Suppression and Enhancement in Parametric Two-Photon Resonant Nondegenerate Four-Wave Mixing via Quantum Interference *

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Quantum interference may lead to suppression and enhancement of the two-photon resonant nondegenerate four-wave mixing signal in a cascade four-level system. Such phenomena are demonstrated in Ba through inducing atomic coherence between the ground state $6s^2$ and the doubly excited autoionizing Rydberg state $6pnd$. This method can be used as a new spectroscopic tool for measuring the transition dipole moment between two highly excited atomic states.

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Quantum interference, one of the basic features of quantum mechanics, has attracted much attention since the pioneering works of Harris and his collaborators. The interest in these phenomena is, on the one hand, due to the fundamental nature of coherence effects and, on the other hand, due to possible applications. Phenomena such as electromagnetically induced transparency (EIT), enhanced index of refraction and lasing without population inversion have been predicated theoretically and demonstrated experimentally.[1] EIT is also accompanied by large frequency dispersion and therefore, can slow and even stop the light pulse.[2−6]

One important application of EIT is its ability to enhance the efficiencies of nonlinear optical processes.[7−10] In contrast, Agarwal and Harshwardhan [11] showed that in a ladder-type four-level system, two-photon excitation may be selectively suppressed or enhanced by coupling the intermediate state to a higher-excited state and creating interference among the dressed-state transitions. The suppression of two-photon absorption in a ladder-type four-level system has been observed by Gao *et al.*[12] Similar two-photon suppression in a standard Λ-type system has been demonstrated in cold Rb atoms.[13]

In this Letter, we report suppression and enhancement of the signal in parametric two-photon resonant nondegenerate four-wave mixing (NFWM) via quantum interference. Our scheme is in sharp contrast to that proposed by Agarwal and Harshwardhan.[11] In the scheme of Ref. [11], the authors used a coupling laser to couple the intermediate state with some other states. The inhibition and enhancement of two-photon absorption are due to the quantum interference between two excitation paths, which took place through dressed intermediate states. In contrast, instead of coupling the intermediate state, here we couple the upper state with some other states so as to induce dressed states in the upper level of the two-photon transition. Quantum interference in parametric two-photon resonant NFWM is then investigated.

Another interesting thing is that most of the EIT-based experiments deal with single-electron atoms, where an electron is coherently forced to a superposition state. In contrast, here we devote to the quantum interference in two-electron atoms. We can induce atomic coherence between a doubly excited autoionizing Rydberg state and the ground state via three-photon resonant excitation. Quantum interference can be observed in this system even without using stabilized tunable lasers.

The idea is illustrated by considering the example of the relevant energy levels of Ba (Fig. 1(b)), where the ground state $6s^2$ ($|0\rangle$), the intermediate state $6s6p$ ($|1\rangle$), the Rydberg state $6snd$ ($|2\rangle$) and the doubly excited autoionizing Rydberg state $6pnd$ ($|3\rangle$) formed a cascade four-level system. The states between $|0\rangle$ and $|1\rangle$, between $|1\rangle$ and $|2\rangle$ and between $|2\rangle$ and $|3\rangle$ are coupled by dipolar transitions with resonant frequencies Ω_1, Ω_2 and Ω_3 and dipole moment matrix elements μ_1, μ_2 and μ_3, respectively. We first consider a two-photon resonant NFWM in the $|0\rangle$-$|1\rangle$-$|2\rangle$ cascade three-level system. As shown in Fig. 1(a), beams 2 and $2'$ have the same frequency ω_2 and a small angle exists between them; beam 1 with frequency ω_1 is propagating along the opposite direction of beam 2. We assume that $\omega_1 \simeq \Omega_1$ and $\omega_2 \simeq \Omega_2$, so that ω_1 drives the transition from $|0\rangle$ to $|1\rangle$, while ω_2 drives the transition from $|1\rangle$ to $|2\rangle$. In this case, the simultaneous interactions of atoms with beams 1 and 2 can

