Theory of Ideal Four-Wave Mixing in Bose-Einstein Condensates

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Abstract

Starting from a second-quantized Hamiltonian of many-particle systems, we derive a set of time evolution equations for four-wave mixing (4WM) processes of coherent matter waves, which is analogous to those in optical 4WM except that the spatial variable is being replaced by time. Several interesting problems in 4WM such as the phase-matching condition, the effects of relative phase difference, and the conversion efficiency are then discussed in detail. We also show that the main features in recent 4WM experiment [Deng \textit{et al}, Nature 398, 218 (1999)] can be understood within the present simplified model.

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1 Introduction

The experimental realizations of Bose-Einstein condensation (BEC) in trapped dilute alkali atomic vapours [1] and the atom “laser” [2] have made possible the experimental study [3] of four-wave mixing (4WM) of coherent matter-waves. As a typical effect of nonlinear atom optics [4], the possibility of 4WM with matter waves of Bose-Einstein condensate (BEC) can be deduced from the similarity between the Maxwell equations for lasing action in nonlinear optical media [5] and the Gross-Pitaevskii (GP) equation [6] widely used in studying the properties of macroscopic quantum systems such as BECs.

In the past few years, 4WM in BECs and related problems have been studied intensively [3,7–16]. Early in 1995, Goldstein \textit{et al.} had started to investigate

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the problem of phase conjugation\cite{7} in BECs, including the case of multi-component BECs\cite{8}. A 4WM experiment using three colliding BEC wavepackets with different central momenta was proposed by Trippenbach et al.\cite{9}. Recently, Deng et al. successfully carried out 4WM experiment in sodium condensates\cite{3} by means of Bragg diffraction technology\cite{17}. In addition, stimulated by the work of Ho and coworkers\cite{10} and Ohmi and Machida\cite{11} on the Bose condensates with internal degrees of freedom, Law et al. studied quantum spin-mixing in spinor BECs\cite{12} using an algebraic method developed in problems related to cavity QED\cite{18}, and Goldstein and Meystre\cite{13} developed a quantum theory of atomic 4WM involving both the internal and spatial degrees of freedom. Rzążewski et al. investigated the fluctuations in the populations of atoms in 4WM wavepackets due to the quantum mechanical nature of the mean-field BEC wavefunctions\cite{14}. Although the experimental results turn out to be consistent with the numerical simulations by Trippenbach et al.\cite{9} based on the GP equation, further theoretical studies are still needed in order to understand 4WM in BECs more clearly. Without invoking the undepleted pump approximation, Wu et al.\cite{15} calculated analytically the 4WM in the experimental condition of Ref.\cite{3}. Based on the slowly-varying-envelope approximation, Trippenbach et al.\cite{16} developed a three-dimensional quantum mechanical description for 4WM of wavepackets created from BEC.

The main purpose of the present work is to study the ideal 4WM with coherent matter waves from BEC in which only four plane waves with definite wavevectors and frequencies are involved. We organize the paper as follows. A set of time evolution equations for 4WM are derived in Sec.2 starting from a Hamiltonian for many-particle systems in the second quantized form rather than the original GP equation. Section 3 gives a short discussion on the phase-matching condition for ideal 4WM. In Sec.4, we present results demonstrating the effects of relative phase difference and the collapse and revival (C&R) behaviour of ideal 4WM. The problem of conversion efficiency and the main characters of recent 4WM experiment\cite{3} are investigated in Sec.5. Sec.6 gives a brief summary.

2 Time evolution equations for 4WM

To study 4WM with matter waves, we consider the dynamics of $N$ interacting identical bosons. For simplicity, we omit the internal degrees of freedom. In standard experimental setups for studying 4WM, the confining potential is removed, and the Hamiltonian can be generally written in terms of bosonic creation and annihilation operators as\cite{19}

$$
\mathcal{H} = \sum_k \varepsilon_k \hat{a}_k^\dagger \hat{a}_k + \sum_{k_1+k_2=k_3+k_4} \frac{U}{2V} \hat{a}_{k_1}^\dagger \hat{a}_{k_2}^\dagger \hat{a}_{k_3} \hat{a}_{k_4},
$$

