Nonlinear Optical Processes Using Electromagnetically Induced Transparency

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We show that by applying a strong-coupling field between a metastable state and the upper state of an allowed transition to ground one may obtain a resonantly enhanced third-order susceptibility while at the same time inducing transparency of the media. An improvement in conversion efficiency and parametric gain, as compared to weak-coupling field behavior, of many orders of magnitude is predicted.

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It is well known by those practicing the techniques of nonlinear optics that the power which may be generated in a frequency summing process, or the gain which may be obtained in a parametric process is determined by the interplay of the nonlinear and linear susceptibilities.[1,2] In general, as an atomic transition to the ground state is approached, the nonlinear susceptibility is resonantly enhanced, but at the same time the media exhibits a rapidly increasing refractive index and becomes opaque.

In this Letter we show how it is possible to create nonlinear media with resonantly enhanced nonlinear susceptibilities and at the same time induce transparency and a zero in the contribution of the resonance transition to the refractive index. An energy-level diagram for a prototype system is shown in Fig. 1. We apply a strong electromagnetic coupling field of frequency ω_c between a metastable state $|2\rangle$ and a lifetime-broadened state $|3\rangle$, and generate the sum frequency $\omega_d=\omega_a+\omega_b+\omega_c$. We assume that $|1\rangle$-$|3\rangle$ is a resonance transition and that in the absence of ω_c, radiation at ω_d is strongly absorbed.

When the Rabi frequency of the coupling field exceeds the Doppler width of the $|1\rangle$-$|3\rangle$ transition, the media becomes transparent on line center. This transparency occurs because of the destructive interference of the split (Autler-Townes) components of the $|1\rangle$-$|3\rangle$ transition. Though one might expect that this interference would also negate the nonlinearity that causes the generation of ω_d, this is not so; because of a sign change in the dressed eigenvectors, for generated frequencies lying between the Autler-Townes components, there is a constructive rather than a destructive interference in the nonlinear susceptibility.

Before proceeding we note earlier work: The use of electromagnetic fields to create transparency has been reviewed by Knight.[3] Armstrong and Wynne[4] observed that Fano-type interferences between photoionization and autoionization are not mirrored in $\chi^{(3)}$ profiles; Pavlov *et al.*[5] observed an enhancement of sum frequency generation by inducing a Fano-type state into the continuum. The work described here does not involve photoionization. State $|3\rangle$ may decay radiatively, or by autoionization, but if it decays by autoionization, then this work neglects the direct coupling of states $|1\rangle$ and $|2\rangle$ to the continuum.

In the following paragraphs we first consider the dressed susceptibilities of a single atom and thereafter include the effects of collisional and Doppler broadening.

We assume that an electromagnetic field with the frequencies ω_a, ω_b, ω_c, and ω_d is applied to the atom and calculate the total dipole moment at ω_d. This dipole moment may be expressed in terms of a linear and a third-order susceptibility. These susceptibilities depend on the magnitude of the coupling field ω_c and in this sense are dressed by the field. The pertinent quantities are defined as

$$E(t)=\mathrm{Re}\left\{\sum_{k=a}^{d}E(\omega_k)e^{j\omega_k t}\right\},\quad P(t)=\mathrm{Re}\left\{\sum_{k=a}^{d}P(\omega_k)e^{j\omega_k t}\right\},$$
$$P(\omega_d)=\epsilon_0\chi_D^{(1)}(-\omega_d,\omega_d)E(\omega_d)+\tfrac{3}{2}\epsilon_0\chi_D^{(3)}(-\omega_d,\omega_a,\omega_b,\omega_c)E(\omega_a)E(\omega_b)E(\omega_c)\,. \tag{1}$$

