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Ultraviolet single-photons on demand and entanglement of photons with a large frequency difference

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We investigate an efficient scheme for generating ultraviolet single-photons (∼300 nm). The scheme combines the highly efficient single-photon four-wave mixing scheme and fast developing quantum dot single-photons on demand source technology. We show that near maximum, entanglement between two well matched ultraslowly propagating single-photon wave packets can be achieved. This study may lead to research and development opportunities in highly efficient entanglement schemes using photons of very large frequency difference, quantum information processing, and single-photon metrology and single-photon counting sensors in the uv spectra region.

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Generating single-photons "on demand" represents the ultimate control of the light emission process. Impressive progress has been made in the past several years in obtaining such sources using single quantum dot based technology [1–3] and photonic band-gap materials [4]. The intense effort is motivated by the emerging field of quantum information science [5], which requires deterministic single-photon sources. Indeed, with nearly 100% conversion efficiency [3], such single-photon sources may be used in broad fields of research and technology ranging from single-photon quantum key distributions [6], quantum circuitry for single-photon switching [7], to single uv photon imaging technology [8].

In this work, we investigate a highly efficient ultraviolet single-photon "on demand" scheme using the highly efficient single probe photon four-wave mixing (FWM) technique [9,10]. The motivation for this study is twofold. First, a highly efficient single-photon source in the uv region is not available and its entanglement properties and potentials should be explored [11]. Secondly, there has been no study on a high-efficiency scheme to entangle photons of very different frequencies, and a study that provides basic understandings of this process is desirable. Aside from these fundamental physics viewpoints, a well characterized single uv photon source is also desirable for the fast development of single-photon counting sensors working in the uv spectra region. Such a detection system, although insensitive to the entanglement properties of the uv photon, requires well characterized single-photon responsivity in the region of 180—800 nm. Furthermore, fast developing GEM-based single uv photon multiplier technology for uv imaging [8] requires a well characterized single uv photon source as a metrological calibration source to provide a well traceable metrological standard. These considerations have prompted us to investigate efficient single uv photon generation schemes and to study the fundamental physical properties of such systems.

The scheme reported here has four unique features. (1) It provides the first single-photon "on demand" source in ultraviolet spectrum region and the deterministic properties of the generated ultraviolet photons are dictated by the deterministic properties of the single-photon input source. (2) It does not require high-energy, short-pulsed pump lasers as in the case of the familiar down-conversion scheme to produce single-photons. In fact, using a well-characterized single-photon on demand source, our scheme is capable of generating single ultraviolet photons (≃300 nm) using highly stable commercial infrared (∼800 nm) diode lasers with near 100% photon flux conversion efficiency. This may significantly improve the stability and controllability of the system, both of which are critically important to applications in quantum information processing. (3) Under suitable conditions, the generated ultraviolet photons can travel in the medium with ultraslow group velocities (e.g., $v_g/c \sim 10^{-4}$ or less), and both the ultraviolet and infrared photon wave packets maintain a well matched temporal profile as that of the input single-photon wave packet. It has been established that such ultraslow propagation velocities significantly increase the interaction time between single-photon wave packets and may significantly enhance certain nonlinear processes [9,10,12–15]. These enhancement effects may lead to controlled interaction between photons, which is a main challenge in quantum information processing. Finally, (4) it opens the possibility of achieving very efficient quantum entanglement of photons with very large frequency differences within a small propagation distance. We will show that it is possible to achieve nearly maximum entanglement of two quantized electromagnetic fields with very large frequency differences for an extended period of time permitted by the ultraslow propagation of single-photon wave packets. This may lead to the possibility of transferring quantum entanglement properties from multiple low-frequency carriers to a single ultraviolet carrier. In addition to the above described

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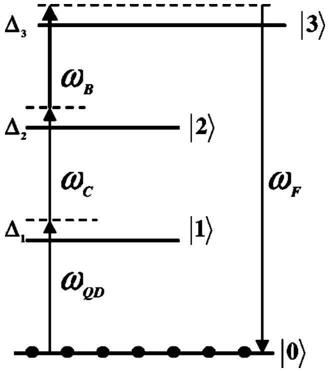

