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<th>Wave mixing of hybrid Bogoliubov modes in a Bose-Einstein condensate</th>
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Mode-mixing of coherent excitations of a trapped Bose-Einstein condensate is modeled using the Bogoliubov approximation. Calculations are presented for second-harmonic generation between the two lowest-lying even-parity $m=0$ modes in an oblate spheroidal trap. Hybridization of the modes of the breather ($l=0$) and surface ($l=4$) states leads to the formation of a Bogoliubov dark state near phase-matching resonance so that a single mode is coherently populated. Efficient harmonic generation requires a strong coupling rate, sharply-defined and well-separated frequency spectrum, and good phase matching. We find that in all three respects the quantal results are significantly different from hydrodynamic predictions. Typically the second-harmonic conversion rate is half that given by an equivalent hydrodynamic estimate.

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I. INTRODUCTION

Observations of coherence of matter wave fields such as four-wave mixing [1], squeezing [2], and harmonic generation [3], in atomic Bose-Einstein condensates have recently been reported. The well-defined phase of the condensate means that small amplitude quasiparticle excitations can be produced that combine, through elastic coherent (phase preserving) atomic collisions, to produce frequency-mixed modes. We analyze, within the Bogoliubov model [4], second-harmonic generation of excitations in trapped spheroidal atomic condensates: a process recently observed in experiment [3]. Efficient harmonic generation is predicated on three elements; a strong coupling rate, a sharply defined and well separated frequency spectrum, and good phase matching. We find that in all three respects the quantal results are significantly different from hydrodynamic predictions [5]. The principal reason is that hybridization of degenerate Bogoliubov states occurs leading to the creation of coherent-matter dark states. Typically, nonhydrodynamic frequency shifts and the hybridization process lead to conversion efficiencies roughly half that given by an equivalent hydrodynamic estimate and create a mode of mixed axial and radial symmetry. The presence of a dark Bogoliubov mode near the wave-mixing resonance means that a single isolated mode can be excited rather than the doublet and therefore better control of single-mode harmonic generation may be possible.

Consider a condensate of a large finite number of atoms $N$, each of mass $m$, trapped by a spheroidal potential. The angular frequency of the trap along the polar $z$-axis is $\omega_z$, and the corresponding radial frequency is $\omega_r$, with the trap aspect ratio $\lambda = \omega_z / \omega_r$. Then the trapping potential can be written: $V_{\text{trap}} = 1/2 m \omega_z^2 (r^2 + \lambda^2 z^2)$. In the limit of small amplitude excitations, the acoustic equation is separable and the axial, radial, and angular symmetries provide good quantum numbers [6,7]. Let the azimuthal angular momentum quantum number be denoted by $m$. The two lowest-frequency excitations of symmetry $m=0$, are a quadrupole mode, that will act as the pump with frequency $\omega_\perp$, and the breathing mode $\omega_\perp$. In the hydrodynamic limit [8] the frequencies are given by: $\omega_{1,2}^\perp = \sqrt{9\lambda^4 - 16 \lambda^2 + 16}$. The nonlinear interactions caused by collisions create mode mixing. This coupling is most strongly pronounced when the scattering amplitude is large and temporal phase-matching occurs, i.e., when second-harmonic resonance arises: $\omega_{2} = 2 \omega_1$. The corresponding values of $\lambda$, the trap aspect ratio, are thus $\lambda = 1/6 \sqrt{77 \pm 5 \sqrt{145}} \approx 0.683$ and $1.952$ [8].

The effect was first observed in an oblate trap by Hechenblaikner and co-workers [3] for the following parameters: $N \approx 20000$ atoms of $^8$Rb, $T < 0.5T_c$, with $\omega_z = 2 \pi \times 126$ Hz. In measurements carried out within the range $1.6 < \lambda < 2.8$, strongly enhanced second-harmonic generation was found when $\lambda = 1.93 \pm 0.02$. The corresponding chemical potential of this condensate $\mu$, such that $\mu \gg \hbar \omega_z$, lies within the range where hydrodynamic theory should be valid. Indeed, both the frequency of the breathing mode $\omega_\perp$ [6] and the resonant aspect ratio $\lambda$, measured by experiment are in very good agreement with linear hydrodynamic theory [8]. Subsequently a hydrodynamic model of the nonlinear mixing of quasiparticles [4] was applied to estimate the rate of second-harmonic generation [9] and gave results consistent with experiment. However, this consistency hides potentially important quantal features not previously considered or observed. This paper examines the process of mode mixing and harmonic generation using a detailed quantal treatment of the process [4] taking into account atom number and quantum pressure corrections.

