The ratio of the powers in the sideband +1 to that in the desired sideband is

$$\frac{P_{+1,20}}{P_{-1,00}} = \frac{1}{128}\left(\frac{\lambda L}{a^2}\right)^2\left[1+\frac{8}{\pi}-8\left(\frac{2}{\pi}\right)^2\right] = 2.38\times 10^{-3}\left(\frac{\lambda L}{a^2}\right)^2. \quad \text{(A.20)}$$

One quick estimate of the magnitude of the term (A.20) may be made by noting that

$$\frac{\lambda}{\pi w_0^2} \equiv \frac{1}{b} \quad \text{(A.21)}$$

where b is Kogelnik's b-parameter. Hence the power ratio (A.20) may be kept under control as long as the coupling distance $L = \pi/2c$ obeys the relationship.

$$\left(\frac{\lambda L}{\pi a^2}\right) = (L/b)\left(\frac{w_0}{a}\right)^2 < 1. \quad \text{(A.22)}$$

It should be noted that in these calculations, we have neglected the phase shift exp $[j(m+n+1)\tan^{-1}(z/b)]$ associated with Hermite Gaussian mode number and the phase shift due phase front curvature both of which multiply u_ν and u_0 [10]. However, in (A.3) the phase front curvature factor cancels because it is independent of the mode number. Ignoring the factor exp $[j(m+n+1)\tan^{-1} z/b]$ results in an overestimate of the power generated in the modes $m \neq 0, n \neq 0$.

References

[1] G. Magerl and E. Bonek, "A 2.75 GHz tunable CO_2-laser-infrared source," *J. Appl. Phys.*, vol. 47, pp. 4901–4903, Nov. 1976.

[2] W. E. Bicknell, L. R. Tomasetta, and R. H. Kingston, "10.6 μm FM-chirp radar using narrow-band correlation detection," *IEEE J. Quantum Electron.*, vol. QE-11, pp. 308–311, June 1975.

[3] G. M. Carter, "Tunable high-efficiency microwave frequency shifting of infrared lasers," *Appl. Phys. Lett.*, vol. 32, pp. 810–812, June 1978.

[4] C. F. Buhrer, D. Baird, and E. M. Conwell, "Optical frequency shifting by electro-optic effect," *Appl. Phys. Lett.*, vol. 1, pp. 46–49, Oct. 1962.

[5] C. F. Buhrer, L. R. Bloom, and D. H. Baird, "Electro-optic light modulation with cubic crystals," *Appl. Opt.*, vol. 2, pp. 839–846, Aug. 1963.

[6] J. P. Campbell and W. H. Steier, "Rotating-waveplate optical-frequency shifting in lithium niobate," *IEEE J. Quantum Electron.*, vol. QE-7, pp. 450–457, Sept. 1971.

[7] C. F. Buhrer, "Wideband electro-optic light modulation utilizing an asynchronous traveling-wave interaction," *Appl. Opt.*, vol. 4, pp. 545–550, May 1965.

[8] A. Yariv, "Coupled-mode theory for guided-wave optics," *IEEE J. Quantum Electron.*, vol. QE-9, pp. 919–933, Sept. 1973.

[9] J. E. Kiefer, T. A. Nussmeier, and F. E. Goodwin, "Intracavity CdTe modulators for CO_2 lasers," *IEEE J. Quantum Electron.*, vol. QE-8, pp. 173–179, Feb. 1972.

[10] A. Yariv, *Introduction to Optical Electronics*. New York: Holt, Reinhart and Winston, 1971, p. 40.

Transient Four-Wave Mixing and Real Time Holography in Atomic Systems

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Abstract—**The problem of transient four-wave mixing with noncoincident optical pulses is analyzed using the formalism of the time evolution and the density matrix operators. The results are relevant to problems involving real time holography and wave conjugation. The treatment establishes a bridge between the conventional formalisms of nonlinear optics and of photon echoes.**

THE subject of conjugate wave generation by nonlinear optical mixing has been the subject of considerable recent interest [1]-[5]. The formal analogy between the case of degenerate four-wave mixing and holography [6] has suggested the possibility of using four-wave mixing for real time holographic applications.

