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# Derivation of nonlinear optical conductivity by using a reduction identity and a state-dependent projection method

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## Abstract

A new nonlinear optical conductivity formula for a system of electrons interacting with phonons was derived using a reduction identity and a state-dependent projection technique introduced by the authors. The results include a general formula for the nonlinear optical conductivity of the general rank and the linear, first-order nonlinear and second-order nonlinear conductivity are calculated in terms of the linewidth. The linewidth term includes the electron and phonon distribution functions properly. Therefore, it is possible to explain the phonon emission and absorption in all electron transition processes in an organized manner.

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## 1. Introduction

The transport behavior of electrons in semiconductors is characterized by scattering mechanisms, including electron–electron, electron–phonon and electron–impurity interactions. However, the electron–electron and electron–impurity interactions may be neglected if the number density of electrons is very low, as in ordinary semiconductors, and the temperature is in the middle range (50–200 K). Hence, the dominant scattering mechanism is an electron–phonon interaction, which may be dealt with as a perturbation. Therefore, a study of the optical transitions in electron systems is important for determining the electronic structures of solids because the absorption lineshapes are quite sensitive to the type of scattering mechanisms affecting the behavior of carriers. However, most theories have been limited to linear scheme [1–15].

The field of nonlinear optical phenomena [16–39] was initiated by the experimental work by Franken *et al* [17] on optical second-harmonic generation and the theoretical work by Bloembergen *et al* [18, 19] on optical wave mixing. Although there are many approaches to these phenomena, such as the time-dependent perturbation theory [21, 24, 25], density-functional theory [30], Boltzmann equation method [26–28, 31], *K*-operator technique [29] and two-temperature model [16, 22, 32], we have been interested in the quantum-statistical projection technique [12–14, 33]. In this method, the resolvent factor contained in the conductivity tensor was expanded using projectors, yielding quite many useful formulae. Note that quantum transport theory is a very powerful tool for examining the microscopic scattering phenomena of the electron systems.

There are two types of projection techniques: the single-electron projection technique [1–6, 15] and the many-electron projection technique [7, 9, 12–14, 33]. The latter is more general than the former because the many-body formalism in solids generally cannot be reduced to a single-body one. The many-body projectors are classified into two categories: the first being state independent [7, 9, 12] and the second being state dependent [13, 14]. The state-independent method is applicable only to cases where the spacing between the nearest levels is constant, such as cyclotron resonance phenomena. However, it is unsuitable to a system with nonuniform energy separation, such as a quantum square well. Therefore, the state-dependent projection method is less limited than the state-independent one.

This study applied the state-dependent projection method and the general form of the reduction identity, introduced by the authors [13], to derive rigorous explicit expressions for the linear, first-order nonlinear and second-order nonlinear optical conductivity tensors for an electron system interacting with the phonons. As the higher order calculation is more general, how the reduction identity is utilized in this problem will be shown in detail only in section 6, in which the second-order nonlinear part is dealt with.

## 2. Reduction identity

We start with the following equation for the arbitrary operators $X$ and $A$:

$$T_R\{\rho(H)[LX, A]\} = T_R\{\rho(H)(HXA - XHA - AHX + AXH)\}, \qquad (2.1)$$

where $T_R$ means the many-body trace with the basis states $|\Psi\rangle = \left(a_\alpha^+\right)^{n_\alpha}\left(a_\beta^+\right)^{n_\beta}\cdots|\Psi_0\rangle$, $n_\alpha, n_\beta, \ldots$ being the number operators for the occupied states $\alpha, \beta, \ldots$, $|\Psi_0\rangle$ is the vacuum state, $a_\alpha^+$ denotes the creation operator for an electron in the $|\alpha\rangle$ state, $\rho(H)$ is the density operator, $L$ is the Liouville operator corresponding the Hamiltonian of the system, $H$, which is defined as $LX \equiv [H, X]$ for an arbitrary operator, $X$, and $[A, B]$ is the commutator. Considering $[H, \rho(H)] = 0$ and $T_R(ABC) = T_R(BCA)$ for the first and fourth terms on the right-hand side, the zeroth-order reduction identity can be obtained as

$$\begin{aligned} T_R\{\rho(H)[LX, A]\} &= T_R\{\rho(H)(XAH - XHA + AHX - HAX)\} \\ &= -T_R\{\rho(H)[X, LA]\}, \end{aligned} \qquad (2.2)$$

which is useful because $L$ and $X$ are separated (see the following sections). Similarly, for arbitrary operators $A$ and $B$,

$$\begin{aligned} T_R\{\rho(H)[[LX, A], B]\} = T_R\{\rho(H)(XABH - BHXA - AHXB + BAHX \\ - XHAB + BXHA + AXHB - HBAX)\}, \end{aligned} \qquad (2.3)$$

where $[H, \rho(H)] = 0$ and $T_R(ABC) = T_R(BCA)$ were considered for the first and eighth terms on the right-hand side, respectively. The first-order reduction identity can be

obtained by adding $T_R\{\rho(H)(-XAHB + HBXA + HAXB - BHAX + XAHB - HBXA - HAXB + BHAX)\}$, which is null, to equation (2.3) and rearranging the terms as follows:

$$T_R\{\rho(H)[[LX, A], B]\} = -T_R\{\rho(H)[[X, LA], B]\} - T_R\{\rho(H)[[X, A], LB]\}. \tag{2.4}$$

Therefore, the general form of the reduction identity can be obtained as (appendix A)

$$\begin{aligned} T_R\{\rho(H)[\cdots[[LX, A_1], A_2], \cdots, A_n]\} &= -T_R\{\rho(H)[\cdots[[X, LA_1], A_2], \cdots, A_n]\} \\ &\quad - T_R\{\rho(H)[\cdots[[X, A_1], LA_2], \cdots, A_n]\} \\ &\quad \cdots - T_R\{\rho(H)[\cdots[[X, A_1], A_2], \cdots, LA_n]\}. \end{aligned} \tag{2.5}$$

Hereafter, this identity is called the KC reduction identity to be discerned from the other forms and will be used to derive the nonlinear optical conductivity.

