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Efficient frequency up-conversion in resonant coherent media

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We demonstrate an efficient frequency up-conversion based on generation of large atomic coherence in a cascade system. Two infrared, low power laser fields tuned to the vicinity of the two-photon transition in Rb vapor were converted spontaneously into infrared and blue radiation. Extension of the technique into other spectral regions using highly excited states seems feasible.

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Effects of atomic coherence and interference are currently becoming an important tool in resonant nonlinear physics [1]. These effects have been demonstrated to yield resonant enhancement of second harmonic generation [2], four-wave mixing [3], and frequency conversion without stringent phase-matching requirements [4–9]. Nonlinear phenomena induced by tiny optical fields due to extraordinarily large Kerr nonlinearities has been observed already [10] and are expected to yield a new regime of quantum nonlinear optics [11].

In this paper we demonstrate that techniques of atomic coherence and interference can be used for efficient frequency up-conversion of low power, continuous wave (cw) laser fields. Specifically, we describe an experiment in which two infrared laser beams have been efficiently converted into far infrared and blue radiation in a fully resonant wave mixing process.

The present approach is related to but different from earlier studies involving lasers without inversion [13], frequency conversion [4], and parametric oscillators driven by maximal coherence, as well as Raman generation by phased and antiphased states in molecules [6]. Here we operate in a very different parameter domain involving relatively weak laser fields and much closer to the single-photon resonance; most importantly, however, we work with a cascade atomic configuration. Finally, we note that the effects related to the present observation have been seen using excitation with short laser pulses [12]. The earlier experiments were transient in nature and used significantly higher levels of power than those used here.

Consider a four-state cascade atomic medium [Fig. 1(a)] in which $|b\rangle \rightarrow |a\rangle \rightarrow |c\rangle$ transitions are driven by a pair of external resonant fields with Rabi frequencies Ω_1 and Ω_2. In systems of this kind the dephasing rate of the upper state τ_b is much slower than the corresponding rate of the intermediate states τ_a, τ_d. Whenever this is the case optical fields can excite a large atomic coherence on the dipole-forbidden transition $|b\rangle \rightarrow |c\rangle$, thereby transferring a large fraction of population into the excited state. This allows one to create a large population inversion on an infrared ($|c\rangle \rightarrow |d\rangle$) transition by exciting the upper state. At the same time the $|b\rangle \rightarrow |c\rangle$ coherence results in a reduction of resonant absorption for driving fields and modifies the corresponding refractive index. When the coherence is created in a medium with a sufficiently large density-length product, a correlated pair of fields with frequencies corresponding to the optical transitions of the second cascade $|c\rangle \rightarrow |d\rangle \rightarrow |b\rangle$ can be spontaneously generated in a very efficient nonlinear mixing process.

We work in atomic ^{87}Rb in which the ground state $|b\rangle = |5S_{1/2}\rangle$ is coupled to the slowly decaying $|c\rangle = |5D_{5/2}\rangle$ level (lifetime τ_c is 240 ns) by two infrared lasers at 780 nm and 776 nm. The intermediate state $|a_1\rangle = |5P_{3/2}\rangle$ has a lifetime of $\tau_{a1} = 32$ ns. The uppermost level of the cascade has a dipole-allowed path to the $|d\rangle = |6P_{3/2}\rangle$ state ($\tau_d = 112$ ns). This transition has a very strong dipole moment, but a very slow radiative decay due to a long wavelength (5.5 μm). Finally, the state $|6P_{3/2}\rangle$ has a dipole-allowed transition to the ground state with a wavelength in the blue spectral region (420 nm). In what follows we show that it is the interplay between population inversion in the infrared transition and large two-photon coherence that results in the generation of infrared and blue radiation. Specifically, spectral components of the amplified spontaneous emission at 5.5 μm beat with those of two-photon coherence resulting in generation of the 420 nm field with a large nonlinear gain.

