$$\mathrm{P}_{xp}^{(1)}(\omega_p)+\mathrm{P}_{xp}^{NL}(\omega_p)=\varepsilon_0[n_x^2(\omega_p)-1]\mathrm{E}_{xp}(\omega_p)=\varepsilon_0[(n_0^2-1)$$
$$+\sum_{m=1}\frac{1}{4^m}\frac{(2m+1)!}{m!m!}\tilde{\tilde{\chi}}_{(2m+2)x}^{(2m+1)}(-\omega_p)\,|\,\mathrm{E}_x(\omega)\,|^{2m}]\mathrm{E}_{xp}(\omega_p); \tag{15}$$

$$\mathrm{P}_{yp}^{(1)}(\omega_p)+\mathrm{P}_{yp}^{NL}(\omega_p)\varepsilon_0[n_y^2(\omega_p)-1]\mathrm{E}_{yp}(\omega_p)=\varepsilon_0[(n_0^2-1)$$
$$+\sum_{m=1}\frac{1}{4^m}\frac{(2m+1)!}{m!m!}\tilde{\tilde{\chi}}_{yy,(2m)x}^{(2m+1)}(-\omega_p)\,|\,\mathrm{E}_x(\omega)\,|^{2m}]\mathrm{E}_{yp}(\omega_p). \tag{16}$$

For the pump beam, the ω_p on the input side is replaced by another ω and hence there are m + 1 + ω 's but still m -ω's so that

$$\mathrm{P}_x^{(1)}(\omega)+\mathrm{P}_x^{NL}(\omega)=\varepsilon_0[n_x^2(\omega)-1]\mathrm{E}_x(\omega)=\varepsilon_0[(n_0^2-1)$$
$$+\sum_{m=1}\frac{1}{4^m}\frac{(2m+1)!}{(m+1)!m!}\tilde{\tilde{\chi}}_{(2m+2)x}^{(2m+1)}(-\omega)\,|\,\mathrm{E}_x(\omega)\,|^{2m}]\mathrm{E}_x(\omega). \tag{17}$$

For $\mathrm{P}_y^{NL}(\omega)$, the first 2 *y's* belong to the probe beam and there are still m ω's and m -ω's, just like in $\mathrm{P}_{yp}^{NL}(\omega_p)$ so that

$$\mathrm{P}_y^{(1)}(\omega)+\mathrm{P}_y^{(NL)}(\omega)=\varepsilon_0[n_y^2(\omega)-1]\mathrm{E}_y(\omega)=\varepsilon_0[(n_0^2-1)$$
$$+\sum_{m=1}\frac{1}{4^m}\frac{(2m+1)!}{m!m!}\tilde{\tilde{\chi}}_{yy,(2m)x}^{(2m+1)}(-\omega)\,|\,\mathrm{E}_x(\omega)\,|^{2m}]\mathrm{E}_y(\omega). \tag{18}$$

4. Total nonlinear birefringence

It is clear from Eqs. (15)–(18), that in order to find the birefringence, the relationship between the nonlinear susceptibilities $\tilde{\tilde{\chi}}_{(2m+2)x}^{(2m+1)}(-\omega)$ and $\tilde{\tilde{\chi}}_{yy,(2m)x}^{(2m+1)}(-\omega)$ must be found. This depends on the symmetry properties of the medium. Even for isotropic media these are relatively complicated calculations and hence they are summarized in the Appendix along with some general results valid for all frequencies. Making the results specific to the non-resonant, isotropic medium case, Eq. (A17) is

$$\begin{aligned}\tilde{\tilde{\chi}}_{(2m+2)x}^{(2m+1)}(-\omega_p)&=(2^m+1)\tilde{\tilde{\chi}}_{yy,(2m)x}^{(2m+1)}(-\omega_p);\\ \tilde{\tilde{\chi}}_{(2m+2)x}^{(2m+1)}(-\omega)&=(2^m+1)\tilde{\tilde{\chi}}_{yy,(2m)x}^{(2m+1)}(-\omega).\end{aligned} \tag{19}$$

