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NONLINEAR IONIC POLARIZATION*

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ABSTRACT

The derivation of an explicit expression for the ionic contributions to the nonlinear electric polarization for second and third harmonic generation of light is presented. The ionic polarizations obtained are to first order in the anharmonic potential energy in the dipole moment approximation. In contrast to the usual semiclassical or quantum-mechanical perturbation approach, we have used many-time temperature-dependent Green's functions.

I. INTRODUCTION

The theory of second harmonic generation (SHG) of light involves the process of annihilation of two light quanta and the creation of one new photon with twice the energy. This effect was first observed by Franken et al.[1] in 1961 and stimulated considerable interest in nonlinear optical effects. The availability of intense light fluxes from laser sources makes possible the experimental observation of such higher-order effects in the laboratory.

Second harmonic generation of light was first discussed from a theoretical point of view by Armstrong et al.[2] and Kleinman[3] in 1962; the method used in those investigations was that of quantum-mechanical perturbation theory. A descriptive review of optical harmonics and nonlinear phenomena has been given by Franken and Ward.[4] (Also see the recent paper by Kielich.[5])

One interesting aspect of nonlinear optical problems is calculating the nonlinear electric polarization for SHG and third harmonic generation (THG) of light. In general, this polarization results from contributions due to electronic motion within atoms (or molecules) and forced vibrations of ions in an anharmonic potential.

In this paper, we use the simplified model proposed by Armstrong et al.[1] and calculate the ionic contributions (ionic polarization) to the nonlinear electric polarization by use of many-time temperature dependent Green's functions. It is found that the nonlinear ionic polarization in terms of the many-time Green's functions gives rise to expressions for both SHG and THG contributions. In Sec. II, the nature of the model used and the appropriate Green's functions are discussed. The calculation of the nonlinear ionic polarization is presented and discussed in Sec. III.

II. HAMILTONIAN FOR THE SYSTEM

Following Armstrong et al.,[1] a given molecule in the isotropic system is treated as a collection of three independent one-dimensional anharmonic oscillators. The Hamiltonian of this simplified system in the absence of radiation is

$$H = \frac{1}{2} \sum_{i=x,y,z} \left(\frac{p_i^2}{m_i} + m_i \omega_{0i}^2 r_i^2 \right) - \frac{1}{2} \sum_{i=x,y,z} \lambda_i r_i^3 - \frac{1}{4} \sum_{i=x,y,z} \eta_i r_i^4 . \tag{1}$$

The last two terms in Eq. (1) represent the anharmonic part of the potential in which the ions oscillate. The interaction of the system with radiation in the electric dipole approximation is given by

$$H' = \sum_{i=x,y,z} e_i r_i E_i e^{-iEt} , \tag{2}$$

where

$$E = \omega + i\epsilon . \tag{3}$$

The general equation of motion for many-time Green's functions for nonlinear transport coefficients in the energy representation is[6]

$$nEG_{n+1}(B; A)_E = \frac{1}{2\pi} G_n([B,A]; A)_E + G_{n+1}([B,H]; A)_E , \tag{4}$$

where

$$G_1([B,A]; A)_E = \langle [B,A] \rangle_E , \tag{5}$$

and

$$G_2(B;A)_E = \langle\langle B; A \rangle\rangle_E \ . \tag{6}$$

An ensemble average is indicated by $\langle \cdots \rangle$, and $\langle\langle \cdots \rangle\rangle$ denotes Fourier transform of the many-time Green's function.[7]

In our specific case, the contributions of the x component of second- and third-order polarizability due to the x component of the electric field will be calculated. The appropriate Green's functions are

$$G_{\substack{n+1 \\ \epsilon \to 0}}(x; x)_\omega = \begin{cases} n = 2; \ \lambda_x \neq 0; \ \eta_x = 0 & \text{(SHG)} , \\ n = 3; \ \lambda_x \neq 0; \ \eta_x \neq 0 & \text{(THG)} . \end{cases} \tag{7}$$

Equation (4) now becomes

$$n\omega G_{n+1}(x; x)_\omega = \frac{1}{2\pi} G_n([x, x]; x)_\omega + G_{n+1}([x, H_x]; x)_\omega , \tag{8}$$

where

$$H_x = \frac{P_x^2}{2m_x} + m_x \omega_{0x}^2 x^2 - \frac{\lambda_x}{3} x^3 - \frac{1}{4} \eta_x x^4 . \tag{9}$$

