0 follows from Eq. (2.4) as a consequence of the conservation law (1.7), having, in the stationary case and in dimensionless variables, the form

$$N_0 + F(0) = G. \tag{2.5}$$

The solution of (2.4) satisfying the obvious boundary condition

$$F(1) = 0, \tag{2.6}$$

has the form

$$F(x) = {}^1\!/_2\{G + D + x - [(G + D + x)^2 + 4GD(x - 1)]^{1/2}\}. \tag{2.7}$$

Therefore, using (2.5), we find that

$$N_0 = {}^1\!/_2[G - D + |G - D|] \tag{2.8}$$

and the critical value of excitation intensity is

$$G_c = D. \tag{2.9}$$

In the Fröhlich model the threshold condition (2.9) is preserved, if the excitation by an arbitrary form is inhomogeneous in momentum space. It is satisfied for the average value

$$G = {}^1\!/_2 d\tau \int g(\varepsilon)\varepsilon^{d/2-1}d\varepsilon.$$

The threshold steady-state boson concentration

$$n_c = V_0^{-1}\tau_{phon}/\tau \tag{2.10}$$

corresponds to the threshold excitation intensity (2.9). The physical meaning of this relationship is obvious: the longer the boson lifetime, the faster they cool off on phonons and the higher the degree of "quantum nature" ($V_0 \propto \lambda_s^d$), the lower the Bose condensation threshold. Equation (2.10) gives the correct (to within a numerical factor depending on the form of W) value of the threshold concentration for $T = 0$ also in the more general case than the Fröhlich model examined (the problem of Sec. 3 can serve as an example). Thus it can be used to evaluate the minimum excitation intensity necessary for observation of a Bose condensate of excitons. In particular, for three-dimensional $n_c \approx 10^{15}\tau_{phon}/\tau$ (cm^{-3}). Characteristic phonon times for $\varepsilon > \varepsilon_s$ are of the order of 10^{-9} s; however, in the low-energy region phonon, processes are slowed by at least a factor of 10^5 (see the Appendix). From this we can conclude that for $\tau \approx 10^{-6}$ s (the characteristic lifetime for indirect excitons) we have $n_c \approx 10^{17}-10^{18}$ cm^{-3} even at $T = 0$. The value of n_c can be somewhat lower due to fast relaxation in the region $\varepsilon > \varepsilon_s$ (several values are obtained at the end of Sec. 2). But even when this is taken into account, large values are obtained, which means that Bose condensation of a dilute exciton (or biexciton) gas by a phonon cooling mechanism alone is, most often, not possible.

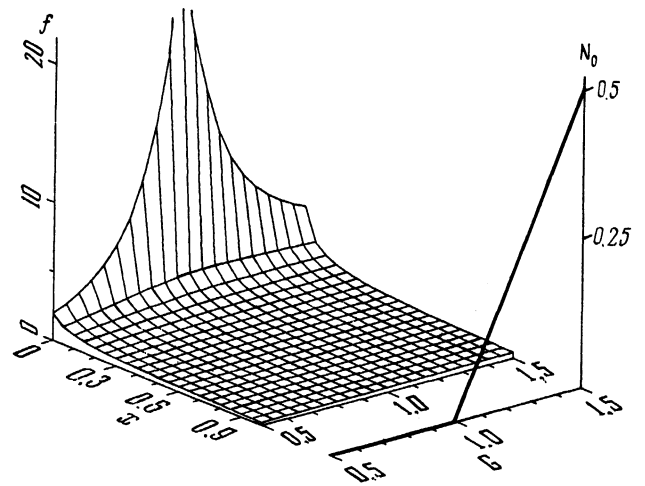Turning to the Fröhlich model, we cite the form of the

FIG. 1. The distribution (2.11) for $D = 1$ and different G; at the right the $N_0(G)$ dependence is shown.

continuous part of the steady-state distribution:

$$f_\varepsilon = -F'|_{x=\varepsilon^{d/2}} = {}^1\!/_2\{(\varepsilon^{d/2} + G + D + 2GD)[(\varepsilon^{d/2} + G + D)^2 + 4GD(\varepsilon^{d/2} - 1)]^{-1/2} - 1\}. \tag{2.11}$$

This distribution is shown for various G and $D = 1$ in Fig. 1. To the side, the $N_0(G)$ dependence of Eq. (2.8) is shown (in arbitrary units). At the Bose condensation threshold the continuous part of the distribution function has an integrable singularity at the point $\varepsilon = 0$:

$$f_\varepsilon|_{G=G_c} \propto \varepsilon^{-d/4}.$$

The fact that, after formation of the condensed mode, the continuous part of the distribution function again becomes finite at $\varepsilon = 0$ is a specific feature of the Fröhlich model at $T = 0$ [see formula (1.9) and the subsequent discussion].

