PHYSICAL REVIEW A VOLUME 39, NUMBER 5 MARCH 1, 1989

Quantum coherent states and the second-order susceptibility

Richard T. Hammond
Code 5052, Naval Air Development Center, Warminster, Pennsylvania 18972

(Received 9 November 1988)

The second-order susceptibility is calculated in a time-dependent, fully quantized system using coherent states. This is done for the two different interaction Hamiltonians of quantum optics, and the results are compared and discussed. Also, the contribution from the second harmonic originally present in the incident beam is separated out and its effect is assessed.

I. INTRODUCTION

One of the most common nonlinear effects in use today is the second-harmonic generation of coherent light, which was first reported in 1961.[1] This effect is concisely described by the second-order susceptibility, $\chi^{(2)}$, which has been extensively described in the literature.[2-4] It is the purpose of this paper to calculate $\chi^{(2)}$ in a fully quantized time-dependent treatment using coherent states, and to show some new results from this treatment which are not generally reported in the literature.

Coherent states may be defined as the eigenfunctions of the annihilation operator a_σ, i.e.,

$$a_\sigma|\alpha_\sigma\rangle=\alpha_\sigma|\alpha_\sigma\rangle \ , \tag{1}$$

where $|\alpha_\sigma|^2$ represents the mean number of photons in the coherent state $|\alpha_\sigma\rangle$ of frequency ω_σ. Coherent states are used here because these best describe laser radiation, which is usually used to generate second harmonics. Some novel effects predicted by the use of quantum coherent states have been described elsewhere.[5,6]

A convenient way of calculating second-order effects makes use of the density matrix ρ. With this the expectation value of the polarization vector $\mathbf{P}$ may be found from

$$\langle \mathbf{P}\rangle=\mathrm{Tr}(\rho\mathbf{P}) \ , \tag{2}$$

from which the second-order part may be extracted. From this, $\chi^{(2)}$ is found (Sec. II). Time-dependent calculations will be performed, so the second-order part of $\mathbf{P}$ can be used to assess time dependence of the second harmonic.

In using the above formalism, the interaction Hamiltonian of the electron and the field must be prescribed. There are two well-known forms of this interaction. One is obtained through minimal coupling and is given by $-e\,\mathbf{A}\cdot\mathbf{p}/m$ and the other is given by $-\boldsymbol{\mu}\cdot\mathbf{E}$, where $\mathbf{A}$ is the vector potential and $\mathbf{E}$ is the electric field, and the other variables describe the electron where $\mathbf{p}$ is the momentum and $\boldsymbol{\mu}=e\mathbf{r}$ is the dipole moment. The interaction term that is described by the field may be obtained from the potential term via a unitary transformation. Over the years there has been much controversy concerning which of these forms is the better.[7-10] In this paper, $\chi^{(2)}$ will be calculated for each interaction form. It will be shown that the correct form to use depends upon which basis functions are chosen, and that the $-e\,\mathbf{A}\cdot\mathbf{p}/m$ form is the more natural choice. It will be seen that the forms of these results are different.

Another topic which is investigated in this paper is the following. In practice, no incident beam is truly monochromatic. Long pulses may be well approximated by a monochromatic beam but very short pulses cannot. In many derivations of $\chi^{(2)}$, the incident (classical) field is considered as a Fourier sum (or transform), so that some of the contribution to $\chi^{(2)}$ comes from the second harmonic originally present in the beam. Therefore different experimenters can measure different values of $\chi^{(2)}$ depending on the shape of their pulse. These effects will become pronounced for very short pulses. To assess these effects, it will be assumed that, at the initial time t_0, the radiation part of the density operator consists of a mixed state. From this, the contribution from the second harmonic (originally present in the beam) on $\chi^{(2)}$ can be determined. It will be shown that $\chi^{(2)}$ is actually reduced in value by an amount proportional to the ratio of the power in the second harmonic to that of the fundamental. Throughout the paper, naturalized ($\hbar=c=1$) Gaussian units are used.

