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Growth kinetics in exciton systems

Jian Hua Yao, Hong Guo, and Martin Grant
Centre for the Physics of Materials, Physics Department, Rutherford Building, McGill University, 3600 rue University, Montréal, Québec, Canada H3A 2T8

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We study the kinetics of first-order phase transitions in exciton systems in both two and three dimensions, where the volume fraction of the condensing droplets is small but finite. The solution of our mean-field equations gives explicit self-consistent expressions for the droplet distribution function, the nucleation rate, and the time evolution of both the average droplet size and the total number of droplets. The results are compared with those of the classical Ostwald ripening problem.

I. INTRODUCTION

It is known that bounded electron-hole pairs, called excitons, can be excited optically in a semiconductor.[1] At high temperatures, these pairs behave as a Bose gas of free excitons.[2] At high density, the gas ionizes into a fermionic system of electron-hole plasma, and a metal-insulator transition occurs. Below a critical temperature, a first-order phase transition may take place, which is analogous to a liquid-gas transition, except that now particles have a finite lifetime due to recombination of the electron-hole pairs. The two-phase coexistence consists of a low density exciton gas and a high density fermionic "liquid" phase. This two-phase coexistence is a nonequilibrium steady state, but its phase diagram is very similar to that of liquid-gas systems.[3–6]

Nucleation phenomena in this nonequilibrium phase transition have been studied both experimentally and theoretically.[3,5,7–16] On the experimental side, many interesting physical quantities have been measured, such as the mass of excitons, the effective surface tensions of the droplets, the critical temperatures, the average finite lifetimes of excitons, and the average radius of exciton droplets. On the theoretical side, several theories have been proposed to study this nonequilibrium phase transition.[9,17–21]

A particularly interesting question is whether one can apply the classical theory of nucleation and growth of a liquid-gas transition to exciton systems, and what modifications one must make for such an application. In this direction, a Becker-Döring–type theory was proposed by Silver.[17] In his theory, electron-hole condensation was treated as a stochastic process where such effects as the finite lifetime of the condensing particles, effective surface tension of the droplets, and impurities were included. The main difference between the classical Becker-Döring theory[22,23] and that of Silver for exciton condensation is that the latter has two additional terms in the equation of motion for the droplet distributions. One term accounts for the evaporation-condensation mechanism induced by the nucleation centers; the other accounts for the decay of excitons due to their finite lifetime and for the creation of new excitons by optical pumping.

An alternative to the cluster dynamics of the Becker-Döring theory is to apply a field theoretic method to the nonequilibrium metastable states. For exciton systems, some initial work in this direction has indeed been carried out,[17] using techniques directly borrowed from studies of liquid-gas transitions.[24] As a starting point, hydrodynamic equations similar to those of Langer and Turski[24] for the classical system were proposed. Scattering of the excitons by, say, phonons, was modeled by a damping term added to the equation. The effects of creation and recombination of excitons were modeled by appropriate terms included in the continuity equation. The predictions of this theory were not dramatically different from those of the Becker-Döring theory, but this formulation may allow studies of cases with large supersaturations. Using this formalism, quantities such as the distribution function of droplet size, the evaporation and recombination of excitons, and the exciton nucleation and decay currents, were computed numerically.

While some interesting results and understanding were gained through these studies of the nucleation and growth in exciton systems, improvements and systematic investigations can now be made due to recent progress in the theoretical understanding of phase transition kinetics. In particular, since the growth of an exciton droplet depends on whether its size is larger or smaller than a critical size, the process cannot be entirely stochastic as assumed in some of the earlier theories.[17] The growth dynamics in the metastable region during a first-order phase transition is well-known to be described by the Lifshitz-Slyozov theory[25] in the limit of very small volume fraction of the condensing phase, and one would also like to generalize this theoretical framework to exciton systems. We also note that in the limit that the lifetime of excitons approaches infinity, one must recover the Lifshitz-Slyozov theory. However, the earlier theories mentioned above do not satisfy this condition. The kinetics of first-order phase transitions often shows self-similar behavior, i.e., various correlation functions develop into scale-invariant forms.[26] However, so far this idea has not been exploited in theoretical studies of droplet growth in exciton systems. Finally, there are many recent experimental works which have studied the electrical and optical properties of excitons confined in very narrow quantum wells.[27] Thus it is important to develop a theory to study the growth kinetics of exciton droplets in both two and three dimensions. The goal of the present paper is to do so, and thereby motivate experiments on these issues.