For the pump-probe geometry in the non-resonant limit, Eq. (19) is inserted into Eqs. (16) and (18) to give

$$\mathrm{P}_{yp}^{(1)}(\omega_p)+\mathrm{P}_{yp}^{NL}(\omega_p)\ =\varepsilon_0[n_y^2(\omega_p)-1]\mathrm{E}_{yp}(\omega_p)=\varepsilon_0[(n_0^2-1)$$
$$+\sum_{m=1}\frac{1}{4^m}\frac{(2m+1)!}{(2^m+1)m!m!}\tilde{\tilde{\chi}}_{(2m+2)x}^{(2m+1)}(-\omega_p)\,|\,\mathrm{E}_x(\omega)\,|^{2m}]\mathrm{E}_{yp}(\omega_p), \tag{20}$$

$$\mathrm{P}_y^{(1)}(\omega)+\mathrm{P}_y^{(NL)}(\omega)=\varepsilon_0[n_y^2(\omega)-1]\mathrm{E}_y(\omega)=\varepsilon_0[(n_0^2-1)$$
$$+\sum_{m=1}\frac{1}{4^m}\frac{(2m+1)!}{(2^m+1)m!m!}\tilde{\tilde{\chi}}_{(2m+2)x}^{(2m+1)}(-\omega)\,|\,\mathrm{E}_x(\omega)\,|^{2m}]\mathrm{E}_y(\omega), \tag{21}$$

#141116 - $15.00 USD Received 14 Jan 2011; revised 28 Feb 2011; accepted 3 Mar 2011; published 21 Mar 2011

so that both the x- and y-components of the nonlinear polarization are given in terms of the same susceptibilities. Noting that $n_{2m}(-\omega_p;\omega)=(m+1)n_{2m}(-\omega;\omega)$ from Eq. (13) and combining Eqs. (15), (17), (20), and (21) now leads directly to

$$n_x^2(\omega_p)=n_0^2\{1+[\sum_{m=1}\bar{A}_m I^m]\};\quad n_y^2(\omega_p)=n_0^2\{1+[\sum_{m=1}\frac{1}{(2^m+1)}\bar{A}_m I^m]\},$$
$$n_x^2(\omega)=n_0^2\{1+[\sum_{m=1}\frac{1}{m+1}\bar{A}_m I^m]\};\quad n_y^2(\omega)=n_0^2\{1+[\sum_{m=1}\frac{1}{(2^m+1)}\bar{A}_m I^m]\}, \qquad (22)$$

in which the coefficient $\bar{A}_m$ is given by

$$\bar{A}_m=2\frac{\bar{n}_{2m}(-\omega_p;\omega)}{n_0},$$
$$\bar{A}_m=\frac{1}{2n_0}\frac{1}{2^m n_0^m c^m \varepsilon_0^m}\frac{(2m+1)!}{m!m!1!}\tilde{\chi}^{(2m+1)}_{(2m+2)x}(-\omega_p)=\frac{1}{2n_0}\frac{(m+1)}{2^m n_0^m c^m \varepsilon_0^m}\frac{(2m+1)!}{m!m!1!}\tilde{\chi}^{(2m+1)}_{(2m+2)x}(-\omega). \qquad (23)$$

This form was chosen so that for the individual nonlinearities m

$$\Delta\bar{n}_x^{(m)}(\omega_p)=\bar{n}_{2m}(-\omega_p;\omega)I^m. \qquad (24)$$

In order to make contact with the experimental data in reference 1 we focus on the nonlinear refractive indices for the pump-probe case so that the nonlinear birefringence is given by

$$\bar{n}_x(\omega_p)=n_0\sqrt{1+[\sum_{m=1}\bar{A}_m I^m]};\ \bar{n}_y(\omega_p)=n_0\sqrt{1+[\sum_{m=1}\frac{1}{(2^m+1)}\bar{A}_m I^m]}; \qquad (25)$$
$$\Delta n_{bir}^{NL}(\omega_p)=n_x(\omega_p)-n_y(\omega_p).$$

The expansion of $\sqrt{1+b}$ for small b is well known from textbooks [13], to be:

$$\sqrt{1+b}=\sum_{s=0}^{\infty}\frac{(-1)^s(2s)!}{(1-2s)(s!)^2 4^s}b^s=1+\frac{1}{2}b-\frac{1}{8}b^2+\frac{1}{16}b^3-\frac{5}{128}b^4+\frac{7}{256}b^5-.. \qquad (26)$$

Therefore

$$\Delta\bar{n}_{bir}^{NL}(\omega_p)=n_0\sum_{s=0}^{\infty}\frac{(-1)^s(2s)!}{(1-2s)(s!)^2 4^s}([\sum_{m=1}\bar{A}_m I^m]^s-[\sum_{m=1}\frac{\bar{A}_m}{2^m+1}I^m]^s). \qquad (27)$$