B. Steady-state distribution in a finite volume. It is very interesting to analyze the properties of the Fröhlich model up to the transition to the statistical limit—at $T = 0$ it is also exactly soluble.

Having eliminated the quantity

$$\sum_{0 < q < p_{max}} f_q$$

from the first of equations (1.1) and the conservation law (1.7), we obtain a quadratic equation for the quantity $N_0(\Omega) = f_0\Omega^{-1}$, where $\Omega = V/V_0$:

$$N_0^2(\Omega) - (G - D - \Omega^{-1})N_0(\Omega) - G(1 + D)\Omega^{-1} = 0. \tag{2.12}$$

Therefore

$$N_0(\Omega) = {}^1\!/_2\{G - D - \Omega^{-1} + [(G - D - \Omega^{-1})^2 + 4\Omega^{-1}G(1 + D)]^{1/2}\}. \tag{2.13}$$

As should happen, in the statistical limit $\Omega \to \infty$ the smooth function (2.13) changes to one with a kink [Eq. (2.8)], characteristic of a phase transition; the criterion for a macroscopic system is that

$$V \gg V_0 \propto \lambda_s^d.$$

C. Transition processes. The question of the time necessary to establish a steady-state distribution in a system with Bose condensation has been analyzed in Ref. 15. Although a

system of stable bosons was studied there, the basic conclusions remain valid for bosons with a finite lifetime. That is, the closer a level is to zero (at which condensation takes place), the longer it takes to establish a stationary population. If at the initial time the system has no Bose condensate, then the steady-state value of its density is established, formally, at infinite time (that is, it diverges in the statistical limit). We will illustrate this with an example from the Fröhlich model. To do this, it is simplest to solve the first of Eqs. (1.1) with the initial conditions

$$f_0|_{t=0} = 0, \qquad (2.14)$$

$$V^{-1} \sum_{0 < q < p_{max}} f_q|_{t=0} = G. \qquad (2.15)$$

Condition (2.14) guarantees the absence of a condensate initially, and conditions (2.15) allows Σ_q^f to be eliminated from Eqs. (1.1) for f_0 as in the derivation of (2.12). We assume that G is the stationary value of the total concentration, established over a time τ [see Eq. (1.7)] which is small compared with the time to establish the steady-state value N_0. Therefore the use of (2.15) does not alter the dependence in question over times of interest to us. The temporal equation for $N_0(\Omega)$ then has the form

$$\tau \frac{\partial N_0(\Omega)}{\partial t} = -N_0^2(\Omega) + (G - D - \Omega^{-1}) N_0(\Omega) + G(1+D)\Omega^{-1}, \qquad (2.16)$$

with

$$N_0(\Omega)|_{t=0} = 0. \qquad (2.17)$$

If, in Eq. (2.16), we transform at once to the statistical limit $\Omega \to \infty$, we find that the only solution satisfying the boundary condition (2.17) is $N_0 = 0$. For $G < G_c = D$ this solution is stable, and for $G > G_c$, unstable, in the sense that for a small change in the initial conditions, N_0 departs from the zero solution and for $t \to \infty$ goes to the stationary value (2.8). To estimate the time scale for the approach to stationary value, it is necessary to solve Eq. (2.16) for finite Ω. This solution has the form

$$N_0(\Omega, t) = N_1 N_2 \{ \exp[(N_1 - N_2)t/\tau] - 1 \} / \{ N_2 \exp[(N_1 - N_2)t/\tau] - N_1 \}. \qquad (2.18)$$

