A general theory of inhomogeneous broadening for nonlinear susceptibilities: the second hyperpolarizability

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Abstract—A general theory of inhomogeneous broadening is rarely applied to nonlinear spectroscopy in lieu of either a simple Lorentzian or Gaussian model. In this work, we generalize all the important third-order nonlinear susceptibility expressions obtained with sum-over state quantum calculations to include Gaussian and stretched Gaussian distributions of Lorentzians. This theory gives a better fit to subtle spectral features — such as the shoulder of the electroabsorption peak, and is a more accurate tool for determining transition moments from spectroscopy experiments.

I. INTRODUCTION

SUM-over states quantum perturbation treatments of the $b$th-order nonlinear susceptibility tensor, $\xi_{ij...k}^{(b)}$, in the dipole approximation yields a sum of terms of the form:

$$\xi_{ij...k}^{(b)} \propto \sum_n \sum_m \sum_l \sum_{i\sigma} \sum_{j\sigma} ... \sum_{k\sigma} \frac{(\mu_i)_{nm} (\mu_j)_{nm} ... (\mu_k)_{nm}}{-(\omega_{ng} - i\Gamma_{ng} - \omega_1)(\omega_{ng} - i\Gamma_{ng} - \omega_1 - \omega_2)...},$$

where $(\mu_i)_{nm}$ is the nm-matrix element of the $i$th component of the electric dipole operator, $\omega_{nm}$ the transition frequency (energy) between states $n$ and $m$, $\omega_1$ the frequency of the $i$th optical field, and $\Gamma_{ng}$ the phenomenological damping factor. The numerator is a product of $b + 1$ transition moments and the denominator a product of $b$ energy terms. For an isolated molecule, the damping factor $\Gamma_{ng}$ is inversely proportional to the lifetime of state $n$ and is a measure of the width of the peak in the spectrum of $\xi_{ij...k}^{(b)}$ associated with a transition between state $n$ and the ground state $g$.

In this work, we take into account the interaction of molecules with their surroundings using a stochastic model as we have reported for the first- and second-order susceptibility. The technique is similar to Stoneham’s approach for the linear susceptibility and Toussaere’s calculation of the hyperpolarizability, who both used Gaussian statistics. In our work, we generalize the statistics to stretched exponentials, which are known to better model the interaction between a molecule and a system that is characterized by a distribution of sites such as a host polymer.

In our treatment of inhomogeneous broadening, each molecule in an ensemble is then viewed as having a different transition frequency (energy), $\omega_{ng}$. For the stretched Gaussian, the probability distribution is of the form

$$f_{ng}(\delta \omega_{ng}) = \frac{1}{N(\gamma_{ng}, \beta)} \exp \left[- \left(\frac{\delta \omega_{ng}}{\gamma_{ng}}\right)^{2\beta}\right],$$

where $\delta \omega_{ng} = \omega_{ng} - \bar{\omega}_{ng}$, $\bar{\omega}_{ng}$ is the mean value of the transition frequency, $N(\gamma_{ng}, \beta)$ the normalization factor, $\gamma_{ng}$ the linewidth of the distribution and $\beta$ is the distribution of sites parameter. For most systems, $\beta = 0$ for an infinitely broad distribution and $\beta = 1$ for a single characteristic width. The susceptibility will then be of the form,

$$\sum_{\delta \omega_{mg}} d(\delta \omega_{mg}) \int_{-\delta \omega_{ng}}^{\delta \omega_{mg}} f_{mg}(\delta \omega_{mg}) f_{ng}(\omega_{ng}) d\omega_{ng} ...$$

Note that $N(\gamma, \beta)$ depends on $\beta$, and will be written as

$$N(\gamma_{ng}, \beta) = \gamma_{ng} \sqrt{\pi} B(\beta),$$

where,

$$B(\beta) = \left[ \frac{1}{\gamma_{ng} \sqrt{\pi}} \int_{-\infty}^{\infty} \exp \left[- \left(\frac{\delta \omega_{ng}}{\gamma_{ng}}\right)^{2\beta}\right] d(\delta \omega_{ng}) \right]$$

II. THIRD-ORDER ENERGY DENOMINATORS

Similar to first- and second-order processes, the SOS Lorentzian energy denominators for third-order processes are

$$D_{lm}^{(3)}(\omega_{1g}; \omega_1, \omega_2, \omega_3) = S_{1,2,3} \times$$

$$\left\{ \left[ (\Omega_{lg} - \omega_\sigma)(\Omega_{lg} - \omega_3)(\Omega_{ng} - \omega_1) \right]^{-1} + \left[ (\Omega_{lg} - \omega_3)(\Omega^*_{ng} + \omega_2)(\Omega_{ng} - \omega_1) \right]^{-1} + \left[ (\Omega_{lg} + \omega_\sigma)(\Omega_{lg} + \omega_3)(\Omega^*_{ng} + \omega_1) \right]^{-1} + \left[ (\Omega^*_{lg} + \omega_3)(\Omega_{ng} - \omega_2)(\Omega^*_{ng} + \omega_1) \right]^{-1} \right\},$$

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In our notation, \( D_{lmn}^{(3)}(-\omega; \omega_1, \omega_2, \omega_3) = S_{1,2,3} \times \)
\[
\left\{ \begin{array}{l}
\frac{1}{(\Omega_{ig} - \omega)(\Omega_{ng} - \omega_1 - \omega_2)(\Omega_{ng} - \omega - \omega_1)} + \\
\frac{1}{(\Omega_{ig} + \omega)(\Omega_{ng} - \omega_1 - \omega_2)(\Omega_{ng} - \omega - \omega_1)} + \\
\frac{1}{(\Omega_{ig} + \omega)(\Omega_{ng} + \omega_1 + \omega_2)(\Omega_{ng} - \omega - \omega_3)} + \\
\frac{1}{(\Omega_{ig} + \omega)(\Omega_{ng} + \omega_1 + \omega_2)(\Omega_{ng} + \omega + \omega_1)} \end{array} \right\} ,
\tag{7}
\]
In our notation, \( D_{lmn}^{(3)}(-\omega; \omega_1, \omega_2, \omega_3) \) represents interactions, which involve only one-photon states, and \( D_{lmn}^{(2)}(-\omega; \omega_1, \omega_2, \omega_3) \) represents interactions that involve both one- and two-photon states.

It is significantly more difficult to calculate the third-order IB theory because of the triple product of Lorentzian terms in the denominator. In order to transform Equation (9) is multiplied by the stretched Gaussian function to perform the following partial fraction expansion,
\[
\int_{-\infty}^{\infty} \frac{T}{(z - t)^3} dt = \int_{-\infty}^{\infty} \frac{iT}{(z - t)^2} dt - \int_{-\infty}^{\infty} \frac{T}{(z - t)^2} dt ,
\tag{13}
\]
where \( \beta = 1 \), except for the denominator. We use integration by parts to re-express the denominator to first-order in \((z - t)\).