## 3. Ensemble average of the current density operator

This paper considers the electron–phonon system, in which the total Hamiltonian $H(t)$ and the corresponding Liouville operator $L(t)$ can be split into two parts, respectively, as follows:

$$H(t) = H_{eq} + H_{int}(t), \tag{3.1}$$

$$L(t) = L_{eq} + L_{int}(t). \tag{3.2}$$

Here $H_{eq}$ is the Hamiltonian of electrons interacting with phonons in thermodynamic equilibrium, which is given by

$$H_{eq} = H_d + V = \sum_\alpha E_\alpha a_\alpha^+ a_\alpha + \sum_q \hbar\omega_q b_q^+ b_q + \sum_q \sum_{\alpha,\mu} C_{\alpha,\mu}(q) a_\alpha^+ a_\mu (b_q + b_{-q}^+), \tag{3.3}$$

where $a_\alpha^+(a_\alpha)$ and $b_q^+(b_q)$ denote the creation (annihilation) operator for an electron in the $|\alpha\rangle$ state with energy $E_\alpha$ and the creation (annihilation) operator for a phonon in the $|q\rangle$ state, respectively; $|q\rangle \equiv |\mathbf{q}, s\rangle$; $\mathbf{q}$ is the phonon wave vector and $s$ is the polarization index; $\hbar\omega_q$ is the phonon energy and $C_{\alpha,\mu}(q)$ is the electron–phonon interaction matrix element. In equation (3.1), the interaction term with the time-dependent external electric field of amplitude $F_j$ and angular frequency $\omega$, $H_{int}(t)$, is given by

$$H_{int}(t) = e \sum_{j=1,2,3} \sum_{\alpha,\beta} (r_j)_{\alpha\beta} a_\alpha^+ a_\beta F_j \exp(i\omega t) + \text{c.c.}, \tag{3.4}$$

where $r_1$, $r_2$ and $r_3$ denote the $x$, $y$ and $z$ components of the electron position vectors, respectively, $(X)_{\alpha\beta} \equiv \langle\alpha|X|\beta\rangle$ for an arbitrary operator $X$, and c.c. means the complex conjugate.

On the other hand, the total density operator $\rho(t)$ follows the Liouville equation as follows:

$$i\hbar \frac{\partial \rho(t)}{\partial t} = L(t)\rho(t). \tag{3.5}$$

A formula for the nonlinear optical conductivity can be derived by expanding it as $\rho(t) = \rho_{eq} + \rho_{int}(t)$, where $\rho_{eq} \equiv \rho(H_{eq})$ is the density operator for a system in thermal equilibrium and $\rho_{int}(t)$ is the perturbed term by the time-dependent external field. Inserting $\rho(t)$ into equation (3.5), we obtain (see [33] for details)

$$\begin{aligned} \rho_{int}(t) &= \sum_{n=1}^\infty \frac{1}{(i\hbar)^n} \int_0^\infty dt_1 \int_0^\infty dt_2 \cdots \int_0^\infty dt_n \exp(-iL_{eq}t_1/\hbar) L_{int}(t - t_1) \exp(-iL_{eq}t_2/\hbar) \\ &\quad \times L_{int}(t - t_1 - t_2) \cdots \exp(-iL_{eq}t_n/\hbar) L_{int}(t - t_1 - \cdots - t_n)\rho_{eq} \\ &\equiv \rho^{(1)}(t) + \rho^{(2)}(t) + \cdots + \rho^{(\infty)}(t), \end{aligned} \tag{3.6}$$

where $\rho^{(n)}(t)$ involves $L_{int}(t)$ $n$ times.

Using equation (3.6), the ensemble average of the $i$th component of the current density operator **J** is given by

$$\langle J_i \rangle_{\text{ens}} = \sum_{n=1}^{\infty} \langle J_i^{(n)} \rangle_{\text{ens}} = \sum_{n=1}^{\infty} T_R \{ \rho^{(n)}(t) J_i \}, \tag{3.7}$$

where $i \equiv x, y, z$ and the many-electron current operator $J_i$ can be written in terms of the single-electron current operator $j_z = (\text{i}e\hbar/m_c)\partial/\partial z$ as $J_i = \sum_{\alpha\beta}(j_i)_{\alpha\beta}a_\alpha^+ a_\beta$, where $m_c$ is the effective mass of the electron.

