### Population dynamics of a Bose gas near saturation

D. W. Snoke and J. P. Wolfe

Physics Department and Materials Research Laboratory, University of Illinois at Urbana-Champaign, 1110 West Green Street, Urbana, Illinois 61801

(Received 22 August 1988)

At low density a gas of bosons achieves a classical equilibrium distribution of energies within just a few characteristic scattering times. At sufficiently high densities, stimulated emission comes into play, increasing the rate of scattering into low-energy states. This results in the well-known Bose-Einstein equilibrium distribution. In this paper we show—by numerical calculation—that for a weakly interacting Bose gas the number of characteristic scattering times required to achieve an equilibrium Bose-Einstein distribution increases with increasing degeneracy. However, we find that the characteristic scattering time for the degenerate gas is significantly shortened by the stimulated emission, with the net result that equilibration times may be comparable to the classical case. These results have a bearing on recent experiments that produced degenerate Bose distributions of excitons. A basic question from these experiments is whether Bose-Einstein condensation can occur within the particle lifetime. Our calculations indicate that condensation may take place. We also show in this paper that exciton-phonon interactions will not significantly increase the rate of approach to condensation.

#### I. INTRODUCTION

The basic result of Bose-Einstein statistics is the occupation number for bosons in equilibrium,

$$f(\mathbf{k}) = f(\mathbf{E}(\mathbf{k}); \mu, T) = \frac{1}{e^{(E-\mu)/k_B T} - 1}, \qquad (1)$$

where  $E(\mathbf{k})$  is the single-particle energy for state  $\mathbf{k}$ ,  $\mu$  is the chemical potential, and T is the temperature. This result follows<sup>1</sup> from postulating a process of stimulated emission, i.e., that the probability of a particle scattering into a state  $\mathbf{k}$  is proportional to  $P = [1+f(\mathbf{k})]$ , in contrast to the Fermi-Dirac case of repressed emission,  $P = [1-f(\mathbf{k})]$ . The chemical potential  $\mu$  in Eq. (1) is determined by the condition  $N = \int f(E;\mu,T)D(E)dE$ , where N is the total number of particles in volume V, and D(E) is the density of states, which is proportional to  $VE^{1/2}$  for constant potential energy.

A direct consequence of the form of f, for fixed temperature, is that above a certain critical density all added particles must enter the ground state. For constant potential energy, this critical density is given by

$$n_{c} = N_{c} / V = 2.612g (m / 2\pi\hbar^{2})^{3/2} (k_{B}T)^{3/2}$$
$$= CT^{3/2} , \qquad (2)$$

where m is the particle mass and g is the spin degeneracy. Superfluid helium is generally believed to manifest this effect, known as Bose-Einstein condensation, even though helium is a strongly interacting liquid.

In the above formulas, chemical and thermal equilibrium are assumed. For the case of liquid helium, the assumption of equilibrium seems satisfied because <sup>4</sup>He atoms, which do not decay, undergo roughly 10<sup>11</sup> interparticle scattering events per second, assuming classical hard-sphere scattering. In other boson systems, however, non-equilibrium effects may have significant effect. For excitons in a semiconductor, the lifetime of the particle may only be a few hundred scattering times. Spinpolarized hydrogen also has a finite lifetime to nonpolarized states.<sup>2</sup> Naively judging from classical behavior, hundreds of scattering times may seem more than adequate time to establish equilibrium. The case of Bose condensation is unique, however, since a macroscopic number of particles must enter a single quantum state, which by random scattering processes alone is highly improbable. Therefore we need not assume that the time for Bose condensation is short.

Recently, experimental data have been reported<sup>3</sup> which have raised the question of whether condensation may indeed take longer than the particle lifetime under some conditions, even though the excitons undergo many interparticle collisions during their lifetimes. Excitons in a semiconductor such as Cu<sub>2</sub>O have been shown theoretically<sup>4</sup> and experimentally to act as bosons under certain conditions of density and temperature. In recent experiments,<sup>3</sup> at low density, the exciton gas exhibited equilibrium near the lattice temperature. As the exciton density increased to approach the Bose-Einstein condensation boundary given in Eq. (2), highly degenerate Bose-Einstein momentum distributions were observed, but a significant condensate fraction did not appear. Instead, the gas increased its temperature to accommodate extra particles, reaching temperatures well above the lattice temperature. The temperature of the excitons at high densities is affected by the balance of a number of processes, including the acoustic and optic phonon emission rates and the Auger recombination rate.<sup>5</sup> Still, the close proximity to the phase boundary over the wide range in density strongly suggests that the quantum statistics of

1 /1 \

the near-condensate region plays an important role in the temperature rise.