On substituting H_x from Eq. (9) into Eq. (8) and evaluating the required commutation relations, we obtain

$$n\omega G_{n+1}(x; x)_\omega = \frac{i}{m_x} G_{n+1}(P_x; x)_\omega ; \qquad \hbar \equiv 1 , \tag{10}$$

where

$$n\omega G_{n+1}(P_x; x)_\omega = -\frac{i}{2\pi} \delta(G_n) - i m_x \omega_{0x}^2 G_{n+1}(x; x)_\omega + i\lambda_x G_{n+1}(x^2; x)_\omega + i \eta_x G_{n+1}(x^3; x)_\omega . \tag{11}$$

A delta function of the form in Eq. (11) will be used throughout this paper; it is defined by

$$\delta(G_n) = \begin{cases} 1 \; ; & n = 1 \\ 0 \; ; & n \neq 1 \end{cases} . \tag{12}$$

The system of dependent equations in Eqs. (10) and (11) will be solved for $G_{n+1}(x;\, x)_\omega$ by evaluating the higher-order Green's functions, $G_{n+1}(x^2;\, x)_\omega$ and $G_{n+1}(x^3;\, x)_\omega$, to zeroth order in λ_x and η_x respectively.

The explicit expression for $G_{n+1}(x^2;\, x)_\omega$ to zeroth order in λ_x is (see Appendix A for details)

$$G_{n+1}(x^2;\, x)_\omega = \frac{2\delta(G_k)}{(2\pi m_x)^2[(k\omega)^2 - \omega_{0x}{}^2][(n\omega)^2 - 4\omega_{0x}{}^2]} \left\{ 1 + \frac{2}{(n\omega)(k\omega)} \right\}$$

$$+ \frac{4\delta(G_k)}{(2\pi m_x)^2(k\omega)[(n\omega)^2 - 4\omega_{0x}{}^2]} , \tag{13}$$

where

$$k = n - 1 . \tag{14}$$

For the special case of second harmonic generation, we have ($n = 2$; the lowest order nonlinear effect)

$$G_3(x^2;\, x)_\omega \approx \frac{1}{2(2\pi m_x)^2(\omega^2 - \omega_{0x}{}^2)^2} , \tag{15}$$

where terms involving $1/\omega^2$ have been neglected. The system of equations for the required Green's functions, Eqs. (10) and (11), reduces to

$$G_3(x;\, x)_\omega = \frac{1}{2}\,\frac{1}{(2\pi)^2}\,\frac{\lambda_x}{m_x^{\,3}}\,\frac{1}{(\omega^2 - \omega_{0x}^{\;2})^2(\omega_{0x}^{\;2} - 4\omega^2)} \;. \tag{16}$$

The nonlinear ionic polarization in this case is

$$P_{xx}^{\;NL}(2\omega,\ \text{ionic}) = \frac{\lambda_x}{2(2\pi)^2}\left(\frac{e_x}{m_x}\right)^3 \frac{E_x^{\;2}\; e^{-2i\omega t}}{(\omega^2 - \omega_{0x}^{\;2})^2(\omega_{0x}^{\;2} - 4\omega^2)} \;. \tag{17}$$

The expression for the ionic contribution to the electric polarization in SHG, Eq. (17), was obtained by Armstrong et al.[1] using quantum mechanical perturbation theory and by Tanaka et al.[6] using many-time temperature-dependent Green's functions.

We now extend the Green's function method and obtain the ionic contribution to the electric polarization in THG. In this case, we must retain the η_x term in Eq. (11) and calculate $G_4(x;\, x)_\omega$.

For $n = 3$, Eqs. (10) and (11) reduce to

$$3\omega\, G_4(x;\, x)_\omega = \frac{i}{m_x}\, G_4(P_x;\, x)_\omega \tag{18}$$

and

$$3\omega\, G_4(P_x;\, x)_\omega = -i\, m_x\, \omega_{0x}^{\;2}\, G_4(x;\, x)_\omega + i\, \eta_x\, G_4(x^3;\, x)_\omega \;. \tag{19}$$

The details for obtaining $G_4(x^3;\, x)_\omega$ are given in Appendix B. From Eq. (B12), we note that

$$G_4(x^3;\, x)_\omega = \gamma'\, G_3(x^2;\, x)_\omega + \Delta'(xP_x)\beta_1' - \Delta'(P_x^{\;2})\alpha_1' + i\Delta'(P_x)\, G_4(P_x;\, x)_\omega + \Delta'(x)\, G_4(x;\, x)_\omega \;. \tag{20}$$