## 4. Linear optical conductivity

The first-order term of $\langle J_i \rangle_{\text{ens}}$ in equation (3.7) is given by

$$\langle J_i^{(1)} \rangle_{\text{ens}} \equiv \sum_j \sigma_{ij}(\omega) F_j(\omega), \tag{4.1}$$

where the linear term of the optical conductivity tensor for the incident wave of a frequency $\omega$, $\sigma_{ij}(\omega)$, is defined as

$$\sigma_{ij}(\omega) \equiv e \lim_{s \to 0^+} \sum_{\delta_1,\delta_2} \sum_{\alpha_1,\alpha_2} (j_i)_{\delta_1\delta_2}(r_j)_{\alpha_1,\alpha_2} A_{\alpha_1\alpha_2}^{(0)\delta_1\delta_2}(\bar{\omega}). \tag{4.2}$$

Here,

$$A_{\alpha_1\alpha_2}^{(0)\delta_1\delta_2}(\bar{\omega}) \equiv T_R \{ \rho_{\text{eq}} [ K(\bar{\omega}) a_{\delta_1}^+ a_{\delta_2}, a_{\alpha_1}^+ a_{\alpha_2} ] \}, \tag{4.3}$$

where $\bar{\omega} \equiv \omega + \text{i}s (s \to 0^+)$ and $K(\omega) \equiv (\hbar\bar{\omega} - L_{\text{eq}})^{-1}$.

In order to calculate $A_{\alpha_1,\alpha_2}^{(0)\delta_1,\delta_2}(\bar{\omega})$, the zeroth state-dependent projectors, $P_0$ and $Q_0$, for an arbitrary operator, $X$, can be defined as follows:

$$P_0 X \equiv \frac{\langle X \rangle_\alpha}{\langle a_{\delta_1}^+ a_{\delta_2} \rangle_\alpha} a_{\delta_1}^+ a_{\delta_2}, \qquad Q_0 \equiv 1 - P_0, \tag{4.4}$$

where

$$\langle X \rangle_\alpha \equiv T_R \{ \rho_{\text{eq}} [ X, a_{\alpha_1}^+ a_{\alpha_2} ] \}. \tag{4.5}$$

In equations (4.4) and (4.5), the projection operators are state dependent, i.e. these operators project an arbitrary operator, $X$, into the operator $a_{\delta_1}^+ a_{\delta_2}$, which depends on the states $\delta_1$ and $\delta_2$. On the other hand, state-independent projection operators were used in a previous paper [33], i.e. those operators project $X$ into the state-independent current density operator, $J_k$.

The previous result [33] is applicable only to the case in which $L_d J_k = c$-number $\times J_k$ is satisfied or $Q_0 L_d J_k = 0$ where $J_k$ is the many-electron current operator. The cyclotron phenomenon belongs to this category because for the current operator $J_\pm = J_x \pm \text{i}J_y$, we have $L_d J_+ = \hbar\omega_c J_+$, where $\omega_c$ is the resonance frequency. In this paper, however, the above criterion is always satisfied because $L_d a_\alpha^+ a_\delta = (E_\alpha - E_\delta) a_\alpha^+ a_\delta$ or $Q L_d a_\alpha^+ a_\delta = 0$, so the present method is more general and the condition that $Q_0 L_d J_k = 0$ in [33] has nothing to do with the present result. Since the derivation of higher order terms is more general, we will show the detailed procedure of utilizing the reduction identity in section 6 only, in which the second-order nonlinear term is calculated. Thus, here we will not show the detailed calculations in order to save space.

In equation (4.3), applying the identity $1 = P_0 + Q_0$ on the right-hand side of the Liouville operator, $L_{\text{eq}}$, in $K(\omega)$ as $L_{\text{eq}} = L_{\text{eq}}(P_0 + Q_0)$, considering $L_{\text{eq}}$ as $L_{\text{eq}} = L_d + L_v$, where $L_d$

and $L_v$ are the Liouville operators corresponding to the Hamiltonian $H_d$ (diagonal part) and $V$ (nondiagonal part), respectively, and using the identity

$$\frac{1}{A-B} = \frac{1}{A} - \frac{1}{A}B\frac{1}{A-B}, \tag{4.6}$$

we obtain, after systematic calculations as in [33],

$$\sigma_{ij}(\omega) = e \sum_{\delta_1,\delta_2} \frac{(j_i)_{\delta_1\delta_2}(r_j)_{\delta_2\delta_1}(f_{\delta_1} - f_{\delta_2})}{\hbar\bar{\omega} - E_{\delta_1\delta_2} - \Gamma^{(0)}_{\delta_2\delta_1}(\bar{\omega})}, \tag{4.7}$$

where $f_i$ is the Fermi distribution function for an electron with energy $E_i$. In equation (4.7), the real parts of the self-energy (Lamb frequency shift) are not included because we consider a very weak scattering. Note that those parts are quite small in comparison with $E_{\delta_1} - E_{\delta_2}$ in the quantum limit. In equation (4.7), the zeroth-order linewidth (or damping term), $\Gamma^{(0)}_{\delta_1\delta_2}$, is given by the following:

$$\begin{aligned}
\Gamma^{(0)}_{\delta_1\delta_2}(\bar{\omega})(f_{\delta_1} - f_{\delta_2}) = \sum_q \sum_\lambda \Big[ & \left|C_{\delta_2\lambda}(q)\right|^2 G^{(+)}_{\delta_1\lambda}(\bar{\omega})\{W_+(\delta_1,\lambda) - W_-(\lambda,\delta_1)\} \\
& - \left|C_{\delta_2\lambda}(q)\right|^2 G^{(-)}_{\delta_1\lambda}(\bar{\omega})\{W_+(\lambda,\delta_1) - W_-(\delta_1,\lambda)\} \\
& + \left|C_{\lambda\delta_1}(q)\right|^2 G^{(+)}_{\lambda\delta_2}(\bar{\omega})\{W_+(\lambda,\delta_2) - W_-(\delta_2,\lambda)\} \\
& - \left|C_{\lambda\delta_1}(q)\right|^2 G^{(-)}_{\lambda\delta_2}(\bar{\omega})\{W_+(\delta_2,\lambda) - W_-(\lambda,\delta_2)\}\Big]. 
\end{aligned} \tag{4.8}$$