In the present work, we address the question of how a nonequilibrium nearly-ideal Bose gas approaches quantum saturation and condensation. We present the results of two numerical models, first, the time evolution of the energy distribution of a weakly interacting Bose gas, and second, the time evolution of the energy distribution of an ideal Bose gas with inelastic interaction with a phonon bath.

## **II. PREVIOUS WORK**

Two recent theoretical works have specifically addressed the question of the time evolution of a weakly interacting Bose gas.<sup>6-8</sup> Before discussing the modeling we have done, we briefly review the scope of each of these.

Inoue and Hanamura<sup>6</sup> considered the evolution of an ideal Bose gas of excitons coupled to a *phonon* bath with infinite heat capacity. Considering only phonon scattering with small energy exchange, and using several other approximations, they obtained a numerically solvable Fokker-Planck equation. Although their discrete energy points were not close enough to model a true condensation, they found a lower limit on the time to reach condensation of several phonon scattering times, on the order of nanoseconds for excitons at low temperatures in some semiconductors. Despite the degree of approximation, this study showed that the time for condensation can possibly be comparable to the carrier lifetime.

Levich and Yahkot<sup>7</sup> studied the same problem, except that they considered an ideal Bose gas coupled to a *Fermi* heat bath with infinite heat capacity. They analytically calculated that the time for condensation in this situation should be *infinite*. By including weak boson-boson interactions but excluding terms which led to spectral "peak broadening," they later<sup>8</sup> found a solution which led to an explosive appearance of a condensate, but they concluded that this effect could have been an artifact of their approximations.

In each of the above calculations, the time evolution of the bosons was expressed in terms of a Boltzmann equation of the form

$$\frac{dn(\mathbf{k})}{dt} \equiv \int F(\mathbf{k}, \mathbf{k}_1, \dots, \mathbf{k}_{d-1}; n(\mathbf{k}), n(\mathbf{k}_1), \dots, n(\mathbf{k}_{d-1})) \times d^3k_1 \cdots d^3k_{d-1}$$
$$\equiv I , \qquad (3)$$

where F is some function of the instantaneous number densities  $n(\mathbf{k}_i)$  and wave vectors  $\mathbf{k}_i$ , i = 1, ..., d, for d incoming and outgoing wavevectors in a given scattering process. This equation was then manipulated through various approximations to take on the following form:

$$\frac{dn(E)}{dt} = G(E, n(E, t), t) , \qquad (4)$$

where G is some explicit function. This equation could then be solved analytically for certain cases, but in general it was solved numerically. Another approach—the one we take here—is solve Eq. (3) directly, by evaluation of the integral I. Once the rate of change in  $n(\mathbf{k})$  has been calculated for all points on some grid in  $\mathbf{k}$  space, then  $dn(\mathbf{k})$  can be added to each of the  $n(\mathbf{k})$  for some small time step dt, and the integral I can be recalculated. Following this iterative process, the distribution can be determined for all times.

The problem with this iterative approach is that for even a small number of k-points, the integral I can become formidable; for instance, for two-body elastic collisions, finding the rates at all k, at a single point in time, involves integration over four wavevectors in three dimension, which naively would require a number of calculations which goes as the twelfth power of the number of k-points! We can make this problem tractable, however, first by mathematically reducing I to an integration over only three energies, and second by utilizing the power of modern array processors. In doing so, we can model the time evolution of the nonequilibrium boson gas without any additional approximations.

#### III. MODEL OF THERMALLY ISOLATED BOSE GAS WITH INTERPARTICLE SCATTERING

As in the above references,<sup>6-8</sup> we use the randomphase approximation, which is valid for a weakly interacting, noncondensed Bose gas. Excitons in Cu<sub>2</sub>O are expected to interact like hard spheres of radius < 10 Å (Ref. 9), therefore up to densities of  $10^{20}$  cm<sup>-3</sup> an exciton gas in Cu<sub>2</sub>O may be treated as weakly interacting. The random-phase approximation will break down when coherence appears; however, it will be valid to very near condensation.

