Generalized positive *P* representation with third-order noise in intracavity coherent photoassociation of Bose-Einstein condensates

M. K. Olsen,[1,2] L. I. Plimak,[1,3] and M. J. Collett[1]
[1]*Department of Physics, University of Auckland, Private Bag 92019, Auckland, New Zealand*
[2]*Insituto de Física, Universidade Federal Fluminense, Boa Viagem 24210-340, Rio de Janeiro, Brazil*
[3]*Fachbereich Physik, Universität Kaiserslautern, D-67663 Kaiserslautern, Germany*
(Received 3 April 2001; revised manuscript received 19 July 2001; published 6 November 2001)

We investigate the process of intracavity photoassociation of Bose-Einstein-condensed atoms to form a molecular condensate. As shown previously, this process can only be successfully described by a quantum treatment of all the interacting fields. We extend our previous work by representing the full quantum aspects of the problem using an extension of the positive *P* representation to model non-Wiener noises. This allows the mapping of a generalized Fokker-Planck equation with third-order derivatives onto a set of coupled stochastic difference equations. We also investigate parameter regimes not covered previously, as well as the effects of spontaneous dissociation of the condensed molecules.

DOI: 10.1103/PhysRevA.64.063601 PACS number(s): 03.75.Fi, 32.80.−t, 42.50.Ct, 42.65.−k

I. INTRODUCTION

There are now a number of dynamical situations that have been investigated for dilute gas Bose-Einstein condensates (BEC) that are described incorrectly in a mean-field approach using Gross-Pitaevskii (GPE)-type equations [1]. Although it is obvious by definition that a GPE-type approach can predict little about the quantum statistics of a process, in these situations it does not even make successful predictions for the mean-field behavior. These begin with the evaporative cooling by which a BEC is produced [2] and include molecular association of an atomic BEC using both laser light [3,4] and Feshbach resonances [5], as well as the photodissociation of a molecular condensate [6].

What all these works have in common is that a full quantum treatment of all the interacting fields is still not necessary. For example, in evaporative cooling, the radio frequency scalpel that is used to remove the hotter atoms need not be treated quantum mechanically to obtain good results. In the treatments of photoassociation, Góral *et al.* [3] use a multimode approach but in a semiclassical, linearized way. Hope and Olsen [4] treat the laser field classically while Holland *et al.* [5] treat the molecules classically. In the work on photodissociation, a process that will not begin in a mean-field description, Poulsen and Mølmer [6] treat the molecular field as infinite, analogously to the undepleted pump approximation of quantum optics. This approximation necessarily means that any results are at best valid for short times only.

In a previous paper, we have shown that there is a simple dynamical process for which a quantum treatment of all the interacting fields is necessary; namely, the intracavity coherent photoassociation of an atomic condensate to form a molecular condensate [7]. For this system, we have demonstrated that there are parameter regimes in which the quantum solutions, obtained using positive *P* representation equations truncated at second order, give qualitatively different results to the semiclassical mean-field equations. In this paper, we extend our analysis in two ways. We map the full problem, without truncation, onto stochastic difference equations, and we include a phenomenological treatment of spontaneous dissociation of the molecules.

The equation of motion for the pseudoprobability distribution of the system in question is not of the standard Fokker-Planck form, as it contains third-order derivatives. Although formal methods are known for dealing with these [8], they are not easy to use except in some special cases [9,10]. An approximation that is commonly made, especially in the Wigner representation, is to truncate the equation at second order; this is exactly how we proceeded in [7]. This approximation has been shown to be accurate for the dynamics and quadrature variances of second-harmonic generation (SHG) [11] and for calculating first-order correlation functions in trapped BEC [12], although it is not accurate for the calculation of higher-order correlations in traveling-wave SHG [13] and may give misleading results for the optical parametric oscillator [14–16]. This truncation is usually justified by claiming that the coefficients of the third-order terms are smaller than the other coefficients in the equation, which is certainly the case in our present paper. However, unless we know the exact solutions, obtained by including the third-order terms, this remains an uncontrolled approximation. In this paper, we extend the positive *P* representation to include third-order noises, using methods described elsewhere [17–20]. This allows us to examine the validity of our previous truncation.

II. THE SYSTEM

The system we consider is with a trapped atomic condensate held in an electromagnetic cavity (see Fig. 1). Our for-

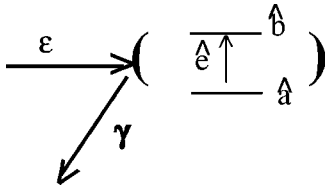

FIG. 1. Schematic of the condensate, represented by the operators $\hat{a}$ and $\hat{b}$, inside the electromagnetic cavity with field operator $\hat{e}$. The classical cavity pumping is represented by ϵ and the cavity loss rate is represented by γ.

malism is applicable to both microwave and optical transitions. The empty cavity is resonant at the frequency of the transition between atomic and molecular states of the condensate. Here, we make the approximation that all three fields may be represented as single modes, which is reasonable as long as we are considering short interaction times where the kinetic energy may be ignored. This approximation has previously been shown to be valid in an analysis of molecular formation using Bose-stimulated Raman adiabatic passage [21], where a single-mode approach was found to capture the relevant physics over short-time scales. As we have also found this approach to be qualitatively accurate for traveling-wave superchemistry over short-time scales, and the most interesting physics happens over the first few cavity lifetimes, we feel that this approximation is justified here. We also ignore the vibrational and rotational levels of the molecular state, as the energy spacing between these is more than the laser linewidth. We also make the normal zero-temperature approximation of quantum optics, as condensates exist at temperatures of the order of nanokelvins.