Here

$$G^{(\pm)}_{\alpha\beta}(\bar{\omega}) \equiv (\hbar\bar{\omega} - E_{\alpha\beta} \pm \hbar\omega_q)^{-1} \tag{4.9}$$

and

$$W_\pm(\alpha,\beta) \equiv (N_q + 1/2 \pm 1/2)f_\alpha(1 - f_\beta) \tag{4.10}$$

where $N_q$ is the Planck distribution function for a phonon with energy $\hbar\omega_q$. The physical meaning of equation (4.8) is clear. For example, the first term represents the transition of the electron from states $\delta_1$ to $\lambda$ with phonon emission. Here $1 + N_q$ is the condition for phonon emission, and $f_{\delta_1}(1 - f_\lambda)$ is the condition for the transition $\delta_1 \to \lambda$. The denominator enforces energy conservation in the transition, i.e. $E_{\delta_1} = E_\lambda + \hbar\omega_q + \hbar\bar{\omega}$. The other seven terms in equation (4.8) can be explained in a similar manner. The linear part given by equation (4.8) has been reported to give a good interpretation of the intraband transition in a two-dimensional electron system [14].

According to the Pauli exclusion principle, the population of electrons (fermions) and phonons (bobons) should appear independently. Thus, the Fermi functions should be multiplied by the Planck distribution functions in the formalism. Some other results [7, 9, 23] with a sum of two functions such as $(N_q + 1/2 \pm 1/2 \mp f_\gamma)$ fail to meet this criterion. This problem can be avoided with proper use of the reduction identity.

## 5. First-order nonlinear optical conductivity

To derive the first-order nonlinear optical conductivity formula, we consider the second-order term of $\langle J_i \rangle_{\text{ens}}$ in equation (3.7) given by

$$\langle J_i^{(2)} \rangle_{\text{ens}} \equiv \sum_{j,k} \sigma_{ijk}(\omega_{12})F_j(\omega_1)F_k(\omega_2), \tag{5.1}$$

where the first-order nonlinear optical conductivity tensor for the incident waves of frequencies $\omega_1$ and $\omega_2$, $\sigma_{ijk}(\omega_1, \omega_2)$, is defined as

$$\sigma_{ijk}(\omega_{12}) \equiv e^2 \lim_{s \to 0^+} \sum_{\delta_1, \delta_2} \sum_{\alpha_1, \alpha_2} \sum_{\beta_1, \beta_2} (j_i)_{\delta_1 \delta_2} (r_j)_{\alpha_1 \alpha_2} (r_k)_{\beta_1 \beta_2} A^{(1)\delta_1 \delta_2}_{\alpha_1 \alpha_2 \beta_1 \beta_2}(\bar{\omega}_{12}). \quad (5.2)$$

Here

$$A^{(1)\delta_1 \delta_2}_{\alpha_1 \alpha_2 \beta_1 \beta_2}(\bar{\omega}_{12}) \equiv T_R \{ \rho_{eq} [ K(\omega_1) [ K(\omega_{12}) a^+_{\delta_1} a_{\delta_2}, a^+_{\alpha_1} a_{\alpha_2} ], a^+_{\beta_1} a_{\beta_2} ] \}, \quad (5.3)$$

where $\bar{\omega}_i \equiv \omega_i + is$ and $\omega_{12} \equiv \omega_1 + \omega_2$.

Equation (5.3) is calculated by defining the first state-dependent projectors, $P_1$ and $Q_1$, for an arbitrary operator, $X$, as follows:

$$P_1 X \equiv \frac{\langle X \rangle_{\alpha \beta}}{\langle a^+_{\delta_1} a_{\delta_2} \rangle_{\alpha \beta}} a^+_{\delta_1} a_{\delta_2}, \qquad Q_1 \equiv 1 - P_1, \quad (5.4)$$

where

$$\langle X \rangle_{\alpha \beta} \equiv T_R \{ \rho_{eq} [ K(\omega_1) [ X, a^+_{\alpha_1} a_{\alpha_2} ], a^+_{\beta_1} a_{\beta_2} ] \}. \quad (5.5)$$

The present projection method for the first-order nonlinear optical conductivity is more general than that of [33] by the same reason as in the linear case. In equation (5.3), applying the identity $1 = P_1 + Q_1$ on the right-hand side of the Liouville operator $L_{eq}$ in $K(\bar{\omega}_{12})$ as $(\hbar \bar{\omega}_{12} - L_{eq})^{-1} = (\hbar \bar{\omega}_{12} - L_{eq} P_1 - L_{eq} Q_1)^{-1}$ and using equation (4.6), we obtain, after systematic calculations as in [33],