Heer and McManamon [7] first pointed out that wavefront correction and phase conjugation can take place in a photon-echo geometry involving forward traveling noncoincident four-wave mixing. Shiren [8] later showed that a backward conjugate echo can result when two of the noncoincident input pulses are opposite to each other.

The purpose of this note is to explore further these ideas and to develop a perturbation theory that can be applied to more general situations. This theory results in a formulation of the problem that is similar to the perturbational description of CW nonlinear optical mixing [9]. It also includes the effects of collisional relaxation (T_1, T_2) on the process of conjugate echo formation.

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We will first consider an ensemble of two-level atoms. The resonant levels are E_s and $E_m (E_m > E_s)$ and we will, for the moment, assume that no collisions exist so that the relaxation times are taken as infinite. The atoms are subjected to three pulses of duration δ of optical radiation of the same frequency ω but traveling along arbitrary directions

$$\bar{E}_i(\bar{r}, t) = \tfrac{1}{2}\bar{E}_i(t) \exp\,[i(\omega t - \bar{k}_i \cdot \bar{r})] + \text{c.c.}$$
$$i = 1, 2, 3. \tag{1}$$

The pulse amplitude envelopes $E_i(t)$ are shown in Fig. 1. The pulses may or may not coincide. We seek to solve for the polarization induced in the atomic medium which is third order in the applied fields. The pulse areas are taken to be much smaller than π. To establish a bridge between the problem described above and conventional nonlinear optics (where the three fields exist simultaneously) we will solve the problem using a perturbation expansion of the time evolution operator [10]. The wave function at time t is related to its value at an earlier time, say t_0, according to

$$\Psi(t) = \exp\left[-i\frac{\hat{H}_0}{\hbar}(t - t_0)\right]\Psi(t_0)$$
$$- \frac{i}{\hbar}\left\{\int_{t_0}^{t}\sum_g |g><g| \exp\,[-i\omega_g(t - t')]\,\hat{V}(t')\right.$$
$$\left.\cdot \exp\left[-i\frac{\hat{H}_0}{\hbar}(t' - t_0)\right] dt'\right\}\Psi(t_0)$$
$$= [\hat{U}^{(0)}(t - t_0) + \hat{U}^{(1)}(t - t_0)]\,\Psi(t_0) \tag{2}$$

where $\omega_g \equiv E_g/\hbar$ the total Hamiltonian is the sum of the unperturbed value $\hat{H}_0$ and the perturbation $\hat{V}(t)$ and

$$\hat{V}(t) = -\hat{\bar{\mu}} \cdot \bar{E}(\bar{r}, t) \tag{3}$$

with $\hat{\bar{\mu}}$ being the atomic dipole moment operator and where $\bar{E}(\bar{r}, t)$ is treated as a classical variable. Only the zero and first-order terms in the expansion of the time evolution operator are included.

In the limit of small "area" pulses $(\mu E\delta/\hbar) \ll 1$ and negligible dephasing during the pulse $(\omega_m - \omega_s - \omega)\delta \ll 1$ the operator $\hat{U}^{(1)}(t - t_0)$ of (2), which represents the effect (to first order in E) of a pulse at t_i on the wavefunction, becomes

$$\hat{U}^{(1)}(t_i) = \frac{i\delta}{2\hbar} \exp\,[\pm i(\omega t_i - \bar{k}_i \cdot \bar{r})] \sum_g |g><g|E_j^{\pm}\hat{\mu}_j$$
$$g = m \text{ or } s,\ E_j^- \equiv E_j^*,\ E_j^+ \equiv E_j,\ j = x, y, \text{ or } z. \tag{4}$$

We take $(\hat{\mu}_j)_{ms} = \mu_{jms}$ for $m \neq s$ and zero otherwise. It follows that if at t_i the atom is in the ground state $|s>$ then $\hat{U}^{(1)}(t_i)$ causes it to make a transition to the excited state $|m>$ and vice versa. In the first case, we use the lower (-) superscript while in the second case we use the upper (+) superscript. The choice of $E_j^+(\equiv E_j)$ or $E_j^-(\equiv E_j^*)$ is due to the fact that only terms with near vanishing resonance denominators are retained in the derivation leading to (4). (This is equivalent to the rotating wave approximation.)

