Simultaneous Multiphoton-Multiatom Processes in Atomic Gases under Laser Fields

Authors: Yongle Yu, Yongle Yu

Date: 2025-04-07T00:00:00+00:00

Abstract

We investigate simultaneous multiphoton-multiatom processes in atomic gases exposed to laser fields under specific frequency conditions, where multiple atoms are simultaneously excited through the absorption of one laser photon each. These processes represent natural high-order quantum electrodynamics (QED) effects that occur independently of inter-atomic interactions. A characteristic length scale emerges, governing the physical range over which these phenomena manifest. We propose experiments to demonstrate the fundamental aspects of these collective QED processes.

Full Text

Preamble

Simultaneous Multiphoton-Multiatom Processes in Atomic Gases under Laser Fields

Yongle Yu*
State Key Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Wuhan Institute of Physics and Mathematics, Chinese Academy of Science, West No. 30 Xiao Hong Shan, Wuchang, Wuhan, 430071, China

yongle.yu@wipm.ac.cn

Abstract

We investigate simultaneous multiphoton-multiatom processes in atomic gases exposed to laser fields under specific frequency conditions, where multiple atoms are simultaneously excited through the absorption of one laser photon each. These processes represent natural high-order quantum electrodynamics (QED) effects that occur independently of inter-atomic interactions. A characteristic length scale emerges, governing the physical range over which these phenomena

manifest. We propose experiments to demonstrate the fundamental aspects of these collective QED processes.

Quantum electrodynamics (QED) serves as the cornerstone for understanding the interaction between matter and light, providing profound insights into atomic and subatomic processes. When atoms are exposed to coherent electromagnetic fields, such as those produced by lasers, a wide range of phenomena emerges, spanning from single-photon absorption to intricate multiphoton transitions. Among these, multiphoton processes [1–5]—where a single atom simultaneously absorbs multiple photons—represent high-order QED effects that have been extensively studied, leading to significant applications in nonlinear optics, laser physics, precision spectroscopy, and biological imaging. However, relatively less attention has been directed toward another class of high-order QED processes: those involving the simultaneous transitions of multiple atoms, each absorbing a laser photon with a frequency that deviates significantly from its respective atomic transition frequency. These processes, characterized by joint atomic behavior, occur naturally within the framework of QED and represent an intriguing area for exploration in the interaction between light and matter.

In this paper, we investigate some fundamental aspects of these multiphoton-multiatom (MPMA) processes. Notably, we highlight that such processes do not require mediation by interatomic interactions. Furthermore, we demonstrate that the transition rates of these processes can be substantially enhanced in the presence of a large number of atoms, which could lead to new possibilities for studying ultra-weak atomic phenomena. We also propose experiments to directly test several theoretical findings presented in this work.

To provide a general illustration of multiphoton-multiatom processes, we begin with an analysis of a two-photon-two-atom process. A number of theoretical studies have addressed two-photon-two-atom processes, including those in [6–11]. Notably, two pioneering experimental works [12, 13] observed such processes and offer comparative insights into their analysis. Furthermore, an analogous two-photon-two-molecule process has also been reported [14]. Consider a system of two non-interacting atoms, with one atom being of species A and the other of species B, exposed to a homogeneous laser field (see Fig. 1 [Figure 1: see original paper]). Each atom is assumed to be a two-level system, and the transition between its levels occurs via a dipole interaction with the electromagnetic field. Denote the ground state of the A-species atom by |g_a with an energy _a^g, and its excited state by |e_a with an energy _a^e. For the B-species atom, the ground state |g_b has an energy of _b^g, and the excited state |e_b has an energy of _b^e. The angular transition frequency of the A-species atom is ω_a = (_a^e - _a^g)/ , where is Planck' s constant, while the angular transition frequency of the B-species atom is ω_b = (_b^e - _b^g)/ . Although we introduce two distinct atomic species for formal simplicity, the multiphoton-multiatom process can also be realized using a gas composed of a single atomic species. In such cases, two different excitation states of the same species, each with a distinct transition frequency, can be selectively targeted, as demonstrated

in the experiment reported in [13].

The angular frequency of the laser is denoted by Ω_L, set to be Ω_L = (ω_a + ω_b)/2. It is assumed that ω_a and ω_b differ significantly. Under this configuration, one can easily note that if both atoms are initially in their ground states, neither atom can individually absorb a single photon due to the mismatch between Ω_L and the respective transition frequency ω_a or ω_b. However, a joint excitation of the two atoms can occur through simultaneous absorption of one photon by each atom, satisfying energy conservation in the two-atom process.