$$
\begin{aligned}
A^{(0)\delta_1 \delta_2}_{\alpha_1 \alpha_2 \beta_1 \beta_2}(\bar{\omega}_{12}) = {} & \frac{(f_{\delta_1} - f_{\alpha_2}) \delta_{\delta_1 \beta_2} \delta_{\alpha_2 \beta_1}}{\hbar \bar{\omega}_1 - E_{\delta_1 \alpha_2} - \Gamma^{(0)}_{\delta_1 \alpha_2}(\bar{\omega}_1)} \frac{\delta_{\delta_2 \alpha_1}}{\hbar \bar{\omega}_{12} - E_{\delta_1 \delta_2} - \Gamma^{(11)}_{\alpha_2 \delta_1 \delta_2}(\bar{\omega}_{12})} \\
& - \frac{(f_{\alpha_1} - f_{\delta_2}) \delta_{\delta_2 \beta_1} \delta_{\alpha_1 \beta_2}}{\hbar \bar{\omega}_1 - E_{\alpha_1 \delta_2} - \Gamma^{(0)}_{\alpha_1 \delta_2}(\bar{\omega}_1)} \frac{\delta_{\delta_1 \alpha_2}}{\hbar \bar{\omega}_{12} - E_{\delta_1 \delta_2} - \Gamma^{(12)}_{\alpha_1 \delta_1 \delta_2}(\bar{\omega}_{12})},
\end{aligned} \quad (5.6)
$$

where

$$
\begin{aligned}
\Gamma^{(11)}_{\alpha_2 \delta_1 \delta_2}(\bar{\omega}_{12})(f_{\alpha_2} - f_{\delta_1}) & \equiv V^{(11)}_{\alpha_1 \alpha_2 \beta_1 \beta_2}(\delta_1 \delta_2; \bar{\omega}_{12})(f_{\alpha_2} - f_{\delta_1}) \\
& = \sum_q \sum_\lambda \big[ |C_{\lambda \delta_1}(q)|^2 G^{(+)}_{\lambda \alpha_2}(\bar{\omega}_1) \{ W_+(\lambda, \alpha_2) - W_-(\alpha_2, \lambda) \} \\
& \quad - |C_{\lambda \delta_1}(q)|^2 G^{(-)}_{\lambda \alpha_2}(\bar{\omega}_1) \{ W_+(\alpha_2, \lambda) - W_-(\lambda, \alpha_2) \} \\
& \quad + |C_{\delta_2 \lambda}(q)|^2 G^{(+)}_{\delta_1 \lambda}(\bar{\omega}_1) \{ W_+(\delta_2, \lambda) - W_-(\lambda, \delta_2) - W_+(\lambda, \delta_2) \\
& \quad + W_-(\delta_2, \lambda) \} - |C_{\delta_2 \lambda}(q)|^2 G^{(-)}_{\delta_1 \lambda}(\bar{\omega}_1) \{ W_+(\lambda, \delta_1) \\
& \quad - W_-(\delta_1, \lambda) - W_+(\delta_1, \lambda) + W_+(\lambda, \delta_1) \} \big],
\end{aligned} \quad (5.7)
$$

$$
\begin{aligned}
\Gamma^{(12)}_{\alpha_1 \delta_1 \delta_2}(\bar{\omega}_{12})(f_{\alpha_1} - f_{\delta_2}) & \equiv V^{(12)}_{\alpha_1 \alpha_2 \beta_1 \beta_2}(\delta_1 \delta_2; \bar{\omega}_{12})(f_{\alpha_1} - f_{\delta_2}) \\
& = \sum_q \sum_\lambda \big[ |C_{\delta_2 \lambda}(q)|^2 G^{(+)}_{\alpha_1 \lambda}(\bar{\omega}_1) \{ W_+(\alpha_1, \lambda) - W_-(\lambda, \alpha_1) \} \\
& \quad - |C_{\delta_2 \lambda}(q)|^2 G^{(-)}_{\alpha_1 \lambda}(\bar{\omega}_1) \{ W_+(\lambda, \alpha_1) - W-(\alpha_1, \lambda) \} \\
& \quad + |C_{\lambda \delta_1}(q)|^2 G^{(+)}_{\lambda \delta_2}(\bar{\omega}_1) \{ W_+(\lambda, \delta_2) - W_-(\delta_2, \lambda) - W_+(\lambda, \alpha_1) \\
& \quad + W_-(\alpha_1, \lambda) \} - |C_{\lambda \delta_1}(q)|^2 G^{(-)}_{\lambda \delta_2}(\bar{\omega}_1) \{ W_+(\delta_2, \lambda) - W_-(\lambda, \delta_2) \\
& \quad - W_+(\alpha_1, \lambda) + W_-(\lambda, \alpha_1) \} \big].
\end{aligned} \quad (5.8)
$$

For the detailed calculations, see section 6 in which the second nonlinear part is dealt with. In equations (5.7) and (5.8), the results near the resonance points, $\hbar\omega_{12} \approx E_{\beta_2\beta_1}$, were considered, and we have ignored the terms including $C_{\alpha\alpha}(q)$ and $(\hbar\omega \pm \hbar\omega_q)^{-1}$ because we are interested in the resonance phenomena for the electron system interacting with phonons. The present results (equations (5.6)–(5.8)) are similar to those reported by Suzuki and Ashikawa [29]. However, the present results are different because they include the electron and phonon distribution functions differently. Note that the result given in [29] includes the sum of the two distribution functions. Therefore, the present results can offer a more reasonable interpretation, such as in the linear part.