In this approximation we write the probability of two bosons with wavevectors  $\mathbf{k}_1, \mathbf{k}_2$  scattering to  $\mathbf{k}_3, \mathbf{k}_4$  as

$$S(\mathbf{k}_{1}, \mathbf{k}_{2}; \mathbf{k}_{3}, \mathbf{k}_{4}) = M^{2} \delta(\mathbf{k}_{1} + \mathbf{k}_{2} - \mathbf{k}_{3} - \mathbf{k}_{4}) \delta(E_{1} + E_{2} - E_{3} - E_{4}) \times f(\mathbf{k}_{1}) f(\mathbf{k}_{2}) [1 + f(\mathbf{k}_{3})] [1 + f(\mathbf{k}_{4})], \qquad (5)$$

where M is the matrix element of the interaction. We assume here M constant, but the exact form of M turns out not to be important as long as it remains finite at all k.  $f(\mathbf{k})$  is the number of particles in state  $\mathbf{k}$ ; the (1+f) terms give the effect of stimulated emission for bosons, as opposed to (1-f) for fermions. The total scattering rate *into* state  $\mathbf{k}$  can then be expressed as

$$\Gamma_{i}(\mathbf{k}) = \int d^{3}k_{1}d^{3}k_{2}d^{3}k_{3}S(\mathbf{k}_{1},\mathbf{k}_{2};\mathbf{k}_{3},\mathbf{k}) .$$
 (6)

The scattering rate *out* of state **k**,  $\Gamma_o(\mathbf{k})$ , is the same expression, but with the integrand  $S(\mathbf{k}, \mathbf{k}_1; \mathbf{k}_2, \mathbf{k}_3)$ . One can check that the distribution function given in Eq. (1) is the equilibrium solution simply by substituting it into the equation for equilibrium,  $\Gamma_i(\mathbf{k}) = \Gamma_o(\mathbf{k})$ .

As shown in the Appendix, the ninth-order integrals for  $\Gamma_i$  and  $\Gamma_o$  can be simplified by integrating out all the delta functions and angles, leaving the integral only over energies. We then obtain We obtain a similar expression for  $\Gamma_o$ . For the dispersion relation  $E = \hbar^2 k^2 / 2m$ , the derivatives in the denominator are constant. Interactions will in general change the form of the dispersion relation, and in principle we could use a density-dependent dispersion relation without greatly increasing the difficulty of the calculation. Our present calculations use a constant-mass dispersion relation with no renormalization; this is consistent with the weak-interaction assumption.

In this simulation, we place an initial nonequilibrium distribution of particles on a grid of discrete energy points and calculate  $\Gamma_i$  and  $\Gamma_o$  at each point using Eq. (7) and the similar expression for  $\Gamma_o$ . In a single iteration, the change in the number of particles with energy E is given by

$$dN(E) = \underline{x}N \cdot \left[ \frac{\Gamma_i(E)}{\sum_k \Gamma_i} - \frac{\Gamma_o(E)}{\sum_k \Gamma_o} \right], \qquad (8)$$

where N is the total number of particles, and  $\underline{x}$  is an overall multiplicative factor. x is determined by the condition that the total change in the spectrum,  $\sum_k dN(E)$ , must be small compared to N.  $\underline{x}\tau$  is the time step for a single iteration, where  $\tau$  is a characteristic scattering time for particles in the gas.  $\tau$  depends on the matrix element M, the gas density and average energy, and—as we shall see—the degeneracy of the gas.

In principle, one could use a grid of evenly-spaced energy points; however, as condensation nears, the distribution becomes sharply peaked at low energies, requiring a high-density grid. Since computation time goes as the cube of the number of grid points, if an evenly-spaced grid is used, a near-condensate with a narrow peak will require far too much time. We used a nonuniform grid of points with many points near E = 0. To eliminate round-off errors, the particle energies for each calculation were picked randomly within each energy interval (E(i-1), E(i)). As a measure of the error in our program, an equilibrium distribution with  $\mu = -0.001 k_B T$ , which ideally should not change at all, was allowed to scatter. The relative change  $\sum_k dN(E)/N$  was less than one part in a thousand per iteration, for  $\underline{x} = 1$ . The calculations for the evolution of a nonequilibrium system, placed on a 256-point grid of energy points and evolved through 100 iterations, took eight hours on an FPS 264 array processor.

## **IV. RESULTS**

We first model the behavior of a low-density, nearly classical gas, thermally isolated from its surroundings. Figure 1 shows the evolution of such a system after one, two, and four interparticle scattering times, where one "scattering time" is defined as  $(1/\underline{x})$  iterations, the time for all N of the particles to scatter once. An interesting

result is that regardless of the initial distribution, the particles move to the classical Maxwell-Boltzmann distribution in less than five scattering times. In this particular case, the initial distribution corresponds to a uniform occupation number for k-states up to  $E_0$  and zero occupation number of k-states above this energy. The average particle energy in this system is

$$\overline{E} = \left( \int_0^{E_0} E^{3/2} dE \right) / \left( \int_0^{E_0} E^{1/2} dE \right) = 0.6E_0 .$$
(9)

We can express this average energy in terms of the equilibrium temperature of a classical gas  $T_0$  given by  $\overline{E} = 1.5k_BT_0$ , so that  $E_0 = 2.5k_BT_0$ . For a low-density



