We investigate theoretically the four-wave mixing of optical and matter waves resulting from the scattering of a short light pulse off an atomic Bose-Einstein condensate, as recently demonstrated by D. Schneble et al. [Science 300, 475 (2003)]. We show that atomic “pair production” from the condensate results in the generation of both forward- and backward-propagating matter waves. These waves are characterized by different phase-matching conditions, resulting in different angular distributions and temporal evolutions.

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The interaction between optical fields and atomic Bose-Einstein condensates has attracted much recent attention, due to its importance in the preparation, manipulation, and detection of condensates, as well as because of its interest in fundamental studies of the nonlinear interaction between Maxwell and Schrödinger waves. A number of phenomena have already been studied both theoretically and experimentally, including matter-wave superradiance, coherent matter-wave amplification, and Bragg spectroscopy.

In a trialblazing experiment, a cw laser beam shined on a cigar-shaped condensate resulted in the generation of a fun-like pattern of momentum sidemodes of the condensate. The initiation of this pattern can be understood in terms of a four-wave mixing process involving two optical fields – the laser field and a so-called end-fire mode; and two matter-wave modes – the condensate and a mode of momentum such that energy-momentum conservation (or phase matching) is satisfied. We recall here that the end-fire mode, first predicted by Dicke in his study of superradiance, corresponds to the privileged direction for spontaneous emission along the long axis of the condensate. Because of momentum conservation, it is clear that the generated sidemodes must be in the forward direction at a 45° angle between the direction of the incident laser and the long axis of the condensate. The subsequent generation of further sidemodes results simply from wave mixing involving an already excited sidemode instead of the initial condensate at rest.

Recently, that same MIT group reported the results of an experiment where the incident cw laser was replaced by an optical pulse. This led to the remarkable result that for short enough pulses, backward-scattered atoms (with a momentum component antiparallel to the direction of the pump field) were also observed. Moreover, the backward peaks exhibited a slightly different angular distribution compared with the forward peaks. The qualitative difference in diffraction patterns for the short- and long-pulse regimes was attributed to the transition from the Raman-Nath to the Bragg regime of diffraction, that is, to the onset of energy-momentum conservation for long enough interaction times.

In this Letter, we present a theoretical description of this experiment based on an extension of the quasi-mode approach of Ref. 2. We give a full dynamical treatment of both the vacuum photon modes and the condensate sidemodes that interprets the diffraction pattern in terms of atom-photon wave mixing and show explicitly that the counter-propagating (backward and forward) matter-wave quasi-modes result from the quantum-correlated parametric excitation of atomic pairs. The difference in their angular distribution and dynamics results from their distinct phase-matching conditions.

A general theoretical framework to describe the interaction of ultracold atoms with light waves was presented by Zhang and Walls in Ref. 12. In the situation at hand, the atomic system, assumed to be at zero temperature, interacts with a far off-resonant classical laser field of (real) Rabi frequency $\Omega_L$, wave vector $\mathbf{k}_L$ and frequency $\omega_L = c k_L$, as well as with a continuum of electromagnetic field modes of wave vector $\mathbf{k}$ and polarization $\lambda$ characterized by the bosonic annihilation operators $B_{\mathbf{k}\lambda}$. In the case of large atom-laser detunings $\Delta$, we are justified in adiabatically eliminating the excited atomic levels, leaving us with just the bosonic ground-state matter-wave field operator $\psi_1(\mathbf{r},t)$. We furthermore perform the rotating wave approximation and express the dynamics of the coupled atoms-radiation system in a frame rotating at the pump laser frequency to find

$$i\hbar \frac{\partial \psi_1}{\partial t} = H_0(\mathbf{r})\psi_1 + \frac{\hbar \Omega_L}{2\Delta} \sum_{\mathbf{k},\lambda} [g_{\mathbf{k}\lambda}^* B_{\mathbf{k}\lambda} + \text{c.c.}] \psi_1,$$

$$i\hbar \frac{\partial B_{\mathbf{k}\lambda}}{\partial t} = \hbar \omega_{\mathbf{k}} B_{\mathbf{k}\lambda} + \frac{\hbar \Omega_L}{2\Delta} g_{\mathbf{k}\lambda}^* e^{-i\omega_L t} \int d^3r e^{i(k-k_L)\cdot r} \psi_1^\dagger(\mathbf{r},t)\psi_1(\mathbf{r},t),$$

where $g_{\mathbf{k}\lambda} = i \sqrt{2\pi \omega_L/\hbar V} \langle d | e_{\mathbf{k}\lambda} \rangle$ is the coupling strength of the atom with the corresponding vacuum mode ($V$ is the quantization volume, $e_{\mathbf{k}\lambda}$ the photon polarization unit vector and $d$ the atomic dipole moment). In Eq. (1), the atomic Hamiltonian $H_0(\mathbf{r})$ includes the usual kinetic and trapping potential terms, the ac Stark shift arising from the pump laser as well as nonlinear atom-atom interactions resulting e.g. from two-body collisions.