The Hamiltonian of this atomic system and the laser field can be written as:

$$\hat{H} = \hat{H}_a + \hat{H}_b + \hbar\Omega_L \hat{a}^\dagger \hat{a} + \hat{d}_a \cdot \hat{E} + \hat{d}_b \cdot \hat{E}.$$

Here, $\hat{H}_a = \epsilon_a^g |g_a\rangle\langle g_a| + \epsilon_a^e |e_a\rangle\langle e_a|$, and $\hat{H}_b = \epsilon_b^g |g_b\rangle\langle g_b| + \epsilon_b^e |e_b\rangle\langle e_b|$, represent the atomic Hamiltonians of the A-species atom and the B-species atom respectively. $\hat{a}(\hat{a}^\dagger)$ is the annihilation (creation) operator of the laser photon. The operators $\hat{d}_a$ and $\hat{d}_b$ are the dipole moments of the A-species atom and the B-species atom, respectively. $\hat{E}$ is the quantum operator of the laser' s electric field. The dipole moment operators can be written as:

$$\hat{d}_k = \langle e_k | e\hat{r}_k | g_k \rangle |g_k\rangle\langle e_k| + \langle g_k | e\hat{r}_k | e_k \rangle |e_k\rangle\langle g_k| \quad (k = a, b).$$

Here, e is the electric charge of the electron, and $\hat{r}_a$ and $\hat{r}_b$ are the position operators of the electron in the A-species atom and the B-species atom, respectively. $\hat{E}$ can be approximately written in the form:

$$\hat{E} = i(\ )^{1/2} \mathbf{e}(\hat{a} e^{-i\Omega_L t} - \hat{a}^\dagger e^{i\Omega_L t}),$$

where V is the volume of the laser field, ϵ_0 is the permittivity of free space, and $\mathbf{e}$ is the polarization vector.

Assume the initial state of the whole system, including the laser field, is $|\Psi_i\rangle = |g_a\rangle|g_b\rangle|N_\gamma\rangle$ ($|N_\gamma\rangle$ represents the quantum state of the laser field with N_γ laser photons). At time t, the probability of the system being in the excited state $|\Psi_f\rangle = |e_a\rangle|e_b\rangle|N_\gamma - 2\rangle$ is $P_{ex} = |\langle\Psi_f| e^{-i\int^t \hat{H}dt'/\hbar} |\Psi_i\rangle|^2$, which will not be zero in principle. In the interaction picture, the third term of the perturbation expansion of $e^{-i\int^t \hat{H}dt'/\hbar}$ can couple $|\Psi_i\rangle$ to $|\Psi_f\rangle$ through virtual intermediate states.

Defining the following intermediate states: $|\Psi_a\rangle = |e_a\rangle|g_b\rangle|N_\gamma - 1\rangle$, $|\Psi_b\rangle = |g_a\rangle|e_b\rangle|N_\gamma - 1\rangle$. The two-photon excitations can occur through two virtual quantum transition paths, which are $|\Psi_i\rangle \to |\Psi_a\rangle \to |\Psi_f\rangle$ and $|\Psi_i\rangle \to |\Psi_b\rangle \to |\Psi_f\rangle$ (see Fig. 2 [Figure 2: see original paper]). These transitions correspond to a simultaneous two-photon-two-atom process in QED, where the energy of the system is not conserved for the virtual intermediate states.

The transition rate for the simultaneous two-atom excitation can be calculated using the usual perturbation method. After applying the rotating wave approximation, it can be written in the following form:

$$W_{2p2m} = \left| \sum_{\kappa} \frac{\langle \Psi_f | H_{int} | \Psi_\kappa \rangle \langle \Psi_\kappa | H_{int} | \Psi_i \rangle}{E_i + N_\gamma \hbar \Omega_L - E_{\Psi_\kappa}} \right|^2 \rho(E_f) \Big|_{E_f = \epsilon_a^e + \epsilon_b^e + 2\hbar\Omega_L}.$$

Here, $H_{int} = \hat{d}_a \cdot \hat{E} + \hat{d}_b \cdot \hat{E}$, is the coupling between the atoms and laser light. The state $\Psi_\kappa = \Psi_a$ or Ψ_b, is one of the intermediate states with energy $E_{\Psi_\kappa} = \epsilon_\kappa + (N_\gamma - 1)\hbar\Omega_L$ ($\kappa = a, b$). The factor $\rho(E_f)$ corresponds to the level density of the two-atom system and relates to the level density of each atom [15].