Here N_1 and N_2 are solutions of Eq. (2.12); that is, the function (2.13) and its counterpart with a minus sign in front of the quadratic bracket. for $\Omega \gg 1$ and $G > D$ we have $N_1 = G - D$, and $N_2 \propto \Omega^{-1}$. From this we see that the characteristic time to reach the stationary solution (2.16) is

$$t_0 = \tau (N_1 - N_2)^{-1} \ln|N_1/N_2| \propto \tau \ln \Omega. \qquad (2.19)$$

Thus, in the transition to the statistical limit this value diverges logarithmically. Logarithmic divergence is a specific feature of the Fröhlich model. If, for instance, W has the form (1.8) with $\nu > 0$, then t_0 diverges according to a power law. This protraction of the transition to a steady-state population of the fundamental level is connected with the fact that the Bose condensate density in the initial state was zero. In the opposite case the characteristic transition time for the process is τ.

From the results obtained here it follows that for pulsed excitation, Bose condensation of particles with a finite life-

time formally does not, in general, take place, no matter how high the excitation intensity. In a real experimental situation, it is necessary to choose the length and amplitude of the excitation pulse according to the spectral resolution attainable in studying the distribution function in order to observe Bose condensation.

D. Modified Fröhlich model. Let the boson generation intensity be constant in the region $p < p_{max} = (\bar{x})^d p_s, \bar{x} > 1$, and the matrix element for phonon interaction depend on the first argument in a stepwise fashion; thus the scattering time in the region $p < p_s$ significantly exceeds the scattering time for $p > p_s$:

$$W(\mathbf{p}, \mathbf{q}) = \begin{cases} w_1, & p < p_s, \\ w_2, & p_s < p < p_{max}, \end{cases} \qquad (2.20)$$

where $w_1 \ll w_2$ (that is, $\tau_{phon,1} \gg \tau_{phon,2}$ or $D_1 \gg D_2$). Such a model permits evaluation of the effect of Bose condensation of the region of momentum space with fast relaxation (which has been neglected in this work). Obviously this effect is maximum in the steady-state case, since the fast relaxation region is quickly depleted through pulsed uniform excitation—earlier than population inhomogeneity can grow in the slow relaxation region.

If we introduce the function $F(x)$, $0 < x < \bar{x}$ (in the form (2.2), but integrated to $\bar{x}$), it is not difficult to obtain equations replacing (2.3), (2.4) and easily integrable. For example, for $1 < x < \bar{x}$ the equation for $F(x)$ coincides with (2.4) (in which D must be replaced by D_2 and the inhomogeneous term by $GD_2/\bar{x}$), and the boundary condition has the form $F(\bar{x}) = 0$. Therefore for $1 < x < \bar{x}$ we have

$$F(x) = \frac{1}{2} \{ G + D_2 + x - [(G + D_2 + x)^2 + 4GD_2(x/\bar{x} - 1)]^{1/2} \}. \qquad (2.21)$$

As before, when the critical excitation intensity is reached Bose condensation occurs in the system. For $G \geqslant G_c$, the relation

$$D_1^{-1}[F(0) - F(1)] + D_2^{-1}F(1) = 1. \qquad (2.22)$$

holds in the place of (2.3). For $G \leqslant G_c$ we have

$$F(0) = G. \qquad (2.23)$$

The value of G_c is found from the condition that at $G = G_c$ both these relations are satisfied; using (2.21) to calculate $F(1)$, we have

$$G_c = \frac{1}{2} D_1^{-1} \bar{x}^{-1} \{ Q + [Q^2 - 4D_1^2 \bar{x}^2 (D_2^2 + D_2 - D_1)]^{1/2} \} \qquad (2.24)$$

[where $Q = (D_1 - D_2)^2 - \bar{x}(D_1 - D_2 - 2D_1D_2)$]. It is easy to see that the existence of a fast relaxation region leads to substantial reduction of the Bose condensation threshold. For example, if $\bar{x} \gg D_1 \gg 1 \gg D_2$ holds, then in place of (2.10), we find

$$n_c = V_0^{-1}(\tau_{phon,1}/\tau)^{1/2} \qquad (2.25)$$

for the threshold exciton concentration. Since, however, the excitons do not arise immediately, but in the final stages of the fast relaxation process, the correct value must lie in the interval between the values of (2.10) and (2.25).