In the absence of electromagnetic fields, the conden-

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sate at zero temperature is taken to be in the ground state \(\varphi_0(\mathbf{r})\) of \(\hat{H}_0\), with \((\hat{H}_0 - \hbar \mu)\varphi_0(\mathbf{r}) = 0\), \(\mu\) being the chemical potential. The interaction of this condensate with light shifts the atomic momentum by the photon recoil, and transfers condensate atoms to states of the form \(\varphi_q(\mathbf{r})\exp(i\mathbf{q} \cdot \mathbf{r})\). For condensates large compared to an optical wavelength, these states are still approximate eigenstates of \(\hat{H}_0\), with eigenenergy shifted by the recoil frequency \(\omega_q = \hbar q^2 / 2M\), i.e., \(\hat{H}_0 [\varphi_q(\mathbf{r})\exp(i\mathbf{q} \cdot \mathbf{r})] \approx \hbar (\mu + \omega_q)\varphi_q(\mathbf{r})\exp(i\mathbf{q} \cdot \mathbf{r})\), an approximation similar to the slowly varying envelope approximation in optics. Following Ref. [2], this suggests expanding \(\psi_1(\mathbf{r}, t)\) as

\[
\psi_1(\mathbf{r}, t) = e^{-i\omega t} \sum_q \varphi_q(\mathbf{r}) e^{i(\mathbf{q} \cdot \mathbf{r} - \omega_q t)} c_q,
\]

where the atomic quasi-mode field operators approximately obey bosonic commutation relations, \([c_q, c_{q'}^\dagger] = \delta_{q,q'}\). Inserting this expansion into Eq. (1) gives

\[
i\hbar \dot{c}_q = \frac{\hbar \Omega_L}{2\Delta} \sum_{k,\lambda} \sum_{q_1} \left[ g_{k\lambda}^* B_{k\lambda} e^{-i\omega_{kL} t} \Pi_{q-q}(k) + g_{k\lambda} B_{k\lambda} e^{i\omega_{kL} t} \Pi^*_{q-q'}(k) \right] e^{i(\omega_q - \omega_{q'}) t} c_{q'},
\]

where \(\Pi_q(k) = \int d^3r \left| \varphi(r) \right|^2 \exp[-i(q + k - k_L) \cdot r]\) is the spatial Fourier transform of the condensate density profile. Consistently with the slowly-varying approximation implicit in the expansion (4), it satisfies the approximate relation \(\Pi_q(k)\Pi^*_q(-k) \approx |\Pi_q(k)|^2 \delta_{q,q'}\), a condition that indicates that the discretization of \(q\) is such that there is negligible overlap between shifted momentum wave functions. This property is used repeatedly in the following.

We now proceed by formally integrating Eq. (4) to find

\[
B_{k\lambda}(t) = B_{k\lambda}(0) e^{-i\omega_{kL} t} - \frac{\Omega_L}{2\Delta} g_{k\lambda}^* e^{-i\omega_{kL} t} \int_0^t dt' e^{-i(\omega_k - \omega_{kL})\tau} \int d^3r e^{-i(k - k_L \cdot r)} \Pi^*_q(r, t - \tau) \psi_1^\dagger(r, t - \tau)
\]

\[
= B_{k\lambda}(0) e^{-i\omega_{kL} t} - \frac{\Omega_L}{2\Delta} g_{k\lambda}^* e^{-i\omega_{kL} t} \sum_{q_1, q_2} \Pi_{q_1-q_2}(k) \delta(\omega_k - \omega_{q_1} - \omega_{q_2}) e^{i(\omega_{q_1} - \omega_{q_2}) t} c_{q_1}^\dagger(t) c_{q_2}(t),
\]

where we have used Eq. (3) and have adopted the Markov approximation [13] by replacing \(c_{q_1}^\dagger(t - \tau)\) and \(c_{q_2}(t - \tau)\) with \(c_{q_1}^\dagger(t)\) and \(c_{q_2}(t)\) respectively, and setting the upper limit of integration over \(\tau\) to infinity [14].