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induce atomic coherence between $|0\rangle$ and $|2\rangle$ through two-photon excitation. This coherence is then probed by beam $2'$ and the NFWM signal of frequency ω_1, which propagates along the opposite direction of beam $2'$ approximately, is generated as a result. To study the quantum interference in this system, we apply a coupling laser (beam 3) of frequency ω_3 ($\simeq \Omega_3$), which propagates along the direction of beam 2, to drive the transition from $6snd$ to the autoionizing state $6pnd$. This is the isolated core excitation,[14] in which the Rydberg outer electron is basically a spectator when the core electron is excited from $6s$ to $6p$. In the bare-state picture, atomic coherence is induced between the ground state $6s^2$ and the autoionizing Rydberg state $6pnd$. On the other hand, in the dressed-state picture (Fig. 1(c)), the isolated core excitation creates dressed atomic states $|+\rangle$ and $|-\rangle$, which are the superposition of the states $6snd$ and $6pnd$.

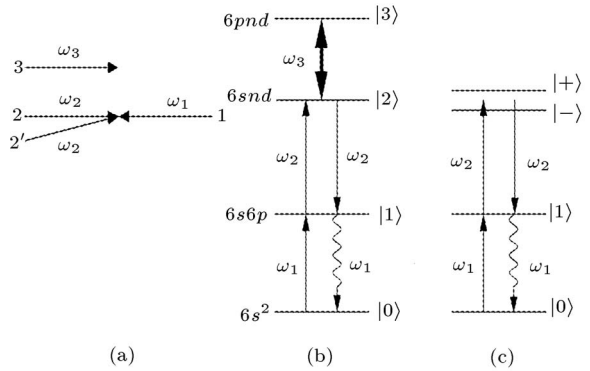


Fig. 1. (a) Schematic of two-photon resonant NFWM with coupling field. (b) Energy-level diagram for quantum interference in two-photon resonant NFWM. (c) Dressed-state picture.

Let Δ represent various detunings: $\Delta_1 = \Omega_1 - \omega_1$, $\Delta_2 = \Omega_2 - \omega_2$, $\Delta_3 = \Omega_3 - \omega_3$. After a canonical transformation the effective Hamiltonian is

$$\begin{aligned} H =& \hbar\Delta_1|1\rangle\langle 1| + \hbar(\Delta_1+\Delta_2)|2\rangle \\ &\cdot\langle 2| + \hbar(\Delta_1+\Delta_2+\Delta_3)|3\rangle\langle 3| \\ &- (\mu_1 E_1|0\rangle\langle 1| + \mu_2 E_2|1\rangle \\ &\cdot\langle 2| + \mu_3 E_3|2\rangle\langle 3| + \text{H.c}). \end{aligned} \tag{1}$$

Here $E_1 = \varepsilon_1 e^{i\boldsymbol{k}_1\cdot\boldsymbol{r}}$, $E_2 = \varepsilon_2 e^{i\boldsymbol{k}_2\cdot\boldsymbol{r}} + \varepsilon_2' e^{i\boldsymbol{k}_2'\cdot\boldsymbol{r}}$ and $E_3 = \varepsilon_3 e^{i\boldsymbol{k}_3\cdot\boldsymbol{r}}$ are the complex incident laser fields with frequencies ω_1, ω_2 and ω_3, respectively; while $\boldsymbol{k}_1$, $\boldsymbol{k}_2$, $\boldsymbol{k}_2'$ and $\boldsymbol{k}_3$ are the wavevectors of beams 1, 2, $2'$ and 3, respectively. The nonlinear polarization responsible for the two-photon resonant NFWM signal is proportional to the off-diagonal density matrix element ρ_{10}. We assume, as usual, that ε_1, ε_2 and ε_2' are weak, whereas the coupling field ε_3 can be of arbitrary magnitude. Thus ρ_{10} needs to be calculated to the order $\varepsilon_1\varepsilon_2\varepsilon_2'$, but to all orders in ε_3. This can be carried out by using density matrix equations with relaxation terms properly included:

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}[H,\rho] + \left(\frac{d\rho}{dt}\right)_{\text{relax}}, \tag{2}$$

from which we obtain

$$\begin{aligned} \rho_{10}(\boldsymbol{r}) =& -i\Big\{G_1 G_2 (G_2')^* [i(\Delta_1+\Delta_2+\Delta_3)+\Gamma_{30}] \\ &\cdot \exp[i(\boldsymbol{k}_1+\boldsymbol{k}_2-\boldsymbol{k}_2')\cdot\boldsymbol{r}]\Big\} \\ &\cdot\Big\{(i\Delta_1+\Gamma_{10})^2\{[i(\Delta_1+\Delta_2)+\Gamma_{20}] \\ &\cdot[i(\Delta_1+\Delta_2+\Delta_3)+\Gamma_{30}]+|G_3|^2\}\Big\}^{-1}, \end{aligned} \tag{3}$$

where Γ_{n0} is the transverse relaxation rate between states $|n\rangle$ and $|0\rangle$; the coupling coefficients $G_1 = \mu_1\varepsilon_1/\hbar$, $G_2 = \mu_2\varepsilon_2/\hbar$, $G_2' = \mu_2\varepsilon_2'/\hbar$ and $G_3 = \mu_3\varepsilon_3/\hbar$. According to Eq. (3), the NFWM signal propagates along the direction $\boldsymbol{k}_1 + \boldsymbol{k}_2 - \boldsymbol{k}_2'$.

In our experiments, barium vapour was contained in a stainless steel heat pipe with temperature around 660°C and with the argon buffer gas pressure at 1 torr. Three dye lasers L1, L2 and L3, which were pumped by the second or third harmonics of a Quanta-Ray Nd:YAG laser, had linewidth 0.007 nm and pulse width 5 ns. All the incident beams were linearly polarized in the same direction. They were focused to spots with diameters of approximately 2.0 mm. L1 was tuned to 553.5 nm, the wavelength of the $6s^2\ ^1S_0$–$6s6p\ ^1P_1$ transition. The blue laser L2 was used to drive the transition from $6s6p\ ^1P_1$ to a Rydberg $6snd\ ^1D_2$ level. It was split into beams 2 and $2'$ by beam splitters, and the two beams intersected in the oven with a small angle (0.5°) between them. On the other hand, L3 was scanned in the vicinity of the $6s_{1/2} - 6p_{3/2}$ resonance lines of the Ba^+ ion ($\lambda_3 \sim$455 nm) to reach the $6p_{3/2}nd$ autoionizing states.

We first kept ω_1 fixed and scanned ω_2 over the whole tunable range of the second dye laser. During the scanning no re-adjustment of the optical alignment is required because phase matching can be achieved for a very wide frequency range.[15] To avoid strong absorption at the resonant frequency of the transition from $6s^2\ ^1S_0$ to $6s6p\ ^1P_1$, here the wavelength of L1 was detuned from the exact resonance by 0.06 nm. Figure 2 presents the NFWM signal intensity versus the wavelength λ_2 of L2, which shows Rydberg series of Ba. We then set λ_2 to be 424.4 nm so that the Rydberg state $6s19d\ ^1D_2$ was chosen, and scanned ω_3 in the vicinity of the $6s_{1/2}$–$6p_{3/2}$ resonance lines of the Ba^+ ion. Figure 3(a) presents the NFWM signal intensity as a function of Δ_3 at exact two-photon resonance. In this experiment, the energies of beam 1 and the second dye laser L2 were $< 1\mu$ J and 20 μ J, respectively; while the energy of the coupling field (beam 3)

was 6 μJ. As one can see, the two-photon resonant NFWM signal intensity is suppressed when the frequency of the coupling laser field is scanned across the resonant frequency of the $6s19d\ ^1D_2$–$6p_{3/2}19d$ transition. We use Eq. (3) for the data analysis because the contribution from the Doppler effect to the linewidth of the dip is only $0.05\,\mathrm{cm}^{-1}$. The solid curve is the theoretical curve with parameters: $|G_3| = 1.3\,\mathrm{cm}^{-1}$, $\Delta_1 + \Delta_2 = 0\,\mathrm{cm}^{-1}$ and $\Gamma_{20} = 0.3\,\mathrm{cm}^{-1}$, which is limited by the laser linewidths. On the other hand, the transverse relaxation rate $\Gamma_{30} = 16\,\mathrm{cm}^{-1}$ is dominated by the autoionization rate of the state $6p_{3/2}19d$.