II. THEORY

The formalism for the wave-mixing processes in Bose condensed gases was developed by Morgan and co-workers.
At low temperatures $T/T_c \ll 0.5$ the dynamics of Bose condensed gases are dominated by single quasiparticle excitations. The atom-atom interactions are represented by an $s$-wave pseudopotential corresponding to a scattering length $a_s$. The spectrum and mode densities of collective excitations can be obtained from the Hartree variational principle

$$\delta \int dt \{ \psi^\dagger [H_0 + \frac{1}{2} g \psi^2 \psi - i \hbar \partial_t] \psi \} = 0,$$  

(1)

where $g = (4 \pi \hbar^2/ma_s)^2 N a_s$, $H_0 = - (\hbar^2/2m_o) \nabla^2 + V_{trap} - \mu$, and $\mu$ plays the role of a Lagrange multiplier implying that the number of atoms $N$ is approximately constant. The condensate and excited modes can be described by the linear response ansatz [4] where

$$\psi(r,t) = b_0(t) \phi(r) + \sum_{j>0} [b_j(t) u_j(r) e^{-i\omega_j t} + b_j^*(t) v_j^*(r) e^{+i\omega_j t}],$$  

(2)

and where $\phi$ represents the highly occupied ($N \gg 1$) condensate; that is, $|b_0| \approx 1$ while $b_j \ll 1$, $j > 0$. From the variation $\delta \phi^*$, and linear expansion in the small parameters $b_j, b_j^*$ taken as constant, the stationary Gross-Pitaevskii equation and Bogoliubov equations follow:

$$H_0 \phi + g|\phi|^2 \phi = 0,$$  

(3)

with $(\phi, \phi) = 1$. The Bogoliubov modes are solutions of the coupled linear equations

$$(H_0 + 2g|\phi|^2) u_j + g \phi^2 v_j = \hbar \omega_j u_j,$$  

(4)

$$H_0 + 2g|\phi|^2) v_j + g \phi^2 u_j = - \hbar \omega_j v_j.$$  

(5)

We take the conventional normalization of modes: $(u_j, u_j) = (v_j, v_j) = 1$. Although the spectrum $\omega_j$ is unique, the mode amplitudes may contain arbitrary components of the kernel, the $\omega = 0$ component. After diagonalization, it is convenient to enforce orthogonality [4]. Gram-Schmidt orthogonalization avoids the eigenmodes overlapping the condensate: $u_j \rightarrow u_j - (\phi, u_j) \phi$ and $v_j \rightarrow v_j + (u_j, \phi) \phi^*$. This guarantees gauge invariance, that is adding an arbitrary constant to the external potential will not mix condensate and excited modes, while preserving the orthogonality relations. Within the Bogoliubov approximation it is assumed that the condensate density and phase do not vary with time. In fact, density and phase fluctuations will arise from second-order mixing of the quasiparticle states [4] and thus only affect the wave-mixing process at the third and fourth order of perturbation theory. Such an approximation would not be valid for strongly perturbed condensates over long time scales [4,10]. Since the trapped condensate is at rest there is no phase gradient within the condensate and its wave function and those of the quasiparticles can be taken as real functions. Suppose that the condensate (mode 0) contains $N$ particles, and a single quasiparticle mode $i$ of excitation is weakly populated. Then mode-mixing collisions of the type, $i + i \rightarrow 0 + j$, and all crossing symmetries, will be significant if phases (energies) match and/or the scattering amplitude (vertex factor) is large.