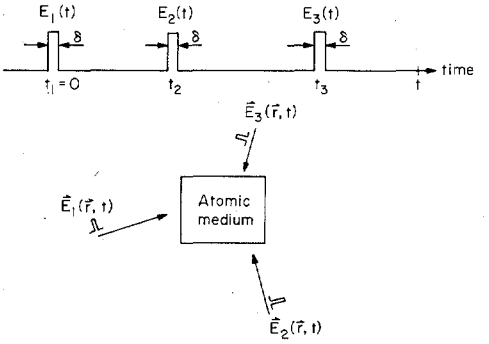

Fig. 1. The geometry for the interaction of the three-pulse sequence with the atomic medium.

We define the eigenfunction $\Psi^{(1,0,3)}$, as an example, as that component of $\Psi(t)$ that is due to "scattering" from $|s>$ to $|m>$ at t_1 (by E_1) and from $|m>$ back to $|s>$ at t_3 (by E_3), i.e.,

$$\Psi^{(1,0,3)}(t) = \hat{U}^{(0)}(t - t_3)\hat{U}^{(1)}(t_3)\hat{U}^{(0)}(t_3 - t_1)\hat{U}^{(1)}(t_1)|s>. \tag{5}$$

We can now use the formalism to calculate the j component of the induced atomic dipole moment $\langle\mu_j(t)\rangle$ following the sequence of three pulses. Specifically we will look first for a dipole moment which is proportional to $E_1^*E_2E_3$. Such a dipole moment results from $\langle\Psi^{(0,0,3)}|\mu_j|\Psi^{(1,2,0)}\rangle$ and from $\langle\Psi^{(0,2,0)}|\mu_j|\Psi^{(1,0,3)}\rangle$. To calculate the first term, we apply (4) to obtain

$$\Psi^{(1,2,0)} = \left(\frac{i\delta}{2\hbar}\right)^2 \mu_{1ms}\mu_{2sm}E_1^*E_2 \exp\,\{-i[(\omega t_1 - \bar{k}_1 \cdot \bar{r})$$
$$+ \omega_m(t_2 - t_1) - (\omega t_2 - \bar{k}_2 \cdot \bar{r}) + \omega_s(t - t_2)]\}|s> \tag{6}$$

$\Psi^{(1,2,0)}$ thus represents a component of $\Psi(t)$ which underwent a scattering $|s> \rightarrow |m>$ absorbing a photon from E_1 followed by $|m> \rightarrow |s>$ scattering due to the emission of a photon to E_2. Similarly

$$\Psi^{(0,0,3)} = \frac{i\delta}{2\hbar}\mu_{3ms}E_3^* \exp\,\{-i[(\omega t_3 - \bar{k}_3 \cdot \bar{r}) + \omega_m(t - t_3)$$
$$+ \omega_s(t_3 - t_1)]\}|m>. \tag{7}$$

When the time t_1 is taken, without loss of generality, as zero, the induced dipole moment at $t > t_3$ is

$$\langle\mu_j\rangle = \langle\Psi^{(0,0,3)}(t)|\mu_j|\Psi^{(1,2,0)}(t)\rangle$$
$$= \frac{i\delta^3}{8\hbar^3}\mu_{1ms}\mu_{2sm}\mu_{3sm}\mu_{jms}E_1^*E_2E_3 \exp\,\{i[\omega t - (-\bar{k}_1$$
$$+ \bar{k}_2 + \bar{k}_3) \cdot \bar{r} + (\omega_{ms} - \omega)(t - t_2 - t_3)]\} \tag{8}$$

where $\omega_{ms} \equiv \omega_m - \omega_s$.