In the experiment [Fig. 1(b)] radiation from two extended cavity diode lasers ECDL1 and ECDL2 was combined on a polarizing beam splitter and focused before entering a 12 cm long Rb cell. The output power of each laser was about 10–15 mW, and spot sizes were about 400 μm in the cell

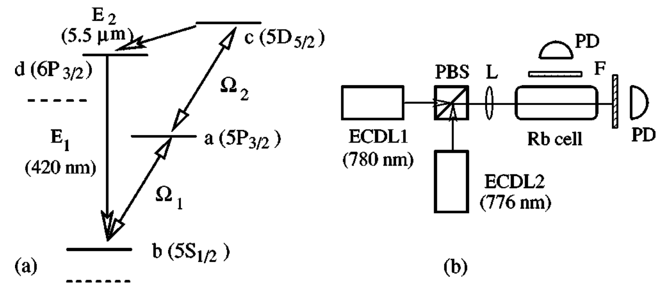

FIG. 1. (a) ^{87}Rb atoms in double-cascade configuration interacting with four optical fields. (b) Schematic of experimental setup. ECDL1,2 are lasers, PB is beam splitter, and PD is photodetector.

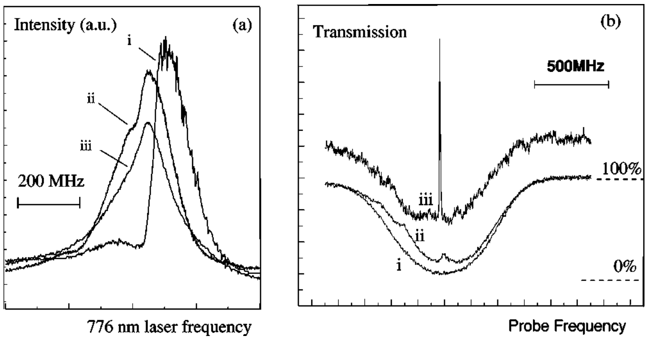

FIG. 2. (a) Generated intensity of blue [420 nm, curves (i), (ii)] and infrared [5.5 μm, curve (iii)] radiation as a function of drive (776 nm) laser tuning. The 780 nm driving laser is tuned to the blue side of the $F=2\rightarrow F''=3$ absorption line of ^{87}Rb (detuning from the line center is about 150 MHz). Driving power is 10 mW and 11 mW at 780 nm and 776 nm, respectively. (b) Transmission of a weak 420 nm probe beam passing through the optically driven vapor cell. (i) Both driving beams are off. (ii) 780 nm is present. (iii) 780 nm and 776 nm beams are present. Curve (iii) is shifted upward due to spontaneously generated background.

region. The two collinear laser beams passed through a Brewster cell containing either natural or isotopically pure Rb maintained at $T\sim 100\,^\circ$C and were subsequently blocked by filters. Collimated, intense blue radiation as well as infrared radiation propagating along the path of the driving beams were detected and analyzed. Fluorescence from the side of the Rb cell was also monitored simultaneously.

Under conditions corresponding to optimal temperature, alignment, and tuning of the driving beams the output power of the forward blue radiation reached the 15 μW level. We note that this corresponds to conversion efficiency that, at the given power level, exceeds that of typical nonlinear crystals by several orders of magnitude. We further note that both processes can be similarly enhanced using build-up cavities. Figure 2 shows typical dependencies of the intensity of forward radiation and orthogonal fluorescence at 420 nm, and the absorption of the 780 nm drive laser as a function of the tuning of the 776 nm drive laser. A large increase of collimated blue radiation is apparent as the 776 nm laser approaches two-photon resonance. Note that, in general, the output intensity of the collimated blue field and the intensity of fluorescence display different dependencies upon the tuning of the driving fields. For example, the curve (i) corresponding to collimated radiation has a strong asymmetry in shape. We further observe that the collimated infrared field is affected only marginally by the presence of the blue generation. In contrast, a very good correlation between infrared field and side fluorescence at 420 nm is evident. At the same time collimated blue generation occurs only when the infrared field is present [14].

The spectral properties of the generated light were studied using Fabry-Pérot reference cavities. The frequency of the generated 420 nm field followed the tuning of either of the driving lasers. The spectral width of the blue radiation was measured to be $\leqslant 3$ MHz, limited by the resolution of the reference cavities [15].