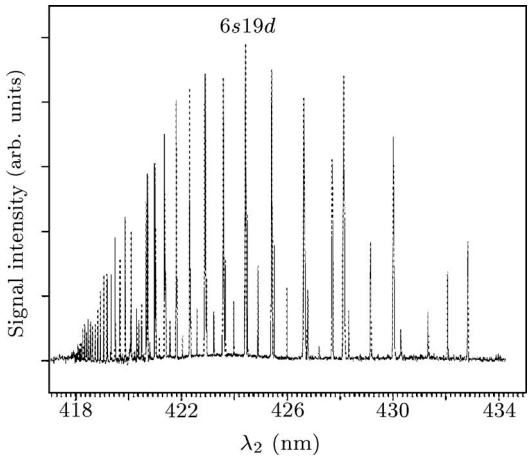


Fig. 2. Two-photon resonant NFWM spectrum.

The coupling field can not only suppress but also enhance the NFWM signal intensity when $\Delta_1 + \Delta_2 \neq 0$. Enhancement can occur for relatively weaker control field strength, so weak so as not to cause the Autler–Townes splitting of the two-photon resonant NFWM spectrum. This feature is displayed in Fig. 3(b), where the Δ_3-dependence of the NFWM signal was measured when the detuning from the two-photon resonance was $\Delta_1 + \Delta_2 = -0.3\,\mathrm{cm}^{-1}$. The solid curve is the theoretical curve with all parameters the same as that used in Fig. 3(a) except $\Delta_1 + \Delta_2 = -0.3\,\mathrm{cm}^{-1}$. The theoretical value of the NFWM signal intensity when the coupling field is absent (dotted line) is also given. It indicates that the NFWM signal exhibits enhancement when $\Delta_3 < -14\,\mathrm{cm}^{-1}$. Physically, in the dressed-state picture (Fig. 1(c)), suppression and enhancement of the NFWM signal are due to the quantum interference between two channels: $\rho_{00}^{(0)} \to \rho_{10}^{(1)} \to \rho_{+0}^{(2)} \to \rho_{10}^{(3)}$ and $\rho_{00}^{(0)} \to \rho_{10}^{(1)} \to \rho_{-0}^{(2)} \to \rho_{10}^{(3)}$.

Effects of quantum interference in two-photon resonant NFWM provide an effective tool for the spectroscopic purpose. This technique is Doppler-free when the incident lasers have narrow bandwidths.[16] Since it involves resonant three-photon excitation in a cascade four-level system, highly excited atomic states can be studied with high sensitivity. We first consider the nonsaturated limit for simplicity. At exact two-photon resonance (i.e. $\Delta_1 + \Delta_2 = 0$) the NFWM signal intensity becomes

$$I(\omega_3) \propto 1 - 2\Big(\frac{\Gamma_{30}}{\Gamma_{20}}\Big)\frac{|G_3|^2}{(\omega_3 - \Omega_3)^2 + \Gamma_{30}^2}, \qquad (4)$$

thus Ω_3 and Γ_{30} can be obtained directly from the ω_3-dependence of the NFWM signal. Moreover, the transition dipole moment between $|2\rangle$ and $|3\rangle$ can be measured in a carefully designed experiment. Specifically, we measure the depth of the dip in the spectrum, which is $F = [I(\omega_3 \to \infty) - I(\omega_3 = \Omega_3)]/I(\omega_3 \to \infty) = 2|\mu_3|^2\epsilon_3^2/\hbar^2\Gamma_{20}\Gamma_{30}$. Since Γ_{20} can be obtained from the two-photon resonant NFWM spectrum in the absence of coupling field, $|\mu_3|$ can be determined if the coupling field spot size and power are accurately measured. For more general cases (i.e. $|G_3|^2 \leq \Gamma_{20}\Gamma_{30}$), $|\mu_3|$ can still be obtained through fitting the ω_3-dependence of the NFWM signal because this spectrum is independent of G_1 and G_2.