The primes on the coefficients in Eq. (20) indicate that the coefficients are evaluated for $n = 3$. For example,

$$\gamma' = \gamma\big|_{n=3} . \tag{21}$$

The unprimed coefficients are defined in Appendix B. On substituting Eqs. (20) and (19) into Eq. (18), we obtain

$$G_4(x; x)_\omega = -\left(\frac{\eta_x \gamma' G_3(x^2; x)_\omega + \Gamma'}{\theta'(\omega)}\right) , \tag{22}$$

where

$$\Gamma' = \eta_x[\Delta'(xP_x)\beta_1' - \Delta'(P_x^2)\alpha_1'] \tag{23}$$

and

$$\theta'(\omega) = 9\omega^2 m_x + 3\omega \eta_x m_x \Delta'(P_x) - \omega_{0x}^2 m_x + \eta_x \Delta'(x) . \tag{24}$$

On multiplying the right-hand side of Eq. (22) by $e_x^4 E_x^3 e^{-3i\omega t}$, we obtain a quantum-mechanical expression for the nonlinear ionic polarization in THG.

APPENDIX A: FINDING $G_{n+1}(x^2;\ x)_\omega$

The details for calculating $G_{n+1}(x^2;\ x)_\omega$ to zeroth order in λ_x are presented in this Appendix. The equation of motion for $G_{n+1}(x^2;\ x)_\omega$ is

$$n\omega G_{n+1}(x^2;\ x)_\omega = \frac{1}{2\pi} G_n([x^2,x];\ x)_\omega + G_{n+1}([x^2,H_x];\ x)_\omega$$

$$= \frac{2i}{m_x} G_{n+1}(xP_x;\ x)_\omega . \qquad \text{(A1)}$$

a. Finding $G_{n+1}(xP_x;\ x)_\omega$

$$n\omega G_{n+1}(xP_x;\ x)_\omega = -\frac{i}{2\pi} G_n(x;\ x)_\omega + \frac{i}{m_x} G_{n+1}(P_x{}^2;\ x)_\omega$$

$$- im_x\ \omega_{0x}{}^2\ G_{n+1}(x^2;\ x)_\omega . \qquad \text{(A2)}$$

b. Finding $G_n(x;\ x)_\omega$

$$k\omega G_n(x;\ x)_\omega = \frac{i}{m_x} G_n(P_x;\ x)_\omega , \qquad \text{(A3)}$$

where $k = n - 1$.

c. Finding $G_n(P_x;\ x)_\omega$

$$k\omega G_n(P_x;\ x)_\omega = -\frac{i\ \delta(G_k)}{2\pi} - i\ m_x\ \omega_{0x}{}^2\ G_n(x;\ x)_\omega . \qquad \text{(A4)}$$

Therefore $G_n(x;\ x)_\omega$ becomes

$$G_n(x;\ x)_\omega = \frac{\delta(G_k)}{2\pi\ m_x[(k\omega)^2 - \omega_{0x}{}^2]} . \qquad \text{(A5)}$$

d. Finding $G_{n+1}(P_x^{\,2};\, x)_\omega$

$$n\omega G_{n+1}(P_x^{\,2};\, x)_\omega = \frac{1}{2\pi} G_n([P_x^{\,2}, x];\, x)_\omega + G_{n+1}([P_x^{\,2}, H_x];\, x)_\omega$$

$$= -\frac{2i}{2\pi} G_n(P_x;\, x)_\omega - 2i\, m_x\, \omega_{0x}^{\,2}\, G_{n+1}(xP_x;\, x)_\omega \,. \tag{A6}$$

Hence,

$$G_n(P_x;\, x)_\omega = \frac{-i}{k\omega}\left\{\frac{\delta(G_k)}{2\pi} + m_x\, \omega_{0x}^{\,2}\, G_n(x;\, x)_\omega\right\}, \tag{A7}$$

$$G_{n+1}(P_x^{\,2};\, x)_\omega = -\frac{2i}{n\omega}\left\{\frac{G_n(P_x;\, x)_\omega}{2\pi} + m_x\, \omega_{0x}^{\,2}\, G_{n+1}(xP_x;\, x)_\omega\right\}, \tag{A8}$$

and

$$G_{n+1}(xP_x;\, x)_\omega = \frac{-\,i(n\omega)}{[(n\omega)^2 - 2\omega_{0x}^{\,2}]}\left\{\frac{1}{2\pi}\left(1 + \frac{2}{(n\omega)(k\omega)}\right) G_n(x;\, x)_\omega \right.$$

$$\left. + \frac{2\delta(G_k)}{(2\pi)^2\, m_x (n\omega)(k\omega)} + m_x\, \omega_{0x}^{\,2}\, G_{n+1}(x^2;\, x)_\omega\right\}. \tag{A9}$$