II. THE POLARIZATION VECTOR

In this section the formalism will be developed for an interacting system without specifying the form of the interaction. From its definition, the equation of motion of the density operator, in the interaction picture (IP), can be derived, and is given by

$$\frac{i\partial\rho}{\partial t}=[V,\rho] \ , \tag{3}$$

where V is the interaction Hamiltonian. From (3) and (2), upon performing repeated integration, and assuming that $\mathbf{P}=-eN\mathbf{r}$ where N is the number density of dipoles, one obtains

$$\langle P\rangle^{(2)}=eN\,\mathrm{Tr}\left[\mathbf{r}\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2[V(t_1)V(t_2)\rho(t_0)-V(t_1)\rho(t_0)V(t_2)+\rho(t_0)V(t_2)V(t_1)-V(t_2)\rho(t_0)V(t_1)]\right] \ , \tag{4}$$

where the trace is over the radiation and atomic variables. In (4), only the second-order part is retained, and in the following it will be understood that only the second-order part is retained. This expression may be simplified by using time-ordering techniques and the cyclic property of the trace, so that, defining

$$I(t)=\int_{t_0}^{t}dt'V(t') , \tag{5}$$

one obtains from (4),

$$\langle \mathbf{P}\rangle = eN\,\mathrm{Tr}\left[[\mathbf{r},I]I\rho(t_0)+\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2V(t_1)V(t_2)[\rho(t_0),\mathbf{r}]\right] . \tag{6}$$

Since the atomic and radiation fields are independent, the density operator at $t=t_0\equiv 0$ may be written in product form, i.e., $\rho(0)=\rho_A(0)\rho_B(0)$. It will be assumed that the material is in some definite state at $t=0$, so that $\rho_A(0)=|i\rangle\langle i|$, but that the incident radiation is not a pure mode, so that $\rho_R=\sum_\sigma p_\sigma|\alpha_\sigma\rangle\langle\alpha_\sigma|$, where p_σ is the probability for the state $|\alpha_\sigma\rangle$. It is easy to generalize to the case of a mixed atomic state as well, but nothing is gained for the purposes here.

The following notation is useful: The coherent state $|\alpha_\sigma\rangle$ is sometimes written as $|\sigma\rangle$ for short, and

$$I_{mn}^{\sigma\beta}=\langle\sigma|I_{mn}|\beta\rangle,\quad I_{mn}=\langle m|I|n\rangle , \tag{7}$$

where $|\beta\rangle$ represents a coherent state and $\{|m\rangle\}$ spans the atomic Hilbert space, and are designated as the eigenfunctions of the interaction-free part of the Hamiltonian given below. With this, (6) becomes

$$\begin{aligned}\langle \mathbf{P}\rangle = eN&\sum_{m,n,\sigma,\beta}p_\sigma\mathbf{r}_{im}(I_{mn}^{\alpha\beta}-I_{im}^{\sigma\beta})I_{ni}^{\beta\sigma}\\ &+eN\sum_\sigma p_\sigma\left\langle\sigma\left|\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2V(t_1)V(t_2)\right.\right.\\ &\qquad\left.\left.\times[\rho(t_0),\mathbf{r}]\right|\sigma\right\rangle .\end{aligned} \tag{8}$$

This is still a cumbersome expression and can be simplified greatly by using the two-level approximation. Assuming only two photon processes occur so that I_{mn} is nonzero only for $m\neq n$, (8) becomes

$$\langle \mathbf{P}\rangle = eN\sum_\sigma p_\sigma\boldsymbol{\Delta}\langle\sigma|I_{if}I_{fi}|\sigma\rangle , \tag{9}$$

where $\boldsymbol{\Delta}=\mathbf{r}_{ii}-\mathbf{r}_{ff}$.