A calculation of the contribution due to $\langle\Psi^{(0,2,0)}|\mu_j|\Psi^{(1,0,3)}\rangle$ gives a result identical to (8). The total value of $\langle\mu_j\rangle$ is thus twice that given by (8). Let the resonance offset parameter be $\Delta \equiv (\omega_{ms} - \omega)$. An inhomogeneous distribution of resonant

frequencies is described by a normalized lineshape function $g(\Delta)$. The induced polarization P_j is obtained by summing (8) over all Δ. The result is

$$P_j = N\int_{-\infty}^{\infty} \langle \mu_j \rangle g(\Delta)\, d\Delta$$

$$= \frac{i\delta^3}{4\hbar^3} N\mu_{1ms}\mu_{2sm}\mu_{3sm}\mu_{jms} E_1^* E_2 E_3 \exp\{i[\omega t - (-\bar{k}_1 + \bar{k}_2 + \bar{k}_3)\cdot \bar{r}]\} S(t - t_2 - t_3) \quad (9)$$

where $S(t) = \int_{-\infty}^{\infty} g(\Delta) e^{i\Delta t}\, d\Delta$ is the Fourier transform of the normalized lineshape function $g(\Delta)$ and N is the dipole moment density.

For an even $g(\Delta)$, the function $S(t - t_2 - t_3)$, hence P_j, is essentially zero except for a duration $\sim 2\pi/\Delta_{1/2}$ centered on

$$t = t_2 + t_3 \quad (10)$$

where $\Delta_{1/2}$ is the width of $g(\Delta)$. During this period the sample will radiate a photon echo provided the phase-matching conditions are satisfied. If the pulses E_2 and E_3 are exactly opposite, $\bar{k}_2 + \bar{k}_3 = 0$, then the radiated echo at $t = t_2 + t_3$ will propagate according to (9) in the direction $-\bar{k}_1$, i.e., the reverse of the direction of E_1. These results have been noted by Shiren [8] who applied the formalism of photon echoes. Our perturbation approach brings out specifically the field dependence $E_1^* E_2 E_3$ which is in a form identical to that used to describe four-wave conjugation in conventional nonlinear optics terminology [10]. It should be recalled however, that here $E_1(t)$, $E_2(t)$, and $E_3(t)$ do not, necessarily, coincide in time.

The output wave radiated by the polarization (9) is the complex conjugate of the first wave $E_1(t)$. In holographic terms we can describe the process as one whereby a "hologram" is written into the atomic medium by pulses $E_1(t)$ and $E_2(t)$ which do not coincide in time. An interrogation pulse at t_3 opposite in direction to $E_2(t)$ gives rise to the backward readout echo at $t = t_3 + t_2$ which is the conjugate replica of $E_1(t)$. If pulses E_2 and E_3 are parallel to each other as well as to pulse E_1, i.e., $\bar{k}_1 = \bar{k}_2 = \bar{k}_3$, then the induced echo has according to (8) a spatial dependence of $\exp[i(\omega t - \bar{k}_1 \cdot \bar{r})]$. It is thus radiated in the forward direction.

The special case of $\bar{E}_2(\bar{r}, t) = \bar{E}_3(\bar{r}, t)$ is of interest. The pulse formed is proportional to $E_1^* E_2^2 \exp\{i[\omega t - (2\bar{k}_2 - \bar{k}_1)\cdot r]\}$ and occurs, according to (10) at $t = 2t_2$. If $\bar{k}_1 = \bar{k}_2$ then the pulse is radiated in the forward direction and can be recognized as the usual forward photon echo [11]. It is interesting to note that it involves a third-order mixing except that two of the fields (E_2 and E_3) are degenerate.

It is of interest to inquire about the possibility of an echo proportional to $E_1 E_2^* E_3$ in which the second pulse is conjugated. An analysis similar to the above reveals that such an echo occurs at $t = t_3 - t_2 < t_3$ and is thus unrealizable except for the special case $t_2 = 0$, i.e., pulses E_1 and E_2 coincide.