(1)
where \( \varepsilon_k = \hbar^2 k^2 / 2M \) is the kinetic energy of a particle in a single-particle state of momentum \( \mathbf{p} = \hbar \mathbf{k} \), \( \mathbf{q} \equiv \mathbf{k}_4 - \mathbf{k}_1 = \mathbf{k}_2 - \mathbf{k}_3 \) denotes the momentum transfer in two-body scattering, and \( M \) denotes the atomic mass. Momentum conservation in the second term results from the calculation of the interaction matrix elements \( U_{k_1,k_3,k_2,k_1} = U_q \delta_{k_1+k_3,k_1+k_2} \), where \( U_q \equiv \int d^3 \mathbf{r} U(\mathbf{r}) \exp(i \mathbf{q} \cdot \mathbf{r}) \) is the Fourier transform of the interaction potential \( U(\mathbf{r}) \). The field operator can be expanded as \( \hat{\Psi}(\mathbf{r},t) = \sum_k \hat{a}_k \exp(i \mathbf{k} \cdot \mathbf{r}) / \sqrt{V} \) with \( V \) denoting the volume of the system.

Within the Heisenberg picture, the wavefunction is independent of time and the dynamical evolution of the system is given by that of the field operators. The annihilation operators satisfy equations of the form

\[
\hat{h} \frac{\partial \hat{a}_k}{\partial t} = (\varepsilon_k + U_0 \hat{n}) \hat{a}_k + \sum_{k_1+k_2-k_3=k_4, q \neq 0} \frac{U_q}{V} \hat{a}_{k_3}^\dagger \hat{a}_{k_2} \hat{a}_{k_1},
\]

where the subscripts \( k_1, k_2, k_3 \) and \( k_4 \) can be any allowed values of wavevectors (or particle momenta), and the operator of the averaged number density of atomic gas is a fixed constant defined by \( \hat{n} \equiv (1/V) \sum_k \hat{a}_k^\dagger \hat{a}_k \).

Guided by results obtained through the experimental realization of BEC and the technique of Bragg diffraction[3,17], we assume that the creation and annihilation operators \( \hat{a}_k^\dagger \) and \( \hat{a}_k \) of a certain mode with macroscopic population of atoms can be treated as a pair of conjugated complex numbers \( a_k^* \) and \( a_k \), and those for the other modes vanished[11,20,21]. Therefore the field operator becomes a macroscopic wavefunction satisfying the GP equation, except that \( U_q \) may depend on \( q \) for scatterings with large momentum transfer[6,22]. Introducing a new variable \( A_k \equiv a_k / \sqrt{V} \) and assuming for simplicity a \( q \)-independent \( U_q \approx 4 \pi a_0 \hbar^2 / M \) with \( a_0 \) denoting the s-wave scattering length, the slowly varying envelope function of the field, \( \hat{A}_k \equiv A_k \exp[i(\varepsilon_k - nU_0)t/\hbar] \) with \( n = \sum_k a_k^* a_k / V \), satisfies a set of coupled nonlinear equations

\[
i \frac{\partial \hat{A}_k}{\partial t} = \frac{2a_0 \hbar}{M} \sum_{k_1+k_2-k_3=k_4, q \neq 0} \hat{A}_{k_3}^* \hat{A}_{k_2} \hat{A}_{k_1} e^{i \Delta \omega t}
\]

with \( \Delta \omega = (\varepsilon_{k_4} + \varepsilon_{k_3} - \varepsilon_{k_2} - \varepsilon_{k_1}) / \hbar \). The momentum conservation indicated in the summation is a consequence of the translational invariance of the system.