For an ideal, purely monochromatic state, $p_\sigma=\delta_{\sigma 1}$ and the sum collapses. This case would represent a good approximation where the pulse is very long. However, pulses are really finite, and recently there has been much interest in ultrashort pulses. In this case, the beam cannot be assumed to be monochromatic and the effect of the various harmonics present in the beam will contribute to $\langle\mathbf{P}\rangle$. This effect will be investigated below. To proceed further, the interaction Hamiltonian must be chosen, which is the subject of the following section.

III. THE $-e\,\mathbf{A}\cdot\mathbf{p}/m$ INTERACTION

The object of this section is to evaluate (9). As stated in the Introduction, there are two forms of the interaction Hamiltonian used in quantum optics. The correct, rigorous way to obtain the Hamiltonian is to replace the derivative by the gauge-covariant derivative, i.e., $\partial_\mu\to\partial_\mu+ie\phi_\mu$, in the Hamiltonian, where ϕ_μ is the electromagnetic four potential. This leads, in the Coulomb gauge, assuming the quadratic term in $\mathbf{A}$ is negligible, to

$$H=H_0-(e/m)\mathbf{A}\cdot\mathbf{p} , \tag{10}$$

where

$$H_0=\mathbf{p}^2/2m+V(r)+(1/8\pi)\int d^3x(\mathbf{E}^2+\mathbf{B}^2) , \tag{11}$$

and where $\mathbf{E}$ represents the transverse part of the field and the last term in (11) becomes $H_F\equiv\sum_\sigma\omega_\sigma a_\sigma^\dagger a_\sigma$, disregarding the zero-point energy. Thus (10) shows that the interaction Hamiltonian is given by $V=-(e/m)\mathbf{A}\cdot\mathbf{p}$. It is assumed that the atomic eigenfunctions $|n\rangle$ are known, so that defining $H_0^A\equiv\mathbf{p}^2/2m+V(r)$, one assumes that $H_0^A|n\rangle=E_n|n\rangle$. The other form of the interaction will be discussed in Sec. IV.

The electromagnetic potential, in the Schrödinger picture, is given by

$$\mathbf{A}=\sum_\nu(2\omega_\nu L^3)^{-1/2}\hat{\mathbf{e}}_\nu(a_\nu e^{i\mathbf{k}_\nu\cdot\mathbf{r}}+a_\nu^\dagger e^{-i\mathbf{k}_\nu\cdot\mathbf{r}}) , \tag{12}$$

and, for later use, the field,

$$\mathbf{E}=\sum_\nu i(\omega_\nu/2L^3)^{1/2}\hat{\mathbf{e}}_\nu(a_\nu e^{i\mathbf{k}_\nu\cdot\mathbf{r}}-a_\nu^\dagger e^{-i\mathbf{k}_\nu\cdot\mathbf{r}}) , \tag{13}$$

where L^3 is the quantization volume. In the following, the dipole approximation is used since only optical frequencies are being dealt with, so that the exponents in (12) and (13) are approximated by unity.

In (9), the terms are given in the IP so that

$$V_{\mathrm{IP}}=-e^{iH_0t}(e/m)\mathbf{A}\cdot\mathbf{p}e^{-iH_0t} . \tag{14}$$

Using this, all interaction terms are put into the IP. One then obtains

$$\begin{aligned}I_{mn}^{\sigma\beta}=-i\Big\langle\sigma\Big|&\sum_\nu(1/2L^3\omega_\nu)^{1/2}\hat{\mathbf{e}}_\nu\cdot\boldsymbol{\mu}_{mn}\\ &\times\int_0^t dt'(a_\nu e^{-i\omega_\nu t'}+a_\nu^\dagger e^{i\omega_\nu t'})\\ &\times e^{i\omega_{mn}t}\omega_{mn}\Big|\beta\Big\rangle ,\end{aligned} \tag{15}$$

where $\omega_{mn}=E_m-E_n$, and $\mathbf{p}_{mn}=i(m/e)\omega_{mn}\boldsymbol{\mu}_{mn}$.