We have discussed above the holographic analogy of the three pulse sequence leading to (9). It is thus important to determine the effect of the population relaxation time (T_1) and dephasing collisions (T_2) on the intensity of the radiated pulse. The induced dipole moment can be alternatively obtained as

$$\langle \mu_j \rangle = \text{Trace}\,(\hat{\rho}\hat{\mu}_j) \quad (11)$$

where $\hat{\rho}$, the density matrix operator, obeys the following equation of motion

$$\frac{d\rho_{ij}^{(1)}}{dt} - i\omega_{ji}\rho_{ij}^{(1)} + \frac{\rho_{ij}^{(1)}}{T_{ij}} = \frac{-i}{\hbar}[\hat{V}(t), \hat{\rho}^{(0)}]_{ij} \qquad i, j = 1, 2. \quad (12)$$

For $i = j$, T_{ij} equals T_1 and $\rho_{ii}^{(0)}$ is determined by the ith energy level thermal equilibrium population. For $i \neq j$, T_{ij} equals T_2 and $\rho_{ij}^{(0)} = 0$. Equation (12) is obtained by using a perturbation expansion of the density matrix operator [12] in terms of the applied fields and we shall look for a term in ρ_{12} that is proportional to the product of the three applied fields.

Upon the incidence of the first pulse, only the off diagonal density matrix elements, ρ_{12} and ρ_{21}, are perturbed to first order in E_1. The driving term on the right-hand side of (12) is proportional to $(\rho_{11}^{(0)} - \rho_{22}^{(0)})E_1(t)$. After the perturbation due to the first pulse, ρ_{12} evolves as $\exp[t(i\omega_{ms} - 1/T_2)]$ and in the limit of small δ

$$\rho_{12}^{(1)}(t_3 > t > t_1) = -i\frac{\mu_{1sm}}{2\hbar}\delta(\rho_{11}^{(0)} - \rho_{22}^{(0)})E_1 \exp[i(\omega t - \bar{k}_1 \cdot \bar{r})] \exp\left[\left(i\omega_{ms} - \frac{1}{T_2}\right)(t - t_1)\right]. \quad (13)$$

The second pulse affects the diagonal elements ρ_{11} and ρ_{22} giving rise to perturbation terms proportional to $E_1^* E_2$. In this case, the driving term in (12) is the product of $E_2(t)$ and the $\rho_{12}^{(1)}(t)$ given in (13). ρ_{11} and ρ_{22} then evolve as $\exp(-t/T_1)$ after the second pulse.

$$\rho_{11}^{(1)}(t > t_2) = \frac{-\mu_{1ms}\mu_{2sm}}{4\hbar^2}\delta^2(\rho_{11}^{(0)} - \rho_{22}^{(0)})E_1^* E_2 \cdot \exp[i\omega(t_2 - t_1) - i(\bar{k}_2 - \bar{k}_1)\cdot \bar{r}] \cdot \exp\left[-\left(i\omega_{ms} + \frac{1}{T_2}\right)(t_2 - t_1)\right] \cdot \exp\left[-\frac{1}{T_1}(t - t_2)\right]. \quad (14)$$

When the third pulse arrives, ρ_{12} and ρ_{21} are affected by driving terms proportional to $E_3(t)(\rho_{11}^{(1)}(t) - \rho_{22}^{(1)}(t))$ when the on diagonal elements are given in (14). Thus ρ_{12} and ρ_{21} are proportional to $E_1^* E_2 E_3$ and evolve as $\exp[t(i\omega_{ms} - 1/T_2)]$ for $t > t_3$. Setting $t_1 = 0$, the final expression for the induced polarization is

$$\bar{P}_j = \frac{i\delta^3}{4\hbar^3} N\mu_{1ms}\mu_{2sm}\mu_{3sm}\mu_{jms} E_1^* E_2 E_3 \exp\{i[\omega t - (-\bar{k}_1 + \bar{k}_2 + \bar{k}_3)\cdot r]\} \times S(t - t_2 - t_3) \exp\left[-\frac{1}{T_2}(t + t_2 - t_3) - \frac{1}{T_1}(t_3 - t_2)\right]. \quad (15)$$

At the height of the echo at time $t = t_2 + t_3$, the effect of relaxation is thus to reduce the radiated field by the factor $\exp[-(1/T_2)(2t_2) - (1/T_1)(t_3 - t_2)]$. It is thus necessary that the time delay t_2 between the first and second pulses be short compared to the homogeneous relaxation time T_2.