FIG. 1. Particle distribution for a boson gas undergoing only interparticle scattering, for the low-density case,  $n = 0.00125n_c^0$ , where  $n_c^0$  is the critical density at temperature  $T_0$ , and  $T_0$  is defined by  $\overline{E} = 1.5k_BT_0$ . The top curve is the initial distribution, and the following curves correspond to the distribution after all the one, two, and four characteristic scattering times. The index  $i_{\text{scatt}}$  is the number of scattering events per particle since the initial creation. The last curve is within 1% of a Maxwell-Boltzmann distribution.

case,  $T_0$  is the temperature which describes the final Maxwell-Boltzmann distribution of the gas. Although the full quantum statistics are included in this calculation, the particle density for this case is only  $n = 0.00125n_c^0$ , where  $n_c^0 = n_c(T_0)$  is the critical density at temperature  $T_0$ . Therefore, quantum effects are not observed.

As the density is increased into the quantum regime, we find that the system takes many more scattering events to come into equilibrium. Figure 2 shows the energy distribution at different times for a Bose gas above critical density with fixed volume, energy, and density, for the same initial energy distribution as the case in Fig. 1. The transients caused by the initial discontinuity at  $E = E_0$  continue up to roughly 20 scattering times. After 80 scattering times, the distribution is still far from condensation.

Upon reflection, it is not surprising that the Bose system takes many scattering events to approach condensation. From Eq. (5) one can show that the scattering rate into the ground state is identically zero for a noncondensed system.<sup>10</sup> The rate into nearby states goes as the magnitude of the wavevector k for constant matrix element M. Terms of  $M^2$  with higher orders of k will contribute to scattering into these states even less.

We find that after the initial transients die away, the distribution fits reasonably well to a single-chemicalpotential Bose-Einstein distribution f(E) as given in Eq. (1); a fit to the distribution at t = 80 is shown as the dotted line in Fig. 2. Each fit has two free parameters, the temperature  $T_s$ , which determines the energy scale, and the ratio  $\alpha_s = -\mu_s/k_BT_s$ , which determines the spectral shape. The fitted values of  $\alpha_s$  and  $T_s$  change in time, moving toward their equilibrium values. In the case shown in Fig. 2, after 80 scattering events the system has reached only  $\alpha_s = 0.024$ , with  $T = 1.625T_0$ . The equilibrium distribution for this case is a 50% condensate ( $\alpha = 0$ ) at a temperature of  $3T_0$ . As the gas is equilibrating its energy spectrum will look similar to that of a much less dense gas in equilibrium at an elevated temperature.

Figure 3 shows the evolution of the fit parameters,





FIG. 2. Particle distribution for a boson gas undergoing only interparticle scattering, for the high density case,  $N = 12.5n_c^0$ , where  $n_c^0$  is the critical density at temperature  $T_0$ , and  $T_0$  is defined by  $\overline{E} = 1.5k_BT_0$ . The top curve is the initial distribution, and the following curves correspond to the distribution after all the particles have scattered 20, 40, and 80 times. The curve at  $i_{\text{scatt}} = 80$  is best fit by a Bose-Einstein distribution of the form of Eq. (1) with  $T_s = 1.625T_0$ ,  $\alpha_s = 0.024$ , shown as the dotted line.

FIG. 3. (a) The temperature  $T_s$ , from best fits of the Bose-Einstein distribution in Eq. (1) vs  $i_{scatt}$ , the number of scattering events per particle, for bosons undergoing only interparticle scattering. Three cases with varying density are shown, all with the same average energy. (b) The best fit value of  $\alpha_s$  vs  $i_{scatt}$  for the same three cases. (c) The values of density *n*, deduced from Eq. (10) using the fit values of  $T_s$  and  $\alpha_s$  for the above three cases.

starting from the same constant-occupation-number distribution, for three different densities at the same average energy. Figure 3(c) shows the "spectroscopic density"  $n_s$ , defined by the relation

$$n_s = g(m/2\pi\hbar^2)^{3/2} \int f(E;\alpha_s,T_s) E^{1/2} dE . \qquad (10)$$

As seen in this figure, in contrast to the two cases below the critical density, above the critical density the spectrum at intermediate times appears to be that of a gas in equilibrium at much lower density—here almost an order of magnitude less. We stress that the number of particles in each calculation is strictly conserved— $n_s$  is merely a deduction from spectral fits. Although  $n_s/n_c^0$ can be greater than 1, the spectrum never appears condensed because the gas temperature is greater than  $T_0$ , which is used to calculate  $n_c^0$  by Eq. (2). That is, the ratio  $n_s/n_c(T_s)$  is always less than one.

