

Suppression of directional light-wave mixing in normal and quantum gases

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We discuss nonlinear optical processes in the presence of quantum gases. We show that, in contrast to a normal gas, where the medium “passively” participates in nonlinear optical processes, quantum gases “actively” interact with the wave mixing process by enforcing the properties arising from condensed matter physics on the generation and propagation of new light fields. This manifestation of condensed matter physics leads to intriguing suppression and enhancement effects in directional wave generation and propagation processes that have no counterpart in a normal gas. This opens a new chapter on light-matter-wave mixing and scattering: nonlinear optics *with* quantum gases.

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I. INTRODUCTION

Quantum interference is one of the most fundamental aspects and predictions of quantum mechanics. In the early days of quantum mechanics the wave nature of the Schrodinger equation inspired phrases such as “wave mechanics” and “interference physics”, even though the term quantum mechanics is currently the widely accepted description of the discipline [1].

In the field of nonlinear optics there is a class of novel effects that is based on quantum interference between different excitation pathways in an atomic or molecular medium resonantly driven by external fields [2]. The key concept is that a particular excited electronic state can be coupled simultaneously by external fields and an internally generated field produced by nonlinear optical-wave generation processes. The internally generated field, which is usually much closer to the single-photon electronic transition, can thus drive the system in a more efficient manner, creating a different and yet very effective excitation pathway which competes with the resonant excitation by the external fields. The interference of these different pathways can lead to profound quantum interference effects that dominate the dynamics of the system response.

One of the most well-known interference effects is the odd-photon destructive interference phenomenon discovered in the early 1980s and extensively studied throughout the 80s and 90s. In a typical three-photon-assisted multiphoton ionization (MPI) process in an inert gas, distinctive and sharp ionization signals were clearly observable at low medium concentrations. The researchers found [3], however, that the resonantly enhanced MPI signal disappeared completely at elevated medium concentrations. This observation, which is contrary to the naive expectation of a higher ion yield with elevated concentrations, was explained later by the quantum destructive interference between the ionization pathway resulting from the external fields and the pathway from the internally generated field. Payne *et al.* [4–6] showed analytically that excitations to the MPI enhancement state by different pathways are 180° out of phase, resulting in total cancellation of the excitation of the state that is actively and dynamically accessed by several strong light fields. Following these pioneering

works, quantum destructive interference in four-wave mixing [7–11], forward hyper-Raman generation [12–14], and optically pumped stimulated emission [15] have all been observed. In addition, suppression of Autler-Townes splittings and large optical shifts [16,17], destructive interference in $(2n + 1)$ -photon multicolor processes [18], and odd-photon process with resonant even-photon intermediate couplings were also demonstrated.

It should be noted, however, that the media in all of the nonlinear optical processes described above share two features. First, these are all single-specie atomic or molecular vapors sometimes mixed with a foreign gas (when the detuning is significantly larger than the resonance linewidth) in order to compensate the large differential dispersion between the pump and the generated fields. Second, Doppler broadenings and thermal motion cannot be neglected. Media with these properties can be collectively referred to as “normal gases”.

Over the past 15 years, a class of exotic media referred to as “quantum gases” has been realized in laboratories worldwide, bringing new and exciting research opportunities to disciplines such as atomic, molecular, and optical physics and condensed matter physics. One well-known example of a quantum gas is the gaseous-phase Bose-Einstein condensate (BEC) [19–21] produced using laser cooling and trapping techniques. Among the various exotic properties of a quantum gas are its very high degree of coherence and therefore extremely narrow motional-state resonance linewidth. This critically important property of the medium has fundamentally altered the field of nonlinear optics and the physics of coherent generation and propagation of new light fields upon which the quantum interference effects described above rest. Given this paradigm shift, one naturally asks if the well-known quantum destructive interference effect observed in normal gases exists and, if so, how it manifests itself in this new class of quantum gases. The purpose of this paper is to answer these fundamental questions.

Recently light-wave mixing, the most widely studied nonlinear optics process in normal gases, has been extended to Bose-condensed media [22]. In this study, multidirectional nonlinear optical processes were examined for the first time, and both coherent wave propagation effects and the

contribution from elementary excitations of condensed matter physics were investigated. The most startling findings were that there was pronounced enhancement or suppression of multidirectional high-gain hyper-Raman emission processes that are not possible in normal gases. This study represents the first achievement that joins nonlinear optics with condensed matter physics of quantum gases, opening a new research field of *nonlinear optics with quantum gases*.

In the present work, we examine the well-known odd-photon destructive interference discovered in the early 1980s in the context of quantum gases. We reveal the fundamental differences in nonlinear optical quantum destructive interference effects in quantum versus normal gases. Our analysis and results clearly indicate that most nonlinear optical processes studied extensively in normal gases must be carefully re-examined in the context of quantum gases which actively participate in and dynamically affect nonlinear optical processes.

II. THEORETICAL CONSIDERATIONS

To illustrate a typical quantum destructive interference effect in wave propagation we consider, without the loss of generality, a third harmonic generation (THG) process [23] with a single pump field $\mathbf{E}_L(\mathbf{k}_L, \omega_L)$ in a two-level system where the ground and the THG electronic states are referred to as $|1\rangle$ and $|4\rangle$ (Fig. 1) [2], respectively. In a normal gas [Fig. 1(a)] all of the fields propagate within the volume swept out by the pump laser beam, however, the generated field $\mathbf{E}_{\text{THG}}(\mathbf{k}_{\text{THG}}, \omega_{\text{THG}})$ can propagate either parallel or antiparallel to the pump beam. We further assume that pump laser fields \mathbf{E}_{L_j} ($j = 1, 2, 3$; for the THG process $\mathbf{E}_{L1} = \mathbf{E}_{L2} = \mathbf{E}_{L3} = \mathbf{E}_L$) are all linearly polarized along the x axis and that all pump frequencies ω_{L_j} ($j = 1, 2, 3$) are sufficiently detuned from any intermediate one-photon resonance so that the dominant contribution is a three-photon excitation of the electronic state $|4\rangle$ where a THG field $\mathbf{E}_{\text{THG}}(\mathbf{k}_{\text{THG}}, \omega_{\text{THG}})$ is generated. In general, the direction of the pump beam is taken to be the z axis, and in a quantum gas [Fig. 1(b)] this pump propagation direction is assumed to coincide with the long axis of the condensate. In order to characterize the excitation of the THG state $|4\rangle$, we now introduce an effective three-photon-coupling Rabi frequency $\Omega_{41}^{(3)}(\omega_L)$. It is also assumed that the one-photon detuning for the THG field is sufficiently large so that $|\delta_4| \gg \Gamma_4$ and $|\delta_4| \gg |\Omega_{41}^{(3)}|$ [see Fig. 1(a)].