In a recent work, we have introduced a theoretical approach to Ostwald ripening for both two- and three-dimensional systems,[28] where screening effects due to interacting droplets are incorporated. The solution of our mean-field equations gives both the droplet distribution function $f(R,t) \propto g(R/\overline{R})/\overline{R}^{D+1}$ and the growth law for the average droplet radius $\overline{R}(t) = [K(\phi)t + \overline{R}^3(0)]^{1/3}$, where $g(z)$ is the scaled distribution function, $K(\phi)$ is the coarsening rate, ϕ is the volume fraction of droplets, and D is spatial dimension. Both the two- and three-dimensional results conform well to numerical simulations and experiments.

In the following we shall extend our method mentioned above[28] to the problem of growth of droplets where the constituent particles have a finite lifetime. In the next section, we establish the basic equations of our theory by considering the effect of finite droplet lifetime. In particular, the finite lifetime of the excitons provides a mechanism for droplets to lose particles, and we assume that the rate of process is proportional to the droplet volume. As a result, droplets cannot grow to form a single macroscopic phase. Furthermore, because of the conservation law and the recombination of excitons inside of droplets, nucleation will not stop even at late times. It is, therefore, necessary to supplement a nucleation term (source term) in the continuity equation. In Sec. III, we will solve the basic equations and discuss the results. Section IV gives a conclusion.

II. THE MODEL

In this section, we will present the model for the droplet growth in an exciton system. To make the derivation clear, we will first briefly review our model for classical Ostwald ripening with finite volume fractions of the minority phase.[28] The appropriate equations for the exciton systems will be a generalization of the classical theory.

Our study makes use of dimensionless variables. Units of length and time are given in terms of the capillary length $l_c = (D-1)\gamma V_m/R'T$ and a characteristic time $t^* = l_c^2/(\mathcal{D}C_\infty V_m)$. These quantities involve the surface tension γ, the molar volume V_m, the gas constant R', the temperature T, the diffusion coefficient $\mathcal{D}$, and the solute concentration in the matrix at a flat interface C_∞. It is also convenient to introduce a dimensionless concentration field $\theta(\mathbf{r}) = [C(\mathbf{r}) - C_\infty]/C_\infty$, where $C(\mathbf{r})$ is the concentration field at point $\mathbf{r}$ outside the droplets. For small volume fraction of the condensing phase, it is reasonable to assume that the droplets are of spherical shape as observed in classical systems.[29]

In the classical nucleation theory, using a mean-field approximation, the growth law of droplet radius R at late times is given by[28,30]

$$\frac{d(vR^D)}{dt} = I(R)[\theta_{\text{av}} - \theta(R)] \ , \tag{1}$$

where $v = \pi^{D/2}/\Gamma(D/2+1)$. To write down this equation, we have assumed that the material flux of the condensing phase which determines the growth rate of the droplet is only proportional to the difference between the *average* bulk concentration θ_{av} and the concentration at the droplet boundary $\theta(R)$. The curvature-dependent coefficient $I(R)$ is unknown *a priori*, but can be determined self-consistently (see below).

The concentration of the condensing phase satisfies a many-droplet diffusion equation. In steady state this equation is written as[31]

$$\nabla^2\theta(\mathbf{r}) = a\sum_{i=1}^{N} B_i\delta(\mathbf{r} - \mathbf{r}_i) \ , \tag{2}$$

where $a = 2\pi^{D/2}/\Gamma(D/2)$, N is the total number of the droplets, $\mathbf{r}_i$ gives the location of the ith droplet, and B_i is the growth or shrink rate of the ith droplet. The conservation of the total concentration requires

$$\sum_{i=1}^{N} B_i = 0 \ . \tag{3}$$

Noticing the similarity of the last two equations to those of the homogeneous electron gas, we introduce a Thomas-Fermi approximation to reduce the steady-state many-body diffusion equation to an one-body Helmhotz equation,[28]