As seen in Fig. 3(a), the equilibrium temperature rises with density. This reflects the fact that for a given temperature the average energy of a Bose gas decreases as the gas becomes more degenerate. Specifically, for a Bose gas at constant potential energy the total number of particles is

$$N = (\text{const})(k_B T)^{3/2} \int f(\epsilon; \alpha) \epsilon^{1/2} d\epsilon$$
 (11)

and the total energy is

$$U = (\text{const})(k_B T)^{5/2} \int f(\epsilon; \alpha) \epsilon^{3/2} d\epsilon , \qquad (12)$$

where the unitless integrals depend only on the quantity  $\alpha = -\mu/k_B T$ . Thus  $\overline{E} = U/N = (k_B T)g(\alpha)$ , where g has the classical value  $\frac{3}{2}$  for  $\alpha \gg 1$ , approaches  $\frac{3}{4}$  for  $\alpha$  near zero, and goes to zero as an increasing fraction of the gas enters the condensate. For constant total energy, then, if the degeneracy increases, so must the temperature. Figure 4 shows the equilibrium temperature versus density of Bose particles at constant average energy.

So far we have described the thermalization process in terms of the number of scattering events per particle. The average time per scattering event for a particle obviously depends on the actual particle density n = N/V, the average energy, and the scattering cross section of the particles. The average scattering time also depends on



FIG. 4. The equilibrium temperature vs the density of bosons, for constant average energy. Also plotted is the critical temperature for Bose condensation, as a function of density.

the degree of degeneracy, since the (1+f) stimulated emission terms enhance the scattering rate. At high quantum degeneracy, the scattering rate increases as  $n^3$ instead of linearly with n as expected from classical statistics. Also, the average scattering rate changes in time as the degree of degeneracy changes during equilibration. In the example of the high-density case shown in Fig. 2, the average scattering time, proportional to the sum of all  $\Gamma_i$ , decreases by almost an order of magnitude between the first and 200th iterations. Figure 5 shows the values of the fitted  $\alpha_s$  and the spectral half-width  $\Delta$  as a function of real time, where we have calibrated the time scale by equating the rate sums  $\sum_k \Gamma_i$  and  $\sum_k \Gamma_o$  at low density to the classical collision rate  $1/\tau_0 = \sigma n \overline{v}$ , where  $\sigma$ is a constant collision cross section, n is the density, and  $\overline{v} = (3k_B T_0/m)^{1/2}$  is the classical average particle velocity. Both the fit value of  $\alpha_s$  and the halfwidth are well described by an exponential decay in time. For the example of excitons in Cu<sub>2</sub>O with density  $n = 10^{19} / \text{cm}^3$ , average energy  $\overline{E} = 1.5k_B T_0$ ,  $T_0 = 4.3$  K, mass of  $3m_0$  (Ref. 11), and scattering cross section 20 Å, the gas takes roughly 100 ps to reach the value of  $\alpha_s = 0.01$ .

At first, it may seem that since  $\alpha_s$  never reaches zero, that the system can never condense. However, from Eq. (1) the occupation number of the ground state is given approximately by  $-1/(\mu/k_BT)\equiv 1/\alpha$ . Therefore, when  $\alpha_s$ reaches values around 1/N, the gas is effectively condensed. Figure 5 shows that  $\alpha_s$  decreases by 2 orders of magnitude in a time  $0.15\tau_0$  with  $\tau_0$  the classical hard sphere scattering time. If the system continues at the same exponential rate it will reach condensation within just a few classical scattering times. We see that although the total number of scattering events to reach equilibrium greatly increases at condensation, the real time required does not, due to the enhancement of the overall scattering rate by the stimulated emission effect.



FIG. 5. (a) The value of  $\alpha_s$  vs time for the case  $n = 12.5n_c^0$ . The classical scattering time is given by  $\tau_0 = 1/n\sigma v$ , where n,  $\sigma$ , and v are defined in the text. (b) The halfwidth of the energy distribution,  $\Delta$ , vs time for the same case.

The application of these results to systems with finite lifetime depends on the mechanism of decay. If the decay rate were enhanced by stimulated emission in the same way as the interparticle collision rate, then the lifetime should remain proportional to the scattering time and condensation would be impossible. In the case of excitons in Cu<sub>2</sub>O, the dominant decay mechanism is believed to be an Auger process<sup>5</sup> in which two particles collide and one is annihilated. Since the final state of the remaining particle is expected to be a high-energy ionized state, the stimulated emission effect should not enhance the decay rate, and the Auger process should simply be proportional to the *classical* collision rate  $1/\tau_0 = \sigma n \overline{v}$ . As shown above, a lifetime of a few classical collision times is adequate for condensation. Therefore, if the Auger cross section is smaller than the collision cross section  $\sigma$ , the system can condense. In fact, the measured cross section for Auger decay of in Cu<sub>2</sub>O is on the order of 2Å (Ref. 5), which is much smaller than the expected hardsphere collision cross section.