In order to fully appreciate the differences in the nonlinear optical responses of quantum versus normal gases, we begin by examining the wave mixing process in a normal gas. It is well known that THG cannot be efficiently generated in a single-component (pure) normal (non-Bose-condensed or quantum degenerate) atomic gas because the differential dispersion experienced by the pump and THG fields having very different wavelengths quenches any efficient coherent generation gain. Consequently, coherent propagation gain is not supported in any direction. To compensate this differential dispersion a foreign gas is usually added so that coherent generation and propagation gain is supported in the forward direction, where the THG field propagates in the direction of the pump laser [24]. Due to the large optical-wave phase mismatch, dispersion compensation provided by the foreign gas is simply not sufficient to support backward THG. In a

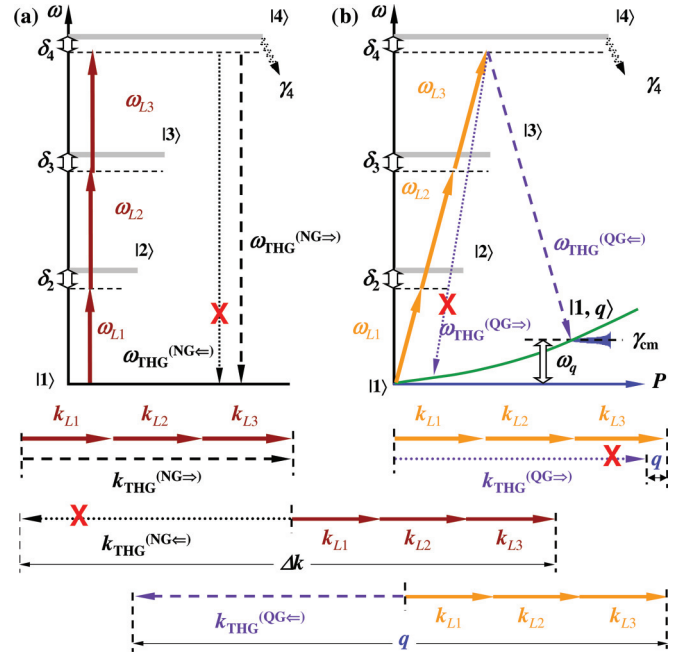


FIG. 1. (Color online) Energy levels, laser couplings, and vector diagrams for forward and backward THG processes in a normal gas (a) and in a quantum gas (b). (a) The vertical dashed arrow represents forward THG emission $[\omega_{\text{THG}}^{(\text{NG} \Rightarrow)}]$, which can be efficiently generated and later leads to the odd-photon destructive interference effect. The vertical dotted arrow with a large cross (X) denotes the backward THG emission process $[\omega_{\text{THG}}^{(\text{NG} \Leftarrow)}]$, which is not supported by propagation gain. (b) The dotted arrow with a large cross (X) denotes the forward emission process $[\omega_{\text{THG}}^{(\text{QG} \Rightarrow)}]$, which is strongly suppressed by the condensate structure factor because of the small quasimomentum transfer. The dashed arrow represents a resonant backward-emitted THG field $[\omega_{\text{THG}}^{(\text{QG} \Leftarrow)}]$, which benefits from extremely efficient hyper-Raman propagation gain.

single-component quantum gas, however, no buffer gas can be added without completely destroying the formation of the quantum gas [25]. Thus, at first glance one would think that THG processes with gain should not be present in quantum gases in any direction.

However, in quantum gases there is a wavelength-dependent differential dispersion compensation mechanism that arises from the intrinsic condensed matter elementary excitation spectrum. A recent study [22] has shown that Bogoliubov dispersion may be able to compensate a residual optical phase mismatch over a broad range of propagation directions. Thus, coherent propagation gain can be achieved in nearly all directions within the medium as long as the medium is fully illuminated by the pump field. In other words, THG is automatically optical-matter-wave phase matched in nearly all directions. It is the condensate's aspect ratio that determines the most efficient propagation direction having maximum gain. This is a very unique and remarkable feature that has no correspondence in normal gases, where coherent propagation gain, if allowed, can only occur efficiently in the direction that achieves the maximum overlap with the pump fields. In the following we examine propagation-based interference effects in normal and quantum gases.

A. The case of a normal gas

In a normal gas with the above-described two-level model, the atomic response and THG wave equations can be expressed as [2]

$$\frac{\partial \rho_{41}}{\partial t} = i\Delta_4 \rho_{41} + i(\Omega_{41}^{(3)} e^{i3k_L z} + \Omega_{41} e^{ik_{\text{THG}} z}), \quad (1a)$$

$$\frac{\partial \Omega_{41}}{\partial z} = i\kappa_{14} \rho_{41} e^{-ik_{\text{THG}} z}, \quad (1b)$$

where $\kappa_{14} = \omega_{\text{THG}}^2 |d_{14}|^2 / (2\epsilon_0 c^2 k_{\text{THG}})$, with d_{14} being the dipole matrix element for the one-photon $|4\rangle \leftrightarrow |1\rangle$ transition. The complex THG detuning is $\Delta_4 = \delta_4 + \Delta_D + i\Gamma_4$, with $\delta_4 = 3\omega_L - \omega_{41} = \omega_{\text{THG}} - \omega_{41}$, Δ_D , and Γ_4 being the laser detuning from the upper electronic state, the Doppler shift, and the Doppler broadened resonance linewidth of the THG state $|4\rangle$, respectively. We have also assumed that $|\delta_4| \gg \Gamma_4$. More rigorously, the right side of Eq. (1b) should be integrated over a velocity distribution, but we have neglected this since it does not affect the main conclusion of the simplified treatment presented here. $\Omega_{41}(\omega_{\text{THG}})$ is the one-photon-coupling Rabi frequency by the THG field. The total momentum-energy mismatch, with nonresonant contributions from a foreign gas included, can be expressed as