We have also performed measurements in which absorption of a weak 420 nm probe field passing through the Rb cell was studied [Fig. 2(b)]. In this experiment the second harmonic of an 840 nm extended-cavity diode laser was generated using a $LiIO_3$ crystal. Typically a few nanowatts of power were obtained (i.e., more than three orders of magnitude less than in the present process). The driving intensities were chosen to be much lower than in the case of spontaneous generation [small spontaneous generation results in unwanted background radiation; see the difference between curves (ii) and (iii) for large detunings in Fig. 2(b)]. In the case when the driving fields were not present about 95% of the blue probe light was absorbed. When a pair of driving beams was applied large amplification was observed. The particular value of the amplification coefficient was limited by saturation of the medium. Whereas the saturation threshold could be increased by increasing the driving field intensities, for stronger fields the spontaneous generation dominates.

We now turn to a theoretical discussion of the present results. In order to understand the origin of the efficient nonlinear generation we consider a situation in which the four-state system [Fig. 1(a)] is interacting with four optical fields. The main features of resonant nonlinear generation can be understood by considering the coherence of the dipole-forbidden $|b\rangle\rightarrow|c\rangle$ transition and the upper state population. In an idealized case when the dephasing of the upper state $|c\rangle$ is negligible and when there is no Doppler broadening, a pair of laser fields Ω_1,Ω_2 can drive the atomic system into a state $|\Psi\rangle=\cos\theta|b\rangle+\sin\theta|c\rangle$, if the sum frequency of the two fields is close to the two-photon resonance in the atomic medium. This state corresponds to a maximal two-photon coherence $|\rho_{bc}|^2=\rho_{bb}\rho_{cc}$ and can result in a large population of the excited state $|c\rangle$. Note that the cascade coherence is a running wave $\rho_{cb}=\bar{\rho}_{cb}\exp(ik_{cb}z)$ with a wave vector k_{cb} which is equal to the sum of the wave vectors of the driving fields, modified by appropriate refractive indices.

Consider now propagation of the pair of optical fields E_1, E_2 in the medium with initial coherence in a cascade transition. The relevant Maxwell equations are

$$\frac{\partial E_1}{\partial z}=\left(\kappa_1\frac{\rho_{dd}-\rho_{bb}}{\Gamma_{db}}+i\,\delta k\right)E_1-\frac{\kappa_1}{\Gamma_{db}}\bar{\rho}_{cb}E_2^*, \tag{1}$$

$$\frac{\partial E_2^*}{\partial z}=\left(\kappa_2\frac{\rho_{cc}-\rho_{dd}}{\Gamma_{dc}}\right)E_2^*+\frac{\kappa_2}{\Gamma_{dc}}\bar{\rho}_{bc}E_1, \tag{2}$$

where ρ_{ii} are populations of atomic states and Γ_{ij} are complex relaxation rates of optical coherences. $\delta k=k_1'+k_2'-k_{bc}$ is the wave vector mismatch. In these equations terms proportional to populations of levels describe linear gain and loss as well as phase shifts, whereas terms proportional to coherence are responsible for nonlinear cross coupling between fields. Both are proportional to coupling constants of the corresponding transitions: $\kappa_1=3/(4\pi)N\lambda_1^2\gamma_{d\rightarrow b}$, $\kappa_2=3/(4\pi)N\lambda_2^2\gamma_{c\rightarrow d}$ (γ_i denote radiative decay rates between levels $|i\rangle\rightarrow|j\rangle$). Note that for the current problem involving a strong infrared and a weak short wavelength transition $\kappa_2\gg\kappa_1$.

Interesting insight into propagation dynamics can be gained by studying the eigenvalues of two normal modes corresponding to Eqs. (1) and (2). In the case when absorption of driving fields can be disregarded they are

$$\Lambda_{1,2}=a\mp\left[a^2-|\rho_{bc}|^2-\kappa_2\frac{\rho_{cc}-\rho_{dd}}{\Gamma_{dc}}\times\left(\kappa_1\frac{\rho_{dd}-\rho_{bb}}{\Gamma_{db}}+i\,\delta k\right)\right]^{1/2}, \tag{3}$$

where

$$a=(1/2)[\kappa_1(\rho_{dd}-\rho_{bb})/\Gamma_{db}+i\,\delta k+\kappa_2(\rho_{cc}-\rho_{dd})/\Gamma_{dc}].$$