Although the numerical calculations described above

were done for particles with no decay, the results of these calculations can be extended in a simple way to systems with generation and decay of particles. We have repeated the above iteration process for a gas in steady state, with equal numbers of particles generated and eliminated each iteration. For particles with lifetime of  $i_x$  scattering times, the distribution reaches a value of  $\alpha$  in steady state which is nearly the same as the fit value  $\alpha_s$  found after  $i_x$  scattering times in the calculation above. Thus these calculations serve as predictions for the real steady-state degeneracy of finite-lifetime particles.

## V. MODEL OF BOSE GAS WITH INELASTIC PHONON SCATTERING

In high density and temperature regimes, interparticle scattering will be the main mechanism by which particles such as excitons initially establish a temperature. At sufficiently low densities, however, collisions with a heat reservoir of phonons will dominate. These collisions have the following form:

$$S(\mathbf{k}_{1},\mathbf{k}_{2},\mathbf{k}_{p}) = M_{p}^{2} \{\delta(\mathbf{k}_{1}+\mathbf{k}_{p}-\mathbf{k}_{2})\delta(E_{1}+E_{p}-E_{3})f(\mathbf{k}_{1})F(\mathbf{k}_{p})[1+f(\mathbf{k}_{2})] + \delta(\mathbf{k}_{1}-\mathbf{k}_{p}-\mathbf{k}_{2})\delta(E_{1}-E_{p}-E_{3})f(\mathbf{k}_{1})[1+F(\mathbf{k}_{p})][1+f(\mathbf{k}_{2})]\}, \qquad (13)$$

where F is the phonon occupation number.  $M_p$ , which typically depends on the phonon momentum  $k_p$ , is the bosonphonon interaction matrix element which determines the overall time scale. Using a procedure very similar to the derivation of Eq. (7) in the Appendix, we integrate out the delta functions and angles, to obtain the scattering rate into state k,

$$\Gamma_{i}(E(\mathbf{k}))dE = \int dE \, k_{p}^{2} dk_{p} M_{p}^{2} \frac{1}{\frac{\partial E}{\partial k^{2}}} \frac{1}{\left|_{k} \frac{\partial E}{\partial k^{2}}\right|_{k_{1}}} \times \{f(E_{1})F(E_{p})[1+f(E)]\Theta(E_{1}+E(k_{p}+k))\Theta(E_{1}-E(k-k_{p}))|_{E_{1}}=E-E_{p}} + f(E_{1})[1+F(E_{p})][1+f(E)]\Theta(E(k_{p}+k)-E_{1})\Theta(E_{1}-E(k-k_{p}))|_{E_{1}}=E+E_{p}}\},$$
(14)

where  $\Theta(E)$  is the Heaviside function. For these calculations we assume  $M_p$  constant. The first term in the brackets corresponds to phonon absorption, while the second corresponds to phonon emission. As in the interparticle scattering case, the scattering rate into the E=0state vanishes for a noncondensed system.

We can use this formula, plus the similar expression for  $\Gamma_0$ , to calculate the scattering rates at every point on a grid of energy points. We iterate as in the interparticle scattering case, except that the phonon distribution is kept fixed at the Planck distribution and thus total particle energy is not conserved. That is, we assume a phonon bath of infinite heat capacity.

Figure 6 shows the halfwidth of the energy distribution versus time for a system with a density slightly above the critical density and initial temperature twice the phonon temperature. In this case, the fit parameter  $\alpha_s$  goes quickly to less than 0.001, but then slows down in its ap-



FIG. 6. The halfwidth of the energy distribution,  $\Delta$  vs number of phonon scattering events, ignoring interparticle scattering, for the case  $n = 1.25n_c^0$ , with initial  $E = 2(1.5k_BT_0)$  and final  $E = 1.5k_BT_0$ , reached after three scattering events.

proach to zero. The fit temperature goes to the bath temperature almost immediately, within a few scattering times.

In the case of  $Cu_2O$ , deformation-potential theory predicts<sup>12</sup> an exciton-phonon matrix element of

$$M_p^2 = \frac{\hbar D^2}{2\rho v_l} \frac{k_p}{V} , \qquad (15)$$

where D is the deformation potential, equal to 2.1 eV for  $Cu_2O$  (Ref. 13),  $\rho$  is the mass density of the crystal,  $v_l$  is the longitudinal acoustic sound velocity, and V is the crystal volume. For a lattice temperature of 2 K, this yields an average phonon scattering time of one nanosecond. This implies that at 2 K, the phonon-exciton scattering rate will be less than the interparticle scattering rate of excitons in  $Cu_2O$  for densities above roughly<sup>18</sup>/cm<sup>3</sup>.