$$\Delta K = \Delta k - n_0(\omega_{\text{THG}}) \frac{\omega_{\text{THG}}}{c}, \quad (2)$$

where $n_0(\omega_{\text{THG}})$ is the index due to the foreign gas at the THG frequency. The optical phase mismatch between the THG field and the pump laser fields is given by

$$\Delta k = n(\omega_L) \frac{3\omega_L}{c} \mp n(\omega_{\text{THG}}) \frac{\omega_{\text{THG}}}{c}, \quad (3)$$

where \mp denote forward or backward propagation of the THG field (i.e., the THG field either co- or counter-propagates with respect to the pump laser field). Equation (2) clearly shows that the purpose of the foreign gas is to provide a dispersion compensation to the optical phase mismatch. Indeed, in the forward direction this can lead to $\Delta K \approx 0$ so that efficient propagation gain is supported in the forward direction (see below).

In time-Fourier transform space, the THG field amplitude can be obtained from Eqs. (1a) and (1b),

$$\Lambda_{41}(z; \omega) = -\Lambda_{41}^{(3)}(0; \omega) \left[\frac{e^{i\Delta K z} - e^{-i\kappa_{14} z / (\Delta_4 + \omega)}}{1 + \Delta K (\Delta_4 + \omega) / \kappa_{14}} \right], \quad (4)$$

where Λ_{41} and $\Lambda_{41}^{(3)}$ are the Fourier transforms of Ω_{41} and $\Omega_{41}^{(3)}$, respectively, and ω is the transform variable.

1. Forward generation

In the forward direction, phase-matched THG can be achieved for $|\delta_4| \gg \Gamma_4$, where $|\Delta k| \ll k$, and the phase mismatch can be compensated through the use of a foreign buffer gas, resulting in $\Delta K \approx 0$. On the other hand, under nearly resonant excitation ($|\delta_4| \sim \Gamma_4$) at elevated concentrations, or after a sufficient propagation distance z , the dominant consequence of a Doppler broadened linewidth is that $\text{Re}[e^{-i\kappa_{14} z / (\Delta_4 + \omega)}] \ll 1$. This is the regime where [see Eqs. (1a) and (4)]

$$\Lambda_{41} \approx -\Lambda_{41}^{(3)}, \quad \text{and} \quad \rho_{41} \rightarrow 0, \quad (5)$$

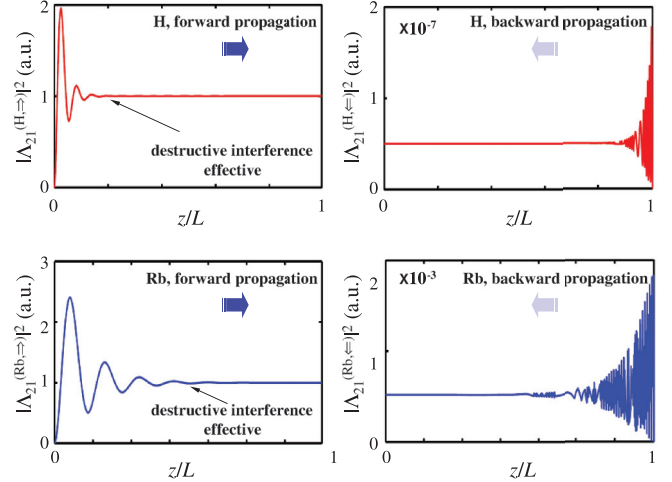


FIG. 2. (Color online) Top: Hydrogen gas. Plot of Eq. (4) for forward emission (left), where optical phase matching $\Delta K \approx 0$ is satisfied (by adding a foreign buffer gas), and backward emission (right), where $\Delta K \approx 6k_L$, which cannot be compensated with a foreign buffer gas. The sample length is $L = 1$ cm and $k_L = 2\pi/\lambda_L$, with $\lambda_L \approx 363$ nm. The density of the gas is about 10^{15} cm^{-3} . Bottom: Rubidium vapor. Plot of Eq. (4) for forward emission (left), where optical phase matching $\Delta K \approx 0$ is satisfied through the use of a foreign buffer gas, and backward emission (right), where $\Delta K \approx 6k_L$, which cannot be compensated with a foreign buffer gas. The sample length is $L = 1$ cm and $k_L = 2\pi/\lambda_L$, with $\lambda_L \approx 2340$ nm. The density of the vapor is 10^{13} cm^{-3} .

indicating that a three-photon destructive interference has occurred and the production of the forward THG field and scattering cease to grow (Fig. 2).

2. Backward generation

In backward THG, Δk is very large and it cannot be compensated for by the dispersion contribution of the foreign gas. This large ΔK leads to fast oscillations that preclude any efficient THG. In other words, there is no efficient coherent propagation gain for the backward THG process (Fig. 2).

B. The case of a quantum gas

As discussed in Sec. I, it has recently been shown that Bogoliubov dispersion arising from condensed matter physics can perfectly compensate the optical-matter-wave phase mismatch in nearly all propagation directions even for a single-component quantum gas. This may lead to the naive conclusion that the above-described three-photon destructive interference will also occur in quantum gases in both the forward and the backward directions. However, as we show below, the nature of fundamental excitations also brings a detrimental impact to directional propagation gain through the condensed matter structure factor. In fact, this structure factor impacts the coherent propagation gain much more severely than the requirement of optical-matter-wave phase matching.