On substituting $G_{n+1}(xP_x;\, x)_\omega$ into Eq. (A1), we obtain

$$G_{n+1}(x^2;\, x)_\omega = \frac{2\,\delta(G_k)}{(2\pi\, m_x)^2[(k\omega)^2 - \omega_{0x}^{\,2}][(n\omega)^2 - 4\omega_{0x}^{\,2}]}\left\{1 + \frac{2}{(n\omega)(k\omega)}\right\}$$

$$+ \frac{4\delta(G_k)}{(2\pi\, m_x)^2 (k\omega)(n\omega)[(n\omega)^2 - 4\omega_{0x}^{\,2}]} \,. \tag{A10}$$

For $n = 2$, we obtain

$$G_3(x^2;\, x)_\omega = \frac{1}{2(2\pi\, m_x)^2\,(\omega^2 - \omega_{0x}{}^2)}\left(1 + \frac{1}{\omega^2}\right) + \frac{1}{2(2\pi\, m_x)^2(\omega^2 - \omega_{0x}{}^2)\omega^2} \,. \qquad \text{(A11)}$$

APPENDIX B: FINDING $G_{n+1}(x^3;\, x)_\omega$

This Appendix is devoted to the presentation of the details of finding $G_{n+1}(x^3;\, x)_\omega$ to zeroth order in η_x. The equation of motion for $G_{n+1}(x^3;\, x)_\omega$ is

$$n\omega G_{n+1}(x^3;\, x)_\omega = \frac{1}{2\pi}\, G_n([x^3, x];\, x)_\omega + G_n([x^3, H_x];\, x)_\omega$$

$$= \frac{3i}{m_x}\, G_{n+1}(x^2\, P_x;\, x)_\omega + \frac{3}{m_x}\, G_{n+1}(x;\, x)_\omega\,. \qquad \text{(B1)}$$

a. Finding $G_{n+1}(x^2\, P_x;\, x)_\omega$

$$n\omega G_{n+1}(x^2\, P_x;\, x)_\omega = \frac{1}{2\pi}\, G_n([x^2\, P_x,\, x];\, x)_\omega + G_{n+1}([x^2\, P_x,\, H_x];\, x)_\omega$$

$$= \frac{-i}{2\pi}\, G_n(x^2;\, x)_\omega + \frac{2i}{m_x}\, G_{n+1}(xP_x{}^2;\, x)_\omega$$

$$+ \frac{1}{m_x}\, G_{n+1}(P_x;\, x)_\omega - i\, m_x\, \omega_{0x}{}^2\, G_{n+1}(x^3;\, x)_\omega\,. \qquad \text{(B2)}$$

On evaluating the various Green's functions involved in $G_{n+1}(xP_x{}^2;\, x)_\omega$, we obtain

$$G_{n+1}(xP_x{}^2;\, x)_\omega = \frac{n\omega}{[(n\omega)^2 - 3\omega_{0x}{}^2]} \left\{ -\frac{2i}{2\pi}\, G_n(xP_x;\, x)_\omega + \frac{3G_n(P_x{}^2;\, x)_\omega}{2\pi\, m_x(n\omega)} \right.$$

$$+ \frac{2i\, \omega_{0x}{}^2\, G_{n+1}(P_x;\, x)_\omega}{n\omega} - 2i\, m_x\, \omega_{0x}{}^2\, G_{n+1}(x^2\, P_x;\, x)_\omega$$

$$\left. - m_x\, \omega_{0x}{}^2\, G_{n+1}(x;\, x)_\omega \right\}\,. \qquad \text{(B3)}$$

When Eq. (B3) is substituted into Eq. (B2), one obtains

$$G_{n+1}(x^2 P_x; x)_\omega = -\frac{i[(n\omega)^2 - 3\omega_{0x}^2]G_n(x^2; x)_\omega}{2\pi(n\omega)[(n\omega)^2 - 7\omega_{0x}^2]} + \frac{4G_n(xP_x; x)_\omega}{2\pi m_x[(n\omega)^2 - 7\omega_{0x}^2]}$$

$$+ \frac{6 i G_n(P_x^2; x)_\omega}{2\pi(n\omega)[(n\omega)^2 - 7\omega_{0x}^2]} + \frac{[(n\omega)^2 - 3\omega_{0x}^2]}{m_x(n\omega)[(n\omega)^2 - 7\omega_{0x}^2]}$$

$$\times \left\{ 1 - \frac{4\omega_{0x}^2}{[(n\omega)^2 - 3\omega_{0x}^2]} \right\} G_{n+1}(P_x; x)_\omega$$

$$- \frac{i m_x[(n\omega)^2 - 3\omega_{0x}^2]\omega_{0x}^2 G_{n+1}(x^3; x)_\omega}{n\omega[(n\omega)^2 - 7\omega_{0x}^2]}$$

$$- \frac{2i\omega_{0x}^2 G_{n+1}(x; x)_\omega}{[(n\omega)^2 - 7\omega_{0x}^2]} . \qquad \text{(B4)}$$