With \( T = \exp(-t^2) \), integrating by parts twice yields:
\[
\int_{-\infty}^{\infty} \frac{T}{(z - t)^3} dt = \int_{-\infty}^{\infty} \frac{iT}{(z - t)^2} dt - \int_{-\infty}^{\infty} \frac{T}{(z - t)^2} dt ,
\tag{13}
\]

where certain terms vanish when the argument of the exponential is small \((\approx -10^3)\) at the lower limit.

Using \((z + t) = (z^2 - t^2)/(z - t)\) and \( \int_{-\infty}^{\infty} t \exp(-t^2) dt = 0 \) to recast Equation (13) into a more convenient form, we get:
\[
\int_{-\infty}^{\infty} \frac{T}{(z - t)^3} dt = (2z^2 - 1) \int_{-\infty}^{+\infty} \frac{T}{(z - t)^2} dt \\
- 2z \int_{-\infty}^{+\infty} T dt, \approx (1 - 2z^2)i\pi W(z) - 2z\sqrt{\pi}.\tag{14}
\]

So, the convolution of the cubic Lorentzian with the Gaussian distribution (with \( \beta = 1 \)) is,
\[
\int_{-\infty}^{\infty} \frac{C_3}{\gamma_{ng} \sqrt{\pi}} f_n (\omega_{ng} - \omega) d\omega_{ng} \\
- \frac{C_3}{\gamma_{ng} \sqrt{\pi}} \left\{ (1 - 2z^2)i\pi W(z) - 2z\sqrt{\pi} \right\} ,
\tag{15}
\]
where \( z = (\omega_{ng} + i\Gamma_{ng} + \omega)/\gamma_{ng} \).

Table II summarizes the third-order fundamental energy denominators for the Lorentzian and IB theories with \( \beta \leq 1 \), and \( \beta = 1 \), respectively. These in conjunction with those derived in Ref. [1] can be used to construct any IB energy denominator for any first-, second-, and/or third-order process.

III. THIRD-ORDER MOLECULAR SUSCEPTIBILITY

As an example of the third-order molecular susceptibility for homogeneous and inhomogeneous models, we use Eq.’s \( (6), (7), (26), (28), (30) \) and \( (32) \), for the respective models, in \( \xi^{(3)}(-\omega; \omega, 0, 0) \)
\[
\frac{1}{\epsilon_0 \beta h^3} \left\{ |\mu_{g1}|^2 |\mu_{12}|^2 D_{121}(-\omega; \omega, 0, 0) - |\mu_{12}|^2 D_{111}(-\omega; \omega, 0, 0) \right\},
\tag{16}
\]
to calculate the imaginary part of \( \xi^{(3)}(-\omega; \omega, 0, 0) \) for a three-level system. Figure II shows the imaginary part of the third-order susceptibility for a system with one one-photon and
TABLE I
FUNDAMENTAL DENOMINATOR CONTRIBUTIONS TO HOMogeneously BROADERened AND INHOMogeneously BROADERened ELECTRonic TRANSITIONS

| Model   | Equation |
|---------|----------|
| L       | \( \frac{C_3}{\gamma_{ng}}(\omega_{ng} + \Gamma_{ng} + \omega) \) |
| IB (\( \beta \leq 1 \)) | \( \frac{C_3}{\gamma_{ng}}\left(1 - W\left(\frac{3}{\gamma_{ng}}\right)\right) \) |
| IB (\( \beta = 1 \)) | \( \frac{C_3}{\gamma_{ng}}\left(2(\omega_{ng} + \Gamma_{ng} + \omega)^2 - \gamma_{ng}^2 \right) \times \)
|           | \( W\left(\frac{-\omega_{ng} + \Gamma_{ng} + \omega}{\gamma_{ng}}\right) \) + \( \frac{2(\omega_{ng} + \Gamma_{ng} + \omega)}{\sqrt{\gamma_{ng}}^2} \) |

Fig. 1. Imaginary part of \( \xi^{(3)}(-\omega; \omega; 0, 0) \) from the generalized IB theory for a one-photon excited state centered at 660 nm (\( \Gamma_{1g} = 10 \text{ meV} \), and \( \gamma_{1g} = 40 \text{ meV} \)) and a two-photon state centered at 595 nm (\( \Gamma_{2g} = 40 \text{ meV} \), and \( \gamma_{2g} = 40 \text{ meV} \)). Three values of \( \beta \) are compared to the Lorentzian theory (\( \Gamma_{1g} = 40 \text{ meV} \), and \( \Gamma_{2g} = 40 \text{ meV} \)). \( \mu_{2g} / \mu_{1g} = 0.4 \) for all the models.

Fig. 2. Absolute value of the imaginary part of \( \chi^{(3)} \) from a quadratic electroabsorption experiment on SiPc/PMMA and least-squares fits using Lorentzian and IB(\( \beta = 1 \)) theories. The fit parameters are from ref. [11].

from quadratic electroabsorption experiments. Also plotted are the Lorentzian (L) and IB theories. A log scale is used to highlight the qualitative and quantitative features of the two models. The fit parameters are from the literature[11]. Like the linear absorption fits in Ref. [1], the IB model fits the data better both quantitatively, roughly a factor of 6 smaller relative error, and qualitatively, especially in the wings of the resonant signal. Neither model fits the data off-resonance because of the large random error associated with the lock-in amplifier signal away from resonance. The error bars cannot be plotted on a log scale because the error range includes negative values in the wings.

IV. COMPARISON OF THEORY TO QUADRAtric ELECTROABSORPTION EXPERIMENTAL RESULTS

We use quadratic electroabsorption spectra to test our models. Details of the experiment and the relationship between Eq. [16] and \( \chi^{(3)}(-\omega; \omega; 0, 0) \) can be found in the literature. Figure 2 compares experimental values of the imaginary part of \( \chi^{(3)} \) for silicon phthalocyanine-methylmethacrylate in polymethylmethacrylate (SiPc/PMMA), which were derived from quadratic electroabsorption experiments. Also plotted are the Lorentzian (L) and IB theories. A log scale is used to highlight the qualitative and quantitative features of the two models. The fit parameters are from the literature[11]. Like the linear absorption fits in Ref. [1], the IB model fits the data better both quantitatively, roughly a factor of 6 smaller relative error, and qualitatively, especially in the wings of the resonant signal. Neither model fits the data off-resonance because of the large random error associated with the lock-in amplifier signal away from resonance. The error bars cannot be plotted on a log scale because the error range includes negative values in the wings.

Nonlinear spectroscopy experiments aim to determine zero frequency nonlinear susceptibilities by extrapolation, which can lead to large uncertainties depending on the quality of the dispersion models. Indeed, Canfield[12], Vigil,[13] and Kruhlak[11] have shown that it is often difficult to reconcile the transition moments as determined by independent means. The wing region near resonance and the shape of the resonance peak may play an important role when using fitting to determine transition moments or for extrapolating to off-resonant values of \( \chi^{(n)} \) from a data set with limited spectral range.