The interaction Hamiltonian for this system in the rotating wave approximation is

$$\mathcal{H}=\frac{i\hbar g}{2}[\hat{a}^{\dagger 2}\hat{b}\hat{e}^{\dagger}-\hat{a}^{2}\hat{b}^{\dagger}\hat{e}]+\hbar\chi_a\hat{a}^{\dagger 2}\hat{a}^{2}+\hbar\chi_b\hat{b}^{\dagger 2}\hat{b}^{2}+i\hbar(\epsilon\hat{e}^{\dagger}-\epsilon^{*}\hat{e})+\Gamma^{\dagger}\hat{e}+\Gamma\hat{e}^{\dagger}, \tag{1}$$

where g represents the effective coupling strength between the condensates and the electromagnetic field, $\hat{a}$ ($\hat{b}$) is the annihilation operator for the atomic (molecular) condensate and $\hat{e}$ is the annihilation operator for the intracavity electromagnetic field. The χ_j represent the self-interaction terms between the atoms or molecules, ϵ represents the classical pumping of the cavity, and Γ is a bath operator for the electromagnetic field.

III. GENERALIZED POSITIVE *P* EQUATIONS

Following the standard methods [22], we find a partial differential equation for the P distribution of this system,

$$\begin{aligned}\frac{\partial P}{\partial t}=&\left\{\frac{\partial}{\partial\alpha}[-ge^{*}\alpha^{*}\beta+2i\chi_a\alpha^{*}\alpha^{2}]+\frac{\partial}{\partial\alpha^{*}}[-ge\alpha\beta^{*}-2i\chi_a\alpha^{*2}\alpha]+\frac{\partial}{\partial\beta}\left[\frac{g}{2}\alpha^{2}e+2i\chi_b\beta^{*}\beta^{2}\right]\right.\\&+\frac{\partial}{\partial\beta^{*}}\left[\frac{g}{2}\alpha^{*2}e^{*}-2i\chi_b\beta^{*2}\beta\right]+\frac{\partial}{\partial e}\left[-\frac{g}{2}\alpha^{*2}\beta+\gamma e-\epsilon\right]+\frac{\partial}{\partial e^{*}}\left[-\frac{g}{2}\alpha^{2}\beta^{*}+\gamma e^{*}-\epsilon^{*}\right]\\&+\frac{1}{2}\left[\frac{\partial^{2}}{\partial\alpha^{2}}(g\beta e^{*}-2i\chi_a\alpha^{2})+\frac{\partial^{2}}{\partial\alpha^{*2}}(g\beta^{*}e+2i\chi_a\alpha^{*2})+\frac{\partial^{2}}{\partial\beta^{2}}(-2i\chi_b\beta^{2})+\frac{\partial^{2}}{\partial\beta^{*2}}(2i\chi_b\beta^{*2})\right.\\&\left.\left.+\frac{\partial^{2}}{\partial\alpha\partial e}(2g\alpha^{*}\beta)+\frac{\partial^{2}}{\partial\alpha^{*}\partial e^{*}}(2g\alpha\beta^{*})\right]-\frac{1}{6}\left[\frac{\partial^{3}}{\partial\alpha^{2}\partial e}(3g\beta)+\frac{\partial^{3}}{\partial\alpha^{*2}\partial e^{*}}(3g\beta^{*})\right]\right\}P(\alpha,\beta,e,t),\end{aligned} \tag{2}$$

where γ represents the loss rate of the optical field from the cavity.

As this equation contains mixed third-order derivatives, it is not amenable to standard phase-space techniques [22], nor to the methods used in Refs. [9,10]. An alternative approach based on techniques of the quantum field theory was introduced in Refs. [17–20]. Following these methods, we may map Eq. (2) onto the following set of coupled stochastic *difference* equations in an extended positive P representation, [with $\alpha(t+\Delta t)=\alpha(t)+\Delta\alpha(t)$, and so on for the other variables]

$$\Delta\alpha=\left[-2i\chi_a\alpha^{\dagger}\alpha^{2}+ge^{\dagger}\alpha^{\dagger}\beta+\frac{\sqrt{g}}{2}\left(e^{\dagger}+\frac{\xi_1+i\xi_2}{\sqrt{2}u}\right)\times(\eta_1+i\eta_3)+\frac{\sqrt{g}}{2}\beta(\eta_1-i\eta_3)+\sqrt{-2i\chi_a\alpha^{2}}\eta_5\right]\Delta t,$$

$$\Delta\alpha^{\dagger}=\left[2i\chi_a\alpha^{\dagger 2}\alpha+ge\alpha\beta^{\dagger}+\frac{\sqrt{g}}{2}\left(e+\frac{\xi_3-i\xi_4}{\sqrt{2}u}\right)(\eta_2-i\eta_4)+\frac{\sqrt{g}}{2}\beta^{\dagger}(\eta_2+i\eta_4)+\sqrt{2i\chi_a\alpha^{\dagger 2}}\eta_6\right]\Delta t,$$

$$\Delta\beta=\left[-2i\chi_b\beta^{2}\beta^{\dagger}-\frac{g}{2}\alpha^{2}e+\sqrt{-2i\chi_b\beta^{2}}\eta_7\right]\Delta t,$$