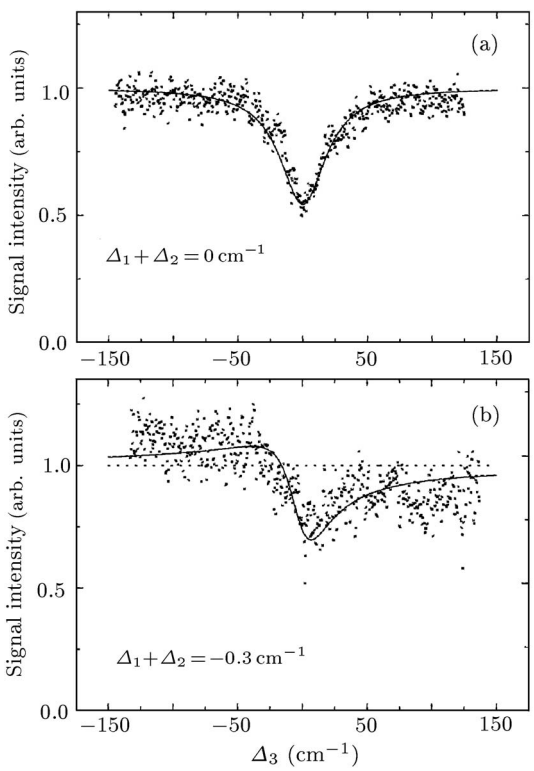


Fig. 3. NFWM signal intensity versus Δ_3 for (a) $\Delta_1 + \Delta_2 = 0\,\mathrm{cm}^{-1}$ and (b) $\Delta_1 + \Delta_2 = -0.3\,\mathrm{cm}^{-1}$. Solid curves are the theoretical curves with parameters $|G_3| = 1.3\,\mathrm{cm}^{-1}$, $\Gamma_{20} = 0.3\,\mathrm{cm}^{-1}$, $\Gamma_{30} = 16\,\mathrm{cm}^{-1}$ and $\Delta_1 + \Delta_2 = 0\,\mathrm{cm}^{-1}$ for (a) and $-0.3\,\mathrm{cm}^{-1}$ for (b). The dotted line in (b) corresponds to the theoretical value of the NFWM signal intensity when the coupling field is absent.

In summary, based on the quantum interference between two channels which are induced by the

coupling field, we have observed suppression and enhancement of two-photon resonant NFWM signals in Ba. This method can be used as a new spectroscopic tool for measuring the transition dipole moment between two highly excited atomic states.

References

[1] Harris S E 1997 *Phys. Today* **50** 36
[2] Hau L V et al 1999 *Nature* **397** 594
[3] Liu C et al 2001 *Nature* **409** 490
[4] Turukhin A V et al 2002 *Phys. Rev. Lett.* **88** 023602
[5] Zibrov A S et al 2002 *Phys. Rev. Lett.* **88** 103601
[6] Tu X et al 2003 *Chin. Phys. Lett.* **20** 1954
[7] Zhang G Z, Hakuta K and Stoicheff B P 1993 *Phys. Rev. Lett.* **71** 3099
[8] Zibrov A S, Lukin M D and Scully M O 1999 *Phys. Rev. Lett.* **83** 4049
[9] Wu Y, Saldana J and Zhu Y 2003 *Phys. Rev.* A **67** 013811
[10] Wu Y, Wen L and Zhu Y 2003 *Opt. Lett.* **28** 631
[11] Agarwal G S and Harshwardhan W 1996 *Phys. Rev. Lett.* **77** 1039
[12] Gao J Y et al 2000 *Phys. Rev.* A **61** 023401
[13] Yan M, Rickey E G and Zhu Y 2001 *Phys. Rev.* A **64** 043807
[14] Gallagher T F 1994 *Rydberg Atoms* (Cambridge: Cambridge University Press)
[15] Fu P, Jiang Q, Mi X and Yu Z 2002 *Phys. Rev. Lett.* **88** 113902
[16] Fu P, Wang Y, Jiang Q, Mi X and Yu Z 2001 *J. Opt. Soc. Am.* B **18** 370