Differences between the IB model and the standard Lorentzian model can be used to determine the reliability of zero-frequency susceptibilities and the uncertainty in transition moments. More importantly, precise modeling aimed at understanding the dispersion of the nonlinear-optical response will need to take into account all possible broadening mechanisms. Because IB theory takes into account the distribution of sites, it may well be the best way to model systems such as doped polymers.

V. CONCLUSION

In conclusion, we have calculated the inhomogeneously broadened third-order nonlinear-optical susceptibilities for a Gaussian and stretched Gaussian distribution of Lorentzians.
The results are applied to the quadratic electro-absorption spectrum of SiPc/PMMA and we find that the Lorentzian fit alone does not fit the data at the wings. The IB theory, however, fits the data over a broader wavelength range and shows that the distribution of sites is nearly Gaussian, implying that interactions between the polymer and dopant are small.

Since broadening of the nonlinear susceptibility is shown to have an important affect the dispersion, the determination of excited state properties of molecules from spectroscopy requires that such a theory be used. So, IB theory using stretched Gaussian statistics may become an important tool for interpreting nonlinear-optical spectroscopy measurements.

ACKNOWLEDGMENTS

We thank the National Science Foundation (ECS-0354736) and Wright Patterson Air Force Base for generously supporting this work.

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APPENDIX A

COMPACT NOTATION

The energy denominators \( D \) for the higher-order susceptibilities are complex combinations of \( W^{(z)}(z) \) or \( W(z) \). We have developed a more compact notation than previously used[11]. For \( \beta \leq 1 \), we have added a subscript to \( \beta \) to indicate the excited state involved in the process and a * on the power of \( W \) to indicate a complex conjugate of the complex argument \( \Omega \). Similarly for \( \beta = 1 \), the subscript on \( W \) indicates the excited state and the superscript * on \( \Omega \) indicates the complex conjugate of \( \Omega \). This allows us to use a simple frequency argument of \( \pm \omega \) that significantly improves the readability of the equations in the extensive appendices that follow. An example of the compact notation is given below:

\[
W^{(1)}_{\beta}(\frac{-\Omega g}{\gamma_g}) \rightarrow W^{(1)*}_{\beta_2}(\omega_3). \tag{17}
\]

Since all arguments are all of the form \( -\Omega_g \pm \omega \) or \( \Omega_g \mp \omega \) and all transitions are from/to the ground state \( (g) \), this form describes all inhomogeneous broadening terms in this paper. Table II summarizes the compact notation.

| \( \beta \) | Ref. | Compact Form |
|---|---|---|
| \( \leq 1 \) | \( W^{(z)}_{\beta}(\frac{-\Omega_g \mp \omega}{\gamma_g}) \) | \( W^{(z)*}_{\beta_2}(\mp \omega) \) |
| \( 1 \) | \( W(\frac{-\Omega_g \pm \omega}{\gamma_g}) \) | \( W(\mp \omega_1) \) |

TABLE II

COMPACT FORM OF \( W^{(z)}(z) \) AND \( W(z) \) UP TO THIRD ORDER \( (z = \{1, 2, 3\}) \).
APPENDIX B
ENERGY DENOMINATORS FOR SELECTED PROCESSES

A. Third-Harmonic Generation

1) \( \beta \leq 1 \):

\[
D_{ln}^{IB}(-3\omega; \omega, \omega, \omega) = \frac{\sqrt{\pi}}{\omega \gamma_{lg}} \left\{ \frac{i}{4\omega} \left[ W_{\beta_m}^{(1)}(-3\omega) - W_{\beta_m}^{(1)}(\omega) - W_{\beta_m}^{(1)*}(\omega) \right]
+ \frac{\Gamma_{lg}}{2(\omega + i\Gamma_{lg})} \left[ \frac{1}{\gamma_{lg}} W_{\beta_l}^{(2)*}(\omega) - \frac{1}{\gamma_{lg}} W_{\beta_l}^{(2)}(-\omega) \right] \right\}
\]

(18)

2) \( \beta = 1 \):

\[
D_{ln}^{IB}(-3\omega; \omega, \omega, \omega) = \frac{\pi^{3/2}}{2\gamma_{lg} \gamma_{mg}^{3/2}} \left\{ \frac{1}{\omega} \left[ W_{\beta_m}^{(1)}(-\omega) W_{\beta_m}^{(1)}(-2\omega) \left[ W_{\beta_l}^{(1)}(-3\omega) + W_{\beta_l}^{(1)*}(\omega) \right]
+ W_{\beta_l}^{(1)*}(\omega) W_{\beta_m}^{(1)}(2\omega) \left[ W_{\beta_l}^{(1)}(\omega) + W_{\beta_l}^{(1)*}(3\omega) \right] \right]\right\}
\]

(19)

\[
D_{l_{mn}}^{IB}(-3\omega; \omega, \omega, \omega) = \frac{1}{\gamma_{lg} \gamma_{mg}^{3/2}} \left\{ \frac{1}{\omega} \left[ W_{\beta_m}^{(1)}(-\omega) W_{\beta_m}^{(1)}(-2\omega) \left[ W_{\beta_l}^{(1)}(-3\omega) + W_{\beta_l}^{(1)*}(\omega) \right]
+ W_{\beta_l}^{(1)*}(\omega) W_{\beta_m}^{(1)}(2\omega) \left[ W_{\beta_l}^{(1)}(\omega) + W_{\beta_l}^{(1)*}(3\omega) \right] \right]\right\}
\]

(20)

B. Quadratic Electrooptic Effect

1) \( \beta \leq 1 \):

\[
D_{ll}^{IB}(-\omega; \omega, 0, 0) = \frac{2\sqrt{\pi}}{\gamma_{lg}} \left\{ \frac{1}{\omega \gamma_{lg}} \left[ W_{\beta_l}^{(2)*}(\omega) - W_{\beta_l}^{(2)}(-\omega) \right]
+ \frac{i\Gamma_{lg}}{\omega(\omega + 2i\Gamma_{lg})} \left[ W_{\beta_l}^{(2)}(0) - W_{\beta_l}^{(2)*}(0) \right]
\right\}
\]

(21)

\[
D_{ln}^{IB}(-\omega; \omega, 0, 0) = \frac{\pi^{3/2}}{2\gamma_{lg} \gamma_{mg}^{3/2}} \left\{ \frac{1}{\omega} \left[ W_{\beta_m}^{(2)}(-\omega) \left[ W_{\beta_l}^{(2)}(-\omega) + W_{\beta_l}^{(2)*}(\omega) \right]
+ W_{\beta_l}^{(2)*}(\omega) W_{\beta_m}^{(2)}(\omega) \left[ W_{\beta_l}^{(2)}(\omega) + W_{\beta_l}^{(2)*}(3\omega) \right] \right]\right\}
\]