3. BOSE CONDENSATION OF THREE-DIMENSIONAL EXCITONS COOLED BY ISOTOPE SCATTERING

We now examine a case which is more complicated than the Fröhlich model—a function W of the form (1.4) with
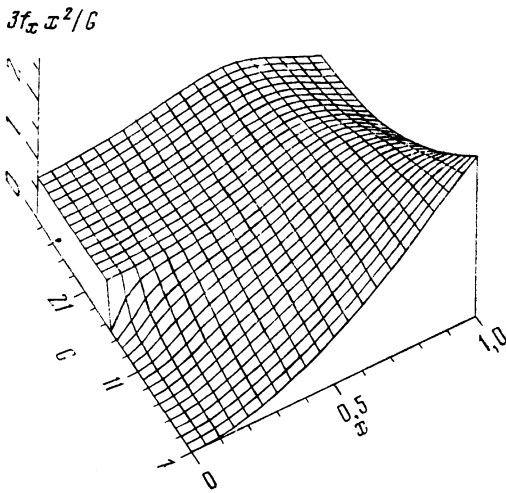
$3f_x x^2/G$

FIG. 2. The normalized distribution function for $D = 3/2$ and different G.

$v = 3$, $d = 3$. It is shown in the Appendix that such a dependence arises when isotope fluctuations of the exciton-phonon interaction constant are taken into account. For this form of W, the steady-state distribution has not been obtained in a precise analytical form; however, it can easily be found numerically.

As in Sec. 2, we will transform to dimensionless variables ε,η. We introduce the function

$$F(\varepsilon) = {}^{3}/_{2} \int_{\eta} f_{\eta}(\eta-\varepsilon)^{3}\eta^{1/2}d\eta, \quad 0<\varepsilon<1. \tag{3.1}$$

Then Eq. (2.3) preserves its form, but in place of (2.4) we get the fourth-order linear differential equation

$$\Phi(\varepsilon)F''''(\varepsilon)-\Phi''''(\varepsilon)[F(\varepsilon)+GD]=0, \tag{3.2}$$

where

$$\Phi(\varepsilon) = A+\varepsilon B+{}^{1}/_{2}\varepsilon^{2}C-\varepsilon^{3}G-({}^{16}/_{105})\varepsilon^{9/2},$$

with boundary conditions:

$$F(1)=F'(1)=F''(1)=F'''(1)=0, \tag{3.3}$$

$$F(0)=D+A, \ F'(0)=B,$$

$$F''(0)=C, \ F'''(0)=6(N_0-G). \tag{3.4}$$

The boundary conditions come directly from the definition of $F(\varepsilon)$ and Eq. (1.6). To obtain the last condition in (3.4) it is necessary to use the conservation law (1.7), which in this case takes the form

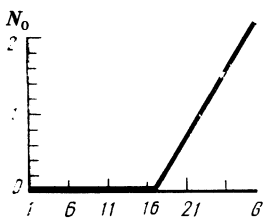$$N_0-{}^{1}/_{6}F'''(0)=G. \tag{3.5}$$

N_0
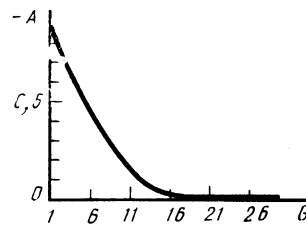

FIG. 3. The $N_0(G)$ dependence for $D = 3/2$.

$-A$

FIG. 4. The $A(G)$ dependence for $D = 3/2$.

So, the nonlinear differential equation (1.6) reduces, for $v = 3$ and in the steady-state case, to a linear differential equation. Nonlinearity is retained in the dependence of the coefficients of this equation on the boundary conditions on F, F', F'' at $\varepsilon = 0$. Incidentally, a similar transformation can be performed for any d and for odd v (the order of the differential equation and the form of the function Φ is changed).[5] For even v the analogous procedure gives a clearly nonlinear equation (we saw the example $v = 0$, the Fröhlich model, in Sec. 2).

Before going to the results, we will briefly describe the numerical calculation procedure. It consists of solving the system (3.4) of four equations in four unknowns A, B, C, and N_0 (for given G and D). The nonlinear functional dependence of F, F', F'', and F''' at $\varepsilon = 0$ on A, B, C, and N_0 is given by the solution of Eq. (3.2) with the initial conditions (3.3).