Allowing for variation of the constants, $b_j$, of the trial function (3) in the variational principle (1), and neglecting transitions far from resonance, we get [4]

$$i \hbar \frac{db_j}{dt} = \sum_{j} g b_j b_j^* b_j e^{-\Delta_{ij}},$$  

(6)

$$i \hbar \frac{db_j}{dt} = \frac{1}{2} g^2 b_j b_j^* e^{\Delta_{ij}},$$  

(7)

where $\Delta_{ij} = \omega_j - 2 \omega_i$ is the detuning. The mode conversion equations are exactly analogous to those in classical nonlinear optics, allowing for two or more excited modes ($j$) to be populated from the pump mode ($i$). The coupling strength (scattering amplitude) for the process $M_{ij}$ is given by [4]

$$M_{ij} = g \int dr \{ \phi^* [2 u_j^* v_j^* u_i + v_j^* v_j^* v_j]$$  

$$+ \phi [2 u_i^* v_i^* v_i + u_i^* v_i^* u_i] \}$$  

and for a condensate with uniform phase throughout, all modes can be written in terms of real functions. In the derivation of Eqs. (6) and (7) a fixed condensate population is assumed consistent with the weak-coupling regime where the quasiparticle amplitudes are small. In the strong-coupling limit, higher-order processes including the depletion of the condensate and the recoupling of the excited modes with the condensate should be taken into account.

III. ENERGY SPECTRUM

The quasiparticle amplitudes and excitation spectra were found through discretization of the set of Eqs. (3)- (5) using the method of Ref. [11]. We define a dimensionless interaction parameter, proportional to the number of atoms condensed: $C = 8 \pi N a_s (\hbar/2m_o \omega_o)^{-1/2}$, so that $C \rightarrow 0$ represents the ideal gas limit. In the hydrodynamic approximation the chemical potential is simply given by: $\mu^h = (15C \chi 4 \pi)^{2/3} \hbar \omega_o$. The hydrodynamic regime can be very roughly characterized by $\chi = (\mu^h / \hbar \omega_o) \gg 1$. An example of the spectra of Bogoliubov states for $C = 1000$ (that is $\chi \approx 7$) is shown in Fig. 1. This would correspond, for example, to $N \sim 4500$ atoms of $^{87}$Rb with scattering length $a \sim 110a_o$ within a trap of frequency $\omega_o = 2 \pi \times 126$ Hz. In the inset of Fig. 1 we plot (for reference) the equivalent $N$-independent hydrodynamic results. The main feature of the quantum spectrum, compared with the hydrodynamic results, is the very large differences in the high-$l$-state frequencies. We find that in the quantum regime these surface (high-$l$) modes, in particular the state labeled $l = 4$, interact with the second-harmonic breather mode and play an important role in harmonic generation efficiency.

For low-$n,l$ states, the quantum corrections for the frequencies are rather small for this number of atoms. For example, at $\lambda = 1$ the solution of Eqs. (4) and (5) for the quad-
rupole state gives $\omega_1 = 1.4808\omega_r$ for $C = 1000$. This changes to $\omega_1 = 1.457\omega_r$ for $C = 2000$, and reaches the hydrodymic limit ($C \rightarrow \infty$) at $\omega_1 = 1.4142\omega_r$. The $n=1,l=0$ breather frequency is $\omega_2 = 2.2101\omega_r$ for $C = 1000$, shifting to $\omega_2 = 2.2195$ for $C = 2000$, tending to $\omega_2 = 2.2361\omega_r$ for $C \rightarrow \infty$. However, while the quantal frequencies for these lowlying modes agree well with hydrodymic theory for $C \sim 1000$, the agreement does not extend to wave-mixing process.

IV. BOGOLIUBOV HYBRID MODES

The quantal breather state undergoes a series of avoided crossings with higher-$l$ states as $\lambda$ varies (Fig. 1). The frequency of the $l=4$ quantal mode is greatly different from the hydrodymic predictions. For example, at $\lambda = 1$ the $l=4,m=0$ mode has angular frequency $\omega = 2.426\omega_r$ ($C = 1000$), and $\omega = 2.291\omega_r$ ($C = 2000$) compared with $\omega = 2.000\omega_r$ as $C \rightarrow \infty$. While at $\lambda = 2$, $\omega = 3.164\omega_r$ ($C = 1000$), and $\omega = 3.042\omega_r$ ($C = 2000$), compared with $\omega = 2.732\omega_r$ as $C \rightarrow \infty$. As a result, hydrodymic theory does not predict a degeneracy of the $l=4$ and monopole states.