(22)
\[ D_{ll}^{IB}(-\omega; \omega, 0, 0) = -\frac{\pi}{\gamma_l \gamma_{ng}} \left\{ W_{\beta_m}^{(1)}(-\omega) \left[ \frac{-1}{\gamma_l} W_{\beta_l}^{(2)}(-\omega) \right] + W_{\beta_m}^{(1)*} \left[ \frac{-1}{\gamma_l} W_{\beta_l}^{(2)*}(\omega) \right] + \frac{1}{\omega} \left( \left\{ W_{\beta_m}^{(1)}(-\omega) - W_{\beta_m}^{(1)*}(\omega) \right\} \left\{ W_{\beta_l}^{(1)}(-\omega) - W_{\beta_l}^{(1)*}(\omega) \right\} \right) \right\} \]
$$D^{IB}_{ln}(-\omega; \omega, 0, 0) = -\frac{2(1 + i \Gamma_{lg})}{(\omega + 2i \Gamma_{lg})} \left[ W_m(-\omega)W_0(0) - W_m^*(\omega)W_0^*(\omega) \right] + \frac{2(1 - i \Gamma_{lg})}{(\omega + 2i \Gamma_{lg})} \left[ W_m^*(\omega)W_0^*(0) - W_m(-\omega)W_0^*(0) \right] + \frac{1}{\omega} \left[ W_m(0)W_0(-\omega) - W_m^*(0)W_0^*(\omega) \right] + \frac{1}{(\omega + 2i \Gamma_{lg})} \left[ W_m^*(0)W_0(-\omega) - W_m(0)W_0^*(\omega) \right]$$ (32)

$$D^{IB}_{lnn}(-\omega; \omega, 0, 0) = -\frac{2i \sqrt{\pi}}{\gamma_{lg}} \left\{ \frac{W_n(0)W_n(0)}{\omega + 2i \Gamma_{lg}} \right\} \left[ \frac{W_n^*(0)W_n^*(0)}{\omega + 2i \Gamma_{lg}} \right]$$ (33)

### C. Electric-Field Induced Second Harmonic Generation

1) \( \beta \leq 1 \):

$$D^{IB}_{ln}(-2\omega; \omega, 0, 0) = \frac{-i \pi^{3/2}}{\gamma_{lg}} \left\{ \left\{ W_{\beta_n}(-\omega)W_{\beta_n}(0) - W_{\beta_n}^*(\omega)W_{\beta_n}^*(0) \right\} \right\}$$ (34)

$$D^{IB}_{ln}(-2\omega; \omega, 0, 0) = \frac{-i \pi^{3/2}}{\gamma_{lg}} \left\{ \left\{ W_{\beta_n}(-\omega)W_{\beta_n}(0) - W_{\beta_n}^*(\omega)W_{\beta_n}^*(0) \right\} \right\}$$ (35)

$$D^{IB}_{ln}(-2\omega; \omega, 0, 0) = \frac{-i \pi^{3/2}}{\gamma_{lg}} \left\{ \left\{ W_{\beta_n}(-\omega)W_{\beta_n}(0) - W_{\beta_n}^*(\omega)W_{\beta_n}^*(0) \right\} \right\}$$ (36)
\[ D_{imn}^{\beta}(-2\omega; \omega, 0) = -i\frac{\pi^{3/2}}{\gamma_l g \gamma_m g} \left\{ W_{\beta_m}^{(1)}(-2\omega) W_{\beta_n}^{(1)}(-\omega) \left[ W_{\beta_l}^{(1)}(-2\omega) + W_{\beta_l}^{(1)*}(0) \right] \right. \\
+ W_{\beta_m}^{(1)*}(\omega) W_{\beta_n}^{(1)*}(2\omega) \left[ W_{\beta_l}^{(1)}(0) + W_{\beta_l}^{(1)*}(2\omega) \right] \\
+ \left. W_{\beta_m}^{(1)}(-\omega) \left[ W_{\beta_l}^{(1)}(-2\omega) + W_{\beta_l}^{(1)*}(\omega) \right] \left[ W_{\beta_n}^{(1)}(-\omega) + W_{\beta_n}^{(1)*}(0) \right] \\
+ W_{\beta_l}^{(1)*}(\omega) \left[ W_{\beta_l}^{(1)}(-\omega) + W_{\beta_l}^{(1)*}(2\omega) \right] \left[ W_{\beta_l}^{(1)*}(\omega) + W_{\beta_l}^{(1)*}(0) \right] \right\} \] (37)

2) \( \beta = 1 \):

\[ D_{il}^{IB}(-2\omega; \omega, 0) = \frac{2i\sqrt{\pi}}{\gamma_l g} \left\{ \frac{2 \Gamma_l g}{\omega(\omega + 2i \Gamma_l g)^2} \left[ (\Omega_{lg}^* + \omega) W_{l}^{*}(\omega) + \frac{2i \gamma_l g}{\sqrt{\pi}} + (\Omega_{lg} - \omega) W_l(-\omega) \right] \right. \\
+ \frac{2 \Gamma_l^2 g}{\omega^2(\omega + 2i \Gamma_l g)^2} \left[ W_l(-\omega) + W_l^*(\omega) - W_l^*(0) - W_l(0) \right] \\
+ \left. \frac{1}{\omega^2} [W_l(-2\omega) + W_l^*(2\omega) - W_l(-\omega) - W_l^*(\omega)] \right\} \] (38)

\[ D_{in}^{IB}(-2\omega; \omega, 0) = -\frac{\pi}{\gamma_l g \gamma_m g} \left\{ \frac{1}{2\omega} \left[ W_n(-\omega) \{ W_l(-2\omega) - W_l(0) \} + W_n^*(\omega) \{ W_l^*(0) - W_l^*(2\omega) \} \right] \\
+ \frac{1}{\omega} \left[ \{ W_n(-\omega) + W_n(0) \} \{ W_l(-2\omega) - W_l(0) \} + \{ W_n^*(\omega) + W_n^*(0) \} \{ W_l^*(0) - W_l^*(2\omega) \} \right] \\
+ \frac{1}{2(\omega + i \Gamma_l g)} \left[ \{ W_n(-\omega) + W_n^*(\omega) \} \{ W_l(-\omega) - W_l^*(\omega) \} \right] \\
+ \frac{1}{\gamma_l g} \left[ \{ W_n(-2\omega) + W_n^*(2\omega) \} \{ W_l(-\omega) - W_l^*(0) \} \right] \\
+ \{ W_n(-\omega) + W_n^*(2\omega) \} \{ W_l(0) - W_l^*(\omega) \} \right\} \] (39)

\[ D_{im}^{IB}(-2\omega; \omega, 0) = -i\pi^{3/2} \frac{\gamma_l g \gamma_m g}{\gamma_l g} \left\{ W_m(-2\omega) W_n(-\omega) [W_l(-2\omega) + W_l^*(0)] + W_m^*(\omega) W_m^*(2\omega) [W_n(0) + W_n^*(2\omega)] \\
+ W_m(-\omega) [W_l(-2\omega) + W_l^*(\omega)] [W_n(-\omega) + W_n(0)] \\
+ W_m^*(\omega) [W_n(-\omega) + W_n^*(2\omega)] [W_l^*(\omega) + W_l^*(0)] \right\} \] (40)

\[ D_{imn}^{IB}(-2\omega; \omega, 0) = -i\pi^{3/2} \left\{ W_m(-2\omega) W_n(-\omega) [W_l(-2\omega) + W_l^*(0)] + W_m^*(\omega) W_m^*(2\omega) [W_n(0) + W_n^*(2\omega)] \\
+ W_m(-\omega) [W_l(-2\omega) + W_l^*(\omega)] [W_n(-\omega) + W_n(0)] \\
+ W_m^*(\omega) [W_n(-\omega) + W_n^*(2\omega)] [W_l^*(\omega) + W_l^*(0)] \right\} \] (41)