$$\Delta\beta^{\dagger}=\left[2i\chi_b\beta^{\dagger 2}\beta-\frac{g}{2}\alpha^{\dagger 2}e^{\dagger}+\sqrt{2i\chi_b\beta^{\dagger 2}}\eta_8\right]\Delta t,$$

$$\Delta e=\left[\epsilon-\gamma e+\frac{g}{2}\alpha^{\dagger 2}\beta+\sqrt{g}\alpha^{\dagger}(\eta_1+i\eta_3)+\frac{u}{\sqrt{2}}(\xi_1-i\xi_2)\right]\Delta t,$$

$$\Delta e^{\dagger}=\left[\epsilon^{*}-\gamma e^{\dagger}+\frac{g}{2}\alpha^{2}\beta^{\dagger}+\sqrt{g}\,\alpha(\eta_{2}-i\eta_{4})+\frac{u}{\sqrt{2}}(\xi_{3}+i\xi_{4})\right]\Delta t, \tag{3}$$

where u is a free parameter (see below). These equations imply an equal discretization of the time axis, with Δt being the step of the time grid. It should be noted here that there is no limit of these equations as stochastic differential equations [20], although this does not prevent their being solved numerically. In the above, all noise sources are real and have the properties

$$\overline{\eta_{i}(t)}=0,\quad \overline{\eta_{j}(t)\,\eta_{k}(t')}=\delta_{jk}\delta(t-t'),$$
$$\overline{\xi_{i}(t)}=0,\quad \overline{\xi_{j}(t)\xi_{k}(t')}=\delta_{jk}\delta(t-t'). \tag{4}$$

The δ function is to be understood in accordance with the time discretization, $\delta(t-t')=\delta_{tt'}/\Delta t$, where $\delta_{tt'}$ is the Kronecker symbol. As in the usual positive P representation, there is a correspondence between the c-number variables $[\alpha,\alpha^{\dagger},\beta,\beta^{\dagger},e,e^{\dagger}]$ and the operators $[\hat{a},\hat{a}^{\dagger},\hat{b},\hat{b}^{\dagger},\hat{e},\hat{e}^{\dagger}]$, although a variable such as $\alpha^{\dagger}$ is not complex conjugate to α (except in the mean over a large number of stochastic trajectories), due to the independence of the noise sources. We should note here that the above equations, although they would exhibit a formal similarity to those used to describe traveling-wave SHG with an additional $\chi^{(3)}$ nonlinearity [23] if the ξ_{j} were deleted, exhibit one important difference. Instead of a constant κ, the effective $\chi^{(2)}$ interaction used in [23], we now have the field dependent ge. Another difference in our present case would be that we now have a term $(g/2)\alpha^{\dagger\,2}\beta$ in the equation for the electromagnetic field.

We also note here that the above equations are by no means a unique mapping of the third-order equation for the pseudoprobability distribution. Although there may be many ways to factorize the diffusion matrix of a normal Fokker-Planck equation, there is an even larger degree of freedom in representing the third-order terms. Essentially, following the methods of Ref. [20], we find that the second-order noise terms proportional to η_{1} and η_{3} in the truncated equations used in Ref. [7] are replaced by

$$\Delta\alpha=[\cdots+p\,\eta+r\,\eta^{*}+\cdots]\Delta t,$$
$$\Delta e=[\cdots+q\,\eta+u\,\eta'^{*}+\cdots]\Delta t, \tag{5}$$

where η and η' are independent complex Gaussian noise sources with the properties

$$\overline{\eta(t)\,\eta(t')}=\overline{\eta'(t)\,\eta'(t')}=0,$$
$$\overline{\eta(t)\,\eta^{*}(t')}=\overline{\eta'(t)\,\eta'^{*}(t')}=\delta(t-t'). \tag{6}$$

This means that $\eta=(\eta_{1}+i\eta_{3})/\sqrt{2}$, cf. Ref. [7], while $\eta'=(\xi_{1}+i\xi_{2})/\sqrt{2}$ is needed for the third-order noises. Setting $\eta'=0$ is equivalent to truncation to second order as in Ref. [7]. The quantities p,r,q, and u obey the relations

$$qr=g\alpha^{\dagger}\beta,$$
$$2pr=g\beta(e^{\dagger}+\eta'/u). \tag{7}$$

At first glance, there seems to be no connection between Eq. (2) for the positive P distribution and Eqs. (5). Consider, however, the characteristic function of the increments, (with all powers of Δt shown explicitly)

$$\Phi(\zeta_{\alpha},\zeta_{e})=\overline{\exp\left(\Delta t\sum_{t}\{\zeta_{\alpha}(t)[p(t)\,\eta(t)+r(t)\,\eta^{*}(t)]+\zeta_{e}(t)[q(t)\,\eta(t)+u(t)\,\eta'^{*}(t)]\}\right)}, \tag{8}$$

where the averaging is over the statistics of the η noises,

$$\overline{[\cdots]}=\prod_{t}\left[\int\frac{d^{2}\eta'(t)e^{-\Delta t|\eta'(t)|^{2}}\Delta t}{\pi}\times\int\frac{d^{2}\eta(t)e^{-\Delta t|\eta(t)|^{2}}\Delta t}{\pi}\right][\cdots]. \tag{9}$$