On returning to Eq. (B1), one finds

$$G_{n+1}(x^3; x)_\omega = \Delta(x^2) G_{n+1}(x^2; x)_\omega + i\Delta(xP_x) G_n(xP_x; x)_\omega$$

$$- \Delta(P_x^2) G_n(P_x^2; x)_\omega + i\Delta(P_x) G_{n+1}(P_x; x)_\omega$$

$$+ \Delta(x) G_{n+1}(x, x)_\omega , \qquad \text{(B5)}$$

where

$$\Delta(x^2) = \frac{3[(n\omega)^2 - 3\omega_{0x}^2]}{2\pi m_x \phi(\omega)} , \qquad \text{(B6)}$$

$$\Delta(xP_x) = \frac{12(n\omega)}{2\pi\, m_x^{\,2}\, \phi(\omega)} , \tag{B7}$$

$$\Delta(P_x^{\,2}) = \frac{18}{2\pi\, m_x\, \phi(\omega)} , \tag{B8}$$

$$\Delta(P_x) = \frac{3[(n\omega)^2 - 3\,\omega_{0x}^{\,2}]}{m_x^{\,2}\, \phi(\omega)} \left\{ 1 - \frac{4\,\omega_{0x}^{\,2}}{[(n\omega)^2 - 3\,\omega_{0x}^{\,2}]} \right\} , \tag{B9}$$

$$\Delta(x) = \frac{3(n\omega)[(n\omega)^2 - 5\,\omega_{0x}^{\,2}]}{m_x\, \phi(\omega)} , \tag{10}$$

and

$$\phi(\omega) = [(n\omega)^4 - 10\,\omega_{0x}^{\,2}(n\omega)^2 + 9\,\omega_{0x}^{\,2}] . \tag{B11}$$

Equation (B5) may be rewritten in the form

$$G_{n+1}(x^3;\, x)_\omega = \gamma\, G_n(x^2;\, x)_\omega + \Delta(x\, P_x)\beta_1 - \Delta(P_x^{\,2})\alpha_1$$

$$+ \; i\, \Delta(P_x)\, G_{n+1}(P_x;\, x)_\omega + \Delta(x)\, G_{n+1}(x;\, x)_\omega , \tag{B12}$$

where

$$\gamma = \Delta(x^2) + \beta_2\, \Delta(xP_x) - \Delta(P_x^{\,2})\alpha_2, \tag{B13}$$

$$\beta_1 = \frac{k\omega\left(1 + \frac{2}{kk'\omega^2}\right) \delta(G_{k'})}{(2\pi)^2\, m_x[(k'\omega)^2 - \omega_{0x}^{\,2}][(k\omega)^2 - 2\,\omega_{0x}^{\,2}]}$$

$$+ \frac{2\, \delta(G_{k'})}{(2\pi)^2\, m_x[(k\omega)^2 - 2\,\omega_{0x}^{\,2}]kk'\omega} , \tag{B14}$$

$$k' = k - 1 = n - 2 , \tag{B15}$$

$$\beta_2 = \frac{k\omega\, m_x\, \omega_{0x}^{\;2}}{[(k\omega)^2 - 2\,\omega_{0x}^{\;2}]} , \tag{B16}$$

$$\alpha_1 = - \frac{2\,\delta(G_{k'})}{(2\pi)^2\, kk'\omega^2} - \frac{2\,\omega_{0x}^{\;2}\,\delta(G_{k'})}{(2\pi)^2\, kk'\omega^2[(k'\omega)^2 - \omega_{0x}^{\;2}]}$$

$$- \frac{2\, m_x\, \omega_{0x}^{\;2}\, \beta_1}{k\omega} , \tag{B17}$$

and

$$\alpha_2 = - \frac{2\, m_x\, \omega_{0x}^{\;2}\, \beta_2}{k\omega} . \tag{B18}$$

FOOTNOTES AND REFERENCES

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