FIG. 1. A lifetime broadened four-level atomic system that interacts with two continuous wave (cw) classical laser fields (frequencies ω_B and ω_C, and Rabi frequencies $2\Omega_B$ and $2\Omega_C$) and a quantized pump field of frequency ω_{QD} to generate a quantized four-wave-mixing field of frequency ω_F.

aspects from a fundamental physics point of view, the system studied may also have potential as a standard single uv photon calibration source for fast developing single uv photon sensor technology. For instance, it can serve as a well-characterized single-photon source for GEM-based single uv photon multipliers as a metrological traceable calibration standard.

The scheme under consideration is a lifetime broadened four-state ladder system (Fig. 1). Here, we use a quantum dot single-photon on demand laser source (Ω_{QD}) as the first step of frequency upconversion process (i.e., $|0\rangle \rightarrow |1\rangle$. We note that the following calculation is independent of the type of single-photon source as long as it is a well-characterized "on-demand" type single-photon source). Two continuous wave (cw) classical fields (Ω_C, Ω_B) complete the remaining two steps up to resonantly reach state $|3\rangle$ where a phase matched FWM field (Ω_F) is originated. We show that with experimentally achievable parameters, a single-photon FWM field in the ultraviolet spectrum region can be very efficiently generated. We further show that under suitable conditions, both the ultraviolet and infrared photon wave packets can travel with well matched ultraslow group velocity.

Our investigation begins with the system Hamiltonian given by

$$\hat{H}/\hbar = -\sum_{j=1}^{3} \Delta_j |j\rangle\langle j| - (\Omega_C |2\rangle\langle 1| + \Omega_B |3\rangle\langle 2| + \text{c.c.}) - (\hat{\Omega}_{QD} |1\rangle\langle 0| + \hat{\Omega}_F |3\rangle\langle 0| + \text{H.c.}). \tag{1}$$

Using Eq. (1), the equations of motion for the atom density operator $\hat{\rho}$ and the quantized electromagnetic fields can be obtained as

$$\frac{\partial \hat{\rho}_{10}}{\partial t} = i(\Delta_1 + i\gamma_1)\hat{\rho}_{10} + i\hat{\Omega}_{QD}\hat{\rho}_{00} + i\Omega_C^*\hat{\rho}_{20} - i\hat{\rho}_{11}\hat{\Omega}_{QD} - i\hat{\rho}_{13}\hat{\Omega}_F, \tag{2a}$$

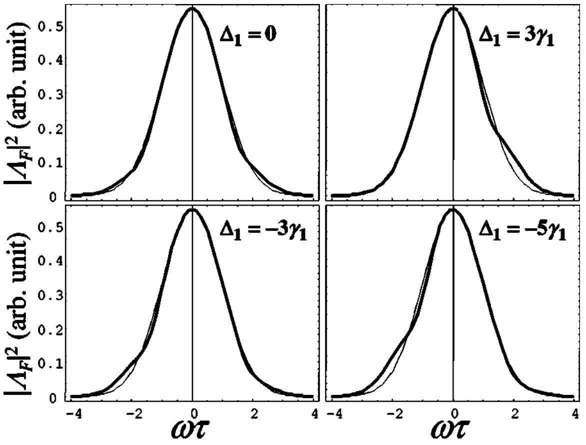

FIG. 2. The absorption coefficients $-\text{Re}[\beta_\pm]$ versus the dimensionless control field Ω_B/γ_2. Parameters used are $\gamma_1\tau \simeq 2\pi \times 5.9 \times 10^6/\text{s}$, $\gamma_2\tau \simeq 2\pi \times 0.8 \times 10^6/\text{s}$, $\gamma_3\tau \simeq 2\pi \times 0.09 \times 10^6/\text{s}$, $\Delta_1 = \Delta_2 = 0$, $\Delta_3 = 2\pi \times 0.02 \times 10^6/\text{s}$, $\kappa_{01} = 2 \times 10^9/(\text{s cm})$, $\kappa_{03} = 2 \times 10^7/(\text{s cm})$, and Ω_C relates to Ω_B by the expression $\kappa_{01}|D_{03}\Omega_B| = \kappa_{03}|D_{01}\Omega_C|$ (for maximum entanglement, we assume $3\omega_{QD} \sim \omega_F$).