The above formalism can be easily applied to describe the interaction between a multilevel atomic system and a multipulse sequence (provided all the pulses have areas very much smaller than π). We shall next consider an interesting case of a three level system with energy levels E_a, E_b, and $E_c (E_c > E_b > E_a)$. The three atomic states are connected by the nonzero dipole matrix elements μ_{ab} and μ_{bc} while μ_{ac} is taken to be zero. The atoms are subjected to three pulses, each of width δ at times t_1, t_2, and t_3, respectively. The first two pulses are of the same frequency $\omega_1 = \omega_2 \simeq \omega_{ba}$, but can have different polarizations and wave vectors. They modulate the atomic state populations, which in turn, scatter the third applied pulse which has frequency $\omega_3 \simeq \omega_{cb}$. The induced dipole moment that involves all the three applied fields is caused by $\langle \Psi^{(0,2,0)} | \mu_j | \Psi^{(1,0,3)} \rangle$ and $\langle \Psi^{(1,0,0)} | \mu_j | \Psi^{(0,2,3)} \rangle$ resulting in

$$\langle \mu_j \rangle = \left(\frac{i\delta}{2\hbar}\right)^3 \mu_{1ab}\mu_{2ba}\mu_{3bc}\mu_{jcb} E_1 E_2^* E_3 \exp\{i[\omega_3 t - (\bar{k}_1 - \bar{k}_2 + \bar{k}_3)\cdot\bar{r} + (\omega_{ba} - \omega_1)(t_2 - t_1) + (\omega_{cb} - \omega_3)(t - t_3)]\} + \left(\frac{i\delta}{2\hbar}\right)^3 \mu_{1ab}\mu_{2ba}\mu_{3bc}\mu_{jcb} \cdot E_1^* E_2 E_3 \exp\{i[\omega_3 t - (-\bar{k}_1 + \bar{k}_2 + \bar{k}_3)\cdot\bar{r} - (\omega_{ba} - \omega_1)(t_2 - t_1) + (\omega_{cb} - \omega_3)(t - t_3)]\}. \quad (16)$$

When summed over the inhomogeneous distribution of the resonant frequencies of the atoms, it is observed that a maximum superradiant superposition of all the individual atomic polarizations is formed at t_3 provided that $t_2 = t_1$. To include the relaxation effect, the density matrix operators are solved and the induced polarization at $t > t_3$ and for the case $t_2 = t_1 = 0$ is

$$\langle \mu_j \rangle = \left(\frac{i\delta}{2\hbar}\right)^3 \mu_{1ab}\mu_{2ba}\mu_{3bc}\mu_{jcb} \exp\left\{-\frac{1}{T_2}(t - t_3) - t_3\frac{1}{T_1} + i(\omega_{cb} - \omega_3)(t - t_3)\right\} \times \{E_1 E_2^* E_3 \cdot \exp\{i[\omega_3 t - (\bar{k}_1 - \bar{k}_2 + \bar{k}_3)\cdot r]\} + E_1^* E_2 E_3 \cdot \exp\{i[\omega_3 t - (-\bar{k}_1 + \bar{k}_2 + \bar{k}_3)\cdot\bar{r}]\}\}. \quad (17)$$

The first term of this polarization gives rise to a radiated field of frequency ω_3 and wave vector $\bar{k}_r$ with $k_r = (\omega_3/c)n(\omega_3)$ provided

$$\bar{k}_1(\omega_1) - \bar{k}_2(\omega_2) + \bar{k}_3(\omega_3) = \bar{k}_r(\omega_3) \quad (18)$$

where $n(\omega_3)$ is the material index of refraction at ω_3. The roles of E_1 and E_2 are reversed in the second term of the induced polarization. In an atomic medium in which $\omega_{ba} = \omega_{cb}$, ω_3 can be made equal to ω_1, and (18) is easily satisfied. In particular, E_1 and E_3 can be chosen to be the two counterpropagating pump fields and a backward conjugate replica of E_2 is produced independent of the direction of $\bar{k}_2$.