From Eq. (2.3) and the first of the conditions (3.4) it follows that the value of A is similar to the chemical potential for an equilibrium Bose gas up to the onset of Bose condensation we have $A < 0$; after Bose condensate formation we have $A = 0$.

We now note some of the numerical results. In Fig. 2, the dependence of the normalized distribution function $3 f_\varepsilon \varepsilon/G$ on the dimensionless momentum $x = \varepsilon^{1/2}$ is shown for $D = 3/2$. At $G = G_c \approx 17$, a phase transition with Bose condensate formation takes place. In Fig. 2 this is manifested in the fact that the asymptote of the function plotted changes as $x \to 0$: up to the phase transition we have $x^2f(x) \propto x^2$, while after it we have $x^2f(x) \propto$ const. In Fig. 3, the corresponding dependence of the Bose condensate density on pump intensity is shown, and in Fig. 4, we show the dependence of the quantity A, which plays the role of a chemical potential. In this model, the critical pumping density in a wide range of
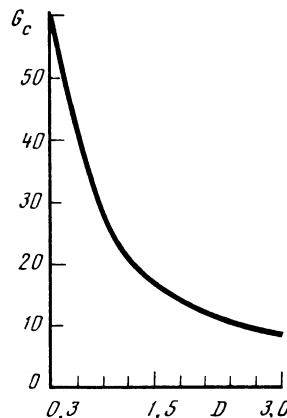
G_c

FIG. 5. The G_c (D) dependence.

values of D satisfies a relationship like Eq. (2.9) to within a numerical factor:

$$G_c \approx 11D$$

[an example of the $G_c(D)$ dependence is shown in Fig. 5].

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APPENDIX
RELAXATION OF SLOW EXCITONS

Phonon emission by slow excitons (with $p < p_s = mS/\hbar$) is presented by the requirement that the momentum and energy conservation laws both hold simultaneously, and phonon relaxation is sharply slowed. However, the energy conservation law is exactly satisfied with regard to the total width of levels participating in excitation scattering. It is also not satisfied if we account for the occasional variation of the exciton-phonon interaction constant in the crystal—for example, due to variation of the isotropic composition, i.e., processes such as isotope scattering. We will evaluate the corresponding contributions to exciton relaxation in the region $p < p_s$.

If we neglect the spread of exciton and phonon levels and assume that the exciton-phonon interaction constant does not depend on position in the crystal, then, as is well known,

$$W(\mathbf{p}, \mathbf{q}) = D^2 (2\hbar\rho S)^{-1} |\mathbf{p}-\mathbf{q}| \delta(\varepsilon_\mathbf{p} - \varepsilon_\mathbf{q} + S|\mathbf{p}-\mathbf{q}|). \quad (A1)$$

Here ρ is the crystal density, and D is the deformation potential constant for excitons. The presence of the δ-function in (A1) causes W to be exactly equal to zero in the $p < p_s$ region.

For $p, q \gtrsim p_s$ we have, in order of magnitude,

$$W \approx W_0 = D^2/(\hbar\rho S^2). \quad (A2)$$

(This value gives the characteristic time for exciton-phonon processes for $p \gtrsim p_s$: $\tau_{\text{phon}} \approx W_0 p_s^3 \approx 10^{-9}$ s.)

Line broadening may be crudely accounted for by substituting for the δ-function in (A1) the expression

$$\gamma(\mathbf{p}, \mathbf{q}) / [(\varepsilon_\mathbf{p} - \varepsilon_\mathbf{q} + S|\mathbf{p}-\mathbf{q}|)^2 + \gamma^2(\mathbf{p}, \mathbf{q})],$$

where γ is the total level width. To evaluate this we can consider that the basic contribution is from broadening of exciton levels connected with the finite exciton lifetime ($\gamma \approx \tau^{-1}$). The phonons participating in scattering are long-wavelength and long-lived and do not contribute to γ. In order of magnitude, we find that for $q, p \lesssim p_s$

$$W \approx \gamma/\varepsilon_s W_0. \quad (A3)$$

Thus, due to the finite level width relaxation takes place in the region $p < p_s$, but is slowed by a factor of $\varepsilon_s/\gamma \approx 10^5$–$10^6$ (for $\gamma \approx 10^5$–10^6 s^{-1}).

