

Light-wave mixing and scattering with quantum gases

L. Deng and E.W. Hagley

National Institute of Standards and Technology, Gaithersburg, Maryland 20899 USA

C.J. Zhu

School of Physical Science and Engineering, Tongji University, Shanghai 200092, China

E-mail: lu.deng@nist.gov

Abstract: We show that optical processes originating from elementary excitations with dominant collective atomic recoil motion in a quantum gas can profoundly change many nonlinear optical processes routinely observed in a normal gas. Not only multi-photon wave mixing processes all become stimulated Raman or hyper-Raman in nature but the usual forward wave-mixing process, which is the most efficient process in normal gases, is strongly reduced by the condensate structure factor. On the other hand, in the backward direction the Bogoliubov dispersion automatically compensates the optical-wave phase mismatch, resulting in efficient backward light field generation that usually is not supported in normal gases.

1. Introduction

Since the invention of the laser in 1960, the field of nonlinear optics has exploded. In a typical nonlinear optical process, often referred to as coherent wave-mixing, several photons are absorbed simultaneously by an atomic or molecular medium, and a photon with a different frequency is produced. Under the right conditions, the generated radiation may be coherently amplified by the quantum mechanical mechanism of stimulated emission identified by Albert Einstein. Today, 95 years after Einstein's prediction, we are surrounded by practical applications of nonlinear optics, from multi-channel telecom lasers to the common laser pointer. Within the past decade, a new type of coherent wave mixing has emerged, which involves the interplay between light and matter waves.

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 [1]. 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 multi-photon ionization (MPI) process in an inert gas, distinctive and sharp ionization signals were clearly observable at low medium concentrations. The researchers found [1], 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.* [2–4] showed analytically that excitations to the MPI enhancement state by different pathways are 180 degree out of phase, resulting in total cancellation of the excitation of the state that is actively and dynamically accessed by several strong light fields.

Recently light-wave mixing, the most widely studied nonlinear optics process in normal gases, has been extended to Bose-condensed media [5–7]. 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*.

2. Theory

We consider the most studied nonlinear optical wave mixing process where the medium is a quantum gas such as a Bose-Einstein condensate or ultra-cold degenerate Fermion gas. Specifically, we focus on the third harmonic generation (THG) process well studied in the case of normal gas for the past 50 years [8]. We show that the THG can be generated in a quantum gas under the circumstances where it would have been impossible in the case of a normal gas.

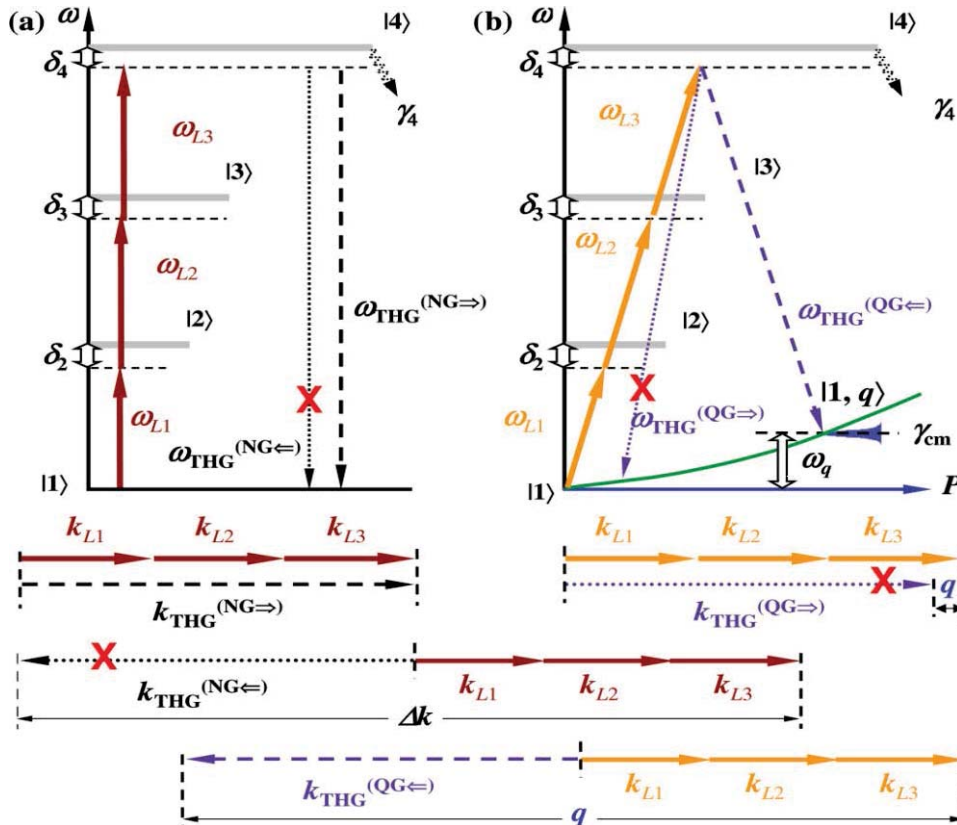


Figure 1: (a) Energy diagram and laser couplings for the THG in a normal gas where atomic center of mass motion is totally negligible. Phase-matched THG can be efficiently generated only in the presence of suitable buffer gas and only in the forward direction. (b) Energy diagram and laser couplings for the THG in a quantum gas where the atomic center of mass motion plays dominant role. Phase-matched THG can be efficiently generated only in the backward direction without the assistance of the buffer gas.