The susceptibilities are calculated from the equations for the time-varying probability amplitudes of a single atom

$$\frac{db_1}{dt}=\frac{j\Omega_{12}}{2}b_2+\frac{j\Omega_{13}}{2}b_3,\quad \frac{db_2}{dt}+j\Delta\tilde{\omega}_{21}b_2=\frac{j\Omega_{12}^*}{2}b_1+\frac{j\Omega_{23}}{2}b_3,\quad \frac{db_3}{dt}+j\Delta\tilde{\omega}_{31}b_3=\frac{j\Omega_{13}^*}{2}b_1+\frac{j\Omega_{23}^*}{2}b_2, \tag{2a}$$

$$\Delta\tilde{\omega}_{21}=\Delta\omega_{21}-\frac{j\Gamma_2}{2},\quad \Delta\tilde{\omega}_{31}=\Delta\omega_{31}-\frac{j\Gamma_3}{2},\quad \Omega_{12}=\sum_i\frac{\Omega_{1i}\Omega_{i2}}{2}\left[\frac{1}{\omega_i-\omega_a}+\frac{1}{\omega_i-\omega_b}\right]. \tag{2b}$$

The Ω_{ij} are the respective Rabi frequencies; Ω_{12} is an effective Rabi frequency which is obtained by summing over intermediate states $|i\rangle$; Γ_2 and Γ_3 are noninterfering decay rates of states $|2\rangle$ and $|3\rangle$, respectively. The quantities $\Delta\omega_{21}=\omega_2-\omega_a-\omega_b-\omega_1$ and $\Delta\omega_{31}=\omega_3-\omega_d-\omega_1$. For use below, $\mu_{lm}=\langle l|er|m\rangle$ are the pertinent matrix elements.

We are interested in steady-state solutions and drop the derivatives in Eq. (2a). We take $|b_1|^2=1$, assume that Ω_{13} and Ω_{12} are very small as compared to Ω_{23}, and solve for b_2 and b_3. The per-atom dipole moment at ω_d is $2\mathrm{Re}[\mu_{31}b_3^* \exp(j\omega_d t)]$, and the dressed susceptibilities, as defined in Eq. (1), are

$$\hat{\chi}_D^{(1)}(-\omega_d,\omega_d)=\frac{|\mu_{13}|^2N}{\epsilon_0\hbar}\left\{\frac{-4\Delta\omega_{21}(|\Omega_{23}|^2-4\Delta\omega_{21}\Delta\omega_{31})+4\Delta\omega_{31}\Gamma_2^2}{(4\Delta\omega_{21}\Delta\omega_{31}-\Gamma_2\Gamma_3-|\Omega_{23}|^2)^2+4(\Gamma_2\Delta\omega_{31}+\Gamma_3\Delta\omega_{21})^2}\right.$$
$$\left.-j\frac{8\Delta\omega_{21}^2\Gamma_3+2\Gamma_2(|\Omega_{23}|^2+\Gamma_2\Gamma_3)}{(4\Delta\omega_{21}\Delta\omega_{31}-\Gamma_2\Gamma_3-|\Omega_{23}|^2)^2+4(\Gamma_2\Delta\omega_{31}+\Gamma_3\Delta\omega_{21})^2}\right\}, \tag{3a}$$

$$\hat{\chi}_D^{(3)}(-\omega_d,\omega_a,\omega_b,\omega_c)=\frac{\mu_{23}\mu_{31}N}{6\epsilon_0\hbar^3(\Delta\tilde{\omega}_{21}\Delta\tilde{\omega}_{31}-|\Omega_{23}|^2/4)^*}\sum_i\mu_{1i}\mu_{i2}\left(\frac{1}{\omega_i-\omega_a}+\frac{1}{\omega_i-\omega_b}\right), \tag{3b}$$

where N is the atom density. The caret on the susceptibilities in Eq. (3) denotes that they do not yet include collisional and Doppler broadening.

The transparency of the media is determined by the imaginary part of $\hat{\chi}_D^{(1)}$, i.e., the inverse absorption length $a=-2\pi\mathrm{Im}[\hat{\chi}_D^{(1)}]/\lambda$. For $\Gamma_2=\Delta\omega_{21}=0$, one finds perfect transparency at the generated sum frequency ω_d [Fig. 2(a)]. In general, the frequency of minimum absorption is $\omega_2+\omega_c$. For $\Omega_{23}\gg\Gamma_3>\Gamma_2$, the width of the transparency hole varies as $|\Omega_{23}|$ and the minimum loss varies as $\Gamma_2/|\Omega_{23}|^2$. In this regime it is the decay rate of state $|2\rangle$ and not of state $|3\rangle$ that determines transparency.