We turn now to the contribution due to isotope scattering. This can be evaluated more exactly; in fact, an exact expression for the function $W(\mathbf{p},\mathbf{q})$ can be obtained.

If the deformation potential constant changes randomly in space, we must use in place of (A1) the expression

$$W(\mathbf{p}, \mathbf{q}) = (2\hbar\rho S)^{-1} \int d\mathbf{k} |\mathbf{k}| K(\mathbf{q}-\mathbf{p}+\mathbf{k}) \delta(\varepsilon_\mathbf{p} - \varepsilon_\mathbf{q} + S|\mathbf{k}|). \quad (A4)$$

Here $K(\mathbf{k})$ is the Fourier transform of the deformation potential function:

$$K(\mathbf{k}) = \int d^3r\, e^{i\mathbf{k}\mathbf{r}} K(\mathbf{r}),$$

$$K(\mathbf{r}) = \overline{D(0)D(\mathbf{r})} - \overline{D}^2. \quad (A5)$$

For the simplest estimates we can assume that

$$K(\mathbf{r}) = \begin{cases} \Delta, & r < r_0, \\ 0, & r > r_0, \end{cases} \quad (A6)$$

where r_0 is the correlation radius of the deformation potential. In addition, we can use the approximation $kr_0 \ll 1$ (since we are interested in small momentum transfers) in calculating (A5), and reduce (A4) to the form

$$W(\mathbf{p},\mathbf{q}) = w|\varepsilon_\mathbf{p} - \varepsilon_\mathbf{q}|^3, \quad (A7)$$

where

$$w = \Delta(3\pi\hbar\rho S)^{-1} r_0^3 S^{-4}. \quad (A8)$$

In order of magnitude,

$$W \approx (\Delta/\overline{D}^2)(p_s r_0)^3 W_0.$$

The quantity $\Delta/\overline{D}^2$ can be evaluated from the following considerations. As is known, the exciton-phonon interaction constant contains $M^{1/2}$ in the denominator, where M is the mass of the nucleus. If follows that $\Delta/\overline{D}^2 \approx (\Delta M/M)^2 \approx 10^{-3}$ holds (here ΔM is the amplitude of variation of M due to variation in the isotopic composition). Since we have $p_s r_0 \ll 1$, relaxation in the $p < p_s$ region is slowed by at least a factor of 10^6 due to isotopic scattering. We see that this contribution to the relaxation is weaker than that from the level spread. But, when the exciton lifetime is sufficiently large, $\tau \approx 10^{-3}$–10^{-4} s, it can be dominant.

[1] At low temperatures the interaction between nonequilibrium current carriers causes a large variety of aggregate states in excitonic material—in the phase diagram, exciton and biexciton gases border on a Fermi electron-hole liquid and a plasma (a discussion of a different phase diagrams can be found in Ref. 10). For experimentally accessible temperatures the transition of the exciton gas to a metallic liquid can occur earlier (for lower densities) than quantum degeneracy. Therefore it is not at all simple to select the conditions for observing Bose condensation of excitons or biexcitons. Besides, upon density increase the fact that excitons are bound states of two fermions begins to be expressed—as a result, the properties of the Bose condensate change substantially.[11] Incidentally, as a rule there is an attraction between excitons, and only condensation of biexcitons is possible (excitons in Cu$_2$O, for instance, are an exception[7]). It is necessary to take special measures to observe Bose condensation of excitons—for example, spin orientation by a magnetic field.[8]

[2] In this work no distinctions are made between excitons and biexcitons, since we are considering only their interaction with phonons. Since the interaction of excitons with photons (polariton effects) has not been accounted for, the results obtained can be applied directly only to indirect excitons. Phonon cooling of direct excitons, taking account of polariton effects, was considered in Ref. 12, but without accounting for the possibility of Bose condensation, in the approximation of small occupation number.

[3] Here, except in the Appendix, the system of units with $\hbar = k_B = 1$ is used.

[4] If excitons were formed immediately, the exchange attraction in the fast cooling process would substantially lower the Bose condensation threshold. At the end of Section 2, several values obtained in the framework of a somewhat modified Fröhlich approximation are presented.

[5)]For example, for $\nu = 1$ an equation of form (3.2) is found to be second-order and can be solved exactly.

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