D. Kerr Effect

1) Intensity dependent Refractive index:

a) \( \beta \leq 1 \):

\[ D_{il}^{IB}(-\omega; \omega, \omega, \omega) = \frac{i\sqrt{\pi}}{\gamma_l g} \left\{ \frac{3}{2} \gamma_l g W_{\beta_l}^{(3)}(-\omega) + \frac{1}{2} \gamma_l g W_{\beta_l}^{(3)*}(\omega) + \omega + 2i \Gamma_l g \frac{1}{2\omega \Gamma_l g} \left[ \frac{1}{\gamma_l g} W_{\beta_l}^{(2)*}(\omega) - \frac{1}{\gamma_l g} W_{\beta_l}^{(2)}(-\omega) \right] \right. \\
+ \frac{2 \Gamma_l^2 g - \omega^2 + 2i \Gamma_l g \omega}{4 \Gamma_l^2 g \omega^2} \left[ W_{\beta_l}^{(1)}(\omega) + W_{\beta_l}^{(1)*}(-\omega) - W_{\beta_l}^{(1)*}(\omega) - W_{\beta_l}^{(1)*}(-\omega) \right] \right\} \] (42)
\[ D_{ln}^{IB}(-\omega; \omega, \omega, -\omega) = \frac{-\pi}{\gamma_g \gamma_{ng}} \left\{ \begin{array}{l} [W_{\beta_n}^{(1)}(-\omega) + W_{\beta_n}^{(1)}(\omega)] \left[ \frac{-1}{\gamma_g} W_{\beta_n}^{(2)}(-\omega) \right] + \left[ W_{\beta_n}^{(1)}(-\omega) + W_{\beta_n}^{(1)}(\omega) \right] \left[ \frac{-1}{\gamma_g} W_{\beta_n}^{(2)}(\omega) \right] \\
\frac{1}{2\Gamma_{ng}} \left[ W_{\beta_n}^{(1)}(-\omega) + W_{\beta_n}^{(1)}(\omega) \right] \left[ W_{\beta_n}^{(1)}(-\omega) + W_{\beta_n}^{(1)}(\omega) \right] \\
+ \frac{1}{2(\omega + i\Gamma_{ng})} \left[ W_{\beta_n}^{(1)}(-\omega) - W_{\beta_n}^{(1)}(\omega) \right] \left[ W_{\beta_n}^{(1)}(-\omega) + W_{\beta_n}^{(1)}(\omega) \right] \\
+ \frac{1}{2\omega} \left[ W_{\beta_n}^{(1)}(-\omega) \right] \{ W_{\beta_n}^{(1)}(-\omega) - W_{\beta_n}^{(1)}(\omega) \} \{ W_{\beta_n}^{(1)}(-\omega) - W_{\beta_n}^{(1)}(\omega) \} \right\} \] (43)

\[ D_{lmn}^{IB}(-\omega; \omega, \omega, -\omega) = \frac{-\pi}{\gamma_g \gamma_{mg} \gamma_{ng}} \left\{ \begin{array}{l} [W_{\beta_m}^{(1)}(-2\omega) + W_{\beta_m}^{(1)}(0)] \left[ \frac{-1}{\gamma_g} W_{\beta_m}^{(2)}(-\omega) \right] + \left[ W_{\beta_m}^{(1)}(-2\omega) + W_{\beta_m}^{(1)}(0) \right] \left[ \frac{-1}{\gamma_g} W_{\beta_m}^{(2)}(\omega) \right] \\
\frac{1}{2\Gamma_{ng}} \left[ \{ W_{\beta_m}^{(1)}(-2\omega) + W_{\beta_m}^{(1)}(0) \} \left\{ W_{\beta_i}^{(1)}(-\omega) - W_{\beta_i}^{(1)}(\omega) \right\} \\
+ \left\{ W_{\beta_m}^{(1)}(-2\omega) + W_{\beta_m}^{(1)}(0) \right\} \left\{ W_{\beta_i}^{(1)}(\omega) - W_{\beta_i}^{(1)}(\omega) \right\} \right] \\
+ \frac{1}{2(\omega + i\Gamma_{ng})} \left[ W_{\beta_m}^{(1)}(0) + W_{\beta_m}^{(1)}(0) \right] \left[ W_{\beta_i}^{(1)}(-\omega) - W_{\beta_i}^{(1)}(\omega) \right] \\
+ \frac{1}{2\omega} \left[ W_{\beta_m}^{(1)}(0) \right] \{ W_{\beta_i}^{(1)}(-\omega) - W_{\beta_i}^{(1)}(\omega) \} \{ W_{\beta_i}^{(1)}(-\omega) - W_{\beta_i}^{(1)}(\omega) \} \right\} \] (44)

\[ D_{lmm}^{IB}(-\omega; \omega, \omega, -\omega) = \frac{-i\pi^{3/2}}{\gamma_g \gamma_{mg} \gamma_{ng}} \left\{ \begin{array}{l} W_{\beta_m}^{(1)}(0) \left[ W_{\beta_n}^{(1)}(-\omega) + W_{\beta_n}^{(1)}(\omega) \right] \left[ W_{\beta_n}^{(1)}(-\omega) + W_{\beta_n}^{(1)}(\omega) \right] \\
+ W_{\beta_m}^{(1)}(0) \left[ W_{\beta_n}^{(1)}(-\omega) + W_{\beta_n}^{(1)}(\omega) \right] \left[ W_{\beta_n}^{(1)}(-\omega) + W_{\beta_n}^{(1)}(\omega) \right] \\
+ W_{\beta_m}^{(1)}(-2\omega) W_{\beta_n}^{(1)}(-\omega) \left[ W_{\beta_n}^{(1)}(-\omega) + W_{\beta_n}^{(1)}(\omega) \right] \\
+ W_{\beta_m}^{(1)}(-2\omega) W_{\beta_n}^{(1)}(\omega) \left[ W_{\beta_n}^{(1)}(-\omega) + W_{\beta_n}^{(1)}(\omega) \right] \right\} \] (45)

b) $\beta = 1$:

\[ D_{ln}^{IB}(-\omega; \omega, \omega, -\omega) = \frac{i\sqrt{\pi}}{\gamma_g} \left\{ \begin{array}{l} 2(\Omega_g - \omega)^2 - \gamma_g^2 W_I(-\omega) + \frac{2(\Omega_g + \omega)^2 - \gamma_g^2 W_I^*(\omega)}{\sqrt{\pi} \gamma_g} \right\} \] (46)

\[ D_{ln}^{IB}(-\omega; \omega, \omega, -\omega) = \frac{-\pi}{\gamma_g \gamma_{ng}} \left\{ \begin{array}{l} W_n(-\omega) + W_n(\omega) \left[ \frac{2(\Omega_g - \omega)}{\gamma_g} W_I(-\omega) + \frac{2i}{\sqrt{\pi} \gamma_g} \right] \\
+ [W_n(\omega) + W_n^*(\omega)] \left[ \frac{2(\Omega_g + \omega)}{\gamma_g} W_I^*(\omega) + \frac{2i}{\sqrt{\pi} \gamma_g} \right] \\
+ \frac{1}{2i\Gamma_{ng}} \left[ W_n(-\omega) + W_n(\omega) - W_n^*(\omega) - W_n^*(\omega) \right] \left[ W_I(-\omega) + W_I^*(\omega) \right] \\
+ \frac{1}{2(\omega + i\Gamma_{ng})} \left[ W_n(-\omega) - W_n^*(\omega) \right] \left[ W_I(-\omega) + W_I^*(\omega) \right] \\
+ \frac{1}{2\omega} \left[ W_n(-\omega) \right] \{ W_I(-\omega) - W_I(\omega) \} \{ W_I^*(-\omega) - W_I^*(\omega) \} \right\} \] (47)
\[
D^{LB}_{l,m}(-\omega_1; \omega_1, \omega, -\omega) = \frac{-\pi}{\gamma_g \gamma_{mg}} \left\{ \left[ W_m(-2\omega) + W_m(0) \right] \left[ \frac{2(\Omega_g - \omega)}{\gamma_g^2} W_l(-\omega) + \frac{2i}{\sqrt{\pi} \gamma_g} \right] \right.
\]
\[
+ \left[ W_m^*(2\omega) + W_m(0) \right] \left[ \frac{2(\Omega_g^* + \omega)}{\gamma_g^2} W_l^*(\omega) + \frac{2i}{\sqrt{\pi} \gamma_g} \right] \right.
\]
\[
+ \frac{1}{2i \gamma_g} \left[ \{ W_m(-2\omega) + W_m^*(0) \} \{ W_l(-\omega) - W_l^*(\omega) \} \right. \left. + \{ W_m^*(2\omega) + W_m(0) \} \{ W_l(\omega) - W_l^*(\omega) \} \right]
\]
\[
+ \frac{1}{2 \omega} \left[ W_m(0) \{ W_l(-\omega) - W_l(\omega) \} + W_m^*(0) \{ W_l^*(\omega) - W_l^*(\omega) \} \right] \left\} \right.
\]
\[
+ \frac{1}{2 \omega} \left[ W_m(-2\omega) \{ W_l(\omega) - W_l^*(\omega) \} + W_m^*(2\omega) \{ W_l^*(\omega) - W_l(\omega) \} \right] \right\}
\]
\[
\left[ W_m(-2\omega) + W_m(0) \right] \left[ \frac{2(\Omega_g^* + \omega)}{\gamma_g^2} W_l^*(\omega) + \frac{2i}{\sqrt{\pi} \gamma_g} \right] \right.
\]
\[
+ \frac{1}{2(\omega + i \Gamma_g)} \left[ W_m(0) + W_m^*(0) \right] \left[ W_l(-\omega) - W_l^*(\omega) \right]
\]
\[
+ \frac{1}{2 \omega} \left[ W_m(0) \{ W_l(-\omega) - W_l(\omega) \} + W_m^*(0) \{ W_l^*(\omega) - W_l^*(\omega) \} \right] \right\}
\]
(48)

2) Pump-Probe:

a) \( \beta \leq 1 \):

\[
D^{LB}_{l,m}(-\omega_1; \omega_1, \omega_2, -\omega_2) = \frac{2i \sqrt{\pi}}{\gamma_g \gamma_{mg} \gamma_{ng}} \left\{ \frac{2 \omega_1}{\gamma_g} \left[ \frac{1}{\gamma_g} W_{\beta_1}^{(2)*}(\omega_1) - \frac{1}{\gamma_g} W_{\beta_1}^{(2)}(-\omega_1) \right] \right.
\]
\[
+ \frac{2(\omega_1^2 - 2 \omega_1 \omega_2 + \omega_2^2 + 2i \Gamma_{lg} \omega_1^2 + 6 \Gamma_{lg}^2 \omega_2^2 - 8 \Gamma_{lg} \omega_1 \omega_2)}{(\omega_1 + \omega_2)^2(\omega_1 - \omega_2)^2(\omega_1 + \omega_2 + 2i \Gamma_{lg})(\omega_1 - \omega_2 + 2i \Gamma_{lg})} \left[ W_{\beta_1}^{(1)}(-\omega_1) + W_{\beta_1}^{(1)*}(\omega_1) \right]
\]
\[
+ \frac{(\omega_1^2 - \omega_1 \omega_2^2 + 3i \Gamma_{lg} \omega_1 \omega_2 - 3 \Gamma_{lg}^2 \omega_2^2 + \Gamma_{lg}^2)}{i \Gamma_{lg} \omega_2(\omega_1 - \omega_2)^2(\omega_1 + \omega_2 + 2i \Gamma_{lg})} \left[ W_{\beta_1}^{(1)}(-\omega_2) + W_{\beta_1}^{(1)*}(\omega_2) \right]
\]
\[
+ \frac{(\omega_1^2 + \omega_1 \omega_2^2 - 3i \Gamma_{lg} \omega_1 \omega_2 - 3 \Gamma_{lg}^2 \omega_2^2 + \Gamma_{lg}^2)}{i \Gamma_{lg} \omega_2(\omega_1 + \omega_2)^2(\omega_1 - \omega_2 + 2i \Gamma_{lg})} \left[ W_{\beta_1}^{(1)}(\omega_1) + W_{\beta_1}^{(1)*}(\omega_2) \right]
\]
\]
(50)