The refractive index of the media is determined by the real part of $\hat{\chi}_D^{(1)}$, $n-1=\mathrm{Re}[\hat{\chi}_D^{(1)}/2]$. On line center at any value of Ω_{23}, $n-1=0$, and the contribution of the resonance to the refractive index is zero. But the media is highly dispersive [Fig. 2(b)].

Figure 2(c) shows the magnitude of $\hat{\chi}_D^{(3)}$. It is resonantly enhanced and has a *constructive* interference at the center of the transparency hole. Calculation shows that $\hat{\chi}_D^{(3)}$ exhibits the symmetry property

$$\hat{\chi}_D^{(3)}(-\omega_d,\omega_a,\omega_b,\omega_c)=\hat{\chi}_D^{(3)}(-\omega_a,\omega_d,-\omega_b,-\omega_c)$$
$$=\hat{\chi}_D^{(3)}(-\omega_b,\omega_d,-\omega_a,-\omega_c).$$

We next consider the effect of dephasing collisions. To do so, Eq. (2a) is recast in density-matrix form and macroscopic collisional terms γ_{12}, γ_{13}, and γ_{23} are added to the off-diagonal equations. Maintaining the same assumptions one may show that the susceptibilities of Eq. (3) remain valid, but with the quantities Γ_2 and Γ_3 replaced by $\Gamma_2+\gamma_{12}$ and $\Gamma_3+\gamma_{13}$, respectively. (The quantity γ_{23} does not appear.) A perturber which shifts the frequency of a bare state $|3\rangle$ atom, in the presence of Ω_{23}, causes the dressed states to shift in opposite directions and results in a zero in the integrated phase perturbation[6] at ω_d. Therefore, transparency is only reduced by collisions which dephase the $|1\rangle$-$|2\rangle$ channel.

To include inhomogeneous or Doppler broadening the susceptibilities of Eq. (3) are integrated over a Gaussian density of states. Since now, $\Delta\omega_{21}$ for individual atoms is not zero, even when $\Gamma_2=0$, complete transparency is no longer obtained.

We turn next to Maxwell's equations. Assuming no depletion of the driving fields ω_a, ω_b, and ω_c, and temporal invariance of the envelope, the sum frequency field ω_d as a function of distance is obtained from

$$\frac{\partial E(\omega_d)}{\partial z}-\frac{\omega}{2c}\mathrm{Im}\chi_D^{(1)}E(\omega_d)+j\left[\frac{\omega}{2c}\mathrm{Re}\chi_D^{(1)}+\Delta k\right]E(\omega_d)$$
$$=-\frac{j3}{4}\frac{\omega}{c}\chi_D^{(3)}E(\omega_a)E(\omega_b)E(\omega_c) \tag{4}$$

with the boundary condition $E(\omega_d)=0$ at $z=0$. The quantity Δk in Eq. (4) contains all of the phase mismatch which results from transitions other than the resonance transition. The susceptibilities in Eq. (4), as described above, now include collisional and Doppler broadening.

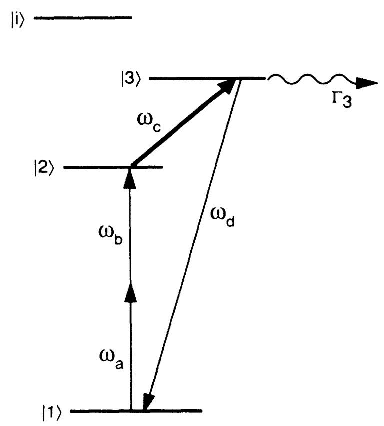

FIG. 1. Energy-level diagram for the sum frequency process $\omega_d=\omega_a+\omega_b+\omega_c$. State $|3\rangle$ is lifetime broadened with a decay rate Γ_3. When a strong field at frequency ω_c is tuned to line center of the $|2\rangle$-$|3\rangle$ transition, the media becomes transparent on the $|1\rangle$-$|3\rangle$ resonance transition. This allows much larger nonlinear $\chi^{(3)}L$ products than are normally possible.