There is an additional complexity in quantum gases of the atomic center-of-mass (CM) motion, which is completely negligible in a normal gas. In fact, the atomic CM motion plays a critical role and fundamentally changes the characteristics of

coherent propagation gain for the THG field [26]. In a normal gas the CM motion of the atoms is completely negligible compared to the large thermal velocity distribution. By contrast, in quantum gases the atomic CM is the dominant feature and therefore must be properly taken into consideration theoretically. We show below mathematically that zero net atomic momentum transfer (also referred to as zero quasimomentum of elementary excitations), while ensuring perfect optical-wave phase matching for forward THG, results in a strong suppression of the forward wave mixing process because the condensate structure factor directly enters the coherent propagation gain coefficient. Thus, simply ensuring light-matter-wave phase matching, as considered in typical nonlinear optics, is no longer sufficient. Properties originating from condensed matter physics now bring about new mechanisms that can significantly impact the light generation process. Indeed, in a quantum gas nonlinear optics and condensed matter physics become intimately coupled and intriguingly intertwined.

In a quantum gas the material equation, corresponding to Eq. (1a) in a normal gas, must be obtained from the Gross-Pitaevskii equation [27] with material density fluctuations included. Instead of the usual atomic wave function $\psi = \sum_j a_j(t) e^{-i\omega_j t} |j\rangle$, the condensate atomic mean-field wave function is given by [27–29]

$$\Psi(\mathbf{r}, t) = e^{i\mu t} \Psi_0(\mathbf{r}) + e^{i\mu t} [u(\mathbf{r}, t) e^{i(\mathbf{q}\cdot\mathbf{r} - \omega_q t)} + v^*(\mathbf{r}, t) e^{-i(\mathbf{q}\cdot\mathbf{r} - \omega_q t)}].$$

Here, $\Psi_0(\mathbf{r})$ is the ground-state condensate wave function in the absence of external fields, and $\mu = g|\Psi|^2$ is the chemical potential. In addition, $\hbar \mathbf{q}$ and $\hbar \omega_q = \hbar^2 q^2 / 2M$ are the quasimomentum and the corresponding atomic CM recoil energy induced by the light scattering process, respectively. This atomic wave function, when inserted into the Gross-Pitaevskii equation, yields the following two material excitation equations of motion that are equivalent to Eq. (1a), but with u and v characterizing the material density fluctuations induced by the light fields [22]:

$$\frac{\partial u}{\partial t} = -\gamma_0 u + i(\omega_q u - Au - Bv - \beta^* \Psi_0 e^{-i\xi}), \quad (6a)$$

$$\frac{\partial v}{\partial t} = -\gamma_0 v + i(\omega_q v + Av + Bu + \beta^* \Psi_0 e^{-i\xi}). \quad (6b)$$

In Eqs. (6a) and (6b) $A = \hbar q^2 / 2M + V_T(\mathbf{r}) + 2g|\Psi_0|^2 / \hbar - \mu$, $B = g\Psi_0^2 / \hbar$, and $\mu = g|\Psi|^2 / \hbar$ is the system chemical potential. $\beta = \Omega_{\text{THG}} \Omega_p^{(3)*} / \Delta_4$ is the effective three-photon excitation Rabi frequency, with Ω_{THG} and Ω_p being the generated and pump-field Rabi frequencies [corresponds to $\Omega_{41}(\omega_{\text{THG}})$ and $\Omega_{41}^{(3)}(\omega_L)$ in the normal gas section]. Furthermore, $V_T(\mathbf{r})$ is the quantum gas trapping potential, $g = 4\pi\hbar^2 a_s / M$, with a_s and M being the s -wave scattering length and the atomic mass, respectively. We have also introduced a multiphoton motional-state resonance linewidth γ_0 which characterizes the damping of elementary excitations. This quantity plays a critical role in high-efficiency wave generation in condensates. For the case of the two-photon process this motion-state resonance line width has already been measured experimentally using the two-laser-beam, coherent motion-state excitation method [30].

It is important to point out that $\xi = (\mathbf{q} - \Delta\mathbf{k}) \cdot \mathbf{r} + (\Delta\omega - \omega_q)t$, with $\Delta\mathbf{k} = \sum_{j=1}^3 \mathbf{k}_{Lj} - \mathbf{k}_{\text{THG}}$ and $\Delta\omega = \sum_{j=1}^3 \omega_{Lj} - \omega_{\text{THG}}$ being the usual optical-wave phase mismatch. This is a generalized phase-matching condition that encompasses both optical waves and elementary excitations.

Before proceeding to the calculation of the coherent propagation gain of the THG field, we briefly comment on the approximations used in the above derivations. In general, the Bogoliubov expansion contains a summation over all possible excitation modes, i.e., $(u, v) \rightarrow (u_n, v_n)$. However, there are several reasons why keeping only a dominant (fundamental) excitation mode as we did is reasonable. First, there is a dominant fundamental Bogoliubov mode that corresponds to the most effective scattering as selected by the gain process and momentum purification process [31]. Other modes do not effectively utilize the high density near the axis and therefore will have much smaller gains. Second, for a uniform gas the dominant momentum transfer is along the long axis and the lowest excitation treatment along this axis is sufficient from both local density approximation and eikonal approximation points of view [28]. Most importantly, however, is the fact that with more quasiparticles included one arrives at a set of linear equations similar to Eqs. (6a) and (6b), but this does *not* introduce any substantial new physics that can alter the conclusion of the treatment. Rather, it only obscures the understanding of the fundamental physical principle. In fact, in a first-order treatment of density fluctuation that is most relevant to light scattering the inclusion of more quasiparticles modes does not lead to any new physics except for an effective correction to the Bogoliubov energy spectrum ω_B (see below) of the primary excitation. Because of the wave propagation gain, this correction can be thought of as a narrow distribution around the sharp fundamental excitation mode we have treated. The key new finding of a resonant denominator by the Bogoliubov excitation remains the same except now this resonance has a small width due to a few nearby spectroscopic excitations. In fact, the angular distribution shown in our work partially reflects this perspective. Thus, it is very reasonable to concentrate on only the most dominant excitation and include a smaller distribution effect. This, by far, gives a clear picture of the underlying physical principle of the process. That is, the Bogoliubov excitation can compensate optical momentum mismatch, leading to an efficient generation in directions that are not possible in a normal gas.