$$\nabla^2\theta(\mathbf{r}) - \xi^{-2}\theta(\mathbf{r}) + \xi^{-2}\theta_{\text{av}} = aB_i\delta(\mathbf{r} - \mathbf{r}_i) \tag{4}$$

in the vicinity of the ith droplet.[28] The screening length ξ is determined by

$$\xi^{-2} = \frac{a}{V'}\int_0^\infty f(R,t)/V(R/\xi,R)dR \ , \tag{5}$$

where $V(R/\xi,R) = \exp(-R/\xi)/R$ for $D = 3$; $V(R/\xi,R) = K_0(R/\xi)$ for $D = 2$, and K_0 is the zeroth order modified Bessel function, $f(R,t)$ is the distribution function of droplet size, normalized to the total number of droplets in the system, and V' is the system volume. According to Eq. (2), the growth law also satisfies the following relationship:[28]

$$\frac{d(vR_i^D)}{dt} = aB_i \ . \tag{6}$$

Using the Gibbs-Thomson boundary condition

$$\theta(\mathbf{r})\Big|_{|\mathbf{r}-\mathbf{r}_i|=R_i} = \frac{1}{R_i} \tag{7}$$

and imposing the concentration far from the droplet $\lim_{r\to\infty}\theta(r) = \theta_{\text{av}}$, the solution of Eq. (4) gives

$$1/R_i = \theta_{\text{av}} - B_iV(R_i/\xi,R) \tag{8}$$

for $i = 1,\ldots,N$. Substituting the solution of Eqs. (3) and (8) into Eq. (6) and comparing with Eq. (1), we obtain the following expressions:

$$I(R) = a/V(R/\xi,R) \tag{9}$$

and

$$\theta_{\rm av} = \int_0^\infty \frac{f(R,t)}{RV(R/\xi,R)}dR \Big/ \int_0^\infty \frac{f(R,t)}{V(R/\xi,R)}dR \ . \tag{10}$$

Finally, the continuity equation can be written as

$$\frac{\partial f(R,t)}{\partial t} + \frac{\partial}{\partial R}[\dot{R}f(R,t)] = 0 \ , \tag{11}$$

and the conservation law Eq. (3) can be rewritten as

$$\frac{v}{V'}\int_0^\infty R^D f(R,t)dR = \phi \ , \tag{12}$$

where ϕ is the volume fraction of droplets and the overdot means derivative with respect to time. Equations (1), (5), (11), and (12) completely specify the droplet growth problem in the classical Ostwald ripening in both two and three dimensions. For further details of various stages of the derivation, see Ref. 28.

The above equations can be generalized to exciton systems in a reasonably simple fashion. First, the finite lifetime of an exciton provides a mechanism for droplets to lose particles. The rate of the reduction is inversely proportional to the lifetime of the exciton τ. We further assume that this rate is also proportional to the droplet volume since a larger droplet has more excitons. Therefore, the growth law for exciton systems should satisfy

$$\frac{dR^D}{dt} = \frac{D}{V(R/\xi,R)}\left\{\frac{\overline{[RV(R/\xi,R)]^{-1}}}{\overline{[V(R/\xi,R)]^{-1}}} - \frac{1}{R}\right\} - \frac{R^D}{\tau} \ , \tag{13}$$

where we have used Eqs. (1), (7), (9), and (10). The quantity with an overbar is defined as

$$\overline{A} \equiv \frac{\int_0^\infty f(R,t)AdR}{\int_0^\infty f(R,t)dR} . \tag{14}$$

Second, we shall study the situation that the system approaches a steady state at large times, i.e., the total number of excitons approaches a constant as $t \to \infty$. This is only possible when the system is continuously illuminated by an external radiation source, such that excitons are created at a rate equal to that for the annihilation of excitons in the droplets. The creation of new excitons leads to a continuous nucleation of droplets even at late times. It is, therefore, necessary to supplement a nucleation source term, $J(R,t)$, in the continuity equation, i.e.,

$$\frac{\partial f(R,t)}{\partial t} + \frac{\partial}{\partial R}[\dot{R}f(R,t)] = J(R,t) \ . \tag{15}$$