It is clear by inspection of Eqs. (3) and (4) that the system dynamics can be interpreted in terms of four-wave mixing processes involving two matter-wave fields and two optical fields. For example, the dynamics of the atomic quasi-mode \(q\) results from the combined effects of the laser and a vacuum field mode together with an additional atomic quasi-mode \(q'\), while the dynamics of a scattered photon mode \(\{k, \lambda\}\) results in turn from the combined effects of the laser field and two atomic modes.

For optical pulses of short duration, we can assume that the condensate population is undepleted and replace the operators \(c_0\) and \(c_0^\dagger\) by the c-number \(\sqrt{N_0}\), where \(N_0\) is the number of condensate atoms. Inserting then Eq. (6) into Eq. (1) results in

\[
\dot{c}_q = A_q(t) c_q + B_q(t) c_{q'} + \Gamma_q(t),
\]

where \(q \neq 0\) and we have only kept the linear terms involving \(c_q\) and \(c_{q'}\) consistently with the undepleted pump approximation. Here

\[
A_q(t) = \frac{N_0 \Omega_L^2}{2\Delta^2} e^{-i\omega_{kL} t} \left[ \left| \Pi_q(k) \right|^2 \delta(\omega_k - \omega_{q_1} - \omega_{q_2}) - \left| \Pi_{-q}(k) \right|^2 \delta(\omega_k - \omega_{q_1} - \omega_{q_2}) \right],
\]

and \(B_q(t) = -A_{-q} \exp(2i\omega_q t)\).

The Langevin noise operators \(\Gamma_q(t)\), which account for the vacuum electromagnetic fluctuations through the initial photon operators \(B_{k\lambda}(0)\) and \(B_{k\lambda}^\dagger(0)\) and are responsible for the initiation of the scattering process [2], have the form \(\Gamma_q(t) = \exp(i\omega_q t)f_q(t) - f_{-q}(t)\), where

\[
f_q(t) = i \sqrt{N_0 \Omega_L^2} \sum_{k,\lambda} g_{k\lambda} B_{k\lambda}(0) e^{-i(\omega_k - \omega_{q_1}) t} \Pi^*_q(k).
\]

The coefficients \(A_q\) and \(B_q\) each contains two terms, corresponding to the four processes illustrated in Fig. 4. The two processes described by \(A_q\) are resonant, while those described by \(B_q\) are characterized by an energy mismatch \(2\hbar \omega_q\), as evident from the expression for \(B_q\). Fig. 4 shows that \(B_q\) describes a pair production process where two condensate atoms are scattered into the quasi-modes \(q\) and \(-q\). Such a process is known in quantum optics to result in the production of entangled or squeezed particle pairs. This process, which leads to the generation of backward-scattered atoms, will clearly occur for interaction times short enough that energy-momentum conservation (or phase-matching) is not yet established. This explains why back-scattering was not observed in the quasi-cw experiments of Ref. [1]. Furthermore, \(A_q(t)\) is positive (negative) for forward- (backward-)scattered atoms. Hence, the onset of backward scattering relies
atom-photon interaction, and $N_M$ and $\gamma$ different from zero for $k$ that the wave numbers of the scattered photons satisfy mentioned earlier implies that $\Pi_q$ is the slowly-varying amplitude approximation here, the two scattering processes represented by the coefficient $A_q (B_q)$. \( ^0q \) and \( ^\pm q \) label the atomic momentum states with \( ^0q \) being the condensate mode. The solid and dashed arrows represent the pump and scattered photon, respectively.

Equations (7) and (8) form a closed set that can be solved numerically. The atomic recoil frequencies $\omega_q$ are many orders of magnitude smaller than the laser frequency $\omega_L$, so that conservation of energy requires that the wave numbers of the scattered photons satisfy $k \approx k_L$. Furthermore, for the large condensates considered here, the slowly-varying amplitude approximation mentioned earlier implies that $\Pi_q (k)$ is only significantly different from zero for $k+q-k_L \approx 0$, indicating momentum conservation for the scattering process. Under such conditions, the atoms are scattered from the condensate in two dipole emission halos, represented schematically by the two circles of radius $k_L$ in Fig. 2.

