Observation of a Red-Blue Detuning Asymmetry in Matter-Wave Superradiance

L. Deng,[1] E. W. Hagley,[1] Qiang Cao,[2] Xiaorui Wang,[2] Xinyu Luo,[2] Ruquan Wang,[2] M. G. Payne,[1] Fan Yang,[3] Xiaoji Zhou,[3] Xuzong Chen,[3] and Mingsheng Zhan[4,5]

[1]Physics Laboratory, National Institute of Standards & Technology, Gaithersburg, Maryland 20899, USA
[2]Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China
[3]School of Electronics Engineering & Computer Science, Peking University, Beijing 100871, China
[4]State Key Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences, Wuhan 430071, China
[5]Center for Cold Atom Physics, Chinese Academy of Sciences, Wuhan 430071, China

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We report the first experimental observation of strong suppression of matter-wave superradiance using blue-detuned pump light and demonstrate a pump-laser detuning asymmetry in the collective atomic recoil motion. In contrast to all previous theoretical frameworks, which predict that the process should be symmetric with respect to the sign of the detuning of the pump laser from the one-photon resonance, we find that for condensates the symmetry is broken. With high condensate densities and red-detuned pump light the distinctive multiorder, matter-wave scattering pattern is clearly visible, whereas with blue-detuned pump light superradiance is strongly suppressed. However, in the limit of a dilute atomic gas symmetry is restored.

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Matter-wave superradiance is coherent, collective atomic recoil motion that was first reported [1] in a Bose-Einstein condensate (BEC) of ^{23}Na atoms illuminated by a single, far red-detuned, long-duration laser pulse. Since its discovery, processes such as short-pulsed, bidirectional superradiance [2], Raman superradiance [3,4], and matter-wave amplification [5,6] have been observed. Also, many theoretical investigations [7–16] have studied this complex light-matter interaction process that is of significant importance to the fields of cold-atom physics, cold-molecular physics, condensed-matter physics, nonlinear optics, and quantum information science.

The widely accepted theory [1] of matter-wave superradiance is based on spontaneous Rayleigh scattering and the buildup of a matter-wave grating that is further enhanced by subsequent stimulated Rayleigh scattering. This intuitive picture, which correctly models late-stage superradiant growth when red-detuned light is used, captures many important aspects of this intriguing process. However, the simple grating viewpoint and most rate-equation-based theories neglect propagation dynamics of the internally generated optical field. In fact, the initial study [1] explicitly states that the optical fields travel at the speed of light in vacuum and therefore do not affect scattering at later times. To date, no report in the literature has contradicted that statement [17]. However, we have recently shown theoretically [18–20] that the internally generated field propagates ultraslowly and plays an important role in the genesis of superradiance with BECs. We also note that most previous theories effectively treat the BEC as a thermal gas by neglecting the extra factor of the mean-field potential in the scattering process due to the exchange term in the Hamiltonian. As we will show, this unique property of BECs profoundly impacts superradiant scattering and leads to the pump-laser detuning asymmetry reported here.

In this Letter, we present the first experimental observation of a red-blue detuning asymmetry in matter-wave superradiance. We demonstrate astonishingly efficient suppression of superradiance when the pump laser is blue detuned that cannot be explained by current theoretical frameworks. However, using our new theoretical framework [18] we propose a possible explanation for the detuning asymmetry based on an induced optical-dipole potential that results from the ultraslow propagation velocity and gain characteristics of the generated field.