The transition rate for the process can be computed and written in the form:

$$W_{2p2m} \approx \frac{2}{7\pi^5} \left(\frac{\Omega_L}{\omega_a} \right) \left(\frac{\Omega_L}{\omega_b} \right) \frac{\gamma_a \gamma_b \Omega_L^2}{|\Omega_L - \omega_a||\Omega_L - \omega_b|} \rho(E_f).$$

Here, $\gamma_a = 4\alpha_e \omega_a^3 |\langle e_a | \hat{r}_a | g_a \rangle|^2 / 3c^2$ and $\gamma_b = 4\alpha_e \omega_b^3 |\langle e_b | \hat{r}_b | g_b \rangle|^2 / 3c^2$, where $\alpha_e = e^2/4\pi\hbar c\epsilon_0 \approx 1/137$ is the fine-structure constant, and c is the speed of light. Additionally, n_{λ^3} represents the number of laser photons within a volume of $\lambda^3 = (2\pi c/\Omega_L)^3$, defined as $n_{\lambda^3} = N_\gamma \lambda^3 / V$, and it is assumed that $N_\gamma \gg 1$.

In this plain perturbation calculation, one can note that W_{2p2m} vanishes exactly since the two terms within the modulus on the right side of the Eq. (6) cancel each other at $\Omega_L = (\omega_a + \omega_b)/2$. This result can be viewed as a quantum interference effect between the two excitation pathways: $|\Psi_i\rangle \to |\Psi_a\rangle \to |\Psi_f\rangle$ and $|\Psi_i\rangle \to |\Psi_b\rangle \to |\Psi_f\rangle$.

However, this cancellation does not imply the complete absence of joint two-atom excitation processes. Instead, two considerations suggest that quantum interference introduces a suppression factor for the transition rate rather than exact vanishing: (i) Higher-order QED contributions: when higher-order QED processes are taken into account, W_{2p2m} no longer vanishes. In particular, the introduction of a finite width for each atom' s excited level modifies the interference terms. By incorporating an imaginary component into ω_a and ω_b in Eq. (6), the terms within the modulus transform as follows:

$$\left| \frac{1}{\Omega_L - \omega_a + i\Gamma/2\hbar} + \frac{1}{\Omega_L - \omega_b + i\Gamma/2\hbar} \right|^2 \approx \frac{\Gamma^2/\hbar^2}{(\Omega_L - \omega_a)^2 (\Omega_L - \omega_b)^2},$$

where Γ represents the (natural) width of the excited level, assumed identical for both atoms for simplicity. In QED, this width arises from a series of repeated virtual processes, wherein an excited atom transitions to the ground state by emitting a virtual photon and reabsorbs it to return to the excited state (see, e.g., [16]). In the context of Eq. (7), the factor $\Gamma^2/\hbar^2(\Omega_L - \omega_b)^2$, typically orders

of magnitude smaller than unity, can be interpreted as the suppression factor associated with the quantum interference. (ii) Effect of detuning: The formal cancellation strictly occurs only at the specific value $\Omega_L = (\omega_a+\omega_b)/2$. However, in practice, Ω_L can be set to cover a range of frequencies, resulting in detuning. Considering $\Omega_L = (\omega_a+\omega_b)/2+\Delta\Omega/2$, where $\Delta\Omega$ is a detuning parameter with a magnitude smaller than or comparable to $\Gamma/\hbar$, the cancellation is no longer exact even in the plain perturbation treatment. In this case, the joint two-atom excitation involves additional processes, such as spontaneous photon emission. For instance, two atoms may jointly transition to excited states by absorbing one laser photon each, followed by the A-species atom emitting a photon and returning to its ground state. The emitted photon has a frequency $\omega_a + \Delta\Omega$ such that energy conservation for the entire process is satisfied. In this scenario, the final state of the system can be expressed as $|g_a\rangle|e_b\rangle|N_\gamma - 2\rangle|\gamma_{sp}\rangle_{\Omega_\gamma=\omega_a+\Delta\Omega}$, where $|\gamma_{sp}\rangle_{\Omega_\gamma}$ represents the quantum state of the emitted photon at frequency Ω_γ. The transition rate of this joint two-atom process involving one-photon emission can be obtained similarly using perturbation theory. One is generally interested in the emitted-photon exclusive transition rate, which is obtained by summing the transition rate over all possible directions of the emitted photon. The exclusive rate near $\Delta\Omega \approx \Gamma/\hbar$ is comparable to the transition rate for exact joint excitation at $\Omega_L = (\omega_a+\omega_b)/2$ (with the finite-width effect included for exact joint excitation). In principle, joint two-atom processes involving emissions of two photons are also possible, but a detailed investigation of these cases is not pursued here. The focus instead is to highlight other fundamental features of two-photon-two-atom processes and general MPMA processes, which remain independent of the specific details of the transition rate.