The nucleation rate $J(R,t)$ can be determined self-consistently by imposing the steady-state condition at large times. Taking a derivative of the volume fraction ϕ, defined in Eq. (12), with respect to time and using the continuity equation (15), we obtain

$$\frac{\partial}{\partial t}\int_0^\infty R^D f(R,t)dR = -\int_0^\infty R^D \frac{\partial}{\partial R}[\dot{R}f(R,t)]dR + \int_0^\infty R^D J(R,t)dR \ . \tag{16}$$

Integrating by parts we obtain

$$\frac{\partial}{\partial t}\int_0^\infty R^D f(R,t)dR = -R^D\dot{R}f(R,t)\Big|_0^\infty + \int_0^\infty \frac{dR^D}{dt}f(R,t)dR + \int_0^\infty R^D J(R,t)dR \ . \tag{17}$$

The first term on the right-hand side of (17) vanishes. Substituting the growth law (13) into the second term, we obtain

$$\frac{\partial}{\partial t}\int_0^\infty R^D f(R,t)dR = \int_0^\infty \frac{D}{V(R/\xi,R)}\left\{\frac{\overline{[RV(R/\xi,R)]^{-1}}}{\overline{[V(R/\xi,R)]^{-1}}} - \frac{1}{R}\right\}f(R,t)dR + \int_0^\infty R^D\left(J(R,t) - \frac{f(R,t)}{\tau}\right)dR. \tag{18}$$

Again, the first term on the right-hand side of Eq. (18) is zero; therefore, Eq. (18) becomes

$$\frac{\partial}{\partial t}\int_0^\infty R^D f(R,t)dR = \int_0^\infty R^D\left(J(R,t) - \frac{f(R,t)}{\tau}\right)dR \ . \tag{19}$$

The left-hand side is a time derivative of the volume fraction, which does not depend on time at late times, thus the simplest self-consistent nucleation rate $J(R,t)$ is

$$J(R,t) = \frac{f(R,t)}{\tau} \ . \tag{20}$$

So far, we have made two assumptions for the model: one is that the rate for droplets to lose a particle is proportional to the volume of the droplets, inversely proportional to the mean lifetime of the excitons, i.e., the term $-vR^D/\tau$ in the growth law equation (13); the other is that the system approaches a steady state at $t \to \infty$, and this is possible when the system is continuously illuminated by an external radiation such that nucleation of new droplets occurs even at late times, which compensates for the loss of excitons due to recombination, i.e., the term $J(R,t) = f(R,t)/\tau$ in the continuity equation (15). These assumptions are physically motivated and allow us to solve the problem in closed form without any further approximation or assumption.

Finally, substituting Eq. (20) into Eq. (15) and rewriting Eq. (13), we obtain the continuity equation and the growth law as follows:

$$\frac{\partial f(R,t)}{\partial t} + \frac{\partial}{\partial R}[\dot{R} f(R,t)] = f(R,t)/\tau \tag{21}$$

and

$$\frac{dR}{dt} = \frac{R^{1-D}}{V(R/\xi, R)} \left\{ \frac{\overline{[RV(R/\xi,R)]^{-1}}}{\overline{[V(R/\xi,R)]^{-1}}} - \frac{1}{R} \right\} - \frac{R}{D\tau} . \tag{22}$$

The second term on the right-hand side of Eq. (22) contributes a decreasing rate for each droplet. This leads to droplet radii being modified by a time-dependent factor, compared with classical Ostwald ripening. Furthermore, Eq. (21) implies that new droplets created by the source term follow the same distribution as old ones. Therefore, we anticipate that the scaled droplet distribution function $g(R/\overline{R})$ retains the same form as that of classical Ostwald ripening, although the growth law must change, since the droplets eventually decay due to exciton recombination. This anticipation can be verified by solving equations (5), (12), (21), and (22).