This result may be used in (9) which gives, upon integration, simplification, and writing $\alpha_\sigma=|\alpha_\sigma|e^{i\xi_\sigma}$,

$$\langle \mathbf{P}^{(2)} \rangle = -(eN/L^3) \sum_{\sigma} p_\sigma (\Delta/\omega_\sigma) |\boldsymbol{\mu}_{if} \cdot \hat{\mathbf{e}}_\sigma|^2 |\alpha_\sigma|^2 \omega_{if}^2 \times \left[\frac{1-\cos(\omega_{if}^{-} t)}{\omega_{if}^{-}\omega_{fi}^{+}} + \frac{1-\cos\omega_{if}^{+} t}{\omega_{if}^{+}\omega_{fi}^{-}} + \frac{\cos(2\xi_\sigma)+\cos(2\xi_\sigma-2\omega_\sigma t)-\cos(2\xi_\sigma+\omega_{if}^{-} t)}{\omega_{if}^{-}\omega_{fi}^{-}} - \frac{\cos(2\xi_\sigma+\omega_{fi}^{-} t)}{\omega_{if}^{-}\omega_{fi}^{-}} \right] , \tag{16}$$

where $\omega_{if}^{\pm} = \omega_{if} \pm \omega_\sigma$.

This result has some interesting features. The time-independent part, which represents the dc rectification value, depends upon the phase ξ_σ of the coherent state. This result has been discussed elsewhere[5] and for the purposes here, the time scale will be shifted so that $\cos(2\xi_\sigma + \omega_{if}^{\mp} t) \rightarrow \cos(\omega_{if}^{\mp} t)$. Consider now that the system is operated far from resonance, so that $\omega_\sigma \gg \omega_{if}$. Retaining only the frequency-doubled part, (16) becomes

$$\langle \mathbf{P}^{(2)} \rangle = (eN/L^3)(\Delta/\omega^3) |\boldsymbol{\mu}_{if} \cdot \hat{\mathbf{e}}_\sigma|^2 \omega_{if}^2 \times [p_1|\alpha_1|^2 - (p_2/2)|\alpha_2|^2] \cos(2\omega t) , \tag{17}$$

where $\omega_2 = 2\omega_1 = 2\omega$.

This result shows that there are two contributions to the second-order part of the polarization vector. If the original laser beam were purely monochromatic then (17), with $p_2 = 0$ and $p_1 = 1$, would represent the pure harmonic conversion. A nonzero value of p_2 corresponds to a second harmonic in the original beam, which, in general, will exist in any pulse, although it will become negligible as the pulse gets long. The point is, this contributes to the second-harmonic conversion and its strength depends on the shape of the original beam. This implies that $\chi^{(2)}$ will have different measured values for differently shaped pulses. As mentioned, this effect will be pronounced only for very short pulses.

In order to calculate $\chi^{(2)}$ from the above, consider the scalar case, so that (the tensor indices on the susceptibility can be resurrected in the usual way)

$$\langle \mathbf{P} \rangle = \mathrm{Tr}(\rho \mathbf{P}) = \chi^{(2)} \mathrm{Tr}(\mathbf{E}^2) , \tag{18}$$

where $\mathrm{Tr}(\mathbf{E}^2)$ may be evaluated using (13). Doing this, one obtains

$$\chi^{(2)} = -eN(\Delta/\omega_\sigma^4) |\boldsymbol{\mu}_{if} \cdot \hat{\mathbf{e}}_\sigma|^2 \omega_{if}^2 \times (1 - p_1|\alpha_1|^2 / 2p_2|\alpha_2|^2) . \tag{19}$$