The leading term (s = 1), expanded up to n_{10} (largest term reported in reference 1), is

$$\begin{aligned}\Delta\bar{n}_{bir}^{s=1}(\omega_p)&=n_0\{\frac{1}{3}\bar{A}_1 I+\frac{2}{5}\bar{A}_2 I^2+\frac{4}{9}\bar{A}_3 I^3+\frac{8}{17}\bar{A}_4 I^4+\frac{16}{33}\bar{A}_4 I^4+\frac{32}{65}\bar{A}_5 I^5\\&=\frac{2}{3}\bar{n}_2(-\omega_p;\omega)I+\frac{4}{5}\bar{n}_4(-\omega_p;\omega)I^2+\frac{8}{9}\bar{n}_6(-\omega_p;\omega)I^3\\&+\frac{16}{17}\bar{n}_8(-\omega_p;\omega)I^4+\frac{32}{33}\bar{n}_{10}(-\omega_p;\omega)I^5\end{aligned} \qquad (28)$$

Terms with $s\geq 2$ contain products of the nonlinear coefficients. Including all of the terms up to I^5,

#141116 - $15.00 USD Received 14 Jan 2011; revised 28 Feb 2011; accepted 3 Mar 2011; published 21 Mar 2011

$$\Delta\bar{n}_{bir}^{NL} = \frac{2}{3}\bar{n}_2(-\omega_p;\omega)I + [\frac{4}{5}\bar{n}_4(-\omega_p;\omega) - \frac{4}{9n_0}\bar{n}_2^2(-\omega_p;\omega)]I^2$$
$$+[\frac{8}{9}\bar{n}_6(-\omega_p;\omega) - \frac{14}{15n_0}\bar{n}_2(-\omega_p;\omega)\bar{n}_4(-\omega_p;\omega) + \frac{13}{27n_0^2}\bar{n}_2^3(-\omega_p;\omega)]I^3$$
$$+[\frac{16}{17}\bar{n}_8(-\omega_p;\omega) - \frac{26}{27n_0}\bar{n}_2(-\omega_p;\omega)\bar{n}_6(-\omega_p;\omega) - \frac{12}{25n_0}\bar{n}_4^2(-\omega_p;\omega)$$
$$+\frac{22}{15n_0^2}\bar{n}_2^2(-\omega_p;\omega)\bar{n}_4(-\omega_p;\omega) - \frac{50}{81n_0^3}\bar{n}_2^4(-\omega_p;\omega)]I^4 \tag{29}$$
$$+[\frac{32}{33}\bar{n}_{10}(-\omega_p;\omega) - \frac{50}{51n_0}\bar{n}_2(-\omega_p;\omega)\bar{n}_8(-\omega_p;\omega) - \frac{44}{45n_0}\bar{n}_4(-\omega_p;\omega)\bar{n}_6(-\omega_p;\omega)$$
$$+\frac{40}{27n_0^2}\bar{n}_2^2(-\omega_p;\omega)\bar{n}_6(-\omega_p;\omega) + \frac{37}{25n_0^2}\bar{n}_2(-\omega_p;\omega)\bar{n}_4^2(-\omega_p;\omega)$$
$$-\frac{67}{27n_0^3}\bar{n}_2^3(-\omega_p;\omega)\bar{n}_4(-\omega_p;\omega) + \frac{847}{972n_0^4}\bar{n}_2^5(-\omega_p;\omega)]I^5.$$

Note that all the numerical pre-factors in this case are all less than 2.5. The products of different nonlinear coefficients are limited to 2 here. However, products of more than two nonlinear coefficients occur for higher-orders in intensity, the first one being $\bar{n}_2\bar{n}_4\bar{n}_6 I^6$. From Eq. (29) it is evident that in a strict mathematical sense the nonlinear birefringence cannot be used as a means to measure the nonlinear coefficients higher than $\bar{n}_2$ .There is no direct correlation between the coefficient $\bar{n}_{2m}$ and the corresponding power of the intensity I^m for $m>1$ due to the existence of the product terms. However, it makes sense to use the simplified notation of Eq. (29) if the relation $\bar{n}_{2m} >> \bar{n}_{2k_1}\bar{n}_{2k_2}...\bar{n}_{2k_{2n}}, m = k_1 + k_2 + ...k_n$ holds.