We note that in all BEC studies where absorption imaging is employed for state analysis, the trap is always turned off right after the light is extinguished. There is a well-defined momentum transfer in this trap-off BEC before the time of flight method which allows momentum analysis can be used.

To first order and within the local density approximation [28] the transverse derivatives with respect to the radial coordinates only result in a small shift in the Bogoliubov energy spectrum and hence can be neglected. Furthermore, the derivative along the long axis is also small since the density distribution along the long axis is approximately constant. This is also the case even with the eikonal approximation [28].

To calculate coherent THG propagation effects in a quantum gas, we consider the Maxwell equation governing the

coherent propagation growth of the THG field [23]:

$$i\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right)E_{\text{THG}} + \frac{1}{2k_{\text{THG}}}\nabla_{\perp}^2 E_{\text{THG}} = P_{\text{THG}}. \quad (7)$$

In this case the polarization source term now includes electronic contributions [the right-hand side of Eq. (1b)] and atomic CM contributions from the quantum gas:

$$\begin{aligned} P_{\text{THG}} &= S_{\text{NG}} + S_{\text{QG}}, \\ &= -\hbar\left(\frac{\kappa_{14}}{d_{41}}\right)\left[\frac{\Omega_{41}^{(3)}e^{i\Delta kz-i\Delta\omega t} + \Omega_{41}}{\Delta_4}\right]|\Psi_0|^2 \\ &\quad -\hbar\left(\frac{\kappa_{14}}{d_{41}}\right)\left[\frac{\Omega_{41}^{(3)}}{\Delta_4}\right](u^* + v^*)\Psi_0e^{-i\xi}. \end{aligned} \quad (8)$$

Here $\Delta_4 = \delta_4 + i\gamma_4$, with γ_4 being the natural linewidth of state $|4\rangle$ (no Doppler broadening is present in a quantum gas). Typically, $|\delta_4| \gg \gamma_4$, so that γ_4 can be neglected. Equations (6)–(8) correspond to Eqs. (1a) and (1b).

Following steps similar to those used in deriving Eqs. (1)–(4) for the normal gas and substituting Eq. (8) into Eq. (7), we obtain the total THG in the time–Fourier transform domain as

$$\begin{aligned} \Lambda_{41}(z; \omega) &= \Lambda_{41}(z; \omega)_{\text{NG}} + \Lambda_{41}(z; \omega)_{\text{QG}} \\ &\approx -\Lambda_{41}^{(3)}(0; \omega)\left[\frac{e^{i\Delta kz} - e^{-i\kappa_{14}z/(\Delta_4 + \omega)}}{1 + \Delta k(\Delta_4 + \omega)/\kappa_{14}}\right] \\ &\quad + \Lambda_{41}^{(0)}\exp\left[i\frac{\kappa|\Psi_0|^2 S(q)z}{\omega_B - \Delta\omega + \omega + i\gamma_0}\right]. \end{aligned} \quad (9)$$

Here $\Lambda_{41}^{(0)}$ is the initial vacuum photon at the THG frequency propagating along the direction of highest optical density. In addition, $\kappa = \kappa_{14}|\Omega_{41}^{(3)}|^2/|\delta_4|^2$ [32].

Under the lowest-order approximation with a uniform total trapping potential (sum of magnetic trapping and mean-field potentials), where $V_T = 0$ and $g|\Psi_0|^2/\hbar = \mu$ [see Eq. (6)], the condensate structure factor $S(q)$ and Bogoliubov elementary excitation spectrum ω_B are given by the Feynman relation [28,33–35]:

$$S(q) = \frac{\hbar q^2}{2M\omega_B(q)}, \quad \omega_B(q) = \sqrt{\frac{\hbar q^2}{2M}\left(\frac{\hbar q^2}{2M} + \frac{2g|\Psi_0|^2}{\hbar}\right)}.$$

The first term in Eq. (9) is exactly Eq. (4) for a normal gas with an added linear absorption but, without any buffer gas, represents the usual electronic contribution. Note that $\Delta k = 3k_L - k_{\text{THG}}$ and $\Delta\omega = 3\omega_L - \omega_{\text{THG}}$ represent the usual optical-wave phase mismatch for the THG process in the *absence* of a foreign buffer gas. Without any buffer gas to assure optical phase matching, Δkz is large even for a condensate only 100 μm in length. This results in a fast oscillation that diminishes contributions of electronic origin regardless of whether the THG is colinear or anticolinear with respect to the pump field. Thus, the first term is negligibly small and can be neglected.

The second term in Eq. (9) arises from the condensed matter physics of quantum gases and it is this term that gives rise to new physics. We note that mathematically the generalized phase-matching condition, $\xi = (\mathbf{q} - \Delta\mathbf{k}) \cdot \mathbf{r} + (\Delta\omega - \omega_{\mathbf{q}})t$, cancels out identically, indicative of automatic phase matching, which is a well-known feature of Raman and hyper-Raman

wave mixing processes. We further point out that there is a resonance denominator that relates the Bogoliubov elementary excitation of condensed matter physics and the energy relation of the optical fields. This feature is the foundation of the new physics that we refer to as *light-wave mixing with quantum gases*. The physical meaning of this contribution from the quantum gas is that the quasimomentum transfer \mathbf{q} and associated elementary excitations will lead to a Bogoliubov dispersion that compensates the energy deficit between the fields involved, resulting in $\omega_B = \Delta\omega$ and, thus, a resonance in the THG gain. While this new Bogoliubov-excitation-based resonance appears to support highly efficient THG gain in any direction, limited only by the optical depth, we show below that the properties arise from condensed matter physics also significantly impact the wave mixing and propagation process, resulting in efficient coherent gain of the THG field *only* in the backward direction, which, in the case of a normal gas, is not allowed even in the presence of a dispersion-compensating foreign buffer gas. This fascinating physics has no counterpart in a normal gas.