The experimental data reported here were obtained using two ^{87}Rb BECs created with very different experimental systems at two independent institutions. In both systems we produced an elongated BEC using standard magneto-optical trapping techniques followed by radio-frequency evaporative cooling. After formation of the BEC a pump laser of selected frequency, polarization, and duration was applied along the BEC's short axis (see Fig. 1). The magnetic trap was then switched off and absorption imaging was employed after a delay sufficient to allow spatial separation of the scattered components. For all data reported the relevant transition was $|5S_{1/2}\rangle - |5P_{3/2}\rangle$ ($|1\rangle - |2\rangle$), the ground electronic state was $F = 2, m_F = +2$, and the detuning was measured with respect to the $F' = 3$ state. We derived the pump laser from a cavity-stabilized diode laser with linear polarization perpendicular to the long axis of the BEC. The detuning asymmetry was investigated from 500 MHz $\leq |\delta|/2\pi \leq$ 4 GHz for both red and blue detunings. Over the range of detunings investigated, superradiance was always strongly suppressed when a pure, high-density BEC was illuminated with blue-detuned light. We note that the

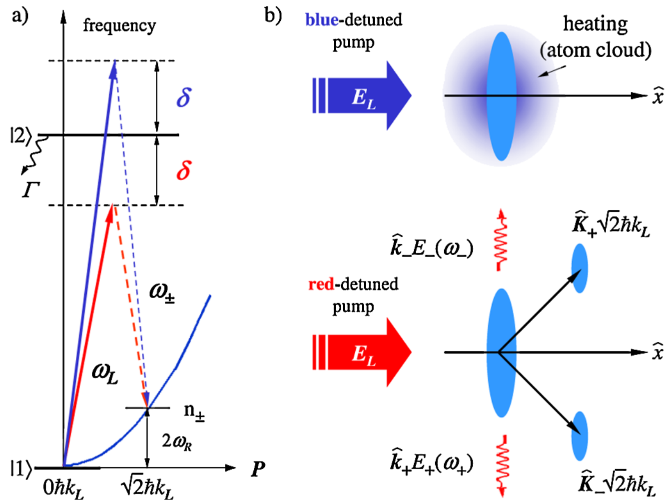

FIG. 1 (color online). (a) Energy levels and laser excitation scheme. The one-photon detuning is $\delta = \omega_L - \omega_{21}$, where ω_L (ω_{21}) is the laser (resonance) frequency, and Γ is the spontaneous emission rate of state $|2\rangle$. (b) Experimental geometry and detuning effect. Upper panel: pump is blue detuned and superradiance is strongly suppressed. Lower panel: pump is red detuned and superradiance is strongly favored. $\hat{K}_\pm$ ($\hat{k}_\pm$) are the unit vectors for the collective atomic recoil (field) modes.

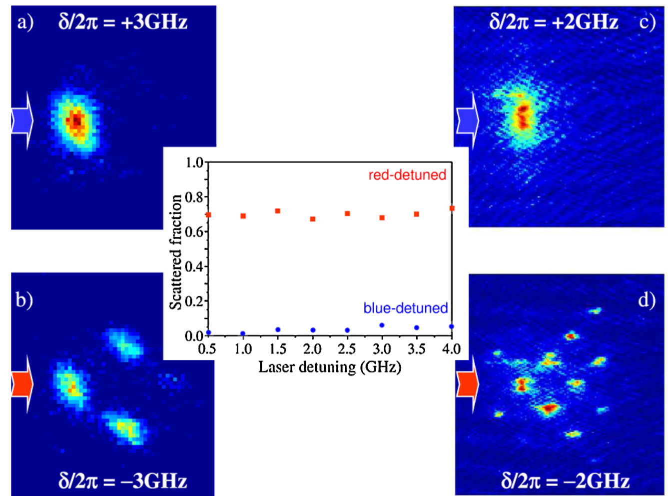

FIG. 2 (color online). TOF absorption images after a 200 μs pump pulse. Left panel image size is 314 μm $\times$ 336 μm, TOF = 15 ms, 5×10^4 atoms, $I_P = 56$ mW/cm^2. The in-trap aspect ratio was about 9 to 1. (a) blue detuning. (b) red detuning. Inset: detuning dependence for (a) and (b) with a constant single-photon scattering rate (uncertainty <10%). Right panel TOF = 20 ms, 2×10^5 atoms, $I_P = 150$ mW/cm^2. (c) blue detuning. (d) red detuning (image size is 914 μm $\times$ 984 μm).

scattering efficiencies for red detunings reported here are also consistent with previous studies [1–3,11,12,21].

