Efficient infrared imaging upconversion via quantum coherence

Robert W. Boyd[a)]
Institute of Optics, University of Rochester, Rochester, New York 14627

Marlan O. Scully
Department of Physics and Institute for Quantum Studies, Texas A&M University, College Station, Texas 77843

(Received 28 August 2000; accepted for publication 9 October 2000)

We show that quantum coherence can be used to achieve marked improvement in the efficiency of the process of infrared upconversion. In one particular example which we analyze in detail, we show that it is possible to convert infrared radiation at a wavelength of 100 μm to the visible with essentially 100% efficiency while maintaining diffraction-limited imaging of the infrared field. © *2000 American Institute of Physics.* [S0003-6951(00)03448-3]

Infrared upconversion[1] is a well-established technique for converting a light field from the infrared portion of the electromagnetic spectrum to the visible, where highly sensitive and low-noise radiation detectors are readily available.[2] This conversion is achieved by generating the sum frequency of the infrared radiation and one or more laser fields through use of second- or third-order nonlinear optical interactions. Infrared upconversion can provide near diffraction-limited performance in converting weak astronomical infrared images to the visible under continuous-wave conditions,[3] and can provide reasonably good conversion efficiency when used with pulsed laser sources.[4] However, the general usefulness of this technique has been limited by the low conversion efficiencies that have in the past been achievable on a steady-state basis when infrared upconversion is implemented using cw laser sources.

In this letter, we propose a technique for infrared upconversion that should be able to produce essentially unit conversion efficiency from the infrared to the visible on a steady state basis. Our proposed method is based on the use of a phase-coherent atomic ensemble (phaseonium) that can be used to render a material system transparent to resonant laser radiation, while retaining the large and desirable nonlinear optical properties associated with the resonant response of a material system.[5] Our method should be generally applicable for upconversion of wavelengths from the near infrared through the submillimeter spectral regions; however, we illustrate this technique for the particular example of conversion of radiation of 100 μm wavelength by a four-wave interaction in atomic sodium vapor. We note that sum-frequency generation resonantly enhanced by sodium Rydberg levels has been observed previously.[6] We note further that recent experiments in Rb^{87} have demonstrated the inverse of the process considered here, that is, the generation of infrared radiation using quantum coherence.[7]

In the 30 yr since the initial demonstrations of infrared upconversion, dramatic improvements have been made in the performance of semiconductor infrared detectors. Nonetheless, the technique proposed here holds considerable promise for use at wavelengths of 100 μm and longer, where improvements in detector perforamce are still being sought.

Let us consider the nonlinear optical interaction shown in part (a) of Fig. 1, where four optical waves interact under nearly resonant conditions in an atomic vapor. Here ω_1 and ω_4 are strong saturating laser fields which partially populate levels a and d and induce a coherence $\rho_{ad}^{(\infty)}$ between them. The infrared field to be detected is designated ω_2, and the upconverted field is at frequency $\omega_3=\omega_1+\omega_2-\omega_4$. We next show that the presence of the strong fields ω_1 and ω_4 significantly enhances the efficiency of the upconversion process. We quantify the atomic response by calculating $\rho_{cd}=\rho_{cd}(\omega_3)$, that is, the amplitude of the density matrix element oscillating at the upconverted frequency ω_3. We calculate the contributions to this quantity that are first order in the weak fields (that is, the infrared field at frequency ω_2 and the upconverted field at frequency ω_3) and correct to all orders in the amplitudes of the strong saturating fields at frequencies ω_1 and ω_4. The equation of motion for ρ_{cd} is given under these conditions by

$$\dot{\rho}_{cd}=-\Gamma_{cd}\rho_{cd}+i\Omega_2\rho_{ad}^{(\infty)}+i\Omega_3\rho_{dd}^{(\infty)}, \tag{1}$$

which leads to the steady-state solution

$$\rho_{cd}=i(\Omega_2/\Gamma_{cd})\rho_{ad}^{(\infty)}+i(\Omega_3/\Gamma_{cd})\rho_{dd}^{(\infty)}. \tag{2}$$

In these equations, we have introduced the complex quantity $\Gamma_{cd}=\gamma_{cd}-i\Delta_3$, where γ_{cd} is the total dephasing rate of the cd coherence and $\Delta_3=\omega_3-\omega_{cd}$ is the detuning. In addition, $\Omega_2=\mu_{ca}E_2/\hbar$ and $\Omega_3=\mu_{cd}E_3/\hbar$ are the coupling strengths induced by the infrared field and upconverted fields respectively, and $\rho_{ad}^{(\infty)}$ is the coherence and $\rho_{dd}^{(\infty)}$ is the population established by the strong saturating fields. These two quantities are given by lengthy expressions that have already appeared in the literature[8] and which will not be reproduced here. However, a crucial aspect of our technique is that $\rho_{ad}^{(\infty)}$ and $\rho_{dd}^{(\infty)}$ are quantities of the order of unity if the fields at frequencies ω_1 and ω_4 are saturating fields. It is for this reason that our method leads to highly efficient upconversion, and also for this reason we prefer to write our results in terms of the quantities $\rho_{ad}^{(\infty)}$ and $\rho_{dd}^{(\infty)}$ rather then in terms of their functional forms. Also, since $\rho_{ad}^{(\infty)}$ and $\rho_{dd}^{(\infty)}$ depend only weakly upon the field amplitudes under saturating condi-

a)Author to whom correspondence should be addressed; electronic mail: boyd@optics.rochester.edu