The degeneracy of the excitons does not affect the phonon-exciton scattering rate very much. From these calculations, when the system reaches the critical density, the phonon-exciton scattering rate increases only about 30% above the rate for a classical exciton gas. Also, the average scattering rate does not vary significantly in time as the system approaches equilibrium. Thus these calculations show that the time for condensation does approach infinity for a system with only particle-phonon interactions. Interparticle scattering must always be the mechanism by which an exciton gas Bose condenses.

#### **VI. CONCLUSIONS**

We have found that, under the random-phase approximation, the weakly interacting Bose gas requires many scattering events per particle to approach equilibrium and condense. Nevertheless, because of the increased scattering rate due to the stimulated emission effect in interparticle scattering, the gas does not take much longer time to condense than it does to reach equilibrium under classical conditions. For the case of excitons in Cu<sub>2</sub>O, it appears that the particle lifetimes are adequate for condensation. Our calculations indicate, however, that exciton-phonon scattering will not help the system condense. The question of whether coherent effects or strong particle interactions initiate condensation on a shorter time scale remain issues for experimental and theoretical investigation.

The excitonic system in  $Cu_2O$  possesses some interesting complications which we did not incorporate into the present calculations. Interconversion between ortho and paraexcitons means that there are actually two separate gas components to consider, presumably in good thermal contact with each other. An Auger recombination process at high densities will not only shorten the excitonic lifetime, but it will also provide an effective heating mechanism for the gas. On the other hand, the emission of optical phonons is a rapid energy loss mechanism which affects the high-energy tail of the kinetic-energy distribution. Also, the exciton is weakly coupled to the electromagnetic field by the polariton effect, which modifies the density of states near k = 0. This effect is small for Cu<sub>2</sub>O—especially paraexcitons—due to the forbidden nature of the band-to-band transitions, but it may affect the detailed kinetics near k = 0. We note that the formalism presented here seems well suited to considering polariton effects, since the scattering formulas do not require a definition of chemical potential  $\mu$ . Finally, the experimental situation involves particle diffusion, which produces inherent spatial inhomogeneities in the gas density. The present calculations best apply to a local region in a gas over which density is relatively uniform.

#### ACKNOWLEDGMENTS

This work is supported by the National Science Foundation Grant No. NSF DMR 87-22761 with facility support by the Materials Research Laboratory Grant No. DMR86-12860. One of us (D.S.) was supported by the University of Illinois as a University Fellow.

# APPENDIX: REDUCTION OF THE SCATTERING RATE INTEGRAL

Let the probability for two bosons wavevectors  $\mathbf{k}_1, \mathbf{k}_2$  to scatter into states  $\mathbf{k}_3, \mathbf{k}_4$  be

$$S(\mathbf{k}_{1},\mathbf{k}_{2};\mathbf{k}_{3},\mathbf{k}_{4}) = M^{2}(|\mathbf{k}_{1}-\mathbf{k}_{3}|) \ \delta(\mathbf{k}_{1}+\mathbf{k}_{2}-\mathbf{k}_{3}-\mathbf{k}_{4})$$
$$\times \delta(E_{1}+E_{2}-E_{3}-E_{4})f(\mathbf{k}_{1})f(\mathbf{k}_{2})$$
$$\times [1+f(\mathbf{k}_{3})][1+f(\mathbf{k}_{4})] .$$

The occupation number f depends only on the magnitude of k. We integrate over  $\mathbf{k}_4$ , eliminating  $\mathbf{k}_4 = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3$ via the momentum delta function, to get

$$\int d^{3}k_{4}S = \delta(E_{1} + E_{2} - E_{3} - E_{4})M^{2}(|\mathbf{k}_{1} - \mathbf{k}_{3}|)$$
  
×  $F(E_{1}, E_{2}, E_{3}, E_{4})|_{E_{4} = E(|\mathbf{k}_{1} + \mathbf{k}_{2} - \mathbf{k}_{3}|)}$ 

where F is the product of the four occupation-number factors. We now define  $\tilde{\mathbf{k}} = \mathbf{k}_1 - \mathbf{k}_3$  and integrate over  $d\Omega_2$ , eliminating  $\cos\theta_2$  via the energy delta function, to get

$$\int d^{3}k_{4} d\Omega_{2}S = 2\pi \frac{1}{\frac{\partial E}{\partial k^{2}}} \frac{1}{|_{k=k_{4}'}} \frac{1}{2k_{2}\tilde{k}} \Theta(E_{4}' - E(\tilde{k} + k_{2}))$$

$$\times \Theta(E(|\tilde{k} - k_{2}|) - E_{4}')$$

$$\times M^{2}(\tilde{k})F(E_{1}, E_{2}, E_{3}, E_{4}')|_{E_{4}'} = E_{1} + E_{2} - E_{3}$$

 $k'_4 = k(E'_4)$  is the magnitude of **k** corresponding to the energy  $E_1 + E_2 - E_3$ .