\[
D^{LB}_{l,n}(-\omega_1; \omega_1, \omega_2, -\omega_2) = \frac{-\pi}{\gamma_g \gamma_{ng}} \left\{ \left[ \frac{-1}{\gamma_g} W_{\beta_n}^{(2)}(-\omega_1) \right] \left[ W_{\beta_n}^{(1)}(-\omega_2) + W_{\beta_n}^{(1)}(\omega_2) \right]
\]
\[
+ \left[ \frac{-1}{\gamma_g} W_{\beta_n}^{(2)*}(\omega_1) \right] \left[ W_{\beta_n}^{(1)*}(\omega_2) + W_{\beta_n}^{(1)*}(\omega_2) \right]
\]
\[
+ \frac{1}{\omega_1 - \omega_2 + 2i \Gamma_{ng}} \left[ W_{\beta_n}^{(1)}(\omega_2) + W_{\beta_n}^{(1)*}(\omega_2) \right]
\]
\[
\times \left[ W_{\beta_n}^{(1)}(-\omega_1) + W_{\beta_n}^{(1)}(\omega_2) - W_{\beta_n}^{(1)*}(-\omega_2) - W_{\beta_n}^{(1)*}(\omega_2) \right]
\]
\[
+ \frac{1}{\omega_1 + \omega_2 + 2i \Gamma_{ng}} \left[ W_{\beta_n}^{(1)}(\omega_1) + W_{\beta_n}^{(1)*}(\omega_1) \right]
\]
\[
\times \left[ W_{\beta_n}^{(1)}(-\omega_2) + W_{\beta_n}^{(1)}(-\omega_2) - W_{\beta_n}^{(1)*}(-\omega_2) - W_{\beta_n}^{(1)*}(\omega_2) \right]
\]
\[
+ \frac{1}{2i \Gamma_{ng}} \left[ W_{\beta_n}^{(1)}(-\omega_1) + W_{\beta_n}^{(1)*}(\omega_1) \right]
\]
\[
\times \left[ W_{\beta_n}^{(1)}(\omega_2) + W_{\beta_n}^{(1)}(\omega_2) - W_{\beta_n}^{(1)*}(\omega_2) - W_{\beta_n}^{(1)*}(\omega_2) \right]
\]
\[
+ \frac{1}{\omega_1 - \omega_2} \left\{ \left[ W_{\beta_n}^{(1)}(-\omega_1) - W_{\beta_n}^{(1)}(\omega_2) \right] \left[ W_{\beta_n}^{(1)}(\omega_1) + W_{\beta_n}^{(1)}(\omega_2) \right]
\]
\[
+ \left[ W_{\beta_n}^{(1)*}(\omega_2) - W_{\beta_n}^{(1)*}(\omega_1) \right] \left[ W_{\beta_n}^{(1)*}(\omega_2) + W_{\beta_n}^{(1)*}(\omega_2) \right] \right\}
\]
\[
+ \frac{1}{\omega_1 + \omega_2} \left\{ \left[ W_{\beta_n}^{(1)}(-\omega_1) - W_{\beta_n}^{(1)}(\omega_2) \right] \left[ W_{\beta_n}^{(1)}(-\omega_1) + W_{\beta_n}^{(1)}(\omega_2) \right]
\]
\[
+ \left[ W_{\beta_n}^{(1)*}(\omega_2) - W_{\beta_n}^{(1)*}(\omega_1) \right] \left[ W_{\beta_n}^{(1)}(\omega_2) + W_{\beta_n}^{(1)}(\omega_2) \right] \right\}
\]
\]
(51)
\[
D_{\text{lin}}^{IB}(-\omega_1; \omega_2, -\omega_2) = \frac{-\pi}{\gamma_\text{Ig} \gamma_\text{ng} \gamma_\text{mng}} \left\{ \left[- \frac{1}{\gamma_\text{Ig}} W_{\beta_I}^{(2)}(-\omega_1) \right] \left[ W_{\beta_m}^{(1)}(-\omega_1 - \omega_2) + W_{\beta_m}^{(1)}(\omega_1 + \omega_2) \right] + \left[ -\frac{1}{\gamma_\text{Ig}} W_{\beta_I}^{(2)*}\right] \left[ W_{\beta_m}^{(1)*}(\omega_1 + \omega_2) + W_{\beta_m}^{(1)*}(-\omega_1 - \omega_2) \right] \right.
\]
\[
+ \frac{1}{\omega_1 - \omega_2 + 2i \Gamma_{\text{Ig}}} \left\{ \left[ W_{\beta_m}^{(1)}(0) + W_{\beta_m}^{(1)*}(-\omega_1 - \omega_2) \right] \left[ W_{\beta_I}^{(1)}(-\omega_1) - W_{\beta_I}^{(1)*}(-\omega_2) \right] + \left[ W_{\beta_m}^{(1)}(0) + W_{\beta_m}^{(1)*}(\omega_1 + \omega_2) \right] \left[ W_{\beta_I}^{(1)}(\omega_1) - W_{\beta_I}^{(1)*}(\omega_2) \right] \right\}
\]
\[
\left. + \frac{1}{\omega_1 + \omega_2 + 2i \Gamma_{\text{Ig}}} \left\{ \left[ W_{\beta_m}^{(1)}(0) + W_{\beta_m}^{(1)*}(-\omega_1 + \omega_2) \right] \left[ W_{\beta_I}^{(1)}(-\omega_1) - W_{\beta_I}^{(1)*}(\omega_2) \right] + \left[ W_{\beta_m}^{(1)}(0) + W_{\beta_m}^{(1)*}(\omega_1 - \omega_2) \right] \left[ W_{\beta_I}^{(1)}(\omega_1) - W_{\beta_I}^{(1)*}(\omega_2) \right] \right\} \right\}
\]
\[
(52)
\]
\[
D_{\text{lin}}^{IB}(-\omega_1; \omega_1, \omega_1, -\omega_2) = \frac{-i \sqrt{3}}{\gamma_\text{Ig} \gamma_\text{ng} \gamma_\text{mng}} \left\{ \left[- \frac{1}{\gamma_\text{Ig}} W_{\beta_I}^{(2)}(-\omega_1) \right] \left[ W_{\beta_m}^{(1)}(-\omega_1 - \omega_2) + W_{\beta_m}^{(1)*}(-\omega_1 + \omega_2) \right] + \left[ -\frac{1}{\gamma_\text{Ig}} W_{\beta_I}^{(2)*}\right] \left[ W_{\beta_m}^{(1)*}(\omega_1 - \omega_2) + W_{\beta_m}^{(1)*}(\omega_1 + \omega_2) \right] \right.
\]
\[
\left. + \frac{1}{2i \Gamma_{\text{Ig}}} \left\{ \left[ W_{\beta_m}^{(1)}(0) + W_{\beta_m}^{(1)*}(-\omega_1 + \omega_2) \right] \left[ W_{\beta_I}^{(1)}(-\omega_1) + W_{\beta_I}^{(1)*}(\omega_2) \right] + \left[ W_{\beta_m}^{(1)}(0) + W_{\beta_m}^{(1)*}(\omega_1 - \omega_2) \right] \left[ W_{\beta_I}^{(1)}(\omega_1) + W_{\beta_I}^{(1)*}(\omega_2) \right] \right\} \right\}
\]
\[
(53)
\]