## 6. Second-order nonlinear optical conductivity

The second-order nonlinear optical conductivity is derived in this section. For that purpose, we consider the third-order term of $\langle J_i \rangle_{\text{ens}}$ in equation (3.7), which is given by

$$\left\langle J_i^{(3)} \right\rangle_{\text{ens}} \equiv \sum_{j,k,l} \sigma_{ijkl}(\omega_{123}) F_j(\omega_1) F_k(\omega_2) F_l(\omega_3). \tag{6.1}$$

Here the second-order nonlinear optical conductivity tensor for the incident waves of frequencies $\omega_1$, $\omega_2$ and $\omega_3$, $\sigma_{ijkl}(\omega_1, \omega_2, \omega_3)$, is defined as follows:

$$\sigma_{ijkl}(\omega_{123}) \equiv e^3 \lim_{s \to 0^+} \sum_{\delta_1,\delta_2} \sum_{\alpha_1,\alpha_2} \sum_{\beta_1,\beta_2} \sum_{\gamma_1,\gamma_2} (j_i)_{\delta_1\delta_2} (r_j)_{\alpha_1\alpha_2} (r_k)_{\beta_1\beta_2} (r_l)_{\gamma_1\gamma_2} A^{(2)}(\bar{\omega}_{123}), \tag{6.2}$$

where $\omega_{123} \equiv \omega_1 + \omega_2 + \omega_3$ and

$$A^{(2)}(\bar{\omega}_{123}) \equiv T_R\left\{ \rho_{\text{eq}}\left[ K(\omega_1)\left[ K(\omega_{12})\left[ K(\omega_{123}) a_{\delta_1}^+ a_{\delta_2}, a_{\alpha_1}^+ a_{\alpha_2} \right], a_{\beta_1}^+ a_{\beta_2} \right], a_{\gamma_1}^+ a_{\gamma_2} \right] \right\}. \tag{6.3}$$

In order to calculate equation (6.3), the second state-dependent projectors $P_2$ and $Q_2$ for an arbitrary operator $X$ are defined as follows:

$$P_2 X \equiv \frac{\langle X \rangle_{\alpha\beta\gamma}}{\langle a_{\delta_1}^+ a_{\delta_2} \rangle_{\alpha\beta\gamma}} a_{\delta_1}^+ a_{\delta_2}, \qquad Q_2 \equiv 1 - P_2, \tag{6.4}$$

where

$$\langle X \rangle_{\alpha\beta\gamma} \equiv T_R\left\{ \rho_{\text{eq}}\left[ K(\omega_1)\left[ K(\omega_{12})\left[ X, a_{\alpha_1}^+ a_{\alpha_2} \right], a_{\beta_1}^+ a_{\beta_2} \right], a_{\gamma_1}^+ a_{\gamma_2} \right] \right\}. \tag{6.5}$$

Applying the identity $1 = P_2 + Q_2$ on the right-hand side of the Liouville operator $L_{\text{eq}}$ in equation (6.3) as $(\hbar\bar{\omega}_{123} - L_{\text{eq}})^{-1} = (\hbar\bar{\omega}_{123} - L_{\text{eq}}P_2 - L_{\text{eq}}Q_2)^{-1}$ and using equation (4.6), we obtain

$$A^{(2)}(\bar{\omega}_{123}) = \frac{\langle a_{\delta_1}^+ a_{\delta_2} \rangle_{\alpha\beta\gamma}}{\hbar\bar{\omega}_{123} - E_{\delta_1\delta_2} - B_2(\bar{\omega}_{123})}, \tag{6.6}$$

where

$$B_2(\bar{\omega}_{123}) \equiv \frac{\hbar\bar{\omega}_{123}}{\langle a_{\delta_1}^+ a_{\delta_2} \rangle_{\alpha\beta\gamma}} \langle K^{(2)}(\bar{\omega}_{123}) L_v a_{\delta_1}^+ a_{\delta_2} \rangle_{\alpha\beta\gamma}. \tag{6.7}$$

In equations (6.6) and (6.7), $\langle a_{\delta_1}^+ a_{\delta_2} \rangle_{\alpha\beta\gamma}$ can be calculated as follows:

$$\langle a_{\delta_1}^+ a_{\delta_2} \rangle_{\alpha\beta\gamma} = A^{(0)\delta_1\alpha_2}_{\beta_1\beta_2\gamma_1\gamma_2}(\bar{\omega}_{12})\delta_{\delta_2\alpha_1} - A^{(0)\alpha_1\delta_2}_{\beta_1\beta_2\gamma_1\gamma_2}(\bar{\omega}_{12})\delta_{\delta_1\alpha_2}. \tag{6.8}$$

Equation (6.7) is calculated further by applying equation (4.6) to $(\hbar\bar{\omega}_{123} - L_{\text{eq}}Q_2)^{-1}$ in $K^{(2)}(\bar{\omega}_{123})$ with $A \equiv \hbar\bar{\omega}_{123}$ and $B \equiv L_{\text{eq}}Q_2$, having

$$B_2(\bar{\omega}_{123}) \langle a_{\delta_1}^+ a_{\delta_2} \rangle_{\alpha\beta\gamma} = D_2(\bar{\omega}_{123}) + E_2(\bar{\omega}_{123}), \tag{6.9}$$

where

$$D_2(\bar{\omega}_{12}) \equiv T_R\{\rho_{eq}[K(\bar{\omega}_1)[K(\bar{\omega}_{12})[L_v a_{\delta_1}^+ a_{\delta_2}, a_{\alpha_1}^+ a_{\alpha_2}], a_{\beta_1}^+ a_{\beta_2}], a_{\gamma_1}^+ a_{\gamma_2}]\}, \tag{6.10}$$

$$E_2(\bar{\omega}_{123}) \equiv T_R\{\rho_{eq}[K(\bar{\omega}_1)[K(\bar{\omega}_{12})[L_{eq}Q_2 K^{(2)}(\bar{\omega}_{123})L_v a_{\delta_1}^+ a_{\delta_2}, a_{\alpha_1}^+ a_{\alpha_2}], a_{\beta_1}^+ a_{\beta_2}], a_{\gamma_1}^+ a_{\gamma_2}]\}. \tag{6.11}$$