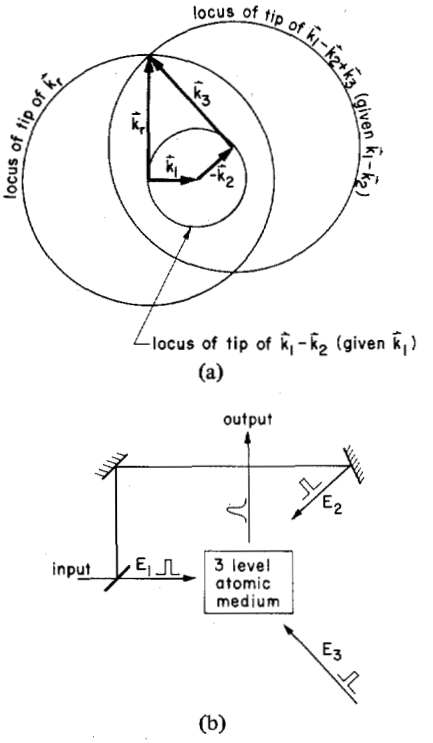

Fig. 2. (a) A graphic construction for satisfying the phase-matching condition $\bar{k}_1(\omega_1) - \bar{k}_2(\omega_2) + \bar{k}_3(\omega_3) = \bar{k}_r(\omega_3)$ in an isotropic medium for $\omega_3 > \omega_2 = \omega_1$; (b) frequency upconverter.

The phase-matching condition of (18) can also be met in a birefringent or an isotropic medium. Fig. 2(a) shows a graphic construction for satisfying the phase-matching condition in an isotropic medium for $\omega_3 > \omega_2 = \omega_1$. Given some choice of $\bar{k}_1(\omega_1)$ and $\bar{k}_2(\omega_1)$ the direction of $\bar{k}_3(\omega_3)$ and the radiated field $\bar{k}_r(\omega_3)$ is determined as shown.

We note that the above described interaction between the three-level system and the pulse sequence can be used as a frequency upconverter. Consider the situation where a light pulse at frequency ω_1 and intensity I_1 is first split into two beams (E_1 and E_2) and incident onto the atomic medium with k-vectors as shown in Fig. 2(b). Upon the incidence, at a later time t_3, of a third pulse at frequency ω_3 and intensity I_3 in the direction determined in Fig. 2(a), an output pulse at frequency ω_3 and of duration $1/\Delta_{1/2}$ will be detected in the direction $\bar{k}_r$. The pulse intensity is proportional to $(I_1)^2 I_3 \cdot \exp(-2t_3/T_1)$. We thus observe that a weak and low frequency input signal can be detected, amplified, delayed (by t_3), and converted to a high frequency output signal, which is emitted as a superradiant pulse along a specific direction. We note that there were other schemes proposed to observe echo phenomena in three-level atomic systems [13].

In conclusion, we have used a simplified time evolution operator formalism to analyze the transient phenomena of four-wave mixing of noncoincident pulses in atomic systems. The relaxation effects are also included by using a density matrix formalism. The treatment is useful (in the small pulse area

limit) in describing photon echoes and thus establishes a bridge between the conventional theories of nonlinear optics and that of coherent pulse excitation of superradiant atomic systems.