5. Comparison with experiments on air

Reference 1 contains data measured in air and its constituents for $n_2(-\omega_p;\omega) \rightarrow n_8(-\omega_p;\omega)$ and also $n_{10}(-\omega_p;\omega)$ for argon. Based on their values, $\bar{n}_{2m}(-\omega_p;\omega) >> \bar{n}_{2q}^r(-\omega_p;\omega)\bar{n}_{2u}^v(-\omega_p;\omega)$ with $m = rq + vu$ and $m \le 5$ is always satisfied in air. Assuming that the only nonlinear mechanism present is the Kerr effect, the nonlinear birefringence is given by the leading term, Eq. (28), which can be expressed as the series

$$\Delta\bar{n}_{bir}^{NL}(\omega_p) = \sum_{m=1} \frac{2^m}{2^m+1}\bar{n}_{2m}(-\omega_p;\omega)I^m \ . \tag{30}$$

This result should be compared with the expansion used by Loriot et al. [1]. Based on a linear extrapolation from the first two terms which Loriot *et al.* obtained from the literature [3,8] they assumed the series

$$\Delta\bar{n}_{bir}^{NL}(\omega_p) = \sum_{m=1}\frac{2m}{2m+1}\bar{n}_{2m}(-\omega_p;\omega)I^m \tag{31}$$

in their analysis of their data. Note that in both series the numerical pre-factors $2^m/(2^m + 1)$ and $2m/(2m + 1)$ respectively converge to unity for large m. A graphical comparison of the two expansions is given in Fig. 2. In Fig. 2(a) we compare the expansion terms as deduced from Eq. (30), $2^m/(2^m+1)$, to the ones derived by Loriot et al. $2m/(2m+1)$. As m is increased their difference is maximized for m = 11. The relative deviation of Loriot *et al.*'s

#141116 - $15.00 USD Received 14 Jan 2011; revised 28 Feb 2011; accepted 3 Mar 2011; published 21 Mar 2011

expansion terms as compared to the analytically derived factors is depicted in Fig. 2(b). For m = 11 the relative error peaks at 6.25%. Furthermore, the Loriot *et al.* formulation systematically underestimates the expansion term coefficients and thus leads to an overestimation of the corresponding $\bar{n}_{2m}(-\omega_p;\omega)$ coefficient for m>2.

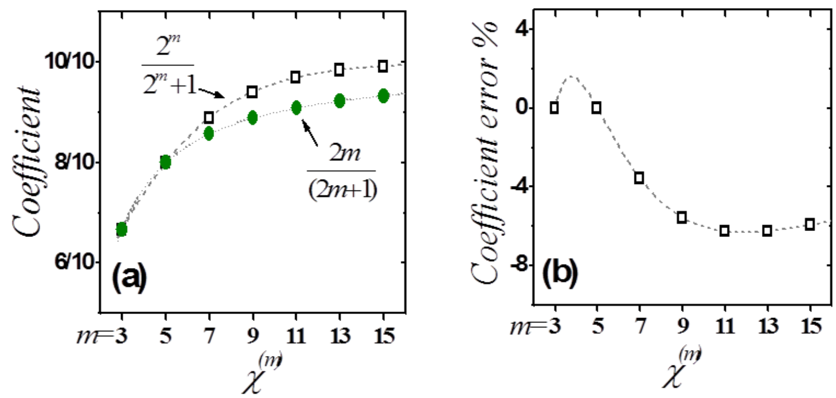

Fig. 2. Comparison between the expansion coefficients estimated by the two models. (a) Coefficients corresponding to $\chi^{(m)}$ terms. (□) analytical model, (•) Loriot et al. estimation, dotted/dashed lines are a guide to the eye. (b) Relative error for the various coefficients of the $\chi^{(m)}$ terms. (Dotted lines are guides to the eye).

6. Conclusions

Expressions for the non-resonant, nonlinear birefringence induced in a probe beam (frequencyω_p) by a strong pump beam of the same frequency in an isotropic medium have been derived for nonlinear Kerr indices $n_{2m}(-\omega_p;\omega)$ for arbitrary m. This was made possible by using combinatorial approaches and by assuming that in isotropic media there is only one unique value for $\chi^{(2m+1)}(-\omega_p)$ for each value of m which was verified previously in the literature for m = 1, 2. Some general relations for arbitrary frequency inputs were also derived.

Because the polarization, linear and nonlinear, induced in a material depends on the square of the refractive index, the nonlinear birefringence was found to depend not only on the intensity-dependent refractive index coefficients $n_{2m}(-\omega_p;\omega)$ but also on the products of the various nonlinear index coefficients. Comparison with existing experiments in air and its constituents showed that the product terms were negligible in that case.

An analytical series was found to describe the nonlinear birefringence. This series was different from that assumed by Loriot et. al based on a linear extrapolation of two points. Since in both cases the individual numerical factors for $n_{2m}(-\omega_p;\omega)$ converged to unity for increasing m, the errors introduced into the analysis of the data were relatively small.