$$\frac{\partial \hat{\rho}_{20}}{\partial t} = i(\Delta_2 + i\gamma_2)\hat{\rho}_{20} + i\Omega_B^*\hat{\rho}_{30} + i\Omega_C\hat{\rho}_{10} - i\hat{\rho}_{21}\hat{\Omega}_{QD} - i\hat{\rho}_{23}\hat{\Omega}_F, \tag{2b}$$

$$\frac{\partial \hat{\rho}_{30}}{\partial t} = i(\Delta_3 + i\gamma_3)\hat{\rho}_{30} + i\Omega_B\hat{\rho}_{20} + i\hat{\Omega}_F\hat{\rho}_{00} - i\hat{\rho}_{31}\hat{\Omega}_{QD} - i\hat{\rho}_{33}\hat{\Omega}_F, \tag{2c}$$

$$\frac{\partial \hat{\Omega}_{QD}}{\partial z} + \frac{1}{c}\frac{\partial \hat{\Omega}_{QD}}{\partial t} = i\kappa_{01}\hat{\rho}_{10},$$

$$\frac{\partial \hat{\Omega}_F}{\partial z} + \frac{1}{c}\frac{\partial \hat{\Omega}_F}{\partial t} = i\kappa_{03}\hat{\rho}_{30}. \tag{2d}$$

Here, $2\Omega_{B(C)}$ and $\omega_{B(C)}$ are the Rabi and optical frequencies of the classical pump field $E_{B(C)}$, $2\hat{\Omega}_{QD(F)} = D_{01(03)}\hat{E}^{(+)}_{QD(F)}/\hbar$ is the Rabi frequency operator for the quantized infrared (FWM) single-photon field $\hat{E}^{(+)}_{QD}$ ($\hat{E}^{(+)}_F$) with frequency ω_{QD} (ω_F), γ_j denotes the decoherence rate of $\hat{\rho}_{j0}$, and $\kappa_{01(03)} = 2N\omega_{QD(F)}|D_{01(03)}|^2/(c\hbar)$ with N and $D_{01(03)}$ being the concentration and dipole moment between states $|0\rangle$ and $|1\rangle$ ($|3\rangle$), respectively. In deriving Eqs. (2a)–(2d), we have assumed that the input infrared field is in a single-photon wave packet state with a shape function $F(t)$ (see below). We have also taken the slowly varying envelope approximation and defined detunings $\Delta_1 = \omega_{QD} - \epsilon_1/\hbar$, $\Delta_2 = \omega_{QD} + \omega_C - \epsilon_2/\hbar$, and $\Delta_3 = \omega_{QD} + \omega_C + \omega_B - \epsilon_3/\hbar$ with ϵ_j being the energy of state $|j\rangle$ ($\epsilon_0 = 0$).

The task of solving Eqs. (2a)–(2d) for the quantized fields starts with the nondepleted ground-state approximation, i.e., $\hat{\rho}_{00} \simeq 1$. We note that the last two terms in Eqs (2a)–(2c) are higher-order terms of small quantities $\hat{\Omega}_{QD}$ and $\hat{\Omega}_F$. For a small signal treatment, we neglect these higher-order terms.