General MPMA processes can be constructed analogously to two-photon-two-atom processes. Consider a system of m atoms ($m = 3, 4, ...$) exposed to a laser field, where one atom is of species A and the remaining $m-1$ atoms are of species B. If the laser field frequency satisfies the condition $m\Omega_L = \omega_a + (m-1)\omega_b$, and the system starts with all atoms in their ground states, then all atoms can be simultaneously excited by absorbing m photons from the laser field. In this m-th order QED process, the system progresses through a sequence of virtual intermediate states, with each atom becoming excited by absorbing one photon.

The transition rate for this process can be estimated using perturbation theory. Similar to a two-photon-two-atom process, quantum interference among different excitation pathways arises, introducing a suppression factor in the transition rate [17]. Moreover, when Ω_L deviates from the exact resonance frequency $\omega_a/m + (m-1)\omega_b/m$, the excitation process in the m-atom system becomes coupled with spontaneous photon emission.

An intriguing aspect of MPMA processes is their ability to facilitate joint quantum behavior among multiple atoms without mediation of any interatomic interactions. Similarly, another remarkable joint quantum phenomenon that occurs without mediated interactions is superradiance, initially theorized by Dicke [18] and later observed in various systems [19–22]. These forms of joint quantum

phenomena defy classical intuition and highlight the extraordinary nature of the quantum framework.

A fundamental aspect of MPMA processes is the subtle emergence of a characteristic length scale. Using the two-atom system as an example, let l_d denote the distance between the two atoms. It is reasonable to anticipate that the MPMA process is influenced by the value of l_d. Specifically, if l_d exceeds a characteristic length, denoted as l_{mpma}, the joint excitation process is likely to become effectively unsupported. Since MPMA processes do not rely on any physical interaction among atoms, l_{mpma} cannot be interpreted as the range of a specific physical force. This initially makes the determination of l_{mpma} seem abstract and elusive. However, valuable insight emerges naturally from the uncertainty principle of quantum mechanics. The MPMA process can be envisioned as a sequence of intermediate virtual transitions, each involving a temporary violation of energy conservation. According to the uncertainty principle, the duration of such a virtual transition is inversely proportional to the magnitude of the energy deviation. By multiplying this timescale by the speed of light, one obtains a spatial range within which this virtual transition can collectively contribute to the joint excitation.

In the two-photon-two-atom quantum process, there are two such virtual transitions. The energy mismatch of each transition is: $\Delta\varepsilon = |\hbar\Omega_L - \hbar\omega_a| = |\hbar\omega_a - \hbar\omega_b|/2$. Defining a length related to this energy mismatch by $l_\Delta = \hbar c/2\Delta\varepsilon$, we can approximate the characteristic length as $l_{mpma} = \alpha l_\Delta$, where α is a parameter of order unity or less. This results in a fundamental relationship between l_{mpma} and the energy mismatch of the process:

$$l_{mpma} = \frac{\alpha c}{\omega_a - \omega_b}.$$

We shall consider the two-photon-two-atom process in an atomic gas with a large number of atoms. As shown earlier, the transition rate W_{2p2m} for a single two-atom system is typically small unless the laser strength reaches an exceptionally high level. This stems from its nature as a high-order perturbation process, further diminished by quantum interference effects. However, in a gas with a large number of atoms, the total transition rate for the two-photon-two-atom process rises substantially.

Take an atomic gas with one A-species atom and N_b B-species atoms, confined to a region smaller than l_{mpma}. Exposed to the same laser field as before, the total rate of the joint two-photon absorption increases by a factor of roughly N_b, corresponding to the number of ways to select one B-species atom from the N_b present.