Since p,q,r, and u do not depend on η, in each time slice in Eq. (8), we may take a simple Gaussian integral over $\eta(t)$. Using Eq. (7) to simplify the result, we recover a Gaussian integral over $\eta'(t)$ per time slice. On taking these integrals and once more using Eq. (7), we arrive at

$$\Phi(\zeta_{\alpha},\zeta_{e})=\exp\left[\Delta t\sum_{t}\left(\frac{\zeta_{\alpha}^{2}}{2}g\beta e^{\dagger}+\zeta_{\alpha}\zeta_{e}g\beta\alpha^{\dagger}+\frac{\zeta_{\alpha}^{2}}{2}\zeta_{e}g\beta\right)\right]. \tag{10}$$

It is now evident that there is a one-to-one correspondence between terms with n derivatives in Eq. (2) and nth order *cumulants* of the increments in Eqs. (5), $n=2,3$. With the exception of Gaussian statistics, there do not exist random variables with finite sets of nonzero cumulants. Luckily, to match Eqs. (2) and (5), we only need cumulants of the increments themselves. Cumulants mixing increments with their complex conjugates are inessential for this matching, and it is easily verified that the set of these cumulants is indeed infinite. We should note here that to specify a finite subset of nonzero cumulants would require a doubling of the

phase space if we were working with a space of normal dimensions such as in the Wigner representation, for example [19]. As we are using the positive-P representation here, we are already in a doubled phase space.

The freedom of choice of the inessential cumulants manifests itself as freedom of choice of the parameters p,q,r, and u. Equations (3) correspond to (with u remaining a free parameter)

$$p=\sqrt{g/2}(e^{\dagger}+\eta'/u), \tag{11}$$

$$r=\beta\sqrt{g/2}, \tag{12}$$

$$q=\alpha^{\dagger}\sqrt{2g}, \tag{13}$$

but there are certainly many other possible choices. For the conjugated equations, everything is conjugated in the positive P representation sense.

In our simulations, the u's were chosen as $u=u^{\dagger}=1/\Delta t^{1/4}$. The powers of Δt are then evenly spread over the third-order contributions in Eqs. (3), which all scale as $\Delta t^{1/4}$. By also rescaling r, we could reduce this to $\Delta t^{1/3}$, which is the best one can do, but we preferred to preserve the conventional scaling of the second-order noises. The scaling of the third-order noises is the formal reason why Eqs. (3) have no limit as stochastic differential equations as $\Delta t\rightarrow 0$ [20]. However, the absence of a continuous limit is not a problem in practice. As these phase-space methods are generally only used when analytical methods are difficult, the equations will almost always be solved numerically by computer. In this case, Δt always remains nonzero and there is no problem (except for the usual ones when integrating positive P equations).

IV. RESULTS

We have solved Eq. (3) numerically for a range of parameters and found behavior of the mean fields that is strikingly different from that found in the usual mean-field approximation, as well as regimes where the mean-field approach is valid. Unlike many situations in quantum optics or in the study of condensates, the stochasticity of the problem may be important even when we do not wish to consider quantum statistical properties.

In our simulations, we begin with an atomic condensate inside an optical cavity that begins to be pumped at $t=0$. Initially, neither molecules nor electromagnetic field are present, with the atomic field being treated as initially in a coherent state. We present the results here of numerical investigations of two different regimes. In what we may consider the strong-interaction regime, the dynamics always exhibits short-time oscillations and photon blockade. In the weak-interaction regime, which may be reached by decreasing the strength of g or the number of atoms, the solutions approach those found by treating all fields semiclassically. The solutions for atom and molecule number are then reminiscent of those found in superchemistry [24] for traveling-wave photoassociation or in traveling-wave second-harmonic generation [11].

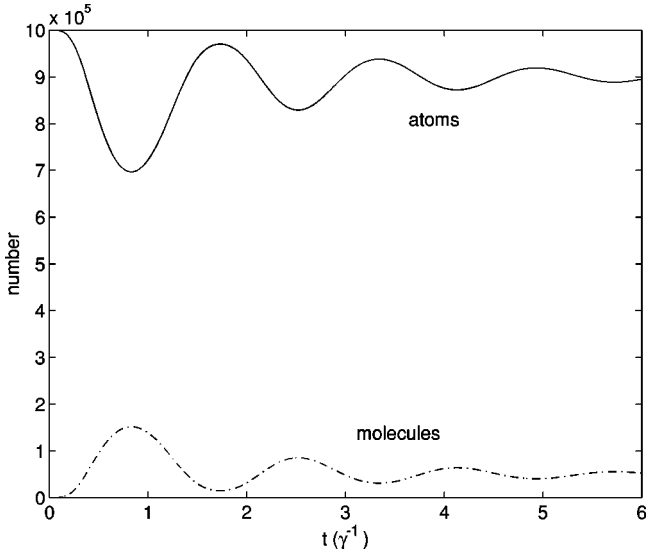

FIG. 2. Occupation numbers of the atomic and molecular condensates as a function of time according to 3×10^5 quantum trajectories. The parameters are $g=10^{-5}$, $|\epsilon|^2=10^6$, $\chi_{a,b}=10^{-9}$, and $|\alpha(0)|^2=10^6$. All quantities plotted in this and the following graphs are dimensionsless.