By taking the time Fourier transform of Eqs. (2a)–(2d), we obtain

$$(\omega+\Delta_1+i\gamma_1)\hat{\alpha}_{10}+\Omega_C^*\hat{\alpha}_{20}=-\hat{\Lambda}_{QD}, \tag{3a}$$

$$\Omega_C\hat{\alpha}_{10}+(\omega+\Delta_2+i\gamma_2)\hat{\alpha}_{20}+\Omega_B^*\hat{\alpha}_{30}=0, \tag{3b}$$

$$\Omega_B\hat{\alpha}_{20}+(\omega+\Delta_3+i\gamma_3)\hat{\alpha}_{30}=-\hat{\Lambda}_f, \tag{3c}$$

$$\hat{L}\hat{\Lambda}_{QD}=i\kappa_{01}\hat{\alpha}_{10},\quad \hat{L}\hat{\Lambda}_F=i\kappa_{03}\hat{\alpha}_{30}, \tag{3d}$$

where $\hat{\alpha}_{j0}$, $\hat{\Lambda}_{QD}$, and $\hat{\Lambda}_F$ are the Fourier transforms of $\hat{\rho}_{j0}$, $\hat{\Omega}_{QD}$, and $\hat{\Omega}_F$, respectively, ω is the Fourier transform variable, and $\hat{L}\equiv\partial/\partial z-i\omega/c$.

The solution to Eqs. (3a)–(3c) can be found as

$$\hat{\alpha}_{10}=\frac{D_{QD}}{D}\hat{\Lambda}_{QD}+\frac{\Omega_C^*\Omega_B^*}{D}\hat{\Lambda}_F, \tag{4a}$$

$$\hat{\alpha}_{30}=\frac{\Omega_C\Omega_B}{D}\hat{\Lambda}_{QD}+\frac{D_F}{D}\hat{\Lambda}_F, \tag{4b}$$

$$\hat{\alpha}_{20}=-\frac{(\omega+\tilde{\Delta}_3)\Omega_C}{D}\hat{\Lambda}_{QD}-\frac{(\omega+\tilde{\Delta}_1)\Omega_B^*}{D}\hat{\Lambda}_F, \tag{4c}$$

where $\tilde{\Delta}_j=\Delta_j+i\gamma_j$ $(j=1,2,3)$, $D=|\Omega_C|^2(\omega+\tilde{\Delta}_3)+|\Omega_B|^2(\omega+\tilde{\Delta}_1)-(\omega+\tilde{\Delta}_1)(\omega+\tilde{\Delta}_2)(\omega+\tilde{\Delta}_3)$, $D_{QD}=(\omega+\tilde{\Delta}_2)(\omega+\tilde{\Delta}_3)-|\Omega_B|^2$, and $D_F=(\omega+\tilde{\Delta}_1)(\omega+\tilde{\Delta}_2)-|\Omega_C|^2$. Substituting Eqs. (4a) and (4b) into Eq. (3d) and noting that there is no generated field at the entrance of the medium, we obtain

$$\hat{\Lambda}_{QD}(z,\omega)=\frac{\hat{\Lambda}_{QD}(0,\omega)[U_+e^{izK_-}-U_-e^{izK_+}]-\hat{\Lambda}_F(0,\omega)[e^{izK_-}-e^{izK_+}]}{U_+-U_-}, \tag{5a}$$

$$\hat{\Lambda}_F(z,\omega)=\frac{\hat{\Lambda}_F(0,\omega)[U_+e^{izK_+}-U_-e^{izK_-}]+U_+U_-\hat{\Lambda}_{QD}(0,\omega)[e^{izK_-}-e^{izK_+}]}{U_+-U_-}, \tag{5b}$$

where $K_\pm=\omega/c+(\kappa_{03}D_F+\kappa_{01}D_{QD}\pm G)/(2D)$, $U_\pm=(\kappa_{03}D_F-\kappa_{01}D_{QD}\pm G)/(2\kappa_{01}\Omega_C^*\Omega_B^*)$, and $G=\sqrt{(\kappa_{03}D_F-\kappa_{01}D_{QD})^2+4\kappa_{03}\kappa_{01}|\Omega_C\Omega_B|^2}$.