Let us consider again an idealized limit when the population in the intermediate levels is negligible, $\rho_{aa}=\rho_{dd}=0$, coherence is maximal, $|\rho_{bc}|^2=\rho_{bb}\rho_{cc}$, and $\delta k=0$. Note that the last condition can easily be achieved due to the large dispersion of the refractive index near two-photon resonance. Such a phase-coherent medium yields two normal modes with propagation constants $\Lambda_1=0$ and $\Lambda_2=a$. The first normal mode corresponds to undistorted propagation of a pair of fields E_1, E_2 with a ratio of amplitudes determined by the Rabi frequencies of the driving fields $E_1/E_2^*=\Omega_1/\Omega_2^*$. Such lossless propagation is known from studies of EIT in a Λ medium, in which case it corresponds to stable propagation of so-called matched pulses [1,17]. The stability is normally ensured due to large absorption of the second eigenmode.

We now show that such a matched-pulse propagation is unstable in the case of a cascade medium. To this end we note that the real part of the second eigenvalue $\mathrm{Re}\kappa_2$ can be large and positive if

$$\mathrm{Re}(\kappa_2)=\frac{\kappa_2\gamma_{dc}}{\gamma_{dc}^2+\Delta_{dc}^2}\rho_{cc}-\frac{\kappa_1\gamma_{db}}{\gamma_{db}^2+\Delta_{db}^2}\rho_{bb}>0. \tag{4}$$

This implies that when the linear gain coefficient for a field E_2 exceeds the linear loss for field E_1 a properly phased pair of fields can be exponentially amplified. The essence of this process is that a light field amplified by a phase insensitive gain mechanism on the $|c\rangle\rightarrow|d\rangle$ transition beats with coherence on the cascade transition and, as a result, generates correlated components of radiation near resonance with the $|d\rangle\rightarrow|b\rangle$ transition. For the present problem involving a strong long-wavelength transition between highly excited states and a weaker transition in the short-wavelength region, this gain coefficient can be very large, $\sim\kappa_2\rho_{cc}/\gamma_{dc}$.

It should be noted that the above regime is very different from that corresponding to the usual two-photon parametric gain [16], which takes place in the off-resonant case, when the coherence and population in the upper state are small. In the latter case of the usual, off-resonant parametric amplification, photons are emitted only in pairs, i.e., infrared photons are generated only if blue photons are present, and vice versa. In the resonant case one of the fields (infrared) is generated regardless of the presence of the second (blue) component.

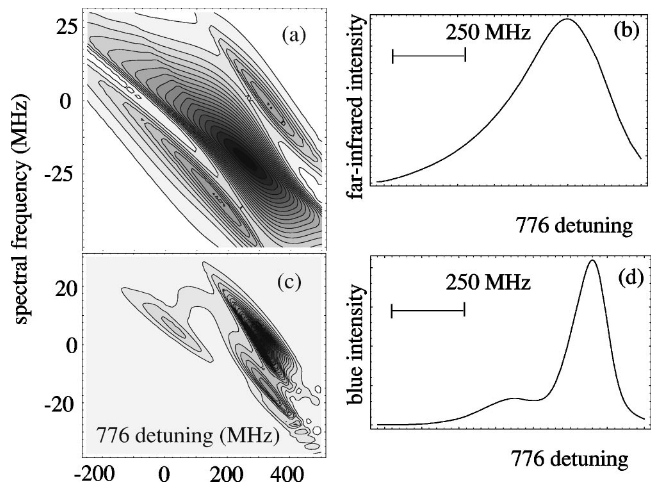

FIG. 3. Theoretically calculated spectra of generated radiation. (a),(c) Contour plots of generated intensity of infrared and blue fields (darker shading corresponds to higher intensity) as a function of spectral frequency of the corresponding component and of detuning of the 776 nm driving laser. (b),(d) Total generated intensity as a function of 776 nm laser tuning.