Contrary to the diminished electronic contribution (i.e., normal gas part), the contribution from the quantum gas has a very different nature. Because of the lack of a foreign gas, a finite CM quasimomentum transfer to the atom is required so that Bogoliubov elementary excitations lead to $\omega_B = \Delta\omega$. Thus, Eq. (9) gives

$$\Lambda_{41}(z; \omega) \approx \Lambda_{41}^{(0)}\exp\left[\frac{\kappa|\Psi_0|^2 S(q)z}{\gamma_0}\left(1 + i\frac{\omega}{\gamma_0}\right)\right], \quad (10)$$

where we have assumed a long pump pulse length τ so that $\gamma_0\tau \gg 1$. It is important to point out that when $S(q) \rightarrow 1$ and $\gamma_0 \rightarrow \gamma_{2\text{ph}}$ (where $\gamma_{2\text{ph}}$ is the two-photon motional state resonance line width), Eq. (10) reduces exactly to the result derived in [36] and [37] for the case of two-photon backward scattering, as it should. Equation (10) is profoundly different from that of a normal gas [see Eq. (4)] in two aspects:

(1) The THG field increases linearly in a normal gas, whereas in a quantum gas it grows exponentially. In fact, in quantum gases, where atomic CM recoil motion is a pronounced feature, all wave mixing processes become Raman or hyper-Raman in nature [2,12–15].

(2) In the case of $|\delta_4| \gg \Gamma_4$, the THG field can only be efficiently generated in the forward direction in a normal gas with a dispersion compensating foreign gas. In a quantum gas, however, phase matching is automatically satisfied as a consequence of the hyper-Raman gain described above, and the condensed matter Bogoliubov dispersion leads to a resonant denominator that allows efficient generation in nearly all directions, limited only by the condensate structure factor (see discussion below) and consideration of the optical depth.

While the first aspect above can be easily understood from the leading term in Eq. (10), the second aspect requires understanding features rooted in condensed matter physics. On the surface, it appears that forward generation should be favored in a quantum gas. However, closer study reveals that condensed matter physics plays a unique role and significantly impacts this nonlinear optical process. We show here that although forward scattering is automatically phase matched

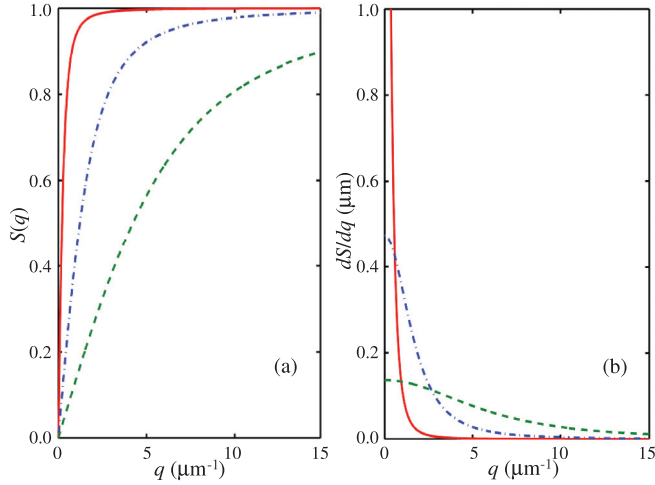


FIG. 3. (Color online) (a) Plot of condensate structure factor for H and Rb (at two densities). (b) Derivatives of the structure factor as a function of quasimomentum transfer. The solid curve is for hydrogen at a density of about 10^{15} cm^{-3} . The dash-dotted curve is the structure factor of a rubidium condensate having the same density as the hydrogen condensate. The dashed curve is the structure factor of a rubidium condensate with a density of 10^{13} cm^{-3} .

and propagates in the direction of greatest optical depth, it is actually strongly suppressed by the condensate's structure factor $S(q)$, which appears in the coherent propagation gain coefficient seen in Eq. (10). From condensed matter physics, it is known that the structure factor changes dramatically when the quasimomentum transfer of the elementary excitation is significantly reduced (see Fig. 3).

C. Forward generation

When the THG field propagates colinearly with respect to the pump field, $q \approx \Delta k = \sum_j^3 k_{Lj} - k_{\text{THG}} \neq 0$, but it is small. This residual momentum transfer arises because of the differential dispersion between the pump and the THG fields (the actual size of this quasimomentum depends on the THG detuning). For the very small q in Fig. 3, it is shown that this small residual quasimomentum by photon exchange during the THG process leads to a drastic reduction in $S(q)$, which significantly quenches the gain coefficient. In this regime of small quasimomentum transfer the scattering is not free-particle-like and the dramatic reduction of $S(q)$ directly impacts the new optical-field generation and coherent propagation processes.

It can also be seen that the structure factor of a hydrogen BEC rises much more rapidly than a rubidium BEC as a function of q . In addition, for high harmonic generation, the residual quasimomentum transfer is dictated by the energy of the mixing wave photons. Consequently, one expects that forward suppression in a hydrogen BEC is much less pronounced than in a rubidium BEC because eventually there will be small fraction of forward emission that becomes amplified. This is one of the many unique aspects of quantum gases that has no correspondence in a normal gas.

We further point out that in the forward direction $S(q)$ also significantly affects the ultraslow group velocity of the

THG field. This results in a rapid increase in group velocity as the momentum transfer decreases, and this further reduces the gain. This negative feedback in the forward emission process vividly demonstrates the important dynamic role played by the SFG field. We emphasize, however, that the forward suppression described here is *fundamentally* different from the forward THG suppression shown in Eq. (5) or the hyper-Raman suppression in a normal gas [2,12–15]. The former requires a foreign gas for dispersion compensation to achieve dynamic excitation suppression, whereas the latter requires a separate wave mixing channel (different excitation pathway) for destructive interference. Both, however, do not invoke condensed matter physics. In the case of a quantum gas the suppression arises solely because of the condensed matter properties of the medium.

D. Backward generation

When the THG field propagates anticolinearly with respect to the pump fields, $q \approx \Delta k = \sum_{j=1}^3 k_{Lj} + k_{\text{THG}}$ is very large. The corresponding Bogoliubov energy is predominately $\hbar\omega_B \approx \hbar^2 q^2 / 2M$, and this renders the scattering process free-particle-like. Under this condition $S(q) \rightarrow 1$, and maximum light-wave gain and collective atomic recoil motion are achieved [see the limit of large q in Fig. 3(a)]. Thus, contrary to a normal gas, where backward generation is *not* supported even in the presence of a foreign gas, backward generation in a quantum gas is the most efficient process and it even benefits by propagating along the largest optical depth of the systems studied in this work.