In Fig. 2, we show the time development of the atomic and molecular fields as the cavity pumping is turned on, for the parameters $g=10^{-5}$,$|\epsilon|^2=10^6$,$\chi_{a,b}=10^{-9}$, and $|\alpha(0)|^2=10^6$, which are all scaled in terms of the cavity loss rate. We have taken the means over 3×10^5 stochastic trajectories, which was more than sufficient to ensure excellent convergence. We can obtain some insight into the behavior exhibited here when we examine the dynamics of the intracavity electromagnetic field, as shown in Ref. [7]. We find an initial build up of intensity in the cavity, with this field also becoming oscillatory and eventually almost vanishing completely. As the cavity continues to be pumped at the same rate, what we see is that it has become opaque. That is, a photon blockade effect is operating [25,26], as seen previously in systems that develop an effective giant $\chi^{(3)}$ nonlinearity. The mean behavior of these three fields is identical to that found in Ref. [7].

For comparison, we showed the solutions of Eq. (3) with the noise terms removed in Ref. [7]. The disagreement between quantum and semiclassical solutions is even more striking than that previously found for pure traveling-wave SHG [11]. One way of explaining the photon blockade effect is by considering that the interaction detunes the cavity. The linearized equation for the electromagnetic field contains a term $(g/2)\alpha^{\dagger\,2}\beta$, which will have some imaginary component due to the self-interaction terms of the atomic and molecular fields. However, this term by itself cannot cause the blockade. What is needed is noise. In this respect, it is interesting to note that solution of the truncated Wigner equations for this system gives the same results as the positive P solutions. This indicates that the noise required need not be deeply quantum, as the truncated Wigner is equivalent to the semiclassical theory of stochastic electrodynamics [27].

In what we call the weak-interaction regime, obtained via either a weaker-coupling constant or a smaller number of

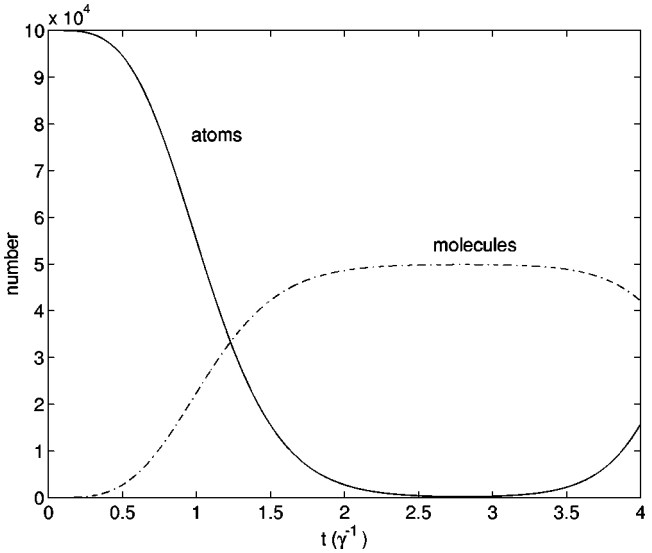

FIG. 3. Occupation numbers of the atomic and molecular condensates calculated quantum mechanically in the weak-interaction regime, with parameters as in Fig. 2, but with $|\alpha(0)|^2=10^5$. The number of trajectories was 2×10^4.

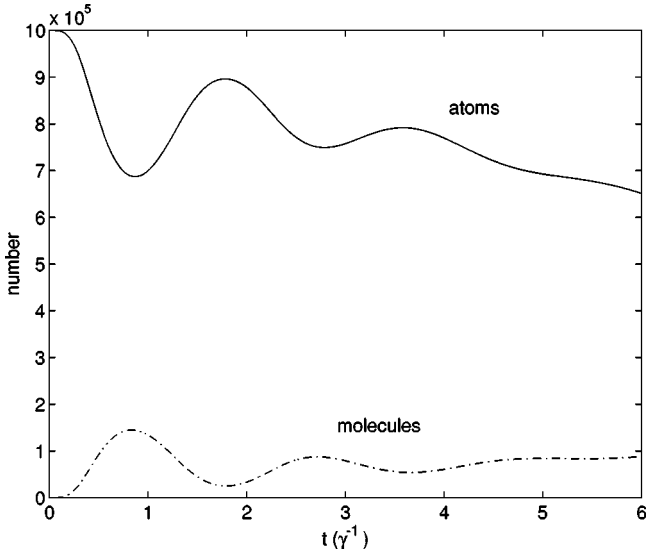

FIG. 4. Occupation numbers of the atomic and molecular condensates calculated quantum mechanically in the strong-interaction regime, with parameters as in Fig. 2, but with $\gamma_b=0.1\gamma$, for 10^4 trajectories.

condensed atoms, the quantum solutions and the semiclassical solutions become indistinguishable. The atomic and molecular numbers undergo giant oscillations reminiscent of traveling-wave second-harmonic generation [11] or semiclassical BEC superchemistry [24] and the optical field attains a steady value as shown in Ref. [7]. The dynamics of the atomic and molecular condensates are as in Fig. 3, beginning with 10^5 condensed atoms. All other parameters are as in the strong-interaction regime. This is consistent with our describing the blockade effect as being due to the noise terms, as the second-order noise in the equations for the electromagnetic field is proportional to the amplitude of the atomic field, as can be seen from Eq. (3).