This enhancement follows from Fermi' s Golden Rule, which implies that the total transition rate is additive over the different final quantum states accessible from the same initial state. The system' s initial state is $\Psi_{ini} = |g_a\rangle|g_{b1}\rangle|g_{b2}\rangle...|g_{bN_b}\rangle|N_\gamma\rangle$, where $|g_{bi}\rangle$ $(i = 1, ..., N_b)$ denotes the

ground state of the i-th B-species atom [23]. Two-photon excitation involves the A-species atom and one B-species atom, yielding distinct final states. For example, if the k_1-th B-species atom is excited, the final state becomes: $\Psi_{ex} = |e_a\rangle|g_{b1}\rangle...|g_{bk_1-1}\rangle|e_{bk_1}\rangle|g_{bk_1+1}\rangle...|g_{bN_b}\rangle|N_\gamma - 2\rangle$, with $|e_{bk_1}\rangle$ denoting its excited state.

Summing over all such final states introduces the N_b factor into the total transition rate. Formally, let $\Psi(t)$ represent the quantum state at a small time t, with $\Psi(0) = \Psi_{ini}$. The second-order perturbation expansion yields: $\Psi(t) = c_0(t)\Psi_{ini} + \sum_{1\le k_1\le N_b} c_{k_1}(t)\Psi_{ex}$. Perturbation analysis reveals that $|c_1(t)|^2 \approx |c_2(t)|^2 \approx ... \approx |c_{N_b}(t)|^2 \approx W_{2p2m}t$. Consequently, the probability of the system remaining unexcited is $P_{un}(t) = |c_0(t)|^2 = 1 - \sum_{1\le k_1\le N_b} |c_{k_1}|^2 \approx 1 - N_{bW_{2p2m}}t$. The rate of change of $P_{un}(t)$ with respect to t corresponds to the total transition rate.

Extending this to an atomic gas with N_a A-species atoms and N_b B-species atoms, the total transition rate scales by $N_{aN}b$. Since N_a and N_b can reach values as large as 10^{14} or higher, simultaneous two-photon absorption can become significant, despite the weakness of a single two-atom transition. For an MPMA process involving m atoms—one of species A and $m-1$ of species B—the enhancement factor scales approximately as $N_{aN}b^{m-1}$. This amplification could, in principle, make extremely weak atomic transitions experimentally accessible, a topic to be explored further in a separate study.

Consider a homogeneous atomic gas with a linear size exceeding l_{mpma}. For a given A-species atom, the number of B-species atoms participating in a two-photon-two-atom transition with it, denoted by N_{bo}, can be approximated as $\rho^3_{bl_{mpma}}$, where ρ_b is the number density of B-species atoms. In this system, the total transition rate of the two-photon-two-atom process (per A-species atom) is proportional to $N_{bo} \approx \rho^3_{bl_{mpma}}$. The characteristic length l_{mpma}, which is given by $\alpha c/(\omega_a - \omega_b)$, plays a crucial role in determining the amplification factor of the transition rate. Interestingly, the experimental observations of the two-photon-two-atom process [12, 13] provide valuable comparative insights. These findings suggest l_{mpma} scales as $c/(\omega_a - \omega_b)$.

In J. C. White' s study [12], a mixture of Ba and Tl atoms was investigated at high temperatures (above 1100°C) and high atomic densities ($10^{16} - 10^{18}$ atoms/cm^3). Here, a Ba atom and a Tl atom simultaneously absorbed two laser photons at $2\pi \times 668.1$ THz, despite this frequency being nonresonant with the isolated transitions of Ba ($6s^2\ {}^1_0S \rightarrow 6p\ {}^1_0P$, $\omega_{Ba} = 2\pi \times 541.4$ THz) and Tl ($6p\ {}^2_{1/2}P \rightarrow 7s\ {}^2_{1/2}S$, $\omega_{Tl} = 2\pi \times 793.8$ THz). This process was observed under laser intensities of $10^8 - 10^9$ W/cm^2. The absorption signal was found to be proportional to the square of the laser intensity, confirming a higher-order process involving the simultaneous absorption of two photons. Additionally, it scaled with the product of the Ba and Tl atom densities, providing clear evidence of joint excitations between atom pairs.