Appendix A. Relationships between the nonlinear susceptibilities

In this Appendix the relations between the $\tilde{\tilde{\chi}}^{(2m+1)}_{(2m+2)x}(-\omega_p)$ and $\tilde{\tilde{\chi}}^{(2m+1)}_{yy,(2m)x}(-\omega)$ are derived, some for arbitrary frequency inputs. Isotropy requires that each coordinate (x and y) comes in pairs. It also requires that the nonlinear polarization should be independent of the orientation of any axis system used. Consider first the *general case* (unrelated to the previous discussion) of three, parallel, co-polarized (along the x-axis) input fields E_1, E_2 and E_3 with different frequencies ω_1, ω_2 and ω_3 producing the field ω_4 via $\chi^{(3)}_{xxxx}(-\omega_4;\omega_1,\omega_2,\omega_3)$. The third order nonlinear polarization (along the x-axis) is

#141116 - $15.00 USD Received 14 Jan 2011; revised 28 Feb 2011; accepted 3 Mar 2011; published 21 Mar 2011

$$\mathrm{P}_x^{(3)}(\omega_4) = \frac{1}{4}\varepsilon_0 \chi_{xxxx}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1)\mathrm{E}_1\mathrm{E}_2\mathrm{E}_3. \tag{A1}$$

Now consider the axis system (*x*', *y*') rotated 45° from the original *x*-axis [8]. The three fields have the following components along the *x*'-axis and *y*'-axis

$$\mathrm{E}_{1x'} = \frac{1}{\sqrt{2}}\mathrm{E}_1;\ \mathrm{E}_{2x'} = \frac{1}{\sqrt{2}}\mathrm{E}_2;\ \mathrm{E}_{3x'} = \frac{1}{\sqrt{2}}\mathrm{E}_3;\ \mathrm{E}_{1y'} = \frac{1}{\sqrt{2}}\mathrm{E}_1;\ \mathrm{E}_{2y'} = \frac{1}{\sqrt{2}}\mathrm{E}_2;\ \mathrm{E}_{3y'} = \frac{1}{\sqrt{2}}\mathrm{E}_3. \tag{A2}$$

For isotropic media, $\chi_{x'x'x'x'}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1) = \chi_{xxxx}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1)$, $\chi_{xxyy}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1) = \chi_{x'x'y'y'}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1),\ = \chi_{yyxx}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1)$ etc., and hence the nonlinear polarization induced along the *x′*-axis is given by

$$\begin{aligned}
\mathrm{P}_{x'}^{(3)}(\omega_4) &= \frac{1}{4}\varepsilon_0[\chi_{xxxx}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1)\mathrm{E}_{1x'}\mathrm{E}_{2x'}\mathrm{E}_{3x'} + \chi_{xxyy}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1)\mathrm{E}_{1y'}\mathrm{E}_{2y'}\mathrm{E}_{3x'} \\
&\quad + \chi_{xyyx}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1)\mathrm{E}_{1x'}\mathrm{E}_{2y'}\mathrm{E}_{3y'} + \chi_{xyxy}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1)\mathrm{E}_{1y'}\mathrm{E}_{2x'}\mathrm{E}_{3y'}] \\
\rightarrow \mathrm{P}_{x'}^{(3)}(\omega_4) &= \frac{1}{4}\varepsilon_0\frac{1}{2\sqrt{2}}[\chi_{xxxx}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1) + \chi_{xxyy}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1) \\
&\quad + \chi_{xyyx}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1) + \chi_{xyxy}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1)]\mathrm{E}_1\mathrm{E}_2\mathrm{E}_3.
\end{aligned} \tag{A3}$$

The nonlinear polarization $\mathrm{P}_{x'}^{(3)}(\omega_4)$ in Eq. (A3) can also be obtained by projecting the nonlinear polarization given by Eq. (A1) onto the *x′*-axis to give

$$\mathrm{P}_{x'}^{(3)}(\omega_4) = \frac{1}{4}\varepsilon_0\frac{1}{\sqrt{2}}\chi_{xxxx}^{(3)}(-\omega_4,\omega_3,\omega_2,\omega_1)\mathrm{E}_1\mathrm{E}_2\mathrm{E}_3. \tag{A4}$$

Since Eqs. (A3) and (A4) *must* give the same result which is valid for any frequencies,

$$\begin{aligned}
\chi_{xxxx}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1) &= \chi_{xxyy}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1) + \chi_{xyyx}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1) \\
&\quad + \chi_{xyxy}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1).
\end{aligned} \tag{A5}$$