b) \( \beta = 1:\)
\[
D_{\text{lin}}^{IB}(-\omega_1; \omega_2, -\omega_2) = \frac{2i \sqrt{\pi}}{\gamma_\text{Ig}} \left\{ \frac{2 \omega_1}{(\omega_1 + 2 \omega_2)(\omega_1 - \omega_2)} \left[ \frac{2(\Omega_{\text{Ig}} - \omega_1)}{\gamma_\text{Ig}} W_I(\omega_1) - 2(\Omega_{\text{Ig}} + \omega_1) \right] \right.
\]
\[
\left. + \frac{2(\omega_1^2 - \omega_2^2)^2 + 2i(\Omega_{\text{Ig}} \omega_2^2 + 8i \Gamma_{\text{Ig}} \omega_1 \omega_2^2 - 8i \Gamma_{\text{Ig}} \omega_1 \omega_2^2)}{(\omega_1 + \omega_2)^2(\omega_1 - \omega_2)^2(\omega_1 + \omega_2 + 2i \Gamma_{\text{Ig}})(\omega_1 - \omega_2 + 2i \Gamma_{\text{Ig}})} \left[ W_I(\omega_1) + W_I^*(\omega_1) \right] \right.
\]
\[
\left. + \frac{8i \Gamma_{\text{Ig}} \omega_1 \omega_2^2 - 3i \Gamma_{\text{Ig}} \omega_1 \omega_2 - 3i \Gamma_{\text{Ig}} \omega_1 \omega_2 + \Gamma_{\text{Ig}}^2 \omega_1}{i \Gamma_{\text{Ig}} \omega_2(\omega_1 - \omega_2)^2(\omega_1 + \omega_2 + 2i \Gamma_{\text{Ig}})} \left[ W_I(\omega_1) + W_I^*(\omega_1) \right] \right\}
\]
\[
(54)
\]
\[
D_{\text{lin}}^{IB}(-\omega_1; \omega_1, \omega_1, -\omega_2) = \frac{-\pi}{\gamma_\text{Ig} \gamma_\text{ng} \gamma_\text{mng}} \left\{ \left[ \frac{2(\Omega_{\text{Ig}} - \omega_1)}{\gamma_\text{Ig}} W_I(\omega_1) + \frac{2i}{\sqrt{\pi} \gamma_\text{Ig}} \right] \left[ W_n(-\omega_2) + W_n(\omega_2) \right] + \left[ \frac{2(\Omega_{\text{Ig}} + \omega_1)}{\gamma_\text{Ig}} W_I^*(\omega_1) + \frac{2i}{\sqrt{\pi} \gamma_\text{Ig}} \right] \left[ W_n^*(\omega_2) + W_n^*(\omega_2) \right] \right.
\]
\[
\left. + \frac{1}{\omega_1 - \omega_2 + 2 \Gamma_{\text{Ig}} \gamma_\text{mng}} \left[ W_I(-\omega_2) + W_I^*(\omega_2) \right] \left[ W_n(-\omega_1) + W_n(\omega_1) - W_n^*(\omega_2) - W_n^*(\omega_1) \right] \right\}
\]
\[
(54)
\]
\[ D_{\text{lim}}^{IB}(-\omega_1; \omega_1, \omega_2, -\omega_2) = \frac{-\pi}{\gamma_1^g \gamma_m g} \left\{ \left[ \frac{2(\Omega_1^g - \omega_1)}{\gamma_1^g} \right] W_i(-\omega_1) + \frac{2i}{\sqrt{\pi \gamma_1^g}} \right\} \left[ W_m(-\omega_1 - \omega_2) + W_m(-\omega_1 + \omega_2) \right] 
+ \frac{1}{\omega_1 - \omega_2 + 2i \Gamma_1^g} \left\{ \left[ W_m(0) + W_m(-\omega_1 - \omega_2) \right] W_i(-\omega_1 - \omega_2) \right. 
\left. + \left[ W_m(0) + W_m^*(\omega_1 + \omega_2) \right] W_i(\omega_2 - \omega_1) \right\} \] 
+ \frac{1}{\omega_1 + \omega_2 + 2i \Gamma_1^g} \left\{ \left[ W_m(0) + W_m(-\omega_1 + \omega_2) \right] W_i(-\omega_1 + \omega_2) \right. 
\left. + \left[ W_m(0) + W_m^*(\omega_1 - \omega_2) \right] W_i(-\omega_1) \right\} 
+ \frac{1}{\omega_1 - \omega_2 + 2i \Gamma_1^g} \left\{ \left[ W_m(0) + W_m(-\omega_1 - \omega_2) \right] W_i(-\omega_1 - \omega_2) \right. 
\left. + \left[ W_m(0) + W_m^*(\omega_1 + \omega_2) \right] W_i(\omega_2 - \omega_1) \right\} 
+ \frac{1}{\omega_1 + \omega_2 + 2i \Gamma_1^g} \left\{ \left[ W_m(0) + W_m(-\omega_1 + \omega_2) \right] W_i(-\omega_1 - \omega_2) \right. 
\left. + \left[ W_m(0) + W_m^*(\omega_1 - \omega_2) \right] W_i(-\omega_1) \right\} 
+ \frac{1}{\omega_1 - \omega_2 + 2i \Gamma_1^g} \left\{ \left[ W_m(0) + W_m(-\omega_1 - \omega_2) \right] W_i(-\omega_1 - \omega_2) \right. 
\left. + \left[ W_m(0) + W_m^*(\omega_1 + \omega_2) \right] W_i(\omega_2 - \omega_1) \right\} 
+ \frac{1}{\omega_1 + \omega_2 + 2i \Gamma_1^g} \left\{ \left[ W_m(0) + W_m(-\omega_1 + \omega_2) \right] W_i(-\omega_1 - \omega_2) \right. 
\left. + \left[ W_m(0) + W_m^*(\omega_1 - \omega_2) \right] W_i(-\omega_1) \right\} \] 
(56)

\[ D_{\text{lim}}^{IB}(-\omega_1; \omega_1, \omega_2, -\omega_2) = \frac{-i \pi^{3/2}}{\gamma_1^g \gamma_m g} \left\{ \left[ W_m(-\omega_1 - \omega_2) + W_m^*(-\omega_1) \right] W_i(-\omega_1) + W_m(-\omega_2) \right\} 
+ W_m(-\omega_1 + \omega_2) \left[ W_i(-\omega_1) + W_i^*(-\omega_2) \right] \left[ W_m(-\omega_1) + W_m(-\omega_2) \right] \] 
+ W_m(0) \left[ W_i(-\omega_1) + W_i^*(-\omega_2) \right] \left[ W_m(-\omega_1) + W_m(-\omega_2) \right] \] 
+ W_m^*(\omega_1 + \omega_2) \left[ W_i^*(\omega_1) + W_i^*(-\omega_2) \right] \left[ W_m(\omega_1) + W_m^*(\omega_1) \right] \] 
+ W_m^*(\omega_1 - \omega_2) \left[ W_i^*(\omega_1) + W_i^*(-\omega_2) \right] \left[ W_m(\omega_2) + W_m^*(\omega_1) \right] \] 
+ W_m^*(0) \left[ W_i^*(\omega_2) + W_i^*(-\omega_2) \right] \left[ W_m(-\omega_1) + W_m^*(\omega_1) \right] \] 
(57)