III. SOLUTION AND RESULTS

Although the basic equations (5), (12), (21), and (22) are very complicated, they are, fortunately, exactly integrable. In particular, we seek a solution to the droplet distribution function $f(R,t)$ in a scaling form. As in the classical Ostwald ripening problem,[23,25,28] a scaling solution of the droplet distribution $f(R,t)$ exists for $\phi \leq 0.085$ in $D=2$ and $\phi \leq 0.06$ in $D=3$. The scaling distribution function $f(R,t)$ can be obtained in a similar way to Ref. 28, satisfying

$$f(R,t) \propto \frac{g(z)}{R^{D+1}(t)} \tag{23}$$

with $z = R/\overline{R}(t)$. The scaled, normalized distribution function $g(z)$ is identical to that of classical Ostwald ripening,[28] obeying

$$g(z) = \begin{cases} \frac{-\lambda D}{w(z,\lambda)} \exp\left[\lambda D \int_0^z w^{-1}(z',\lambda) dz'\right] & \text{if } 0 < z < z_0 \\ 0 \ \text{otherwise} , \end{cases} \tag{24}$$

where

$$w(z,\lambda) = z^{1-D}/V(z/\eta, z)(\sigma - z^{-1}) - \lambda z ,$$

$$\sigma = \int_0^\infty \frac{g(z)}{zV(z/\eta,z)} dz \Big/ \int_0^\infty \frac{g(z)}{V(z/\eta,z)} dz ,$$

$$\eta^{-2} = \frac{\phi D}{\int_0^\infty z^D g(z) dz} \int_0^\infty \frac{g(z)}{V(z/\eta,z)} dz .$$

The separation factor λ and the least upper bound z_0 satisfy

$$w(z_0, \lambda) = 0 \quad \text{and} \quad \left. \frac{dw(z,\lambda)}{dz} \right|_{z=z_0} = 0 .$$

The average droplet radius $\overline{R}(t)$ follows an ordinary differential equation:

$$\frac{\overline{R}^3(t)}{D\tau} + \frac{1}{3} \frac{d\overline{R}^3(t)}{dt} = \lambda , \tag{25}$$

whose solution is

$$\overline{R}(t) = \left\{ \overline{R}^3(0) \exp\left(-\frac{3t}{D\tau}\right) + D\tau\lambda \left[1 - \exp\left(-\frac{3t}{D\tau}\right)\right] \right\}^{1/3} . \tag{26}$$

If $\phi > 0.085$ in $D=2$ or $\phi > 0.06$ in $D=3$, there exists no solution for $f(R,t)$. This gives the valid range of our model.

$\overline{R}(t)$ has been plotted in Fig. 1 for different values of the lifetime τ at $\phi = 0.05$ in $D=3$. The average radius first grows and then saturates at an asymptotic value $(DK\tau/3)^{1/3}$, as determined by Eq. (26), where the coarsening rate $K(\phi) = 3\lambda$ is plotted in Fig. 2. Similar to the classical Ostwald ripening problem, $K(\phi)$ shows considerable dependence on the volume fraction ϕ. Thus the value of $\overline{R}$ in the steady state, i.e., at $t \gg \tau$, depends not only on the finite lifetime τ but also on the volume fraction ϕ. Our conclusion qualitatively differs from Silver's theory[17] in which $\overline{R} \sim \tau$. His result is due to his simple assumption that electron-hole condensation is a stochastic process. Consequently, the probability for a droplet to absorb excitons is proportional to its surface $4\pi R^2$. On the other hand, the probability for a droplet to lose excitons due to the finite lifetime mechanism is proportional to $4\pi R^3/(3\tau)$. Thus the droplet growth rate