The left panel of Fig. 2 shows two time-of-flight (TOF) absorption images of a BEC momentum distribution after application of a pump pulse. For Fig. 2(a), which shows no superradiant scattering, the laser was blue detuned by +3 GHz, whereas for Fig. 2(b) the laser was red detuned by −3 GHz and first-order superradiance is clearly visible. The inset in Fig. 2 is a map of the scattering efficiency for red and blue detunings. These data clearly show that superradiant scattering was strongly suppressed when a blue-detuned pump was applied. The right panel of Fig. 2 displays two TOF images after application of a high-power pump pulse to an elongated BEC using a different experimental apparatus. These images show that growth of higher-order momentum states is subject to a condition similar to the one that leads to suppression of first-order scattering when blue-detuned light is used. We point out that this is consistent with a sequential scattering process where higher-order growth is predicated on the growth of first-order momentum components. The above observations demonstrate the stark contrast in scattering for red- and blue-detuned pump light with BECs, and raise serious challenges to current theoretical frameworks [1,2,7–16] which predict symmetry with respect to the sign of the pump-laser detuning.

When a pump-laser interacts with a BEC it first generates photons by spontaneous Rayleigh scattering, regardless of the sign of the pump-laser detuning. However, even in this early stage the BEC's structure factor [22] imposes additional constraints on the scattering process and slightly suppresses this two-photon channel to about 90% of its free-particle value. Nevertheless, these seed fields may then be amplified by coherent scattering of pump photons via the two-photon process treated in Ref. [18]. Since the initial number of spontaneously scattered photons per unit volume is proportional to the local density, at early times the intensity of these seed fields will directly reflect the local BEC density. However, the velocity of these growing seed fields will be inversely proportional to the local density and the field gain will be an exponential function of density. For sufficiently high spontaneous Rayleigh scattering rates the generated field will grow diabatically with respect to atomic motion, and will result in a non-negligible average optical-dipole potential $\bar{U}_{\rm dipole}$. This induced $\bar{U}_{\rm dipole}$ breaks the detuning symmetry of the original scattering process because for red (blue) detuned light $\bar{U}_{\rm dipole}$ is attractive (repulsive). The important question to ask is how can $\bar{U}_{\rm dipole}$ affect the scattering process?

It has been shown interferometrically [20] that the energy in the light scattering process has an additional mean-field contribution due to the exchange term in the Hamiltonian, $E/\hbar = 4\omega_R + \omega_{\rm MF}$. Here $\omega_{\rm MF} = \bar{U}_{\rm MF}/\hbar = 16\pi\hbar a n_0/(7M)$ is the average mean-field shift where a is the scattering length, M is the atomic mass, n_0 is the peak condensate density, ω_R is the single-photon recoil frequency, and we have neglected the optical index of the medium because of the large detunings in this study. Clearly, the scattering is not free-particle-like because of the additional energy $\hbar\omega_{\rm MF}$. However, with red detunings the induced $\bar{U}_{\rm dipole}$, which is seen by both the original BEC and the scattered atoms, will grow and eventually reach the level of the mean-field potential ($\bar{U}_{\rm dipole} \approx -\bar{U}_{\rm MF}$). Under this condition the net energy available to an atom scattered out of the condensate relative to the unperturbed condensate is simply $E/\hbar = 4\omega_R$, and the scattering becomes "free-particle-like" for all momentum transfers. The attractive $\bar{U}_{\rm dipole}$ can therefore be thought of as a work function for removing atoms from the BEC that is overcome by the additional factor of $\bar{U}_{\rm MF}$ given to the scattering process. Note that only scattered atoms would experience a "flat" potential, and that the host BEC would not be in equilibrium [23]. Satisfying this free-particle-like scattering condition implies that there is no extra energy left for quasiparticle excitations of the host condensate.