Note that *any* isotropic material, for example a mature electron plasma, which exhibits third order effects such as third harmonic generation [14,15] *must* have all of these coefficients non-zero and related as given by Eq. (A5). In the non-resonant limit it can easily be shown that

$$\tilde{\chi}_{xxyy}^{(3)}(-\omega_p) = \tilde{\chi}_{xyyx}^{(3)}(-\omega_p) = \tilde{\chi}_{xyxy}^{(3)}(-\omega_p) \rightarrow \tilde{\chi}_{xxxx}^{(3)}(-\omega_p) = 3\tilde{\chi}_{xxyy}^{(3)}(-\omega_p). \tag{A6}$$

The same result holds for pump beam, i.e. $\tilde{\chi}_{xxxx}^{(3)}(-\omega) = 3\tilde{\chi}_{xxyy}^{(3)}(-\omega)$. Although this result is valid for a single medium, extension to multi-component air is trivial giving

$$\tilde{\tilde{\chi}}_{xxxx}^{(3)}(-\omega_p) = 3\tilde{\tilde{\chi}}_{xxyy}^{(3)}(-\omega_p) \qquad \tilde{\tilde{\chi}}_{xxxx}^{(3)}(-\omega) = 3\tilde{\tilde{\chi}}_{xxyy}^{(3)}(-\omega). \tag{A7}$$

An alternate and more compact approach for arriving at the same result is to again resort to combinatorial mathematics. Since there are three input polarization components, two *y*'-polarized and one *x*'-polarized, which can be permuted among the three input eigenmodes (frequencies), there are 3! possibilities for permuting the corresponding polarization components in $\tilde{\tilde{\chi}}_{xxyy}^{(3)}(-\omega_4;\omega_3,\omega_2,\omega_1)$. Because there must be two identical polarization components (y') and only one x', there are 3!/2!1!unique possibilities and Eq. (A5) can be re-written in the non-resonant limit as

$$\tilde{\tilde{\chi}}^{(3)}_{xxxx}(-\omega_p) = \frac{3!}{2!1!}\tilde{\tilde{\chi}}^{(3)}_{xxyy}(-\omega_p); \quad \tilde{\tilde{\chi}}^{(3)}_{xxxx}(-\omega) = \frac{3!}{2!1!}\tilde{\tilde{\chi}}^{(3)}_{xxyy}(-\omega) \qquad \text{(A8)}$$

The evaluation of the relation between $\tilde{\tilde{\chi}}^{(5)}_{xxxxxx}(-\omega_p)$ and $\tilde{\tilde{\chi}}^{(5)}_{yyxxxx}(-\omega_p)$ (and subsequently the yet higher-order susceptibilities) has additional aspects (relative to the $\chi^{(3)}$ case) associated with the $\tilde{\tilde{\chi}}^{(5)}_{yyxxxx}(-\omega_p) = \tilde{\tilde{\chi}}^{(5)}_{xxyyxx}(-\omega_p) = \tilde{\tilde{\chi}}^{(5)}_{xxyyyy}(-\omega_p)$ etc. terms. Again assuming the general case of five, parallel, co-polarized (along the x-axis) input fields namely E_1, E_2, E_3, E_4, and E_5 with different frequencies ω_1, ω_2, ω_3, ω_4 and ω_5 producing the field ω_6 via $\chi^{(5)}_{xxxxxx}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1)$. This produces the nonlinear polarization (along the x-axis)

$$P^{(5)}_x(\omega_4) = \frac{1}{16}\varepsilon_0\chi^{(5)}_{xxxxxx}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1)E_1E_2E_3E_4E_5. \qquad \text{(A9)}$$

Now consider again the axis system (x', y') rotated 45° from the original x-axis. The five input x-polarized fields again have components along the x'-axis and y'-axis. Note that both mixed polarization terms like $\chi^{(5)}_{xxyyxx}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1)$ as well as $\chi^{(5)}_{xxyyyy}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1)$ contribute to the nonlinear polarization induced along the x'-axis, $P^{NL}_{x'}(\omega_6)$. For the first one, there are 5! input slots for the polarization of which 3 are identical (x') and the two others are also identical (y') and, for the second one, there are 4 (y') identical slots and only the x' is a single slot. Hence the number of unique combinations are 5!/3!2! and 5!/4!1! respectively for $\tilde{\chi}^{(5)}_{xxyyxx}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1)$ and for $\tilde{\chi}^{(5)}_{xxyyyy}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1)$. There are further simplifications in the *non-resonant limit* $\tilde{\tilde{\chi}}^{(5)}_{xxyyxx}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1) = \tilde{\tilde{\chi}}^{(5)}_{xxyxyx}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1)$ etc. so that

$$P^{(5)}_{x'}(\omega_6) = \frac{1}{16}\varepsilon_0\frac{1}{4\sqrt{2}}[\tilde{\tilde{\chi}}^{(5)}_{xxxxxx}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1) + \frac{5!}{3!2!}\tilde{\tilde{\chi}}^{(5)}_{xxyyxx}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1) + \frac{5!}{4!1!}\tilde{\tilde{\chi}}^{(5)}_{xxyyyy}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1)]E_1E_2E_3E_4E_5. \qquad \text{(A10)}$$