In contrast, E. Pedrozo-Peñafiel et al. examined a cold (100 µK), dilute Na gas (10^{12} atoms/cm^3). Laser excitation targeted Na dipole transitions–$3S_{1/2} \rightarrow 3P_{1/2}$ at $2\pi\times508.3$ THz and $3S_{1/2} \rightarrow 3P_{3/2}$ at $2\pi\times508.8$ THz–split by a $2\pi\times0.51$ THz fine-structure gap. The system responded to laser light tuned to the average frequency of these transitions. As in White' s experiments, the excitation signal was proportional to the square of the laser intensity. Using a relatively low laser intensity (on the order of 100 mW/cm^2) and a sample of about 10^9 Na atoms, the two-atom excitation rate was observed to be approximately 1% of the single-atom excitation rate under resonant conditions.

The Tl/Ba gas experiment differs significantly from the Na gas experiment due to its much higher atomic density and laser intensity–the latter being at least 10^9 times greater. A thorough comparison should also account for thermal broadening and laser linewidth, both of which influence the transition rate. In the Tl/Ba gas, thermal broadening exceeded the natural linewidth of dipole transitions by one to two orders of magnitude, while the laser linewidth was 10^3 times broader. Together, these factors suppressed the transition rate by approximately 10^{-4} or so. In contrast, these effects were negligible in the cold Na gas.

The transition rate is proportional to the square of the laser intensity and either the square of the atomic density (Na gas) or the product of atomic densities (Tl/Ba gas). One key to understanding the disparity between the two experiments–particularly the differing laser intensities required–lies in the characteristic length l_{mpma}. This length reflects the energy mismatch of the two-photon-two-atom process, driven by the frequency gap between joint transitions. For the Tl/Ba gas, $l_{mpma} \approx 0.2$ µm (assuming $\alpha \approx 1$ in Eq. (8)), while for the Na gas, $l_{mpma} \approx 100$ µm. Since the transition rate scales with l_{mpma}^3, this results in a rate difference of approximately 10^8, which partially explains why the Tl/Ba experiments require much higher laser intensities to observe joint excitations. Furthermore, the two-photon-two-atom process observed in the dilute Na gas suggests that no physical interatomic interaction is responsible for the process. In this system, the average interatomic spacing is around 1.0 µm, and any residual interatomic interaction is negligible at this distance.

A modified experiment on the two-photon-two-atom phenomenon can be naturally proposed to investigate the significant role of l_{mpma}. In this modified setup, a two-laser excitation scheme replaces the single-laser configuration (see Fig. 3 [Figure 3: see original paper] for the excitation pathways). Using the Tl/Ba gas system as an example, the frequency of one laser (denoted as L1) is detuned from the Tl transition frequency ω_{Tl} by an amount $\delta_{Tl} = |\omega_{Tl} - \Omega_{L1}|$. Simultaneously, the frequency of the other laser (denoted as L2) is detuned from the Ba transition frequency by the same amount in the opposite direction, ensuring the condition $\Omega_{L1} + \Omega_{L2} = \omega_{Tl} + \omega_{Ba}$. In this two-laser scheme, $l_{mpma} = c/2\delta_{Tl}$. By varying Ω_{L1}, l_{mpma} can be easily tuned, allowing for observations of how the detection signal changes with l_{mpma}.

Furthermore, experimental verification of simultaneous three-photon-three-atom processes can be straightforwardly carried out using the two-laser scheme. In the case of the Tl/Ba gas system, the laser frequencies are configured to satisfy either of the following conditions: $2\Omega_{L1} + \Omega_{L2} = 2\omega_{Tl} + \omega_{Ba}$, or $\Omega_{L1} + 2\Omega_{L2} = \omega_{Tl} + 2\omega_{Ba}$. In the three-photon-three-atom processes, the role of l_{mpma} becomes more pronounced compared to the two-photon-two-atom process, as the detection signal scales proportionally to l^6_{mpma} rather than l^3_{mpma}. This heightened sensitivity of the three-photon-three-atom process to variations in l_{mpma} provides a more robust platform for studying its influence.

In conclusion, a simultaneous MPMA process in an atomic gas under laser fields is examined, revealing several distinctive features, including the absence of interatomic interactions and the presence of an intrinsic characteristic length. A proposal for directly demonstrating this characteristic length experimentally is presented. Further studies of this fundamental process will not only enhance our understanding of the interaction between matter and light but also further our exploration of the intriguing quantum world.