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REFERENCES

[1] B. Y. Zel'dovich, V. I. Popovichev, V. V. Ragul'skii, and F. S. Faizullov, *Sov. Phys.–JETP Lett.*, vol. 15, p. 109, 1972.
[2] A. Yariv, *Appl. Phys. Lett.*, vol. 28, p. 99, 1976.
[3] R. W. Hellwarth, *J. Opt. Soc. Amer.*, vol. 67, p. 1, 1977.
[4] D. M. Bloom and G. C. Bjorklund, *Appl. Phys. Lett.*, vol. 31, p. 592, 1977.
[5] A. Yariv and D. M. Pepper, *Opt. Lett.*, vol. 1, p. 16, 1977.
[6] A. Yariv, *Opt. Commun.*, vol. 25, p. 23, 1978.
[7] C. V. Heer and P. F. McManamon, *Opt. Commun.*, vol. 23, p. 49, 1977.
[8] N. S. Shiren, *Appl. Phys. Lett.*, vol. 33, p. 299, 1978.
[9] J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, *Phys. Rev.*, vol. 127, p. 1918, 1962.
[10] A. Messiah, *Quantum Mechanics II*. New York: Interscience, 1962.
[11] I. D. Abella, N. A. Kurnit, and S. R. Hartmann, *Phys. Rev.*, vol. 141, p. 319, 1966.
[12] A. Yariv, *Quantum Electronics*, 2nd ed. New York: Wiley, 1975.
[13] S. R. Hartmann, "H-3–Photon, spin, and Raman echoes," *IEEE J. Quantum Electron.*, vol. QE-4, pp. 802–807, Nov. 1968.

Servo Tuning and Stabilization of Nonlinear Optical Crystals

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Abstract–**A new optical technique for servo tuning and stabilization of angle tuned nonlinear optical crystals is described. This technique has zero insertion loss and works equally well with collimated, focused, or noncollinear fundamental beams. Servo tuning of a Type I KDP nonlinear crystal for summing the output of a tunable dye laser with the Nd:YAG second harmonic and servo stabilization of the production of the Nd:YAG fourth harmonic in Type I KDP are demonstrated.**

NONLINEAR optical crystals are extensively used to extend the wavelength range covered by fixed frequency and tunable lasers [1]-[3]. Coherent radiation at new wavelengths is produced either by direct harmonic generation utilizing a single laser source or by frequency mixing the outputs of two separate lasers. Phase matching, which is necessary for efficient power conversion, is usually achieved by utilizing birefringent nonlinear crystals and adjusting the degree of birefringence to compensate for the index of refraction dispersion between the fundamental and generated wavelengths. Adjustment of the phase matching is accomplished either by changing the crystal temperature ("temperature tuning") or by changing the internal direction of propagation through the crystal with respect to the optic axis ("angle tuning"). Angle tuning, which is usually accomplished by physically rotating the crystal with respect to the fixed direction of propagation of the laser beam, has the advantage of permitting rapid adjustment of phase matching to keep up with variations of the frequencies of the fundamental beams or with laser induced heating of the crystals.

For applications which require either long term stability of the power level of the generated radiation or which require frequency mixing using broadly tunable input lasers, automatic servo control of the angle tuning is desirable. In order to utilize angle tuning for servo control of nonlinear crystals, however, a means must be found to provide a discriminant signal which is sensitive to the *direction* of the deviation from the optimum crystal orientation. For the case of second-harmonic generation of tightly focused beams, Kuhl and Spitschan [4] have developed an elegant scheme wherein the discriminant signal is provided by monitoring the spatial position of the generated beam. Their method is useful for second-harmonic generation of moderate power dye lasers, since a tight focusing geometry is commonly employed to provide maximum generated output. However, when the fundamental beam is collimated or slightly converging, which is usually the optimum case for high power lasers, or when the fundamental beams are noncollinear, the spatial position of the generated beam does not provide an adequate discriminant [5].

In this paper, we report a new optical technique for obtaining a discriminant signal suitable for servo tuning and stabilization of angle tuned uniaxial nonlinear crystals. This technique has zero insertion loss and works equally well with collimated, slightly converging, slightly diverging, or tightly focused funda-

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