The nonlinear polarization $P^{(5)}_{x'}(\omega_6)$ in Eq. A10 can also be obtained by projecting the nonlinear polarization given by Eq. (A9) onto the x'-axis to give

$$P^{NL}_{x'}(\omega_6) = \frac{1}{16}\varepsilon_0\frac{1}{\sqrt{2}}\tilde{\chi}^{(5)}_{xxxxxx}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1)E_1E_2E_3E_4E_5. \qquad \text{(A11)}$$

Again Eqs. (A10) and (A11) must yield identical results and noting again from references 2 and 8 that $\tilde{\tilde{\chi}}^{(5)}_{xxyyxx}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1) = \tilde{\tilde{\chi}}^{(5)}_{xxyyyy}(-\omega_6;\omega_5,\omega_4,\omega_3,\omega_2,\omega_1)$ etc. yields for the cases of interest here in the non-resonant limit

$$\tilde{\tilde{\chi}}^{(5)}_{xxxxxx}(-\omega_p) = 5\tilde{\tilde{\chi}}^{(5)}_{yyxxxx}(-\omega_p); \quad \tilde{\tilde{\chi}}^{(5)}_{xxxxxx}(-\omega) = 5\tilde{\tilde{\chi}}^{(5)}_{yyxxxx}(-\omega). \qquad \text{(A12)}$$

Consider briefly the 7'th and 9'th order susceptibilities. The same procedures as for the 3′rd and 5′th order cases are used. In order to derive the relationship between the different $\tilde{\tilde{\chi}}^{(7)}_{xxxxxxxx}(-\omega_p), \tilde{\tilde{\chi}}^{(7)}_{yyxxxxxx}(-\omega_p)$, etc. seven co-polarized input fields are considered, first in the x,

y, z coordinate system and then in an axis system rotated 45° in the x-y plane. In this case, the mixed polarization terms

$$\chi^{(7)}_{xxyyxxxx}(-\omega_8;\omega_7,\omega_6,\omega_5,\omega_4,\omega_3,\omega_2,\omega_1),\chi^{(7)}_{xxyyyyxx}(-\omega_8;\omega_7,\omega_6,\omega_5,\omega_4,\omega_3,\omega_2,\omega_1)$$

$\chi^{(7)}_{xxyyyyyy}(-\omega_8;\omega_7,\omega_6,\omega_5,\omega_4,\omega_3,\omega_2,\omega_1)$ all contribute to the nonlinear polarizations induced along the x'-axis, $P^{(7)}_{x'}(\omega_8)$. The number of unique combinations are 7!/5!2!, 7!/4!3!and 7!/6!1!respectively for the three cases. Thus again in the non-resonant limit

$$\begin{aligned} P^{NL}_{x'}(\omega_8) &= \frac{1}{64}\varepsilon_0\frac{1}{8\sqrt{2}}[\tilde{\tilde{\chi}}^{(7)}_{xxxxxx}(-\omega_8)+\frac{7!}{5!2!}\tilde{\tilde{\chi}}^{(7)}_{xxyyxxxx}(-\omega_8) \\ &\quad +\frac{7!}{4!3!}\tilde{\tilde{\chi}}^{(7)}_{xxyyyyxx}(-\omega_8)+\frac{7!}{6!1!}\tilde{\tilde{\chi}}^{(7)}_{xxyyyyyy}(-\omega_8)]E_1E_2E_3E_4E_5E_6E_7 \\ &= \frac{1}{64}\varepsilon_0\frac{1}{\sqrt{2}}\tilde{\tilde{\chi}}^{(7)}_{xxxxxx}(-\omega_8)E_1E_2E_3E_4E_5E_6E_7 \end{aligned} \qquad \text{(A13)}$$

Based on the preceding results, only one, unique, nonlinear susceptibility is expected for an isotropic material in the non-resonant limit for each order "$2m+1$" of $\chi^{(2m+1)}$. Therefore all the mixed polarization susceptibilities are equal which gives

$$\tilde{\tilde{\chi}}^{(7)}_{xxxxxxxx}(-\omega_p)=9\tilde{\tilde{\chi}}^{(7)}_{yyxxxxxx}(-\omega_p);\quad \tilde{\tilde{\chi}}^{(7)}_{xxxxxxxx}(-\omega)=9\tilde{\tilde{\chi}}^{(7)}_{yyxxxxxx}(-\omega). \qquad \text{(A14)}$$