In contrast, collision processes with \( \Delta \omega \neq 0 \) are not forbidden in principle. As the system has a fixed number of atoms, the averaged number density is a constant, i.e., \( \sum_k |\hat{A}_k|^2 = n \). We can then define the normalized field amplitudes as \( \xi_k = \hat{A}_k / \sqrt{n} \), which satisfy the normalization condition \( \sum_k |\xi_k|^2 = 1 \), to describe the relative distribution of atoms more conveniently. The time
evolution equations for these normalized amplitudes take on the more compact form of

$$i \frac{\partial \xi_{k_4}}{\partial \tau} = \frac{1}{2} \sum_{\substack{k_1 + k_2 - k_3 = k_4 \quad q \neq 0}} \xi_{k_3}^* \xi_{k_2} \xi_{k_1} e^{i \Omega \tau},$$

(4)

where the re-scaled time $\tau \equiv t/T$ is dimensionless with the characteristic time $T \equiv M/4nq_0h$ being inversely proportional to the number density, and the phase mismatch or ‘energy-loss’ parameter is defined as $\Omega \equiv T \Delta \omega$. Before going into details, some important conclusions can be drawn on the common properties of 4WM with matter waves. The number density of atoms can affect the time evolution of 4WM only by modifying the characteristic time $T$, implying that 4WM evolves faster in situations of higher number density of atoms. In actual experimental situations[3], the characteristic time is estimated to be about $3ms$ (see Sec.5).

The general form of time evolution equations for multi-wave mixing (MWM) is a rather complicated set of coupled nonlinear equations because the subscripts $k_1, k_2, k_3$ and $k_4$ in Eq.(4) can take on any allowed values of wavevectors (or particle momenta). However it can be simplified for an ideal 4WM process to

$$\frac{\partial \xi_1}{\partial \tau} = -i \xi_2^* \xi_3 \xi_4 e^{-i \Omega \tau}, \quad \frac{\partial \xi_2}{\partial \tau} = -i \xi_1^* \xi_3 \xi_4 e^{-i \Omega \tau},$$

(5)

$$\frac{\partial \xi_3}{\partial \tau} = -i \xi_4^* \xi_2 \xi_1 e^{i \Omega \tau}, \quad \frac{\partial \xi_4}{\partial \tau} = -i \xi_3^* \xi_2 \xi_1 e^{i \Omega \tau}.$$  

(6)

This set of equations is analogous to those in optical 4WM except that the spatial variable is replaced by the re-scaled time. In what follows, we will study 4WM of matter waves based on the simplified model described by Eqs.(5) and (6). All numerical calculations are carried out using the 4th order Runge-Kutta method.

### 3 Phase-matching condition for 4WM

The recent 4WM experiment[3] shows that three colliding wavepackets with arbitrary central momenta cannot always create a fourth wavepacket, and a possible 4WM process must satisfy the phase-matching condition, i.e. momentum-energy conservation. The Hamiltonian (Eq.(1)) describes a system satisfying the conservation of total energy, momentum and particle-number. Therefore the phase-matching condition is always understood as $k_4 + k_3 = k_1 + k_2$ and $\varepsilon_{k_4} + \varepsilon_{k_3} = \varepsilon_{k_2} + \varepsilon_{k_1}$. Note that from the dispersion relation of massive
particles $\varepsilon_k = (\hbar k)^2/2M$, the phase mismatch parameter can be written as $\Omega = (k_3 - k_2) \cdot (k_3 - k_1)/8\pi n a_0$. The phase-matching condition, \textit{i.e.} $\Omega = 0$, for matter waves can be shown graphically as in Fig.1.

It has also been emphasized, however, in the discussion after Eq.(1) that the scattering process of bosons \textit{does not} require the conservation of energy. So the constraint of energy conservation deserves some more explanations. In fact, numerical calculations show that the parameter $\Omega \equiv T \Delta \omega$ affects 4WM processes, but only large enough phase mismatch can suppress the creation of fourth wave. Figure 2 shows the population $N_4$ of the created wave as a function of $\tau$ for different values of $\Omega$. For small $\Omega$, $N_4(\tau)$ is similar to that of an ideal 4WM process satisfying the phase-matching condition exactly. The conversion efficiency is lower for higher values of $\Omega$, and no 4WM signal can be observed when $|\Omega| \geq \pi$.