We can now integrate over  $d\Omega_3$ , and convert the integration over  $\cos\theta_3$  to integration over  $\tilde{k}$ . If we assume that E(k) increases monotonically with k, we find

Г

$$\int d^{3}k_{4} d\Omega_{2} d\Omega_{3} S = \frac{(2\pi)^{2}}{4} \frac{1}{\frac{\partial E}{\partial k^{2}}} \frac{1}{|k_{4}k_{4}|} \int_{\tilde{k}_{10}}^{\tilde{k}_{10}} d\tilde{k} M^{2}(\tilde{k}) F(E_{1}, E_{2}, E_{3}, E_{4}')|_{E_{4}'} = E_{1} + E_{2} - E_{3}$$

with

$$\tilde{k}_{up} = \min(k_1 + k_2, k_3 + k_4')$$

and

$$k_{10} = \max(|k_1 - k_2|, |k_3 - k_4'|)$$
.

If  $M^2$  is constant, then the integral over  $\tilde{k}$  becomes just  $\tilde{k}_{up} - \tilde{k}_{lo}$ . Integration over  $d\Omega_1$  at this point results in simply multiplying the above integral by a factor of  $4\pi$ .

- <sup>1</sup>As shown in, for example, D. I. Blokhintsov, *Quantum Mechanics* (Reidel, Dordrecht, 1964), Sec. 120, pp. 493-500.
- <sup>2</sup>I. Silvera and J. T. M. Walraven, Prog. Low Temp. Phys. 10, 139 (1986).
- <sup>3</sup>D. Snoke, J. P. Wolfe, and A. Mysyrowicz, Phys. Rev. Lett. **59**, 827 (1987).
- <sup>4</sup>For a review see E. Hanamura and H. Haug, Phys. Rep. **33**, 209 (1977).
- <sup>5</sup>A. Mysyrowicz, D. Hulin, and C. Benoit a la Guillaume, J. Lumin. 24/25, 629 (1981).
- <sup>6</sup>M. Inoue and E. Hanamura, J. Phys. Soc. Jpn. 41, 771 (1976).
- <sup>7</sup>E. Levich and V. Yakhot, Phys. Rev. B 15, 244 (1977).
- <sup>8</sup>E. Levich and V. Yakhot, J. Phys. A 11, 2237 (1978).
- <sup>9</sup>The pair potential is always repulsive, as shown by F. Bassani and M. Rovere, Solid State Commun. **19**, 887 (1976). The

To calculate the total scattering rate into states with momentum k, we can substitute k for  $k_3$  and E = E(k)for  $E_3$  in the above equation, and integrate over  $k_1$  and  $k_2$ . Utilizing the relationship

$$dE = \frac{\partial E}{\partial k^2} 2k \, dk$$

we recover Eq. (7) in the text. The total scattering rate out of states with momentum k is found by substituting k for  $k_1$ , and integrating over  $k_2$  and  $k_3$ .

n = 1 exciton Bohr radius is 7 Å.

- <sup>10</sup>Three-body collisions of the form  $S(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3; \mathbf{k}_4, \mathbf{k}_5, \mathbf{k}_6)$ = $M'^2 \delta(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3 - \mathbf{k}_4 - \mathbf{k}_5 - \mathbf{k}_6) \quad \delta(E_1 + E_2 + E_3 - E_4 - E_5 - E_6)f(\mathbf{k}_1)f(\mathbf{k}_2)f(\mathbf{k}_3)[1 + f(\mathbf{k}_4)][1 + f(\mathbf{k}_5)][1 + f(\mathbf{k}_6)]$  will not be subject to the same phase-space restrictions as twobody collisions, so that the scattering rate into E = 0 will be finite. The matrix element M' will in general, however, be very small.
- <sup>11</sup>N. Caswell, J. S. Weiner, and P. Y. Yu, Solid State Commun. 40, 843 (1981).
- <sup>12</sup>See, for example, K. Seeger, Semiconductor Physics (Springer, New York, 1973).
- <sup>13</sup>H. Trebin, H. Z. Cummins, and J. L. Birman, Phys. Rev. B 23, 597 (1981).