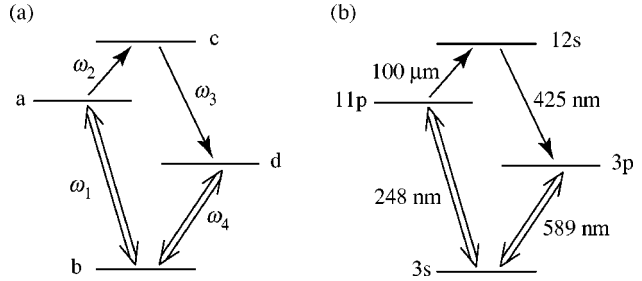

FIG. 1. (a) Use of quantum coherence to enhance the efficiency of infrared upconversion. Here ω_2 represents an infrared field which is converted to the visible at frequency ω_3. The drive fields ω_1 and ω_4 populate levels a and d and create a strong coherence between them, which dramatically increases the efficiency of the upconversion process. (b) A particular example of infrared upconversion, based on a four-wave interaction in atomic sodium vapor.

tions, we prefer to express the nonlinear response not in terms of a third-order susceptibility $\chi^{(3)}$ but rather in terms of effective linear susceptibilities describing the absorption and conversion processes. These quantities are defined through the relation $P(\omega_3)=\chi_{\rm abs}^{(\rm eff)}(\omega_3)E_3+\chi_{\rm con}^{(\rm eff)}(\omega_3)E_2$. Since the polarization is related to the density matrix elements by $P(\omega_3)=N\rho_{cd}\mu_{dc}$, we find that

$$\chi_{\rm con}^{(\rm eff)}(\omega_3)=\frac{iN\mu_{dc}\mu_{ca}\rho_{ad}^{(\infty)}}{\hbar\Gamma_{cd}},$$
$$\chi_{\rm abs}^{(\rm eff)}(\omega_3)=\frac{iN\mu_{dc}\mu_{cd}\rho_{dd}^{(\infty)}}{\hbar\Gamma_{cd}}. \tag{3}$$

The quantity $\chi_{\rm con}^{(\rm eff)}(\omega_3)$ can be alternatively interpreted as a Raman susceptibility as it gives the response at frequency ω_3 in terms of the applied field at frequency ω_2. Of course, the propagation of the signal infrared field will also be influenced by the nonlinear interaction, and we can describe these interactions by analogously defined susceptibilities which are given by

$$\chi_{\rm con}^{(\rm eff)}(\omega_2)=\frac{iN\mu_{ac}\mu_{cd}\rho_{da}^{(\infty)}}{\hbar\Gamma_{ca}},$$
$$\chi_{\rm abs}^{(\rm eff)}(\omega_2)=\frac{iN\mu_{ac}\mu_{ca}\rho_{aa}^{(\infty)}}{\hbar\Gamma_{ca}}, \tag{4}$$

where $\Gamma_{ca}=\gamma_{ca}-i\Delta_2$, and $\Delta_2=\omega_2-\omega_{ca}$. From these susceptibilities, coupled amplitude equations can be derived in the standard manner. We find that

$$\frac{dA_2}{dz}=-\alpha_2A_2+\kappa_2A_3e^{-i\Delta kz},$$
$$\frac{dA_3}{dz}=-\alpha_3A_3+\kappa_3A_2e^{i\Delta kz}, \tag{5}$$

where $\Delta k=k_1+k_2-k_3-k_4$ where $\alpha_i=(2\pi i\omega_i/n_ic)\chi_{\rm abs}^{(\rm eff)}$ and $\kappa_i=(2\pi i\omega_i/n_ic)\chi_{\rm con}^{(\rm eff)}$. These coupled amplitude equations can be solved in closed form, but for our present purposes it is more germane to use these equations to make simple estimates of the efficiency that can be achieved in upconversion enhanced by quantum coherence.

Let us consider the particular example shown in part (b) of Fig. 1, which shows infrared upconversion based on a four-wave interaction in atomic sodium vapor. Here the levels a, b, c, and d of our general formalism correspond to the sodium $11p$, $3s$, $12s$, and $3p$ levels, respectively. This example corresponds to the situation of converting radiation from a wavelength of 100 μm to visible radiation at 425 nm. For simplicity, in our present calculations we ignore the effects of fine and hyperfine structure. From standard references[9] we obtain the oscillator strengths for each of the relevant transition, and when converted to dipole transition moments (assuming z-polarized radiation in all cases) we find that $\mu(3s\rightarrow 3p)=2.467\,ea_0$, $\mu(3s\rightarrow 11p)=0.0121\,ea_0$, $\mu(11p\rightarrow 12s)=21.62\,ea_0$, and $\mu(3p\rightarrow 12s)=0.019\,ea_0$.