Therefore, after some systematic calculations, we obtain (appendix B)

$$B_2(\bar{\omega}_{123})\langle a_{\delta_1}^+ a_{\delta_2}\rangle_{\alpha\beta\gamma} = \frac{-V_{21}/(\hbar\bar{\omega}_1 + E_{\gamma_1\gamma_2}) - V_{22} - V_{23}}{\hbar\bar{\omega}_{12} + E_{\beta_1\beta_2} + E_{\gamma_1\gamma_2}} - V_{24} - V_{25} - V_{26}, \tag{6.12}$$

where

$$V_{21} \equiv T_R\{\rho_{eq}[K^{(d)}(\bar{\omega}_1)[[L_v a_{\delta_1}^+ a_{\delta_2}, a_{\alpha_1}^+ a_{\alpha_2}], a_{\beta_1}^+ a_{\beta_2}], L_v a_{\gamma_1}^+ a_{\gamma_2}]\}, \tag{6.13}$$

$$V_{22} \equiv T_R\{\rho_{eq}[K^{(d)}(\bar{\omega}_1)[K^{(d)}(\bar{\omega}_{12})[L_v a_{\delta_1}^+ a_{\delta_2}, a_{\alpha_1}^+ a_{\alpha_2}], L_v a_{\beta_1}^+ a_{\beta_2}], a_{\gamma_1}^+ a_{\gamma_2}]\}, \tag{6.14}$$

$$V_{23} \equiv T_R\{\rho_{eq}[K^{(d)}(\bar{\omega}_1)[K^{(d)}(\bar{\omega}_{12})[L_v a_{\delta_1}^+ a_{\delta_2}, a_{\alpha_1}^+ a_{\alpha_2}], a_{\beta_1}^+ a_{\beta_2}], L_v a_{\gamma_1}^+ a_{\gamma_2}]\}, \tag{6.15}$$

$$V_{24} \equiv T_R\{\rho_{eq}[K^{(d)}(\bar{\omega}_1)[K^{(d)}(\bar{\omega}_{12})[K^{(d)}(\bar{\omega}_{123})L_v a_{\delta_1}^+ a_{\delta_2}, L_v a_{\alpha_1}^+ a_{\alpha_2}], a_{\beta_1}^+ a_{\beta_2}], a_{\gamma_1}^+ a_{\gamma_2}]\}, \tag{6.16}$$

$$V_{25} \equiv T_R\{\rho_{eq}[K^{(d)}(\bar{\omega}_1)[K^{(d)}(\bar{\omega}_{12})[K^{(d)}(\bar{\omega}_{123})L_v a_{\delta_1}^+ a_{\delta_2}, a_{\alpha_1}^+ a_{\alpha_2}], L_v a_{\beta_1}^+ a_{\beta_2}], a_{\gamma_1}^+ a_{\gamma_2}]\}, \tag{6.17}$$

$$V_{26} \equiv T_R\{\rho[K^{(d)}(\bar{\omega}_1)[K^{(d)}(\bar{\omega}_{12})[K^{(d)}(\bar{\omega}_{123})L_v a_{\delta_1}^+ a_{\delta_2}, a_{\alpha_1}^+ a_{\alpha_2}], a_{\beta_1}^+ a_{\beta_2}], L_v a_{\gamma_1}^+ a_{\gamma_2}]\}, \tag{6.18}$$

which are all calculable forms. Inserting equations (6.8) and (6.12)–(6.18) into equation (6.6) and considering equation (5.6) gives

$$A^{(2)}(\bar{\omega}_{123})$$

$$= \frac{(f_{\delta_1} - f_{\beta_2})\delta_{\delta_1\gamma_2}\delta_{\beta_2\gamma_1}}{\hbar\bar{\omega}_1 - E_{\delta_1\beta_2} - \Gamma_{\delta_1\beta_2}^{(0)}(\bar{\omega}_1)} \frac{\delta_{\alpha_2\beta_1}}{\hbar\bar{\omega}_{12} - E_{\delta_1\alpha_2} - \Gamma_{\beta_2\delta_1\alpha_2}^{(11)}(\bar{\omega}_{12})} \frac{\delta_{\delta_2\alpha_1}}{\hbar\bar{\omega}_{123} - E_{\delta_1\delta_2} - \Gamma_{\beta\delta_1\delta_2}^{(21)}(\bar{\omega}_{123})}$$

$$- \frac{(f_{\beta_1} - f_{\alpha_2})\delta_{\alpha_2\gamma_1}\delta_{\beta_1\gamma_2}}{\hbar\bar{\omega}_1 - E_{\beta_1\alpha_2} - \Gamma_{\beta_1\alpha_2}^{(0)}(\bar{\omega}_1)} \frac{\delta_{\delta_1\beta_2}}{\hbar\bar{\omega}_{12} - E_{\delta_1\alpha_2} - \Gamma_{\beta_1\delta_1\alpha_2}^{(12)}(\bar{\omega}_{12})} \frac{\delta_{\delta_2\alpha_1}}{\hbar\bar{\omega}_{123} - E_{\delta_1\delta_2} - \Gamma_{\beta\alpha_2\delta_1\delta_2}^{(22)}(\bar{\omega}_{123})}$$