4 Dynamics of Ideal 4WM

The time evolution of an ideal 4WM system can be described by Eqs.(5) and (6). This kind of process satisfies not only particle-number conservation but also the conditions

$$\frac{\partial |\xi_4|^2}{\partial \tau} = \frac{\partial |\xi_3|^2}{\partial \tau} = -\frac{\partial |\xi_2|^2}{\partial \tau} = -\frac{\partial |\xi_1|^2}{\partial \tau},$$

(7)

implying that the two pumping waves must provide atoms to amplify the probing and created waves at the same rate. When one of the two pumping waves is exhausted, the another one will take on the role of the probing wave. Figure 3 shows the calculated results for typical collapse and revival (C&R) behaviour of 4WM. Initially, the $N$ atoms are distributed in three states corresponding to the pumping wave 1, pumping wave 2 and probing wave 3. Due to two-body interaction and the bosonic enhancement effect, the atoms in the pumping waves begin to transfer into the states 3 and 4. After a short period of time, all the atoms are transferred out of state 2 and no more atoms are transferred into state 4. The second phrase of 4WM begins at around $\tau \approx 5$. Now waves 3 and 4 play the roles of the pumping waves, and wave 1 acts as a probing wave, wave 2 will become appreciable again until the pumping wave 4 vanishes at about $\tau \approx 10.5$. This completes one period of ideal 4WM.

Another special dynamical character in 4WM processes is the effect of relative phase difference. It is not the phase of each individual wave that matters. Instead if we write $\xi_i = \sqrt{T_i} e^{i \phi_i}$, it is the combination of the phases $\alpha =$
\[ \varphi_4 + \varphi_3 - \varphi_2 - \varphi_1 - \Omega \tau \] that affects 4WM in the way that

\[
\frac{\partial I_1}{\partial \tau} = \frac{\partial I_2}{\partial \tau} = -\frac{\partial I_3}{\partial \tau} = -\frac{\partial I_4}{\partial \tau} = 2A \sin \alpha; \quad (8)
\]

\[
\frac{\partial \alpha}{\partial \tau} = -\Omega + A \left( \frac{1}{I_1} + \frac{1}{I_2} - \frac{1}{I_3} - \frac{1}{I_4} \right) \cos \alpha, \quad (9)
\]

where the amplification factor is defined as \( A \equiv \sqrt{I_1 I_2 I_3 I_4} \). When one of the four waves vanishes, we can set the combined phase \( \alpha \) to be an arbitrary value. Therefore the initial values of the pumping and probing waves do not affect the time evolution of atomic distributions.

5 Conversion efficiency of 4WM

All of the previous works[9,14,16] on the 4WM of matter waves have focussed on the initial rate of growth of the fourth wave because the time of wavepackets interaction in experiment[3] is very short (determined by the size and individual momentum of wavepackets). Initially the rate of growth of the fourth wave is proportional to the product of the intensities of pumping and probing waves. If the experimental setup can be changed in such a way that 4WM can proceed for a much longer period, we need more precise knowledge about the conversion efficiency of 4WM.

In the experiment [3] of 4WM with matter waves in BEC with \( N \approx 1.7 \times 10^6 \) sodium atoms \( (M = 3.848 \times 10^{-23} g \text{ and } a_0 = 28 \text{Å}) \), the pumping (1,2) and probing (3) waves are prepared by Bragg scattering[17]. It was estimated that the population numbers in the three states took on the initial values of \( N_0^1 \approx 6.8 \times 10^5, N_0^2 \approx 6.9 \times 10^5 \) and \( N_0^3 \approx 3.3 \times 10^5 \), respectively. Assuming that 4WM took place in a region of volume \( V = L^3 \) with \( L \approx 0.1 mm \), the characteristic time will be \( T \equiv ML^3/4Na_0h \approx 3 ms \).