V. SPONTANEOUS MOLECULAR DISSOCIATION

One of the approximations made in the previous work [7], and so far in this paper, is that we are considering times over which there will not be significant spontaneous dissociation of the excited molecules. Hence, we have so far ignored this factor in the dynamics. We will now relax this assumption by adding a phenomenological Born-Markovian damping term to the Hamiltonian for the molecular field,

$$\mathcal{H}_{spon}=\Gamma_b^\dagger\hat{b}+\Gamma_b\hat{b}^\dagger. \tag{14}$$

In the normal manner, this results in loss terms $-\gamma_b\beta$ and $-\gamma_b\beta^\dagger$ being added to the equations for β and $\beta^\dagger$. In many situations, dissipation will act to destroy quantum effects, so it is also of interest to see if the discrepancies between the full and semiclassical solutions remain so pronounced with molecular losses included.

In Fig. 4, we show the positive *P* representation result for parameters as in the strong-interaction regime of Fig. 2, but with molecular loss rate $\gamma_b=0.1\gamma$. As optical cavity loss rates are generally of the order of megahertz, this is a physically reasonable choice of spontaneous dissociation rate. What we see is that the behavior has changed, with the atom number undergoing an oscillatory decrease, while the molecule number undergoes an oscillatory increase. The intracavity light field starts to revive as the atom number goes down, with the whole dynamics eventually becoming closer to that of the weak-interaction regime. This is to be expected, as we have less interacting matter inside the cavity as the molecules are damped. However, to develop this picture any further would begin to exceed the limits of our single-mode-type approach. What is interesting is that we can see, by comparing Fig. 4 with Fig. 5, that the semiclassical predictions are still qualitatively wrong. The semiclassical light field again rises monotonically to its steady state value, so

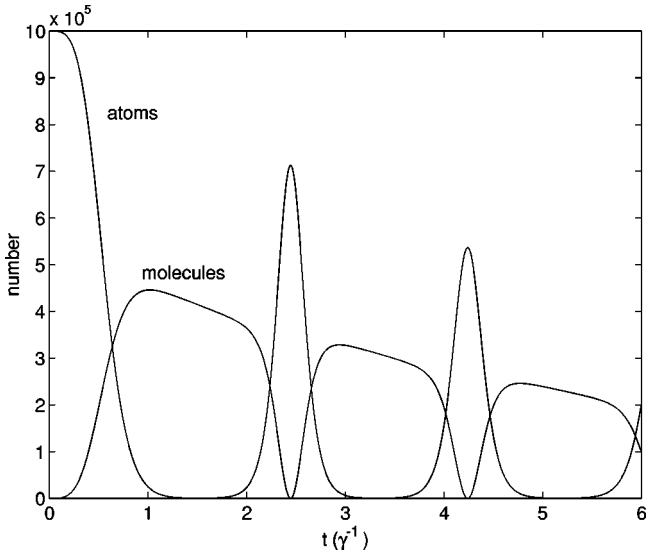

FIG. 5. Occupation numbers of the atomic and molecular condensates as for Fig. 4, but calculated semiclassically.

that the differences should again be easily experimentally detectable.

VI. THIRD-ORDER EFFECTS AND QUANTUM STATISTICS

An interesting question is, what effect does the third-order noise have on this system? The short answer is that we were not able to detect any difference between the full and the truncated calculations for the mean fields, the variances of the intensities, the first- and second-order coherence functions for the three fields and even cumulants up to sixth order [8] in the field amplitudes. Interestingly enough, a Wigner representation truncated to second order also gives essentially the same results for all these quantities.

Upon examination of Eq. (3) we see that the third-order terms (represented by the ξ_j) are needed for spontaneous breakup of a molecule into two condensed atoms and one photon. We therefore investigated the sixth-order field cumulant,

$$\begin{aligned}\langle\langle \hat{a}^{\dagger 2}\hat{a}^2\hat{e}^\dagger\hat{e}\rangle\rangle = &\langle \hat{a}^{\dagger 2}\hat{a}^2\hat{e}^\dagger\hat{e}\rangle - 12\langle \hat{a}^\dagger\hat{a}\rangle^2\langle \hat{e}^\dagger\hat{e}\rangle + 8\langle \hat{a}^\dagger\hat{a}\rangle(\langle \hat{e}\rangle \\ &\times\langle \hat{a}^\dagger\hat{a}\hat{e}^\dagger\rangle + \langle \hat{e}^\dagger\rangle\langle \hat{a}^\dagger\hat{a}\hat{e}\rangle) + 4(\langle \hat{e}^\dagger\hat{e}\rangle\langle \hat{a}^\dagger\hat{a}\rangle^2 \\ &-\langle \hat{a}^\dagger\hat{a}\hat{c}^\dagger\rangle\langle \hat{a}^\dagger\hat{a}\hat{c}\rangle - \langle \hat{a}^\dagger\hat{a}\rangle\langle \hat{a}^\dagger\hat{a}\hat{e}^\dagger\hat{e}\rangle) \\ &+2\langle \hat{a}^{\dagger 2}\hat{a}^2\rangle\langle \hat{e}^\dagger\rangle\langle \hat{e}\rangle - \langle \hat{a}^{\dagger 2}\hat{a}^2\rangle\langle \hat{e}^\dagger\hat{e}\rangle \\ &-\langle \hat{a}^{\dagger 2}\hat{a}^2\hat{e}\rangle\langle \hat{e}^\dagger\rangle - \langle \hat{a}^{\dagger 2}\hat{a}^2\hat{e}^\dagger\rangle\langle \hat{e}\rangle, \end{aligned} \tag{15}$$

where the terms that vanish for our system are not explicitly included.