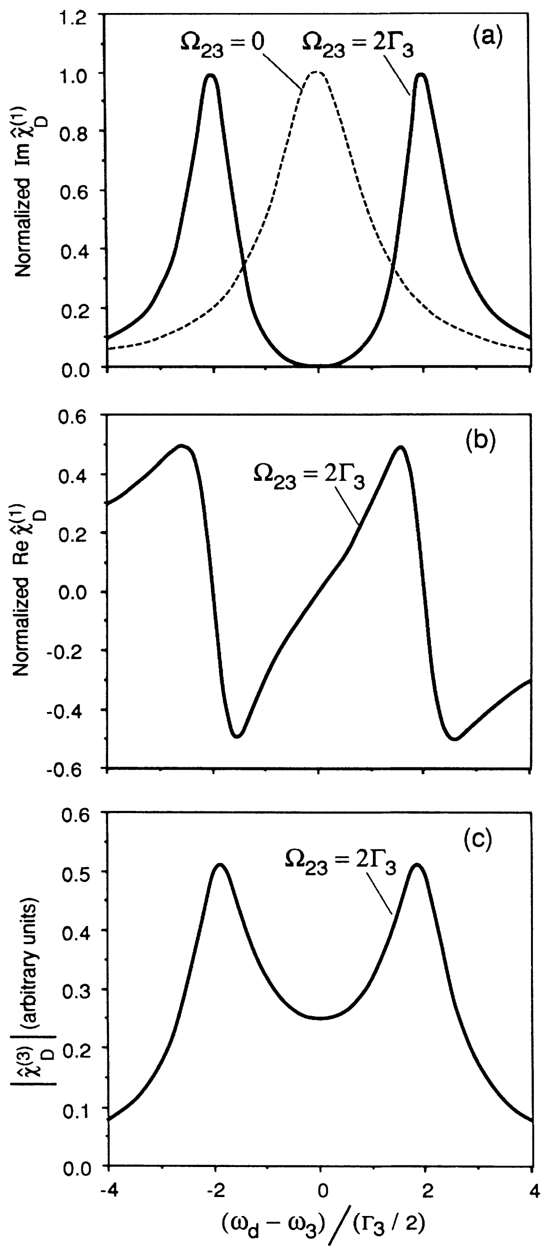

FIG. 2. Real and imaginary parts of $\hat{\chi}_D^{(1)}$ and $|\hat{\chi}_D^{(3)}|$ as a function of normalized detuning from state $|3\rangle$. Both Re$\hat{\chi}_D^{(1)}$ and Im$\hat{\chi}_D^{(1)}$ are normalized to the peak magnitude of Im$\hat{\chi}_D^{(1)}$ when $\Omega_{23}=0$.

The effect of the linear susceptibility on the growth of $E(\omega_d)$ is twofold: Its real part together with Δk causes periodic growth and decay as a function of distance; its imaginary part causes loss and limits the effective length of the nonlinear media.[1,2] We assume that ω_d is at the center of a Doppler-broadened transition, so that Re$\chi_D^{(1)}=0$, and that a phase-matching agent is added to offset Δk. (Or, instead, Δk is offset by detuning ω_d slightly from line center.) The electromagnetic field ω_d now grows toward a steady-state value determined by the ratio of $|\chi_D^{(3)}|$ and Im$\chi_D^{(1)}$.

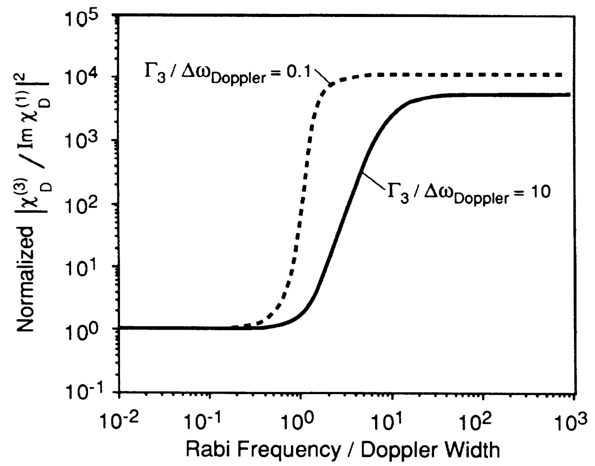

FIG. 3. $|\chi_D^{(3)}/\text{Im}\chi_D^{(1)}|^2$ as a function of the ratio of the Rabi frequency Ω_{23} to the Doppler width of the resonance transition. Normalization is to the small Ω_{23} value of this ratio. For this figure we take $\Gamma_2=0.01\Delta\omega_{\text{Doppler}}$.