This naturally brings us back to the structure factor [22] of a BEC (without $\bar{U}_{\rm dipole}$), which goes to zero for low-momentum scatterings. If we postulate that the free-particle-scattering condition removes the constraint of the host BEC structure factor and allows low-momentum scatterings to occur, then the system would start to behave like an ultracold thermal gas. In this case both two- and potential four-photon [24] processes could occur simultaneously, resulting in very efficient coherent growth of the generated field. We point out that because $\bar{U}_{\rm dipole}$ grows exponentially with density, it becomes more sharply peaked than the Thomas-Fermi density distribution and the resulting transverse optical-dipole force will lead to an increasing transverse velocity spread of the atoms [23]. We speculate that the opening of possible nonlinear gain channels may facilitate triggering bosonic stimulation by scattering more highly monochromatic photons (atoms) along (at 45° to) the long symmetry axis of the BEC where the transverse velocity is minimal and the density is greatest. However, even without invoking nonlinear gain channels the impact of the evolving structure factor on the two-photon channel should be sufficient to explain the asymmetry.

With blue-detuned light the diabatically generated field moves the system further away from free-particle-like scattering because the growing $\bar{U}_{\rm dipole}$ adds to $\bar{U}_{\rm MF}$ rather than canceling it. This would cause the effective structure factor to have an increasingly larger negative impact on the two-photon channel as the optical-dipole potential grows, and would lead to gain clamping. Therefore the two-photon gain channel would become inefficient and potential nonlinear gain channels would remain closed. In addition, the repulsive optical-dipole potential will cause a radially outward-going momentum spread, and this explains the expansion seen in Fig. 2(c).

Although the growing 3D $\bar{U}_{\rm dipole}$ is very difficult to model theoretically, we can estimate its importance [25]. Intuitively, photons emitted along the long axis of the BEC dominate coherent growth because of maximum propagation gain. From Ref. [18], an internally generated field that originates at one end of an elongated condensate and propagates an effective distance α along the long axis generates an optical-dipole potential of

$$U(\alpha)_{\rm dipole} \approx \hbar\left[\frac{3\lambda^2}{8\pi^2}\frac{\Gamma}{\delta}\left(\frac{N_i}{\tau_0 A}\right)e^{2G\alpha}\right].$$

Here λ is the wavelength of the generated field, N_i is the number of initial seed photons, τ_0 is the pulse length of the initial seed photon burst, and A is the BEC cross section. In addition, $G = 4R\kappa_0 n_0/(\gamma_B\Gamma)$ with $\kappa_0 = (2\pi)^2|d|^2/(\hbar\lambda)$ where $|d|$ is the dipole transition matrix element, R is the single-photon scattering rate, and γ_B is the width of the two-photon Bragg resonance involving a pump and a generated photon. For the BEC in Ref. [1] when $N_i \approx 1$, $\bar{U}_{\rm dipole} \approx -\bar{U}_{\rm MF}$ occurs when $R \approx 100$ Hz, in good agreement with the observed threshold scattering rate.