For a normal gas (see Fig. 1a), the THG field can be obtained by solving the Maxwell equation for the THG field amplitude which oscillates at 3ω (here, ω is the angular frequency of the fundamental field, i.e., the pump laser field). By solving the Maxwell equation and the material equation simultaneously one obtains, in the time Fourier transform space, the THG field [1]

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

where ΔK is the general phase mismatch including contributions from both atomic medium and the buffer gas which provide an overall index compensation in order to achieve phase matched coherent constructive interference. Δ_4 is

the third-photon detuning and κ_{14} depends on the atom number density [1]. Without the compensatory contribution from the buffer gas, it is not possible to make $\Delta K \sim 0$ and consequently the efficiency for THG is small. The key feature of this wave mixing process in a normal gas is that the THG field is sinusoidally dependent on the propagation distance in the forward direction (i.e., the direction of the pump laser) when $\Delta K = 0$ is satisfied. Only when the propagation distance $z = L$ is much large than the decay characterized by $|\kappa_{14}L/(\Delta_4 + \omega)| \gg 1$, can one obtain $I_{41}(L; \omega) \propto I_{41}^{(3)}(0; \omega)$.

For a quantum gas (Fig. 1b) the atomic center of mass motion plays a dominant role and therefore overall system momentum transfer must be tracked to single photon recoil. Furthermore, density-density correlation plays a critical role in the overall THG efficiency and the most optimum direction for the phase matched wave to be produced. In the most case of quantum gas such as Bose condensate and degenerate Fermi gas, no buffer gas is allowed and the corresponding index compensation mechanism does not exist. All these novel circumstances associated with a quantum gas indicate that new physics of THG in a quantum gas is anticipated. Starting with the macroscopic atomic mean field wave function the polarization source term in the Maxwell equation for the THG frequency can be separated into the usual electronic contribution and a new term that arises from the elementary excitation rooted in quantum gas density fluctuation. The dominant contribution to the generation of the TH field is this elementary excitation since in the absence of the buffer gas the electronic contribution cannot achieve the phase matched coherent growth condition. Solving the Maxwell equation and the Gross-Pitaevskii equation simultaneously, in the Fourier space the THG field is given by [9,10]

$$\Lambda_{41}(z; \omega) = \Lambda_{41}^{(0)} \exp \left[i \frac{\kappa |\Psi_0|^2 S(q) z}{\omega_B - \Delta\omega + \omega + i\gamma_0} \right]. \quad (2)$$

where $S(q)$ is the quantum gas structure factor and ω_B is the Bogoliubov excitation spectrum ($\kappa |\Psi_0|^2 = \kappa_{14}$). Equation (2) is fundamentally different from the corresponding THG in normal gas given in Eq. (1). The first distinctive difference is that the THG process in a quantum gas is Raman-like (or hyper-Raman-like for multi-photon processes) with the typical exponential gain characteristics. It is this exponential gain characteristics of the THG (and indeed in any wave mixing processes) that prohibits any forward odd-photon destructive interference effect to occur in a quantum gas. These multi-photon interference effects in the forward direction have been widely observed and extensively studied in the last 30 years in the field of nonlinear optics. The second important difference is that in the case of a quantum gas, the dominant scattering and THG is in the backward direction (i.e., opposite the pump laser direction). This is a critically important condensed matter feature that does not have a correspondence in a normal gas and its impact to all nonlinear optical processes in a quantum gas can be seen from the condensate structure factor $S(q)$ contained in the THG gain parameter. It is well-known from condensed matter physics that when the quasi-momentum $q \rightarrow 0$, therefore the forward direction of THG, $S(q) \rightarrow 0$, implying a strong suppression of the forward THG production. On the other hand and for an elongated quantum gas, $S(q) \rightarrow 1$ in the backward direction, indicating that the most efficient direction of THG production, i.e., the overall phase matching direction is the backward direction. It is the quasi-momentum q from the Bogoliubov elementary excitation that provides the necessary momentum conservation that must be met in the wave generation process. In the case of normal gas, however, the backward direction can never be phase matched and efficient generation of THG is strongly prohibited. The third key difference shown in Eq. (2) in the case of a quantum gas is the resonant denominator that contains the Bogoliubov energy spectrum. It is this part of the elementary excitation that ensures the energy conservation, leading to a generalized matter-optical wave mixing phase matching condition. In the latter case, the condensed matter elementary excitations provide a novel phase matching role and all wave mixing processes become Raman/hyper Raman-like.