We now come to a principal result of this Letter: Figure 3 shows the quantity $|\chi_D^{(3)}/\text{Im}\chi_D^{(1)}|^2$ as a function of the ratio of Ω_{23} to the Doppler width of the $|1\rangle$-$|3\rangle$ transition. When Ω_{23} is small as compared to the Doppler width, the media remains opaque and one obtains the result of traditional (small-field) nonlinear optics. As Ω_{23} approaches and exceeds the Doppler width the media becomes transparent. At the same time, as a result of increased Ω_{23}, the magnitude of $\chi_D^{(3)}$ is somewhat reduced. But $|\chi_D^{(3)}/\text{Im}\chi_D^{(1)}|^2$, which (for sufficiently long length) is proportional to the generated output power, increases over its small Ω_{23} value by the square of the ratio of the Doppler width to $\Gamma_2+\gamma_{12}$. If $\Gamma_3<\Delta\omega_{\text{Doppler}}$, then the onset of transparency occurs abruptly as Ω_{23} exceeds $\Delta\omega_{\text{Doppler}}$. If $\Gamma_3>\Delta\omega_{\text{Doppler}}$, then a larger Ω_{23} is required.

The foregoing paragraphs have assumed monochromatic fields at all frequencies. When the electromagnetic field at ω_c has finite linewidth, and a Lorentzian shape, this linewidth in effect dephases the $|2\rangle$-$|3\rangle$ transition;[7,8] the overall improvement which may be obtained is therefore determined by the square of the ratio of the Doppler width to the widest of the decay rate of state $|2\rangle$, the collisional broadening of the $|1\rangle$-$|2\rangle$ transition, or the linewidth of ω_c. The linewidth of the other frequencies will impose additional phase-matching, transparency, and group-velocity limitations.

The transparency which is described here is based on interference and is of a very different nature than that obtained by saturating a class of atoms (hole burning) within an inhomogeneous distribution. Transparency which is obtained by hole burning requires a continual energy input from the electromagnetic field and reduces both Im$\chi^{(1)}$ and $|\chi^{(3)}|$ so as to leave the nonlinearity-effective length product approximately unchanged. For our system, when $\Delta\omega_{21} = \Gamma_2 = 0$, the atomic system does not take energy from the field. Though the susceptibilities of Eq. (3) are only valid in the limit of a very small field at ω_d, at any value of this field the media remains transparent. This transparency is obtained by trapping[3] increasing "coherent" population in state $|2\rangle$.

It is important to note that the contribution to $\chi^{(3)}$ of all perturbation paths[1,2] involving the $|1\rangle$-$|3\rangle$ resonance transition, but not including state $|2\rangle$, are zero. Also, an effect of this type may not be created by splitting state $|3\rangle$, with, for example, a static magnetic field.

Atomic media in the visible or ultraviolet, at densities of 10^{16} atoms cm^{-3}, become opaque at distances on the order of a wavelength. To attain several centimeters of transparency requires a Rabi frequency on the $|2\rangle$-$|3\rangle$ transition of 10 to 50 times the resonance transition Doppler width. This requires power densities of several MW/cm^2, metastability of state $|2\rangle$, and a sufficiently narrow coupling laser linewidth. For these conditions one finds an improvement in conversion efficiency and four-frequency parametric gain, as compared to the traditional (small-field) formulas of nonlinear optics, which often exceed 10^4. Nonlinear susceptibility-length products become comparable to those of second-order (crystalline) media, and offer the possibility of improved nonlinear device operation over a wide range of the electromagnetic spectrum.

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