III. COMMENT ON THE TWO-PHOTON RAMAN-RAYLEIGH SCATTERING PROCESS

Before presenting numerical examples of THG in quantum gases, we comment on two-photon Raman-Rayleigh scattering processes, which have been widely studied over the past 15 years. Generally speaking, these studies can be classified into two categories, based on the methodologies used. The first class includes studies where light-matter interactions are considered from a nonlinear optics viewpoint [36–53]. In such treatments the dipole approximation in the light-matter interaction Hamiltonian is widely used, and the atomic mean-field wave function is described by the Gross-Pitaevskii equation. The propagation of the scattered field is treated by Maxwell's equation analytically [36,37] and numerically [45,50–53]. While these theories can correctly predict the momentum components of the scattered atoms [36–53], the overall time gain [38–53], and the scattered-photon propagation gain [36,37], they completely neglect fundamentally important contributions by condensed matter physics. Indeed, none of these theories can predict the suppression of the forward-scattered field precisely because such prominent features of the process originate from condensed matter physics.

The second class of studies focuses primarily on the condensed matter physics of the atomic response [28,29,33,34,49]. In these treatments the condensate structure factor and related condensed matter quantities are studied from a fundamental excitation viewpoint, but the generation and propagation of the electric field are neglected. Although these theories have

provided important understandings of the light-matter interaction from the perspective of condensed matter physics, they cannot explain very important features of the scattered light field, simply because they do not treat coherent propagation growth of the scattered field. One primary example of the pitfalls of such a condensed matter treatment is its failure to explain the underlying physics behind the forward-scattering process. Even though $q \neq 0$ in the forward direction in a multiphoton wave mixing process such as THG because of the differential dispersion, $q \equiv 0$ does occur in the case of a two-photon Raman scattering process since both the photon momentum and the dispersion relation are automatically conserved and phase matched. We emphasize, however, that the simple hand-waving argument of zero quasimomentum transfer to atoms in such a superinelastic scattering process, while resulting in a reduction in atom scattering (i.e., no atomic CM motion) by the reduced condensate structure factor, is *not* a sufficient argument [33]—nor is any condensed matter argument that is solely based on momentum-energy conservation [49] for the suppression of forward light generation. In fact, in a normal gas in such a geometry where negligible atomic CM motion occurs, the wave generation and propagation process shows the highest efficiency. Suppression of the scattered field by the structure factor arising from Bogoliubov excitations in such a perfect photon momentum conservation case can only be obtained by solving Maxwell's equation for the scattered field with the explicit atomic density treatment as demonstrated by Eqs. (6)–(10) and, also, in Ref. [22]. Only this theory, which combines nonlinear optics with condensed matter physics, can provide an accurate description of the underlying physics of light-matter interactions in the context of quantum gases. Indeed, it is this combined new theory that underscores the importance of the term “nonlinear optics *with* quantum gases”.

IV. NUMERICAL EXAMPLES

The rest of this work focuses on presenting two numerical examples. First, we consider a hydrogen condensate of length $L = 5$ mm with a transverse Thomas-Fermi radius of $r_0 = 10$ μm containing of the order of 10^9 atoms [54]. We take $g/\hbar = 1.64 \times 10^{-17} \text{ m}^3 \text{ s}^{-1}$ (corresponding to a scattering length of $a_S = 0.41a_0$), $\gamma_0/2\pi = 6$ kHz, and $d_{14} = 6.38 \times 10^{-30}$ C m. The pump laser power is chosen such that the three-photon pump rate $R^{(3)}/2\pi = \gamma_4 |\Omega_L^{(3)}|^2 / [4(\delta_4^2 + \gamma_4^2)] \approx 45$ Hz. Accordingly, the pump laser wavelength should be $\lambda_{L1} = \lambda_{L2} = \lambda_{L3} = \lambda_L \approx 363$ nm. In Figs. 4(a) and 4(b) we show forward and backward THG in a hydrogen condensate. In this case the $2P$ -state linewidth is about $\gamma_2/2\pi = 625$ MHz, and we have chosen $\delta_4/2\pi \approx -2$ GHz. This leads to a small nonlinear index change which results in a much smaller $q \approx 3k_L - k_{\text{THG}}$ in the forward direction. Correspondingly, $S(q) \ll 1$ for hydrogen condensates, and therefore a very strong suppression of forward THG [Fig. 4(a)] is expected. In the backward direction q is necessarily large, resulting in $S(q) \rightarrow 1$ and maximum THG [Fig. 4(b)].

It is interesting to point out the substantially narrowed THG-field transverse spread. Initially, the THG transverse distribution is dictated by the initial Thomas-Fermi distribution, which was numerically evaluated by solving the stationary Gross-Pitaevskii equation (solid line in Fig. 4). As the THG

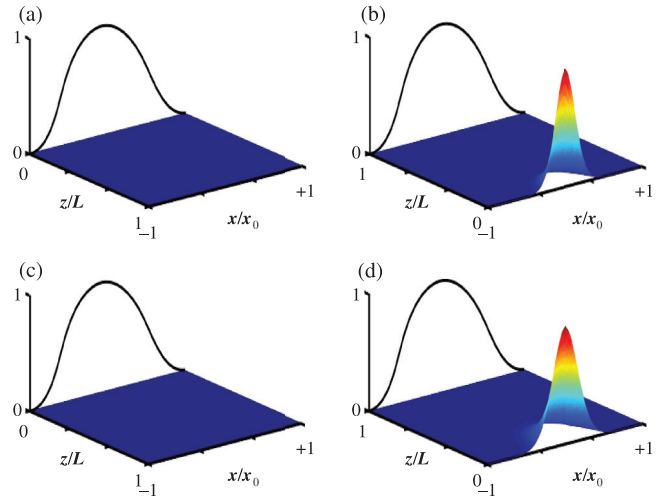


FIG. 4. (Color online) Plot of forward (a) and backward (b) emissions in a hydrogen condensate and forward (c) and backward (d) emissions in a rubidium condensate. The vertical axis denotes the photon numbers of the THG normalized with respect to the backward generation of the corresponding condensate. The curves represent the initial field distribution, which mimics the initial distribution of the condensate obtained by numerical solution of the stationary Gross-Pitaevskii equation. The significant narrowing of the THG transverse distribution in both backward emission processes signifies the hyper-Raman gain with inhomogeneous density distribution.