We focus our attention on the adiabatic regime, where $K_\pm$ and $U_\pm$ can be expressed by a rapidly converging power series of ω. This allows analytical evaluation of Eqs. (5a) and (5b) so that a clear physical picture of the process can be obtained. To ensure such a robust adiabatic process, we assume $|\Omega_C|^2$, $|\Omega_B|^2>\max(|\tilde{\Delta}_1\tilde{\Delta}_2|,|\tilde{\Delta}_2\tilde{\Delta}_3|)$. It can be shown that under these conditions, the linearization treatment introduced before is well justified. When these conditions are satisfied, we find that $U_\pm=W_\pm+\mathcal{O}(\omega)$ and $K_\pm=(K_\pm)_{\omega=0}+\omega/V_{g\pm}+\mathcal{O}(\omega^2)$ can accurately describe the process at hand. After applying the inverse Fourier transform, we obtain from Eqs. (5a) and (5b):

$$\hat{\Omega}_{QD}(z,t)=\frac{[W_+\hat{\Omega}_{QD}(\eta_-)-\hat{\Omega}_F(\eta_-)]e^{z\beta_-}-[W_-\hat{\Omega}_{QD}(\eta_+)-\hat{\Omega}_F(\eta_+)]e^{z\beta_+}}{W_+-W_-}, \tag{6a}$$

$$\hat{\Omega}_F(z,t)=\frac{W_+[\hat{\Omega}_F(\eta_+)-W_-\hat{\Omega}_{QD}(\eta_+)]e^{z\beta_+}+W_-[W_+\hat{\Omega}_{QD}(\eta_-)-\hat{\Omega}_F(\eta_-)]e^{z\beta_-}}{W_+-W_-}, \tag{6b}$$

where $\hat{\Omega}_{QD}(t)\equiv\hat{\Omega}_{QD}(z=0,t)$ and $\hat{\Omega}_F(t)\equiv\hat{\Omega}_F(z=0,t)$ are the quantized infrared and FWM quantized fields at the entrance $z=0$, respectively, $\eta_\pm=t-z/V_{g\pm}$, $\beta_\pm=i(K_\pm)_{\omega=0}$, and

$$\beta_+\simeq i\frac{\kappa_{01}\kappa_{03}\Delta_2}{\kappa_{01}|\Omega_B|^2+\kappa_{03}|\Omega_C|^2}-\frac{\kappa_{01}\kappa_{03}\gamma_2}{\kappa_{01}|\Omega_B|^2+\kappa_{03}|\Omega_C|^2}, \tag{7a}$$

$$\beta_-\simeq-\frac{(\kappa_{01}|\Omega_B|^2+\kappa_{03}|\Omega_C|^2)(B_1+iB_2)}{B_1^2+B_2^2}, \tag{7b}$$

$$\frac{1}{V_g}\equiv\frac{1}{V_{g+}}\simeq\frac{1}{c}+\frac{\kappa_{01}\kappa_{03}}{\kappa_{01}|\Omega_B|^2+\kappa_{03}|\Omega_C|^2}, \tag{7c}$$

$$W_+\simeq\frac{\Omega_B}{\Omega_C^*},\quad W_-\simeq-\frac{\kappa_{03}\Omega_C}{\kappa_{01}\Omega_B^*}. \tag{7d}$$

Here $B_2\equiv|\Omega_C|^2\gamma_3+\gamma_1|\Omega_B|^2$ and $B_1\equiv|\Omega_C|^2\Delta_3+\Delta_1|\Omega_B|^2$. Close inspection of the expressions (7a) and (7b) of $\beta_\pm$ indicates that under the conditions described we have $|\mathrm{Re}[\beta_+]|\ll|\mathrm{Re}[\beta_-]|$ (also see Fig. 2). The key consequence of this relation is that the η_- velocity component in Eqs. (6a) and (6b) decays much faster than the η_+ component. Consequently, after a characteristic propagation distance, the field operators take the form

$$\hat{E}^{(+)}_{QD}(z,t) \simeq \frac{\kappa_{03}|\Omega_C|^2\exp(z\beta_+)}{\kappa_{01}|\Omega_B|^2+\kappa_{03}|\Omega_C|^2}\left[\hat{E}^{(+)}_{QD}\left(t-\frac{z}{V_g}\right) + \frac{\kappa_{01}D_{03}\Omega_B^*}{\kappa_{03}D_{01}\Omega_C}\hat{E}^{(+)}_F\left(t-\frac{z}{V_g}\right)\right], \tag{8a}$$

$$\hat{E}^{(+)}_F(z,t) \simeq \left(\frac{D_{01}\Omega_B}{D_{03}\Omega_C^*}\right)\hat{E}^{(+)}_{QD}(z,t), \tag{8b}$$