The second term in parentheses may be written in terms of the relative power of the second harmonic, originally present in the beam, to the fundamental. To do this, first label each frequency component by $\mathbf{E}_\sigma$. Then, one can show that

$$\frac{\mathrm{Tr}(E_2^2)}{\mathrm{Tr}(E_1^2)} = \frac{p_2|\alpha_2|^2}{p_1|\alpha_1|^2} , \tag{20}$$

and that, if one replaces the trace operation by a time average on the squared classical fields, the amplitudes of which are $\mathcal{E}_1$ and $\mathcal{E}_2$, then one obtains

$$\frac{p_2|\alpha_2|^2}{p_1|\alpha_1|^2} \rightarrow \frac{1}{2} \left| \frac{\mathcal{E}_2}{\mathcal{E}_1} \right|^2 \equiv \frac{R}{2} , \tag{21}$$

where R is the ratio of power. Thus (19), with (21), yields the final form for the value of $\chi^{(2)}$. It is seen that the value of $\chi^{(2)}$ depends upon the amount of second-harmonic power originally present in the beam, and that the effect of the second harmonic is to reduce the value of the second-order susceptibility.

IV. THE $-e\mathbf{r}\cdot\mathbf{E}$ INTERACTION

An alternate, widely used form for the interaction Hamiltonian may be derived through a unitary transformation given by

$$U = e^{ie\mathbf{r}\cdot\mathbf{A}} . \tag{22}$$

The treatment here follows that of Ackerhalt and Milonni,[9] and details may be found there. There are two ways to use this transformation. Consider first that one transforms both the wave function and the variables, such that

$$|n^{\mathrm{new}}\rangle = U^\dagger |n\rangle \tag{23}$$

and

$$\mathbf{p}^{\mathrm{new}} = U^\dagger \mathbf{p} U = \mathbf{p} + e\,\mathbf{A} , \tag{24}$$

and similarly for the rest of the variables. With this, the Hamiltonian becomes

$$H^{\mathrm{new}} = H_0 + H_F - e\mathbf{r}\cdot\mathbf{E} , \tag{25}$$

where a term quadratic in $\boldsymbol{\mu}$ has been dropped.[11] One should note that the interaction-free part of the Hamiltonian is the same as before.

The above equation gives the alternate form of the interaction Hamiltonian, $-e\mathbf{r}\cdot\mathbf{E}$. However, when using this form, one must use the transformed (new) eigenfunctions given by (23). Thus, calling $W_n = E_n + E_F$, where $E_F = \langle \alpha | H_F | \alpha \rangle$,

$$H_0 |\alpha, n^{\mathrm{new}}\rangle = W_n^{\mathrm{new}} |\alpha, n^{\mathrm{new}}\rangle \neq W_n |\alpha, n\rangle . \tag{26}$$

As a matter of fact, writing $|\alpha, n\rangle = |\alpha\rangle |n\rangle$, one obtains from (26)

$$W_n^{\mathrm{new}} = W_n + \langle \alpha, n | U[H_0, U^\dagger] | \alpha, n \rangle . \tag{27}$$

To evaluate the commutator, one uses (11), (12), and (22). Then, using the property

$$[ab, c] = a[b, c] + [a, c]b \tag{28}$$

and deducing the commutation relation for exponents, one may derive, recognizing (13),

$$W_n^{\mathrm{new}} = E_n + E_F + \boldsymbol{\mu}_{nn} \cdot \langle \alpha | \mathbf{E} | \alpha \rangle , \tag{29}$$

where again a term proportional to μ^2 has been dropped.

This result shows that the energy eigenvalues obtained after the unitary transformation are different than those of the original formulation. This does not violate the notion that the physics is preserved under a unitary transformation, however. The crucial point is in identification. To see this, compare (10) and (25). In each case H_0 is the same. This is why there are different eigenvalues of the unperturbed Hamiltonian (recall that the eigenfunction has been transformed). Thus, if one elects to use the $-\boldsymbol{\mu}\cdot\mathbf{E}$ form of the interaction, one cannot use simultaneously the $\{|n\rangle\}$ as the unperturbed eigenfunctions. However, in a given problem, it is usually the $\{|n\rangle\}$ that are known, or can be approximated. For example, in dealing with, say, sodium vapor, or any hydrogenlike vapor, it is the eigenvalues of H_0^A operating on $\{|n\rangle\}$ that are known (the E_n), and not the eigenvalues of H_0 operating on $U^\dagger|\alpha\rangle$. The above result, (29), shows this difference. Thus, using the eigenvalues of $\{|n\rangle\}$, $-e\,\mathbf{A}\cdot\mathbf{p}/m$ is the correct form of the interaction to use. If, on the other hand, the eigenvalues of $U^\dagger|\alpha\rangle$ are known, then $-\boldsymbol{\mu}\cdot\mathbf{E}$ should be used as the interaction Hamiltonian.