Again using the same approach, for the 9'th order susceptibility,

$$\begin{aligned} P^{(9)}_{x}(\omega_{10}) &= \frac{1}{256}\varepsilon_0\frac{1}{16\sqrt{2}}[\tilde{\tilde{\chi}}^{(9)}_{xxxxxxxx}(-\omega_{10})+\frac{9!}{7!2!}\tilde{\tilde{\chi}}^{(9)}_{xxyyxxxxxx}(-\omega_{10})+\frac{9!}{5!4!}\tilde{\tilde{\chi}}^{(9)}_{xxyyyyxxxx}(-\omega_{10}) \\ &\quad +\frac{9!}{3!6!}\tilde{\tilde{\chi}}^{(9)}_{xxyyyyyyxx}(-\omega_{10})+\frac{9!}{1!8!}\tilde{\tilde{\chi}}^{(9)}_{xxyyyyyyyy}(-\omega_{10})]E_1E_2E_3E_4E_5E_6E_7E_8E_9 \\ &= \frac{1}{256}\varepsilon_0\frac{1}{\sqrt{2}}\tilde{\tilde{\chi}}^{(9)}_{xxxxxxxx}(-\omega_{10})E_1E_2E_3E_4E_5E_6E_7E_8E_9. \end{aligned} \qquad \text{(A15)}$$

In the non-resonant limit

$$\tilde{\tilde{\chi}}^{(9)}_{xxxxxxxxxx}(-\omega_{10})=17\tilde{\tilde{\chi}}^{(9)}_{yyxxxxxxxx}(-\omega_{10}) \qquad \text{(A16)}$$

These results suggest simple relations governing the relationship between the susceptibilities, namely

$$\begin{aligned} \tilde{\tilde{\chi}}^{(2m+1)}_{(2m+2)x}(-\omega_p) &= (2^m+1)\tilde{\tilde{\chi}}^{(2m+1)}_{yy,(2m)x}(-\omega_p); \\ \tilde{\tilde{\chi}}^{(2m+1)}_{(2m+2)x}(-\omega) &= (2^m+1)\tilde{\tilde{\chi}}^{(2m+1)}_{yy,(2m)x}(-\omega). \end{aligned} \qquad \text{(A17)}$$

For frequency inputs ω_1, ω_2, ω_3,.. ω_{2m+1} giving an output frequency ω_{2m+2} for isotropic media, the above formulas suggest the following general result:

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$$\tilde{\tilde{\chi}}^{(2m+1)}_{(2m+2)x}(-\omega_{2m+2}) = \frac{1}{2^m}[\tilde{\tilde{\chi}}^{(2m+1)}_{(2m+2)x}(-\omega_{2m+2}) + \frac{(2m+1)!}{(2m)!1!}\tilde{\tilde{\chi}}^{(2m+1)}_{(2)y,(2m)x}(-\omega_{2m+2})$$
$$+\frac{(2m+1)!}{(2m-2)!3!}\tilde{\tilde{\chi}}^{(2m+1)}_{(4)y,(2m-2)x,}(-\omega_{2m+2}) + \frac{(2m+1)!}{(2m-4)!5!}\tilde{\tilde{\chi}}^{(2m+1)}_{(6)y,(2m-4)x}(-\omega_{2m+2})...$$
$$+\frac{(2m+1)!}{2!(2m-1)!}\tilde{\tilde{\chi}}^{(2m+1)}_{(2m-1)y,(2)x}(-\omega_{2m+2})]. \tag{A18}$$

which gives

$$\tilde{\tilde{\chi}}^{(2m+1)}_{(2m+2)x}(-\omega_{2m+2}) = \frac{1}{2^m-1}[\frac{(2m+1)!}{(2m)!1!}\tilde{\tilde{\chi}}^{(2m+1)}_{(2)y,(2m)x}(-\omega_{2m+2})$$
$$+\frac{(2m+1)!}{(2m-2)!3!}\tilde{\tilde{\chi}}^{(2m+1)}_{(4)y,(2m-2)x,}(-\omega_{2m+2}) + \frac{(2m+1)!}{(2m-4)!5!}\tilde{\tilde{\chi}}^{(2m+1)}_{(6)y,(2m-4)x}(-\omega_{2m+2})...$$
$$+\frac{(2m+1)!}{2!(2m-1)!}\tilde{\tilde{\chi}}^{(2m+1)}_{(2m-1)y,(2)x}(-\omega_{2m+2})]. \tag{A19}$$

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