propagates in a condensate which has an inhomogeneous density distribution, the THG field grows more rapidly than a simple exponential gain, causing significant narrowing of the THG-field transverse distribution. This is unique to gaseous phase BECs since the atomic CM motion is a dominant feature. Indeed, it has been shown that the rapid narrowing, and therefore faster-than-exponential-gain growth characteristics, of the THG field leads to a significant transverse compression for red-detuned, long-pulsed pump fields. Under such a strong compression effect atoms can move appreciable distances during the duration of the pump pulse, resulting in further increase in the density near the center along the long axis where THG is most intense. Consequently, the THG field grows even more rapidly where the density is highest. This positive-feedback mechanism for THG production is unique to quantum gases such as a BEC, and it has no counterpart in nonlinear wave generation processes in normal gases.

In the second example we consider a rubidium condensate of length $L = 140$ μm , with a transverse Thomas-Fermi radius of $r_0 = 10$ μm , containing about 10^6 atoms. Here, we take $g/\hbar = 4.85 \times 10^{-17} \text{ m}^3 \text{ s}^{-1}$ (corresponding to a scattering length of $a_S = 100a_0$), with $d_{14} = 3.4 \times 10^{-29}$ C m. The pump laser power is chosen so that the three-photon pump rate is $R^{(3)}/2\pi = \gamma_4 |\Omega_L^{(3)}|^2 / [4(\delta_4^2 + \gamma_4^2)] \approx 20$ Hz. Accordingly, the pump laser wavelength should be $\lambda_{L1} = \lambda_{L2} = \lambda_{L3} = \lambda_L \approx 2340$ nm. In Figs. 4(c) and 4(d) we show forward and backward THG in such a rubidium condensate. In this case the $5P$ -state linewidth is about a factor of 100 times narrower than the $2P$ state of hydrogen. This permits the significantly smaller THG detuning of $\delta_4/2\pi \approx -30$ MHz ($\gamma_4/2\pi \approx 6$ MHz). The corresponding nonlinear index change is fairly large, and this results in a quasimomentum change that is larger than in

the case of hydrogen in the forward direction. However, the suppression due to the corresponding $S(q)$ is still substantial [Fig. 4(c)]. Using the above parameters, we found that there are about 500 times fewer photons generated in the forward direction compared to the number of THG photons generated in the backward direction, where q is very large and $S(q) \rightarrow 1$ results in the highest gain [Fig. 4(d)]. Note that the THG transverse distribution is again narrowed significantly, but it is still a bit broader than in the case of hydrogen. This occurs because the density of the rubidium condensate is lower than that of the hydrogen condensate.

Before we present our conclusions, two important aspects must be pointed out and addressed. First, the “backward” emission and generation discussed in this work are the result of the chosen pumping geometry, in which the pump laser propagates colinearly with respect to the condensate’s long axis. In general, the direction of the enhanced emission of Raman or THG is determined by the orientation of the condensate’s long (high-gain) axis, even if the pump laser propagation direction is not aligned with the long axis. In such cases the maximum gain direction is not in the “backward” direction with respect to the pump laser but, rather, along the long axis of the condensate. However, without a large aspect ratio, and therefore an advantageous direction for maximum gain, the new field will be generated in all directions without the advantage of a momentum-state purification process [31]. In such cases, the generated light will have the full randomness and fluctuations of a spontaneous Raman or hyper-Raman radiation process.

Second, there is a view point that attempts to argue that the treatment presented here, and its two-photon analog, are just simply Bragg scattering. This viewpoint which is often put forth to support the concept of “matter-wave superradiance” [31], first *artificially* renames $(E_L)^3$ as E_{eff} and then argues that, because the Hamiltonian of the THG process can be simplified into a bilinear product of two electric fields as $(E_L)^3 E_{\text{TH}}^* = E_{\text{eff}} E_{\text{TH}}^*$, therefore the process

is just Bragg scattering. We emphasize that such arguments are fundamentally incorrect: (a) traditional Bragg scattering does not invoke coherent propagation growth, nor does it include the initial quantum fluctuations of the system; and (b) the bilinear field operator format appears in many disciplines of physics such as optical physics, condensed matter physics, high-energy physics, and even astrophysics. Even in the optical field, according to this superficial Bragg-scattering-for-all “classification”, Raman scattering and a four-wave mixing process appear to have the “same” bilinear electric-field format, i.e., $E_1 E_R^*$ and $E_1 E_2 E_3 E_{\text{TH}}^* = E_{\text{eff}} E_{\text{TH}}^*$. However, it is well known that these two processes have very different quantum fluctuation, coherence, and even propagation gain characteristics. It is incorrect to claim that Raman scattering and four-wave mixing are just Bragg scattering processes, and it is false to claim that if Hamiltonians share a similar format after some meaningless operator regrouping, they therefore represent the same physics.

V. CONCLUSION

In conclusion, we have presented a detailed study of nonlinear optical processes in the presence of a quantum gas. We have shown that, unlike the case of a normal gas, where the medium *passively* participates in the nonlinear optical process, a quantum gas *actively* interacts with the light-field generation and propagation process by exerting and enforcing effects arising from condensed matter physics. This leads to intriguing new features, such as suppression or enhancement of directional wave generation processes, which have no correspondence in a normal gas. This opens a new chapter on light-matter interactions that we refer to as nonlinear optics *with* quantum gases. Indeed, most nonlinear optical effects with normal gases must be re-examined in the context of quantum gases, where condensed matter physics can lead to novel effects never before realized.

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