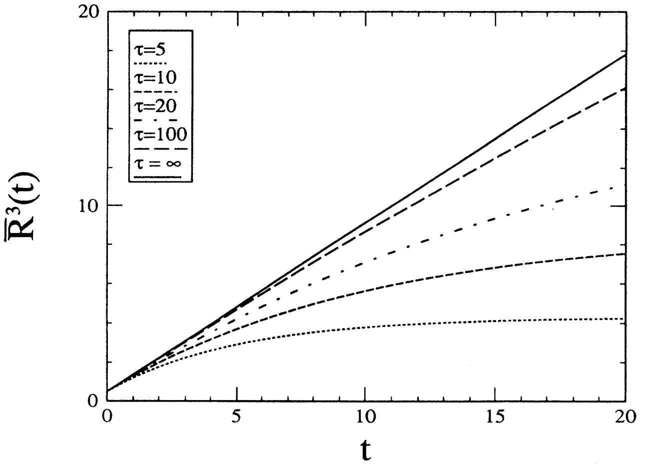

FIG. 1. The time evolution of $\overline{R}^3(t)$ for different finite lifetimes τ, $\phi = 0.05$, and $D = 3$. The results for $D = 2$ are qualitatively the same.

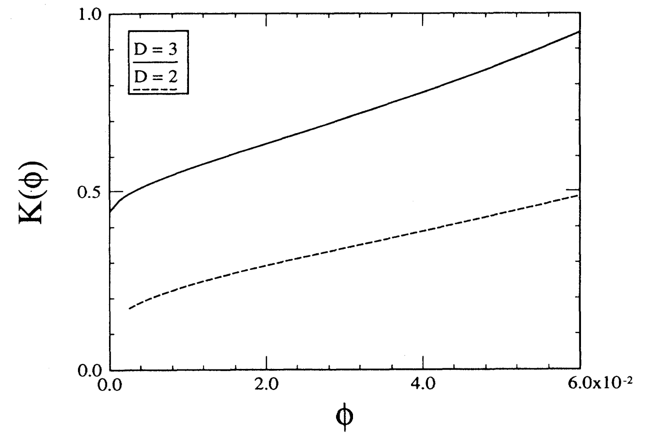

FIG. 2. A plot of the coarsening rate $K(\phi)$ vs the volume fraction ϕ in $D = 2$ and 3.

$d(4\pi R^3/3)/dt \propto 4\pi R^2 - 4\pi R^3/(3\tau)$, and when the system reaches a steady state, $\overline{R} \propto \tau$.

For times $t \ll \tau$, the behavior of $\overline{R}(t)$ is very close to the one-third power law. This is understandable since if the recombination of excitons is negligible, our system is very similar to that of a classical system undergoing Ostwald ripening. Indeed, for very large lifetime τ, or very small time t, Eq. (26) agrees with the result of Lifshitz-Slyozov theory,[28] $\lim_{\tau\to\infty} \overline{R}(t) = [\overline{R}^3(0) + K(\phi)t]^{1/3}$.

The distribution function of droplet size is obtained by inserting $\overline{R}(t)$ and $g[R/\overline{R}(t)]$ into Eq. (23); we obtain

$$f(R,t) \propto \frac{g[R/\overline{R}(t)]}{\overline{R}^{D+1}(t)} . \tag{27}$$

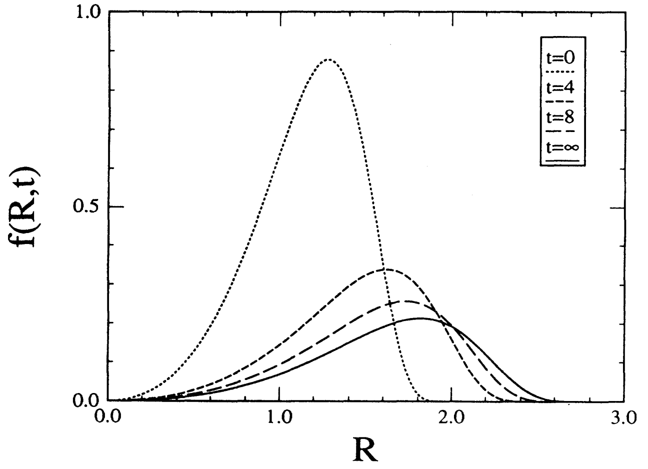

FIG. 3. The time evolution of the distribution function of droplet radii for the volume fraction $\phi = 0.05$, $\tau = 5$, and $D = 3$, where $\overline{R}^3(0) = 1.5$. The results for $D = 2$ are qualitatively the same.