Since this is so important, it will be discussed from another point of view. Again, the treatment of Ackerhalt and Milonni[9] is used. In this case the wave function is not transformed but the Hamiltonian is written in terms of transformed (new) canonical variables:

$$\mathbf{p}_{\rm new}=U\mathbf{p}U^\dagger=\mathbf{p}-e\,\mathbf{A}\ , \tag{30}$$

$$E^j_{\rm new}=E^j+4\pi\mu_i\delta^{ij}_T\ , \tag{31}$$

where δ^{ij}_T is the transverse δ function, and the rest of the variables are invariant under the transformation. Now the Hamiltonian may be written as

$$H=\mathbf{p}^2_{\rm new}/2m+V(r)-\boldsymbol{\mu}\cdot\mathbf{E}_{\rm new}+(1/8\pi)\int d^3x(\mathbf{E}^2_{\rm new}+\mathbf{B}^2_{\rm new})\ , \tag{32}$$

where, again, a term proportional to μ^2 has been dropped. One immediately recognizes the interaction term $-\boldsymbol{\mu}\cdot\mathbf{E}_{\rm new}$. This Hamiltonian is used with the original, untransformed wave functions. This is because (32) is the same as (10), but written in terms of a new, yet equivalent, set of canonical variables. This point is, one may be tempted to use (32), with the untransformed states, to justify the use of $-\boldsymbol{\mu}\cdot\mathbf{E}_{\rm new}$ as the interaction Hamiltonian. This would tacitly assume, however, that $H^A_{0,\rm new}\equiv\mathbf{p}^2_{\rm new}/2m+V(r)$. However, (30) shows that $\mathbf{p}^2_{\rm new}$ already contains the original interaction terms, so one must give up the eigenvalue equation $H_0^A|n\rangle=E_n|n\rangle$. A similar argument holds for the radiation part of the Hamiltonian. The essential point is that if $-\boldsymbol{\mu}\cdot\mathbf{E}_{\rm new}$ is used, then one cannot use the eigenfunctions and values given by $H_0^A|n\rangle=E_n|n\rangle$. When these eigenvalues and eigenfunctions are used, then $-e\,\mathbf{A}\cdot\mathbf{p}/m$ must be used as the interaction Hamiltonian.

Another point that should be mentioned here is that the electric field used in the $-\boldsymbol{\mu}\cdot\mathbf{E}_{\rm new}$ term is not the original electric field, as can be seen from (31). The new, extra part comes from the commutation relation between the electric field and the potential. Therefore this term is purely quantum mechanical in origin. The effect of this new terms has been investigated elsewhere,[6] where it has been shown that it could lead to otherwise forbidden contributions to $\chi^{(2)}$, but is small compared to the usual term, if that term vanishes by symmetry arguments.

To complete this section, $\chi^{(2)}$ will be found using the $-\boldsymbol{\mu}\cdot\mathbf{E}$ form of the interaction. It will be assumed that the eigenvalues and eigenfunctions are those of the transformed state vectors, and not those used previously.