In order to make a detailed comparison between the theoretical predictions and experimental results we have carried out numerical simulations in which the Maxwell equations for four optical fields interacting with four-state atoms have been solved. In the present experimental demonstration the coherence generated is not ideal. In particular, decay from the upper state and Doppler broadening result in a decreased value of this coherence. Due to the Doppler broadening larger values of coherence appear when the driving fields are tuned to the wings of the absorption lines. For typical experimental conditions we calculated both coherence and the upper state population to be on the order of a few percent. To describe the atomic and field dynamics density matrix equations were solved numerically with parameters corresponding to atomic Rb. All saturation effects were included explicitly. To describe the effect of *spontaneous generation*, spontaneous emission must be taken into account. In order to model this effect we include a weak seed field on the $|d\rangle\rightarrow|b\rangle$ transition with initial intensity proportional to the upper state population. For the present experimental conditions we find that the linear amplification factor is on the order of exp(30), in which case the properties of the output radiation are determined by saturation effects and do not depend (to a very good approximation) upon the initial conditions.

Results of the numerical simulations are presented in Fig. 3. These calculations can predict the major qualitative features of the observed line shapes reasonably well. It is remarkable that the spectra of both infrared and blue fields display several peaks corresponding to large amplification. For instance, the infrared spectrum displays three components corresponding to the Autler-Townes splitting. However, it must be emphasized that none of those components is due to parametric two-photon amplification: by artificially excluding the generation of the blue field nearly identical spectra of the field E_2 have been obtained. At the same time, it is clear that the spectrum of blue radiation (E_1) does not follow exactly that of the field E_2, as it should in the case of ideal coherence. Clearly, since coherence is not perfect generation favors some parts of the spectrum over others due to absorption of driving fields and phase-matching considerations. Note that phase matching and hence efficient generation of a certain component frequency can be optimized by small tuning of one of the driving lasers, in agreement with earlier observations [4].

It is important to remark on the saturation behavior of the present system; here it is determined by the depletion of the upper state population which is accompanied by a decrease in the relevant coherence ρ_{bc}. This is different from the effects of competition between amplified spontaneous emission and parametric gain, which dominates in the case of off-resonant excitation. As discussed by Boyd and coworkers this completion mechanism requires the effective two-photon Rabi frequencies on the excitation path $b \rightarrow a \rightarrow c$ and on the generation path $b \rightarrow d \rightarrow c$ to become equal. In the case when input driving fields are very far from the corresponding single-photon resonances [18] and amplified spontaneous emission is resonant, nonlinear generation can be inhibited already for very low generated power. In the present case the input driving fields are themselves close to resonance; hence this competition mechanism will take place only when the generated power becomes comparable to the input power and does not inhibit efficient generation.

One obvious limitation of the present, resonant process is that it is effective in a relatively narrow range of frequencies associated with specific atoms. We note, however, that the decay configuration required to achieve efficient frequency conversion is typical for excited states of many atomic systems. In particular, it is interesting to consider the extension of the present technique to the highly excited Rydberg states. These states are typically very long lived, and yet coupled by strong dipole-allowed transitions in the far infrared or even microwave spectral regions. Such levels can be densely spaced and can potentially be used for stepwise tuning over a large bandwidth. We believe that this lays the ground work for the extension of the present proof-of-principle demonstration for generation of deep uv radiation in a broad class of atomic systems.

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[14] In addition to blue and infrared radiation we also observed weaker components at 2.79 μm and 1.36 μm collimated along the direction of the driving lasers' paths. More experimental detail can be found in A.S. Zibrov *et al.*, in Atomic Physics XVII, edited by E. Arimondo, P. DeNatale, and M. Inguscio, AIP Conf. Proc. No. **551** AIP Melville, NY, (2001).

[15] Noise spectra of the generated radiation were investigated as well. rf amplitude noise spectra displayed two distinguishing features: $\sim$1000-fold enhancement of the noise at low frequencies (<2 MHz) and an additional peak at frequencies ($\sim$10–15 MHz) that were related to the 776 nm driving intensity. The location of this peak scaled as the square root of the 776 nm field intensity and was on the order of Ω_2. Note the corresponding sidebands resulting from the theoretical analysis [Fig. 3(b)].

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