$\kappa_{01}D_{03}/\kappa_{03}D_{01}=\sqrt{\omega_{QD}\kappa_{01}/(\omega_F\kappa_{03})}$. We note that Eq. (8) indicates that the two quantized fields travel with the same group velocity V_g. With appropriate and experimentally achievable parameters (see below), it can be shown that the matched group velocity can be substantially smaller than the speed of light in vacuum. Under the input condition, there exists only an infrared quantum dot single photon with the pulse shape function $F(t)$ of single photons. It is straightforward from Eq. (8) [16] to obtain the state of the total quantized field at the exit $z=L$ as follows:

$$|\Psi\rangle = CF\left(t-\frac{L}{V_g}\right)\left[|1_{QD},0_F\rangle + \left(\frac{\kappa_{01}D_{03}\Omega_B}{\kappa_{03}D_{01}\Omega_C^*}\right)|0_{QD},1_F\rangle\right]\exp(i\theta-\alpha L), \tag{9}$$

where $\theta=\kappa_{01}\kappa_{03}\Delta_2 L/(\kappa_{01}|\Omega_B|^2+\kappa_{03}|\Omega_C|^2)$, $\alpha=\kappa_{01}\kappa_{03}\gamma_2/(\kappa_{01}|\Omega_B|^2+\kappa_{03}|\Omega_C|^2)$, and C is a normalized constant. Equation (10) shows that with appropriate choice of intensities of the two classical fields so that $\kappa_{01}|D_{03}\Omega_B| \sim \kappa_{03}|D_{01}\Omega_C|$, we obtain a nearly maximum entangled photon pair (the entanglement of two frequency modes) with very large frequency difference. Furthermore, it is possible to adjust the probability amplitude of the ultraviolet photon wave packet by varying the intensities of the control fields. This is a remarkable and unique advantage for a single photon on demand source.

We have carried out extensive numerical calculations to establish the validity of the above described analytical treatment by using experimentally achievable parameters. A possible experimental candidate for the proposed system is ultracold ^{85}Rb atoms. We take, for instance, $|0\rangle=|5S_{1/2}\rangle$, $|1\rangle=|5P_{1/2}\rangle$, $|2\rangle=|5D_{3/2}\rangle$, and $|3\rangle=|nP_{3/2}\rangle$ with $n>10$. The respective transitions are $|0\rangle\rightarrow|1\rangle$ at 795 nm ($\gamma_1\simeq 5.9$ MHz), $|1\rangle\rightarrow|2\rangle$ at 762 nm ($\gamma_2\simeq 0.8$ MHz), and $|2\rangle\rightarrow|3\rangle$ at 1.3–1.5μm ($\gamma_3\simeq 0.09$ MHz), all accessible with diode lasers. From Eq. (10) it is easily verified that the same set of parameters as those in Fig. 2 leads to a maximum entanglement between the infrared and the ultra violet photon wave packets. In addition, by choosing intensities of driving fields properly, one can adjust the probability amplitude of the ultraviolet photon wave packet. These are remarkable performances in multi-wave frequency upconversion processes.

The ultra violet single photon on demand using highly efficient single probe photon FWM technique and the newly developed efficient quantum dot single photon on demand source may have wide applications in quantum information processing and photo-sensor metrology. It is a novel scheme for achieving maximum entanglement of two photons with very large frequency difference. The flexibility of being able to adjust the probability amplitude of the ultra violet photon wave packet may lead to new research opportunities in quantum information processing. The well matched group velocity and temporal profile may also find applications in quantum computing, quantum cryptography, entanglement, and single photon quantum key distribution schemes.

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