We evaluate the coefficients in Eqs. (7) and (8), by following the procedure of Ref. [2] to find $A_q = B_q \exp (-i2\omega_q t) = \gamma (\Omega_q - \Omega_{-q}), N_q = 2\gamma \Omega_q$ and $M_q = -\gamma \exp (i2\omega_q t) (\Omega_q + \Omega_{-q})$, where $\gamma = N_0 \Omega_L^2 k_L^2 |d|^2/(4\pi \hbar \Delta^2)$ characterizes the strength of the atom-photon interaction, and

$$\Omega_q = \frac{4\pi}{k^2 \omega \Lambda} \left[ \cos^2 \theta_k + (l/w)^2 \sin^2 \theta_k \right]^{-1/2}.$$

Here, $w$ and $l$ are the dimensions of the cylindrically symmetric condensate along the radial and axial directions, respectively, and $\theta_k$ is the angle between $k= k_L - q$ and the long axis of the condensate. For a cigar-shaped condensate, $l \gg w$, the scattered photons are predominantly along that axis, Dicke’s end-fire mode, so that most of the scattered atoms gain momenta at roughly $45^\circ$ from the long axis of the condensate [1, 2, 11].

Figure 3 shows the angular distribution of the scattered atoms. The forward-scattered atoms form an almost perfect symmetric distribution about the $45^\circ$ angle. This must be contrasted to the backward-scattered atoms, whose distri-
FIG. 3: a) Ratio of the backward- and forward-scattered atom numbers at angle $\phi = 45^\circ$ as a function of time, in units of $1/\omega_r$, where $\omega_r = \hbar k_0^2/(2M)$. The lines from top to bottom correspond to $\gamma = 500$, 250, 150 and 50$\omega_r$. b) Angular distribution in arbitrary units of the forward- (solid line) and backward-scattered (dashed line) atoms. Here $\gamma = 50\omega_r$ and $t = 0.7/\omega_r$. The results are obtained by numerically integrating Eqs. 4 and 5 with initial conditions $n_1 = m_2 = 0$ using a 4th-order Runge-Kutta method.

The experimental observation shows an asymmetry, with more atoms scattered into $\phi > 45^\circ$ than into $\phi < 45^\circ$ (see Fig. 2): Backward scattering favors larger angles because the energy mismatch is smaller for $\phi > 45^\circ$ than that for $\phi < 45^\circ$, since $\Delta E = 2\hbar \omega_p \cos \phi$. A similar asymmetry in the behavior of the forward- and backward-scattered atoms was observed experimentally [11], and was attributed to the fact that the two scattering processes occur mainly at different locations along the condensate. Because the Markov approximation neglects the effects of retardation in the description of the end-fire mode dynamics [10], that mechanism is absent from our analysis. In practice, both phase matching and spatial effects are probably at play, and they could in principle be distinguished by varying the time of flight before detection [17].

In summary, we have presented a theoretical investigation of a zero-temperature condensate interacting with a far off-resonant laser pulse. Our work is valid for interaction times short enough that the condensate remains undepleted and one can retain only linear terms involving atomic modes of finite momenta. Our analysis shows that while the resonant Raman scattering illustrated in the plots $A_{1,2}$ of Fig. 1 is the dominant generation mechanism of forward-scattered atoms, the backward-scattered atoms result from the quantum-correlated pair production processes of plots $B_{1,2}$ in Fig. 1 [13]. The lack of phase-matching for such pair production processes results in distinct angular distributions for the backward- and forward-scattered atomic peaks, in good qualitative agreement with the experiment of Ref. [11].

Schneble et al. interpret the existence of backward-scattered atoms as resulting from Kapitza-Dirac diffraction of matter waves off the optical gratings formed by the pump laser and the end-fire modes [11]. While this point-of-view is consistent with our correlated pair production picture, we emphasize that the strength of the optical grating depends on the intensity of the end-fire modes, which in turn depends on the population of the atomic quasi-modes [see Eq. 3]. It is this interplay of the optical and atomic fields that renders the backward- and forward-scattered atoms correlated. It also shows that the two diffraction processes of the system—optical diffraction off the atomic grating and atomic diffraction off the optical grating—are coherently mixed and hence inseparable. For the same reason, the two counter-propagating optical end-fire modes must also exhibit quantum correlations.

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