Near the avoided crossings in Fig. 1, the Bogoliubov modes mix symmetries. The hybridization of the modes is illustrated by the the excitation functions, $u(r,z)$, shown in Fig. 2; a key finding of this paper. Below the crossing, at $\lambda = 1.35$, the $l=0$ and $l=4$ amplitudes resemble the spherical pattern and the $l$-labeling is certainly appropriate. The radial density of the $l=4$ mode is dominated by the presence of the centrifugal barrier pushing the mean radius towards the surface of the condensate. At $\lambda = 1.60$ (Fig. 2) the $l=0$ and $l=4$ states hybridize and lose the character of conventional classification schemes [6,7]. Above the degeneracy the modes separate and regain their character. Identical features were found at the other crossings near $\lambda = 0.80$ and $\lambda = 2.60$ corresponding to interaction with $l=4$ and $n=1,l=2$ states, respectively. The hybridization, although primarily a quantal feature, persists for much larger numbers of atoms. Considering $C = 2000$, that is $N \sim 10^7$, the second-harmonic resonance occurs at $\lambda_c = 1.980$ and we find a slight displacement of the avoided crossing from $\lambda_c = 1.65$ to $\lambda_c = 1.54$, but hybridization of the type discussed is still present.

V. HARMONIC GENERATION

The results for the coupling strengths are shown in Fig. 3. The data are presented in scaled dimensionless units in order to compare with hydrodymic theory [9]: $m_{ij} = 2(\hbar/m_\omega \omega_r)^{3/2} (\mu^2/\hbar \omega_r) \lambda^{-5/2} M_{ij}$. The states are labeled and identified by the adiabatic noncrossing curves in Fig. 1. At the degeneracy $\lambda \sim 1.65$ the strong mixing of states is reflected in the changes in coupling strength. Coupling to the upper $(\pm)$ hybrid mode drops suddenly at this point due to destructive interference between the mode components. The effect is analogous to an optically dark state, that is a coherent superposition of states such that the dipole moments cancel [12]. The interference effect is still apparent near $\lambda = 2$ when the $(\pm)$-state is predominantly breather-like and resonant with the quadrupole second harmonic. In contrast the
FIG. 3. Coupling strength $m_{ij}$ from the $m=0$ quadrupole state for $C=1000$ as a function of trap aspect ratio $\lambda$: coupling to the ($+$) hybrid mode (short-dashed line), to the ($-$) hybrid mode (long-dashed line), and to the $n=1, l=2$ state (dotted line). The hydrodynamic calculation for the breather mode [9] is shown as the dotted line. Second-harmonic resonance for the ($+$) hybrid mode occurs near $\lambda=0.68$ and $\lambda=1.95$ according to hydrodynamic theory [8] and Fig. 1, as indicated by the vertical lines.

off-resonant ($-$) hybrid, which is surface-like for $\lambda \approx 2$ is strongly coupled near the degeneracy.

Similar effects, including the formation of another dark state, occur at $\lambda=2.6$ corresponding to the crossing between $l=0$ and $n=1, l=2$ as shown in Fig. 3. This highlights the fact that it is not only surface modes which can hybridize with the $l=0$ state, but any degenerate mode with the $m=0$ symmetry. The result of hydrodynamic theory for the quadrupole to breather coupling strength [9] is shown in Fig. 3 as the dotted line. The agreement with the quantal calculation is quite good near $\lambda=1$ but near and beyond the avoided crossing point $\lambda_c=1.65$ the hydrodynamic model does not reflect the rapid quantal variation due to hybridization. At the harmonic generation resonance near $\lambda=2$ the hydrodynamic model severely overestimates the coupling strength of the breather mode by a factor of two, and neglects the contribution of the $l=4$ state.