What we found was that, within the parameter regime where the physical approximations we have made retain some validity, there is no observable difference in the expectation value of this cumulant, whether calculated with or without third-order noises. Where there is a difference is when we turn off the pump and begin with only condensed molecules present. In this case, there is an effect present, but it does not manifest itself until at least twenty cavity lifetimes have passed, which is well beyond the region where our single-mode-type approach can be considered reliable.

Examining the quantum statistics may, however, give some insight into the behavior of the mean fields. In the strong-interaction regime, we find almost no suppression of quantum noise when we consider the quadrature and number variances of the three fields. In fact, the three fields almost always exhibit excess noise, which supports our claim that the detuning effect is noise driven. As can be seen in Fig. 6, which shows the time development of the X_a quadrature variances for the atomic field with and without spontaneous molecular dissociation, there is a very small amount of noise supression at some times. The molecular dissociation acts to destroy even this small amount. Over the time scale shown, the normalized intensity variance, or Fano factor, of the atomic field is identical to the quadrature variance. This is the quadrature of least noise, with all the other quantum correlations we investigated in this regime being well above the standard quantum limit.

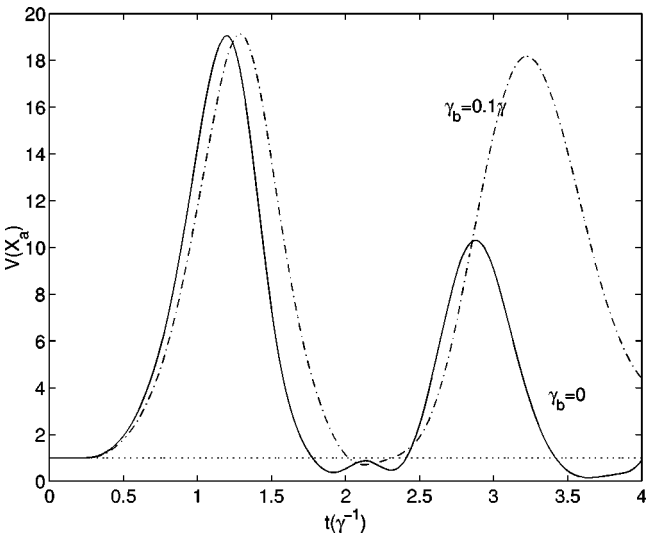

FIG. 6. The X quadrature variances of the atomic field in the strong-interaction regime, with and without molecular loss rate of $\gamma_b = 0.1\gamma$. These results and those of Fig. 7 were calculated using 5×10^5 trajectories. The normalized number variance for this field is identical on this scale. The dashed line represents the standard quantum limit.

On the other hand, in Fig. 7 we show the atomic intensity and X_a quadrature variances in the weak-interaction regime. These are very reminiscent of the variances predicted in traveling-wave SHG with an added χ^3 nonlinearity [23,28]. In that case, the mean intensities were also described well by solution of the mean-field equations. We see that the atomic field develops excess noise as it begins to grow following an almost complete conversion to molecules, due to the semi-spontaneous nature of this process. The molecular field also exhibits statistics reminiscent of traveling-wave SHG. Interestingly enough, it is in this regime that the stochastic inte-

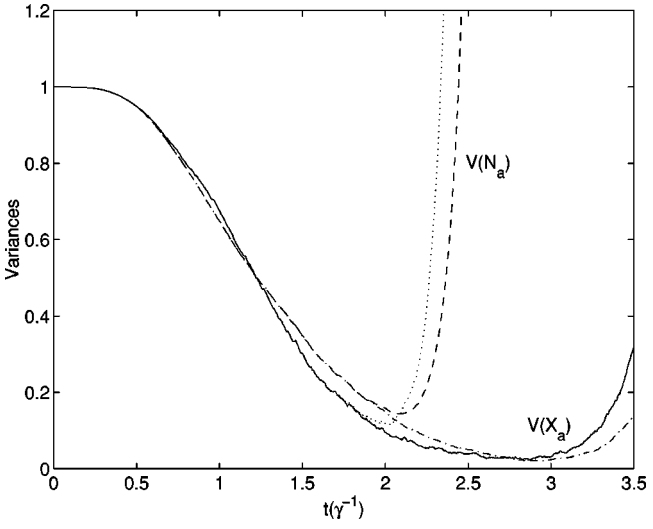

FIG. 7. The atomic field quadrature and number variances in the weak-interaction regime, with and without molecular losses. The continuous and the dotted lines are $V(X_a)$ and $V(N_a)$, respectively, without molecular losses. The dashed line and the dash-dotted line are the same variances with molecular loss rate $\gamma_b = 0.1\gamma$.

gration was least stable, diverging shortly after the maximum time shown in Fig. 3. The modeling of the third-order terms did not make this much worse. In all results of stochastic integration that we show here, the sampling errors were so small over the ensembles that error bars would be barely visible. In the weak-interaction regime, when the divergence appeared, it appeared very rapidly, hinting at a possible exponential divergence in phase space as found with integration of the one-dimensional BEC equations of Ref. [12]. As is common with the positive *P* representation, the addition of the damping term acted to stabilize the numerics, allowing integration over a time period approximately 15% greater before divergences occurred. The only quantum statistical properties that we were not able to calculate accurately were the variances of the electromagnetic field in the strong interaction regime. Even after 10^6 trajectories, these were still so noisy that it was difficult to predict quantative properties. What we can say is that they did not exhibit noise suppression.