The four curves in Fig. 4 show typical results of the conversion efficiency \( N_4/N \) as a function of the re-scaled time \( \tau \) obtained by Eqs.(5) and (6), hence assuming an ideal 4WM process with \( \Omega = 0 \), for different sets of initial conditions. Curve (i) shows the situation in which \( N_0^1 = N_0^2 \gg N_0^3 \), which achieves a maximum efficiency of 50% if 4WM can proceed for a time long enough, e.g., \( t > 10T \). Curve (ii) is calculated using a set of initial conditions similar to the experimental situation. Comparing with curve (i), it demonstrates a much greater initial rate of growth of the fourth wave and a, however, smaller saturation efficiency. Note that a conversion efficiency of 10% is achieved at \( t \approx 1.9T \approx 5.8 ms \), which is consistent with the results reported in Ref.[3]. The initial conditions for curves (iii) and (iv) are chosen so that both of them have
the same initial rate of growth of the fourth wave. Even so, the two curves demonstrate very different evolution processes as a function of $\tau$. Therefore, carefully prepared initial atomic distributions are crucial in achieving a higher initial rate of increase in the efficiency and at the same time a high enough saturation efficiency.

While the initial conditions in Ref.[3] are close to those values in achieving a high efficiency, the observed value of $6 \sim 10\%$ is still rather low. A higher conversion efficiency can then be achieved by increasing the interaction time and/or by shortening the characteristic time by increasing the number density of atoms.

6 Summary

In conclusion, a set of nonlinear equations for the time evolution of multi-wave mixing in BECs is derived. The formalism leads to a physically transparent picture of 4WM with matter waves. The ideal 4WM model is successfully applied to analyze results of 4WM experiments. Factors affecting the value of the conversion efficiency are discussed. The GP equation in momentum space provides us with an efficient way to study the wavepacket effects of 4WM because the four wavepackets are limited to four spots of the momentum space even though they may spread widely in real space.

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The original GP equation was presented for studying the properties of BECs at very low temperature, and only the very low energy scatterings were taken into account. However in 4WM experiments, the momenta of the created matter waves can be so large that the interaction matrix elements can no longer be treated as a constant. In Ref.[16], the authors have discussed the possibility of modifying the GP equation to allow for the incorporation of momentum dependence of the nonlinear parameters.
FIGURE CAPTIONS

Fig.1  The momentum sphere of phase-matching condition for 4WM. The center is located at $\left( \mathbf{k}_1 + \mathbf{k}_2 \right) / 2$, and the radius of the sphere is defined as the magnitude of the vector $\left( \mathbf{k}_1 - \mathbf{k}_2 \right) / 2$. If the momentum of the probing wave points exactly at a point on the surface of the sphere, the largest conversion efficiency of 4WM is achieved.

Fig.2  The conversion coefficient as a function of interaction time for five different values of the phase-mismatch parameter $\Omega = T \Delta \omega$. The time is given in units of the characteristic time as defined in the text. The five curves from top to bottom correspond to $\Omega / \pi = 0, 0.1, 0.2, 0.4, 1$, respectively. The initial conditions for the pumping and probing waves are chosen to be $\xi_1^0, \xi_2^0, \xi_3^0 = 0.68, 0.58, 0.45$.

Fig.3  Typical time evolution curves of the populations of the matter waves involved in a 4WM process. The initial conditions are chosen to be $\xi_1^0, \xi_2^0, \xi_3^0 = 0.68, 0.58, 0.45$.

Fig.4  Typical results of the conversion efficiency as a function of the interaction time for different initial conditions. The initial conditions for curve (i) are $\xi_1^0, \xi_2^0, \xi_3^0 = 0.707, 0.703, 0.243$, which lead to a maximum efficiency of 50% but with a low initial increasing rate. Curve (ii) has initial conditions similar to those in experimental situation: $\xi_1^0, \xi_2^0, \xi_3^0 = 0.637, 0.632, 0.441$. Curves (iii) and (iv) show that in order to achieve an appreciable efficiency, the probing wave can be weak but the pumping waves must be strong. The initial conditions are: (iii) $\xi_1^0, \xi_2^0, \xi_3^0 = 0.804, 0.553, 0.217$ and (iv) $\xi_1^0, \xi_2^0, \xi_3^0 = 0.804, 0.217, 0.553$. 
(\vec{k}_3 - \vec{k}_1) \cdot (\vec{k}_3 - \vec{k}_2) = 0
Deng et al. Fig.2
Deng et al. Fig. 3
Deng et al. Fig.4