For a normal gas, the general phase matching conditions are purely optical and are given by

$$\Delta K = k_{3\omega} - 3k_{\omega} - k_{bg} = 0 \quad \text{and} \quad \omega_{THG} - 3\omega = 0.$$

For a quantum gas the generalized phase matching conditions include fundamental excitation and are given by

$$\Delta K = k_{3\omega} - 3k_{\omega} - q = 0 \quad \text{and} \quad \omega_B(q) - [\omega_{THG} - 3\omega] = 0.$$

In Fig. 2 we show the relative THG field intensity in a normal gas in the forward direction (upper-left) and in the backward direction (upper-right) as the function of THG propagation distance for a typical rubidium vapor at room-temperature. These results show that forward THG is very efficient and later reaches a regime where the further growth of the THG is limited by the well-known three-photon destructive interference effect [1]. In addition, the THG in the backward direction is strongly prohibited.

Also shown in Fig. 2, as the comparison with the normal gas, the relative THG field intensity produced in a quantum gas. Here the rubidium atoms are cooled by laser cooling and evaporative cooling methods to below the momentum state condensation point, forming an exotic coherent matter material referred to as Bose-Einstein condensate. In the forward direction, i.e., the direction of the pump laser the THG is strongly suppressed by the condensate structure factor with small quasi-momentum transfer. In the backward direction, however, the generalized wave mixing and scattering phase matching conditions for the quantum gas are satisfied and the production of THG is most efficient.

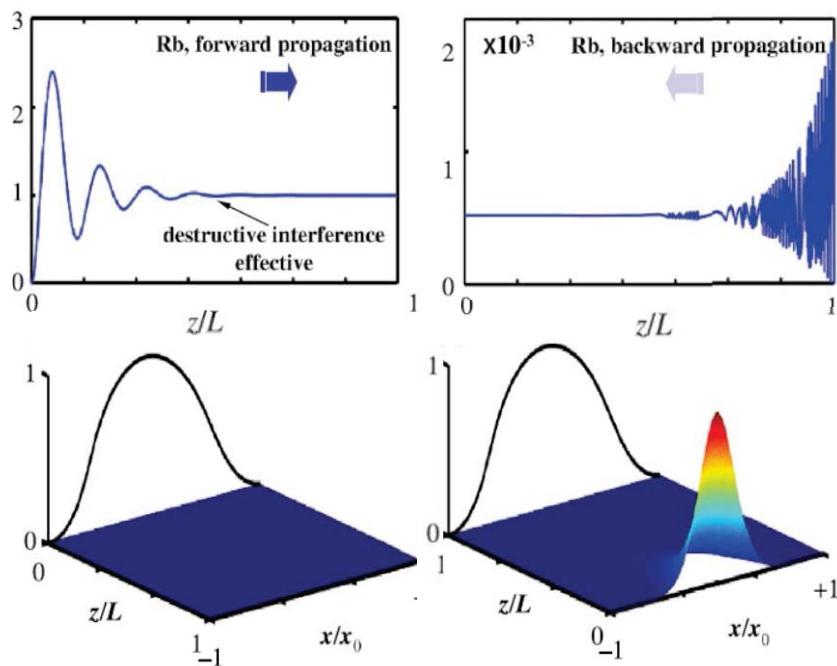


Figure 2. Right Panel: Upper plots: normal gas THG (vertical axis) as a function of normalized propagation distance. Lower plots: quantum gas THG (vertical axis) as a function of normalized propagation distance and transverse radius.

3. Conclusion

In conclusion, unlike the case of a normal gas where the medium passively participates in the nonlinear optical process, a quantum gas the medium actively participates the light generation and propagation process by exerting and enforcing effects arising from condensed matter properties of the medium. This opens a new chapter of light-matter interactions that can only be explained by applying nonlinear optics and condensed matter physics, and hence the new field of nonlinear optics with quantum gases. Indeed, all nonlinear optical processes studied in normal gas and solid-state materials must be revisited in the context of coherent matter wave and quantum gas.

References

- [1] L. Deng, M.G. Payne, and W.R. Garrett, Phys. Rep. 429, 123 (2006).
- [2]. M.G. Payne, W.R. Garrett, and H.C. Baker, Chem. Phys. Lett. 75, 468 (1980)

- [3]. M.G. Payne and W.R. Garrett, *Phys. Rev. A* 26, 356 (1982).
- [4]. M.G. Payne and W.R. Garrett, *Phys. Rev. A* 28, 3049 (1983).
- [5] M.H. Anderson et al., *Science* 269, 198 (1995).
- [6]. K.B. Davis et al., *Phys. rev. Lett.* 75, 3969 (1995).
- [7]. C.C. Bradley et al., *Phys. Rev. Lett.* 78, 985 (1997).
- [8] Y.R. Shen, "The Principles of Nonlinear Optics", (John Wiley & Sons, New York, 1984).
- [9] L. Deng, C.J. Zhu, and E.W. Hagley, *Phys. Rev. Lett.* **110**, 210401 (2013)
- [10]. L. Deng, C.J. Zhu, E.W. Hagley, and W.R. Garrett, *Phys. Rev. A* 88, 043642 (2013)