REFERENCES

[1] M. Goeppert-Mayer, Annalen der Physik 9, 273 (1931).
[2] W. Kaiser and C. G. B. Garrett, Phys. Rev. Lett. 7, 229 (1961).
[3] F. E. Hernández, K. D. Belfield, I. Cohanoschi, M. Balu, and K. J. Schafer, Applied optics 43, 5394 (2004).
[4] Q. Zheng, H. Zhu, S.-C. Chen, C. Tang, E. Ma, and X. Chen, Nature Photonics 7, 234 (2013).
[5] W. Chen, S. Bhaumik, S. A. Veldhuis, G. Xing, Q. Xu, M. Grätzel, S. Mhaisalkar, N. Mathews, and T. C. Sum, Nature communications 8, 15198 (2017).
[6] J. R. Leite and C. B. D. Araujo, Chemical Physics Letters 73, 71 (1980).
[7] D. L. Andrews and M. Harlow, The Journal of Chemical Physics 78, 1088 (1983).
[8] M. H. Nayfeh and G. B. Hillard, Physical Review A 29, 1907 (1984).
[9] M. S. Kim and G. S. Agarwal, Physical Review A 57, 3059 (1998).
[10] A. Muthukrishnan, G. S. Agarwal, and M. O. Scully, Physical Review Letters 93, 093002 (2004).
[11] Z. Zheng, P. L. Saldanha, J. R. R. Leite, and C. Fabre, Physical Review A 88, 033822 (2013).
[12] J. C. White, Optics Letters 6, 242 (1981).
[13] E. Pedrozo-Peñafiel, R. R. Paiva, F. J. Vivanco, V. S. Bagnato, and K. M. Farias, Phys. Rev. Lett. 108, 253004 (2012).
[14] C. Hettich, C. Schmitt, J. Zitzmann, S. Kühn, I. Gerhardt, and V. Sandoghdar, Science 298, 385 (2002).
[15] Formally, the density of states is given by $\rho_f(E_f) = \delta(E_f - \epsilon^e_a - \epsilon^e_b)$. However, due to the inherent interaction between the excited atoms and the quantum electromagnetic field, the excited energy levels of each atom exhibit a resonant-state

nature (see, e.g., [16]), resulting in a finite width. Consequently, the two-atom system also acquires a finite width centered at $E_f = \epsilon_a^e + \epsilon_b^e$, and the density of states could be approximated by: $\rho_f(E_f) \sim \frac{\Gamma_{eff}/2}{(E_f-\epsilon_a^e-\epsilon_b^e)^2+(\Gamma_{eff}/2)^2}$, where Γ_{eff} is comparable to the maximum natural widths of the quantum states $|e_a\rangle$ and $|g_b\rangle$, differing by a factor between 1 and 2. Furthermore, effects such as thermal broadening of the excited states can be incorporated into this density function as well.

[16] J. D.-R. C. Cohen-Tannoudji and G. Grynberg, Atom-Photon Interactions: Basic Processes and Applications (John Wiley & Sons, 1998).

[17] The exact formal cancellation of the transition rate of an m-atom system at the resonant condition can be demonstrated numerically. Moreover, for even m, this precise cancellation can be justified algebraically in a straightforward manner, as the total contribution of all $m!$ excitation pathways exhibits $m!/2$ instances of exact pairwise cancellation.

[18] R. H. Dicke, Physical Review 93, 99 (1954).

[19] A. Goban, C.-L. Hung, J. Hood, S.-P. Yu, J. Muniz, O. Painter, and H. Kimble, Physical Review Letters 115, 063601 (2015).

[20] M. Scheibner, T. Schmidt, L. Worschech, A. Forchel, G. Bacher, T. Passow, and D. Hommel, Nature Physics 3, 106 (2007).

[21] R. DeVoe and R. Brewer, Physical Review Letters 76, 2049 (1996).

[22] N. Lambert, Y. Matsuzaki, K. Kakuyanagi, N. Ishida, S. Saito, and F. Nori, Physical Review B 94, 224510 (2016).

[23] In this analysis, we ignore the atom exchange symmetry, as its effect is negligible when the overlaps between the individual wavefunctions of B-species atoms are minimal, see also [18].

Note: Figure translations are in progress. See original paper for figures.