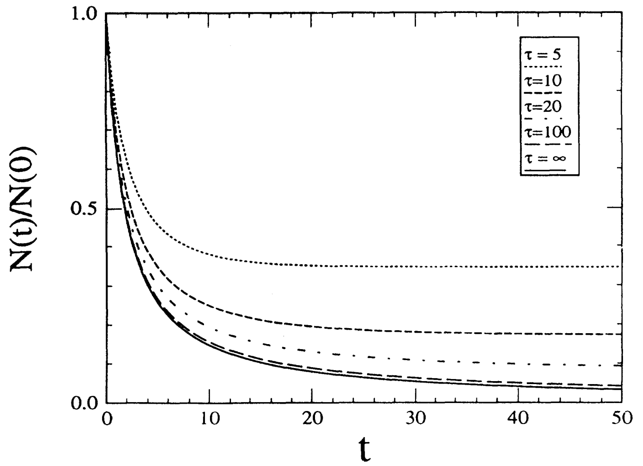

FIG. 4. The time evolution of the total number of droplet for different finite lifetimes τ, $\phi = 0.05$, and $D = 3$, where $\overline{R}^3(0) = 1.5$. The results for $D = 2$ are qualitatively the same.

Figure 3 shows the distribution functions at different times with $\phi = 0.05$ and $\tau = 5$, in $D = 3$. At early times, the area underneath the distribution function curve decays rapidly, and then approaches an asymptotic value. On the other hand, the peak position of the distribution function and the maximal radius $R_{\max}(t)$ increase rapidly, and then saturates. Thus the total number of droplets decreases steadily in time while average droplet size increases, until a balance is arrived between the recombination and creation of excitons, due to finite lifetime τ and continuous illumination of the system, respectively. The total number of droplets $N(t)$ can be easily obtained,

$$N(t) = \int_0^\infty f(R,t)dR = \frac{N(0)\overline{R}^D(0)}{\overline{R}^D(t)} , \tag{28}$$

which is shown in Fig. 4.

Finally, the nucleation rate is obtained as

$$J(R,t) = \frac{f(R,t)}{\tau} \propto \frac{g[R/\overline{R}(t)]}{\tau\overline{R}^{D+1}(t)} , \tag{29}$$

and its late time behavior is given by

$$\lim_{t\to\infty} J(R,t) \propto \frac{g(R/[DK\tau/3]^{1/3})}{\tau(DK\tau/3)^{(D+1)/3}} . \tag{30}$$

We have seen that due to the finite lifetime of the individual excitons, all statistical quantities of interest saturate at late times. While each droplet evolves in time, no droplet can grow to form a single macroscopic phase. This is entirely different from that of the classical theory of nucleation and growth.

IV. CONCLUSIONS

In conclusion, we have extended our theory of classical Ostwald ripening to describe droplet growth in exciton systems, in both two and three dimensions with finite volume (area) fraction. There are two basic assumptions for the model: one is that the rate for droplets to lose particle is proportional to the volume of the droplets, inversely proportional to the mean lifetime of the excitons, i.e., the term $-vR^D/\tau$ in the growth law equation (13); the other is that the system approaches a steady state at $t \to \infty$, and this is possible when the system is continuously illuminated by an external radiation such that nucleation of new droplets occurs even at late times, which compensates the loss of excitons due to recombination, i.e., the term $J(R,t) = f(R,t)/\tau$ in the continuity equation (21). These assumptions are physically motivated and allow us to solve the problem in closed form. In fact, as we have shown in previous sections, the nucleation current $J(R,t)$ is actually determined self-consistently by imposing the steady-state condition at late times, i.e., the form of $J(R,t)$ makes Eqs. (5), (12), (21), and (22) consistent with each other.

Our model predicts that both the distribution function and the nucleation rate have scaling forms as shown in Eqs. (23) and (29). All statistical quantities of interest saturate at late times when the steady state is reached. In particular, the average radius $\overline{R}(t) = (DK\tau/3)^{1/3}$ and the total number $N(t) \approx V'\phi/(DK\tau/3)^{D/3}$ at late times. These results are obtained in closed form. In principle, these quantities can be obtained experimentally, thereby testing our theoretical predictions.

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