Our theoretical formalism presupposes that we are able to saturate the $3s$ to $3p$ and $3s$ to $11p$ transitions. Ideally one would want to saturate the entire Doppler distribution, although the basic interaction described by our formulas can occur as long as at least one Doppler subgroup is saturated. The criterion for saturating one Doppler subgroup is that the Rabi frequency $\Omega/2\pi$ of each saturating field be greater than the 10 MHz homogeneous linewidth of the $3s$ to $3p$ transition. This criterion leads to the requirements that $I_4>13\,\mathrm{mW/cm^2}$ and that $I_1>500\,\mathrm{W/cm^2}$. These criteria are easily met with current cw tunable laser sources. The more restrictive condition that the entire Doppler profile be saturated requires that the Rabi frequency $\Omega/2\pi$ of each saturating field be greater than the approximately 1 GHz inhomogeneous linewidth of the transitions. This condition leads to the requirements that $I_4>130\,\mathrm{W/cm^2}$ and that $I_1>5\,\mathrm{MW/cm}$; it is unlikely that this second condition can be met with current laser sources. The implication of this conclusion is that only some fraction (on the order of 1%) of the atoms within the interaction region can contribute to the quantum-coherence-enhanced upconversion process. Nonetheless, we shall see that high-efficiency upconversion can occur.

In order for upconversion to occur with good efficiency, the infrared and upconverted waves must undergo negligible absorption, appreciable coupling must occur over the length of the interaction region, and the phase matching condition $\Delta\mathbf{k}=0$ must be satisfied. We now address these requirements in turn. For absorption to be negligible, it is necessary that the real parts of the absorption coefficients α_i of the coupled amplitudes Eq. (5) be sufficiently small that negligible absorption occurs over the characteristic distance scale (on the order of $1/|\kappa_3|$) of the interaction. Since both the real and the imaginary parts of the coupling coefficients κ_i lead to the growth of the upconverted field, it is possible to ensure that coupling is much larger than absorption by choosing the detunings of fields 1 and 2 to be sufficiently large. It is also necessary that appreciable conversion occurs over the length of the sodium cell. We have found that these conditions can be met under the following conditions: We assume a sodium number density of $10^{16}\,\mathrm{cm^{-3}}$, with a corresponding effective number density that is 100 times smaller. We also assume that the detunings $\Delta_2/2\pi$ and $\Delta_3/2\pi$ are equal to 1 GHz. We then find that $\mathrm{Re}\,\alpha_2=0.25/\mathrm{cm}$, $\mathrm{Re}\,\alpha_3=6.2\times10^{-5}/\mathrm{cm}$, and $|\kappa_2|=|\kappa_3|=6.2/\mathrm{cm}$. Under these conditions essentially complete conversion of infrared to visible occurs in a path length on the order of 0.5 cm.

The requirements on phase matching are addressed in

the following manner. First, we note that satisfying the longitudinal phase-matching condition is of only secondary importance for an interaction of the sort shown in Fig. 1. The reason is that because of strong saturation of the lower transitions, the drive fields ω_1 and ω_4 will possess their vacuum wave vectors. Also, because of this saturation, the change in wave vectors of the infrared and upconverted waves resulting from the material response will be of the same order of magnitude as the nonlinear coupling between these waves. Thus, to first order, the phase matching condition need not be imposed, because there is no way that the interacting waves can get out of phase with each other in the distance over which they undergo a complete interchange of energy. This circumstance has been described previously in the context of nonlinear optical interactions with maximal atomic coherence.[10] If it is desired that the longitudinal phase matching relation be met precisely, a small angle can be introduced between the propagation directions of the two drive fields to alter the longitudinal components of their wave vectors in order to satisfy the phase matching condition. The transverse phase matching relation ensures that each infrared direction of propagation will produce a unique upconverted direction of propagation. Previous work has shown that diffraction-limited performance can be achieved from the upconversion process.[3,4] In principle, phase matching relations establish a fundamental limitation to the maximum breadth of the spectral bandwidth of an infrared upconverter,[1] although for the particular design described here it is likely that the bandwidth will be set by a decrease in coupling strength with increasing detuning and an increase in absorption with decreasing detuning, leading to a bandwidth on the order of the nominal detuning of the fields from the atomic resonances.

In summary, we have shown that phaseonium can be used to achieve dramatic improvements in the efficiency of the process of infrared upconversion. With these improvements, upconversion could prove to be a generally useful technique for radiation detection, especially in far infrared and at terahertz frequencies.

This work was supported by the ONR, the NSF, and the Welsh Foundation. The authors acknowledge useful discussion with C. R. Stroud, Jr. regarding fundamental aspects of the EIT interaction, with D. Watson regarding current infrared detector technology, and with T. F. Gallagher regarding reliable values of sodium transition moments.

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