In the limit of thermal vapors, where there is no mean-field exchange term in the Hamiltonian and the density distribution is more uniform, the process should not depend on the sign of the detuning. The scattering efficiency will be reduced because of shorter coherence times and lower density, but wave-mixing channels (both linear and nonlinear) will remain open since the scattering would already be free-particle-like in nature. To test this hypothesis we applied a pump-laser pulse to a BEC after adiabatically relaxing the magnetic trapping potential in order to lower the density. In this case wave mixing will occur with both the condensed fraction and the uncondensed fraction that results from the expansion not being completely adiabatic. For the condensed fraction the internal-field generation will be the same as before if R is increased to compensate for the lower density. However, the BEC itself will begin to look more like an ultracold thermal gas since the reduction of $\bar{U}_{\rm MF}$ brings the initial system closer to the free-particle-scattering limit. As the system is expanded to a greater degree, less efficient generation of collective atomic recoil modes from the underlying wave-mixing processes should occur with blue detunings for both the thermal fraction and the BEC itself, and this is consistent with what we observe experimentally. For the upper images in Fig. 3, where the magnetic field was lowered to 50% of its original value, the asymmetry is still pronounced. However, for the lower images, where the magnetic field strength was lowered to 10% of its original value (40% uncondensed), symmetry is beginning to be restored, in agreement with our postulation. We also point out that detuning symmetry should also be restored for BECs in the limit $|\delta| \to \infty$ because $\bar{U}_{\rm dipole} \to 0$. In this limit the original structure factor will be unaffected by field growth and its impact on coherent scattering will therefore be the same for both red and blue detunings. The way in which symmetry is restored may identify the presence of nonlinear processes.

In conclusion, we have demonstrated a red-blue detuning asymmetry in matter-wave superradiance and showed that symmetry is restored in the limit of dilute (thermal) atomic vapors. We also provided a plausible explanation for the symmetry breaking based on the BEC's mean-field potential and an induced $\bar{U}_{\rm dipole}$ to stimulate further studies. We believe that the asymmetry results from early-stage growth of a scattered optical field which causes the system to evolve toward (away from) the free-particle scattering limit with red (blue) detunings. With red detunings this

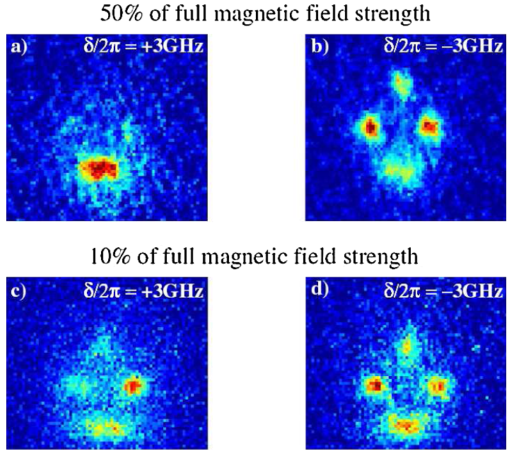

FIG. 3 (color online). Restoration of red-blue detuning symmetry when the magnetic trap is adiabatically relaxed. (a) and (b) $I_P \approx 60\,\mathrm{mW/cm^2}$. (c) and (d) $I_P \approx 110\,\mathrm{mW/cm^2}$. In all images shown the pump duration $= 200\ \mu$s, and the TOF $= 12$ ms. Image size: 620 μm $\times$ 612 μm, number of atoms: 3×10^4.

results in enhanced coherent growth of an ultraslow generated field. However, with blue detunings growth of the generated field is inhibited by the evolving structure factor of the host condensate. At early times in the scattering process, when the genesis of the red-blue asymmetry occurs, the widely accepted grating picture is invalid. However, at late times with red detunings, our model and previous theoretical models converge because the atomic polarization in Maxwell's equation [see Eq. (3) of Ref. [18]] can now be viewed as a grating. We therefore believe that the origin of matter-wave superradiance is fundamentally a multi-matter-optical, wave-mixing process. The suppression of superradiance with blue detunings reported here results from the unique properties of BECs, and will therefore not occur in fermionic or uncondensed bosonic systems. Since the wave-mixing process need not invoke bosonic stimulation, collective atomic recoil motion will occur with fermions [26], but with lower efficiency. Finally, we note that the widely accepted theoretical model of matter-wave superradiance developed over the last decade is incapable of explaining our experimental results because it does not properly address early-stage growth in the scattering process. Since this theoretical framework provides the foundation for many important studies, its revision should be a scientific priority.

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