VII. CONCLUSION

We have described and analyzed a situation in which the Gross-Pitaevski approach does not describe adequately the dynamics of a Bose-Einstein condensate. The differences are not of the order of the inverse of the system size, but are qualitative. We have extended the results of Ref. [7] into different parameter regimes, examining the validity of some of the approximations made in that work. We have seen from numerical investigations that the quantum solutions become closer to the semiclassical solutions as the number of atoms or the coupling decrease. This is a sign of the nonlinearity of the quantum dynamics, where noise-driven correlations are built up between the three fields in a manner that has no semiclassical mean-field description.

We have also shown how the third-order terms that had been dropped from the equations of motion of the earlier work can be modeled. As these terms were found to have no noticeable effect within the limits of our model, the truncation used in the previous work has been shown to be justified, hence, removing a previously uncontrolled approximation. In regimes where the quantum and semiclassical predictions were earlier shown to be different, we have demonstrated that the inclusion of spontaneous molecular dissociation still leaves the quantum solutions qualitatively different. The experimental signature of this difference can be measured in a very simple manner; with photodetection of the light emitted by the optical cavity.

ACKNOWLEDGMENTS

This research was supported by the Marsden Fund of the Royal Society of New Zealand, the Foundation for Research, Science and Technology of New Zealand (UFRJ0001), and the Deutsche Forschungsgemeinschaft under Contract No. FL210/11.

[1] F. Dalfovo, S. Giorgini, L.P. Pitaevskii, and S. Stringari, Rev. Mod. Phys. **71**, 463 (1999).
[2] P.D. Drummond and J.F. Corney, Phys. Rev. A **60**, R2661 (1999).
[3] K. Góral, M. Gajda, and K. Rzążewski, Phys. Rev. Lett. **86**, 1397 (2001).
[4] J.J. Hope and M.K. Olsen, Phys. Rev. Lett. **86**, 3220 (2001).
[5] M. Holland, J. Park, and R. Walser, Phys. Rev. Lett. **86**, 1915 (2001).
[6] U.V. Poulsen and K. Mølmer, Phys. Rev. A **63**, 023604 (2001).
[7] M.K. Olsen, J.J. Hope, and L.I. Plimak, Phys. Rev. A **64**, 013601 (2001).
[8] C. W. Gardiner, *Handbook of Stochastic Methods* (Springer-Verlag, Berlin, 1983).
[9] S.T. Gevorkyan, G.Yu. Kryuchkyan, and K.V. Kheruntsyan, Opt. Commun. **134**, 440 (1997).
[10] S.T. Gevorkyan, Phys. Rev. A **58**, 4862 (1998).
[11] M.K. Olsen, R.J. Horowicz, L.I. Plimak, N. Treps, and C. Fabre, Phys. Rev. A **61**, 021803 (2000).
[12] M.J. Steel, M.K. Olsen, L.I. Plimak, P.D. Drummond, S.M. Tan, M.J. Collett, D.F. Walls, and R. Graham, Phys. Rev. A **58**, 4824 (1998).
[13] M.K. Olsen, L.I. Plimak, M.J. Collett, and D.F. Walls, Phys. Rev. A **62**, 023802 (2000); M.K. Olsen, K. Dechoum, and L.I. Plimak, Opt. Commun. **190**, 261 (2001).
[14] P.D. Drummond and P. Kinsler, Phys. Rev. A **40**, 4813 (1989).
[15] P. Kinsler and P.D. Drummond, Phys. Rev. A **43**, 6194 (1991).
[16] P. Kinsler and P.D. Drummond, Phys. Rev. A **44**, 7848 (1991).
[17] L. I. Plimak, M. J. Collett, D. F. Walls, and M. Fleischhauer, in *Proceedings of the Sixth International Conference on Path Integrals from peV to TeV* (World Scientific, Singapore, 1998), p. 241.
[18] L.I. Plimak, M. Fleischhauer, and D.F. Walls, Europhys. Lett. **43**, 641 (1998).
[19] L. I. Plimak, M. K. Olsen, M. Fleischhauer, and M. J. Collett, Europhys. Lett. (to be published).
[20] L.I. Plimak, M. Fleischhauer, M.K. Olsen, and M.J. Collett, e-print cond-mat/0102483.
[21] J.J. Hope, M.K. Olsen, and L.I. Plimak, Phys. Rev. A **63**, 043603 (2001).
[22] C. W. Gardiner, *Quantum Noise* (Springer-Verlag, Berlin, 1991).
[23] M.K. Olsen, V.I. Kruglov, and M.J. Collett, Phys. Rev. A **63**, 033801 (2001).
[24] D.J. Heinzen, R. Wynar, P.D. Drummond, and K.V. Kheruntsyan, Phys. Rev. Lett. **84**, 5029 (2000).
[25] S. Rebić, S.M. Tan, A.S. Parkins, and D.F. Walls, J. Opt. B: Quantum Semiclassical Opt. **1**, 490 (1999).
[26] M.J. Werner and A. Imamoğlu, Phys. Rev. A **61**, 011801 (1999).
[27] T.W. Marshall, Proc. R. Soc. London, Ser. A **276**, 473 (1963).
[28] V. I. Kruglov and M. K. Olsen, Phys. Rev. A **64**, 053802 (2001).