The same kinds of manipulations that were used before apply now. The polarization vector becomes

$$\langle\mathbf{P}^{(2)}\rangle=(eN/L^3)\sum_\sigma p_\sigma\Delta\omega_\sigma|\boldsymbol{\mu}_{if}\cdot\hat{\mathbf{e}}_\sigma|^2|\alpha_\sigma|^2\times\left[-\frac{1-\cos\omega^-_{if}t}{\omega^-_{if}\omega^+_{fi}}-\frac{1-\cos\omega^+_{if}t}{\omega^+_{if}\omega^-_{fi}}+\frac{\cos(2\xi_\sigma)+\cos(2\xi_\sigma-2\omega_\sigma t)-\cos(2\xi_\sigma+\omega^-_{if}t)}{\omega^-_{if}\omega^-_{fi}}-\frac{\cos(2\xi_\sigma+\omega^-_{fi}t)}{\omega^-_{if}\omega^-_{fi}}\right]. \tag{33}$$

Using the far from resonance condition, and retaining only the frequency-doubled part, $\chi^{(2)}$ becomes, following the same procedure as above,

$$\chi^{(2)}=-eN(\Delta/\omega_\sigma^2)|\boldsymbol{\mu}_{if}\cdot\hat{\mathbf{e}}_\sigma|^2(1-R/2)\ . \tag{34}$$

This evidently differs from (19). This is because the eigenfunctions and eigenvalues used to obtain (34) are different from those used previously, as shown in detail above. In (33), for example, consider the $\omega_{if}=E_i-E_f$. These energy eigenvalues are not those used for the $-e\,\mathbf{A}\cdot\mathbf{p}/m$ interaction. [It is not even certain if the limiting case $\omega_\sigma\gg\omega_{if}$ is valid, even though it was used to obtain (34).] Nevertheless, the point is, (34) is only valid if the eigenfunctions used are the transformed eigenfunctions, $U^\dagger|n\rangle$.

V. SUMMARY

Detailed investigations of two topics concerning the evaluation of the second-order part of the susceptibility are presented for a fully quantized time-dependent system. One of these concerns the contribution to $\chi^{(2)}$ from the second harmonic originally present in the incident beam. No actual beam is truly monochromatic, and for ultrashort pulses monochromicity is not even a good approximation. Therefore it is useful to find out to what extent the second-order part of the original wave contributes to $\chi^{(2)}$. From an empirical standpoint, this would correspond to the possibility that two different experimenters could measure two different values of $\chi^{(2)}$ if they had differently shaped pulses. It is shown that the contribution from the second harmonic scales as the ratio of the power of the second harmonic to that of the fundamental.

The other topic under consideration tackles the old question concerning the correct form of the interaction Hamiltonian to use. For each interaction, $\chi^{(2)}$ is calculated and it is seen that they are different in form. It is shown that the correct form depends upon which eigenfunctions and eigenvalues are used, and that these cannot be mixed. This is shown from two points of view. In one case the Hamiltonian and the state vectors are transformed, producing a new Hamiltonian, which must be used with the new, transformed, eigenfunctions. In the other case, the Hamiltonian was written in terms of a new, canonically equivalent set of variables. Here, what appears to be the interaction-free part of the Hamiltonian really contains an interaction term, and the total Hamiltonian is really the same as the original. Thus if $H_0^A|n\rangle = E_n|n\rangle$ as defined in Sec. II, then $-e\,\mathbf{A}\cdot\mathbf{p}/m$ is the correct form of the interaction to use.

---

[1] P. Franken, A. Hill, C. Peters, and G. Weinreich, Phys. Rev. Lett. **7**, 118 (1961).

[2] N. Bloembergen, *Nonlinear Optics* (Benjamin, New York, 1965).

[3] Y. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984), p. 16.

[4] R. Loudon, *The Quantum Theory of Light* (Clarendon, Oxford, 1985).

[5] R. Hammond, Phys. Rev. A **38**, 4895 (1988).

[6] R. Hammond, J. Opt. Soc. Am. B **5**, 2564 (1988).

[7] E. Power and S. Zienau, Philos. Trans. R. Soc. London, Ser. A **251**, 427 (1959).

[8] E. Power and T. Thirunamachandran, Phys. Rev. A **22**, 2894 (1980).

[9] J. Ackerhalt and P. Milonni, J. Opt. Soc. Am. B **1**, 116 (1984).

[10] L. Mandel, Phys. Rev. A **20**, 1590 (1979).

[11] See Ref. 9, Eq. 2.11.