The solutions of the Eqs. (6) and (7) describing the relative populations in modes $i$ and $j$ are well known [13]. For a resonant two-state model the characteristic coupling time is $T = |\sqrt{2} \hbar / g M_p b_i(0)|$. In terms of the dimensionless matrix elements $m_{ij}^\gamma$, this can be written

$$T = \frac{15\lambda^{-13/60}}{8 \pi m_{ij}^\gamma b_i(0) \omega_r} \left( \frac{15C}{64\pi} \right)^{-3/5}.$$

For $\lambda \approx 1.95$, and $C=1000$ the quantal prediction is $|m_{ij}^\gamma| = 0.013$ compared with the hydrodynamic result [9] $|m_{ij}^\gamma| = 0.028$ (the dotted line in Fig. 3). The difference should be both observable and measurable. Suppose that the condensate is perturbed so that only 5% of atoms were seeded into the pump mode, that is $|b_1(0)|^2 = 0.05$. Then, taking for example a trap containing $N \approx 4500$ Rb atoms with $\omega_r = 2\pi \times 126$ Hz and $\lambda = 1.95$, it follows that the respective coupling times are $T^q = 14.60$ ms compared with $T^h = 6.78$ ms. However, the true situation is more complex since this is not a two-level system. The presence of the hybrid pair changes the coupling times and efficiencies dramatically. A numerical solution of Eqs. (6) and (7) for a pump population of 10% at $\lambda=2$ shows the interplay of the hybrid modes: Fig. 4(a). The nonresonant surface hybrid grows faster due to the larger coupling strength, but is not as efficiently converted as the resonant breather hybrid state. The fundamental mode revives [4] after $\sim 40$ ms for these parameters. Since the two hybrid modes differ in frequency and symmetry this phenomenon should be detectable by observation of the radial and axial density variations as developed in current experiments [3]. In Fig. 4(b) corresponding to $\lambda=1.65$, low conversion efficiency is observed due to poor phase-matching. Moreover, in this case the dark-state hybrid (short-dashed line) is completely suppressed.

VI. CONCLUSIONS

In conclusion, we have analyzed wave-mixing processes in the weak-coupling regime for Bose condensates. We find the hybridization of Bogoliubov modes plays a vital role in mode mixing, both in the coupling strength and resonant frequency. Another consequence is the creation of dark coherent states. The conversion rate for second-harmonic generation to the breather mode is substantially lower than the hydrodynamic theory, while the off-resonant surface mode is converted with almost equal efficiency. This hybridization persists for much larger numbers ($N = 10^9$) of atoms where quantal effects were thought to be negligible. Although the...
hybridization lowers the conversion efficiency in the case we studied, the presence of a dark Bogoliubov mode near the wave-mixing resonance means that a single isolated mode can be excited rather than the doublet and therefore better control of single-mode harmonic generation may be possible.

One important consideration is decoherence due to thermal damping or higher-order excitations of the condensate that impose limits on coherent wave mixing processes [3]. Density fluctuations [4] and phase fluctuations [10] of the condensate arise from second-order mixing of the quasiparticle states. This affects the wave-mixing process at the third and fourth order of perturbation theory, and such corrections would be significant for strongly perturbed condensates over long time scales \(\sim 150\) ms. However, according to experiment, thermal damping is expected to be the most important decoherence process [3] over the time scales of interest \(\sim 50\) ms. A detailed description of condensate excitations including terms beyond the Bogoliubov model in the time domain is provided in Refs. [14,15]. The results presented in our paper would form the basis for further investigation along the lines indicated in this work.

Finally, we note that another potentially interesting case [16] is that of second-harmonic resonant coupling between circulating modes. The \(m = 1, 2\) and \(m = 2, 4\) quadrupoles of an oblate spheroidal condensate can combine to populate \(m = 0, 4\) monopole modes. In this case a crossing of the \(m = 0\) and \(m = 4\) modes occurs at the resonance condition \(\lambda = \sqrt{16/7}\). While the \(m = 0\) and \(m = 4\) modes do not hybridize to first order, the indirect coupling via the pump modes will create mixing. The investigation of this particular degenerate transition would provide further evidence of the effects we have discussed.