$$- \frac{(f_{\alpha_1} - f_{\beta_2})\delta_{\alpha_1\gamma_2}\delta_{\beta_2\gamma_1}}{\hbar\bar{\omega}_1 - E_{\alpha_1\beta_2} - \Gamma_{\alpha_1\beta_2}^{(0)}(\bar{\omega}_1)} \frac{\delta_{\delta_2\beta_1}}{\hbar\bar{\omega}_{12} - E_{\alpha_1\delta_2} - \Gamma_{\beta_2\alpha_1\delta_2}^{(11)}(\bar{\omega}_{12})} \frac{\delta_{\delta_1\alpha_2}}{\hbar\bar{\omega}_{123} - E_{\delta_1\delta_2} - \Gamma_{\alpha_1\beta_2\delta_1\delta_2}^{(23)}(\bar{\omega}_{123})}$$

$$+ \frac{(f_{\beta_1} - f_{\delta_2})\delta_{\delta_2\gamma_1}\delta_{\beta_1\gamma_2}}{\hbar\bar{\omega}_1 - E_{\beta_1\delta_2} - \Gamma_{\beta_1\delta_2}^{(0)}(\bar{\omega}_1)} \frac{\delta_{\alpha_1\beta_2}}{\hbar\bar{\omega}_{12} - E_{\alpha_1\delta_2} - \Gamma_{\beta_1\alpha_1\delta_2}^{(12)}(\bar{\omega}_{12})} \frac{\delta_{\delta_1\alpha_2}}{\hbar\bar{\omega}_{123} - E_{\delta_1\delta_2} - \Gamma_{\beta_1\delta_1\delta_2}^{(24)}(\bar{\omega}_{123})}. \tag{6.19}$$

Here

$$\Gamma_{\beta_2\delta_1\delta_2}^{(21)}(\bar{\omega}_{123})(f_{\beta_2} - f_{\delta_1}) = \sum_q \sum_\lambda [|C_{\lambda,\delta_1}(q)|^2 G_{\lambda\beta_2}^{(+)}(\bar{\omega}_1)\{W_+(\lambda,\beta_2) - W_-(\beta_2,\lambda)\}$$

$$- |C_{\lambda,\delta_1}(q)|^2 G_{\lambda\beta_2}^{(-)}(\bar{\omega}_1)\{W+(\beta_2,\lambda) - W_-(\lambda,\beta_2)\}$$

$$+ |C_{\delta_2,\lambda}(q)|^2 G_{\delta_1\lambda}^{(+)}(\bar{\omega}_{123})\{W_+(\beta_2,\lambda) - W-(\lambda,\beta_2) - W_+(\delta_1,\lambda) + W-(\lambda,\delta_1)\}$$

$$- |C_{\delta_2,\lambda}(q)|^2 G_{\delta_1\lambda}^{(-)}(\bar{\omega}_{123})\{W_+(\lambda,\beta_2) - W_-(\beta_2,\lambda) - W_+(\lambda,\delta_2) + W_-(\delta_1,\lambda)\}], \tag{6.20}$$

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$$\Gamma^{(22)}_{\beta_1\alpha_2\delta_1\delta_2}(\bar{\omega}_{123})\big(f_{\alpha_2} - f_{\beta_1}\big)$$

$$= \sum_{q}\sum_{\lambda}\big[\,\big|C_{\lambda,\delta_1}(q)\big|^2 G^{(+)}_{\lambda\delta_2}(\bar{\omega}_{12})\{W_+(\lambda, \beta_1) - W_-(\beta_1, \lambda) - W_+(\lambda, \gamma_1)$$

$$+ W_-(\gamma_1, \lambda)\} - \big|C_{\lambda,\delta_1}(q)\big|^2 G^{(-)}_{\lambda\delta_2}(\bar{\omega}_{12})\{W_+(\beta_1, \lambda) - W_-(\lambda, \beta_1) - W_+(\gamma_1, \lambda)$$

$$+ W_-(\lambda, \gamma_1)\} + \big|C_{\delta_2,\lambda}(q)\big|^2 G^{(+)}_{\delta_1\lambda}(\bar{\omega}_{123})\{W_+(\beta_1, \lambda) - W_-(\lambda, \beta_1) - W_+(\gamma_1, \lambda)$$

$$+ W_-(\lambda, \gamma_1)\} - \big|C_{\delta_2,\lambda}(q)\big|^2 G^{(-)}_{\delta_1\lambda}(\bar{\omega}_{123})\{W_+(\lambda, \beta_1) - W_-(\beta_1, \lambda)$$

$$- W_+(\lambda, \gamma_1) + W_-(\gamma_1, \lambda)\}\big], \tag{6.21}$$

$$\Gamma^{(23)}_{\alpha_1\beta_2\delta_1\delta_2}(\bar{\omega}_{123})\big(f_{\beta_2} - f_{\alpha_1}\big)$$

$$= \sum_{q}\sum_{\lambda}\big[\,\big|C_{\delta_2,\lambda}(q)\big|^2 G^{(+)}_{\delta_1\lambda}(\bar{\omega}_{12})\{W_+(\gamma_1, \lambda) - W_-(\lambda, \gamma_1) - W_+(\alpha_1, \lambda)$$

$$+ W_-(\lambda, \alpha_1)\} - \big|C_{\delta_2,\lambda}(q)\big|^2 G^{(-)}_{\delta_1\lambda}(\bar{\omega}_{12})\{W_+(\lambda, \gamma_1) - W_-(\gamma_1, \lambda) - W_+(\lambda, \alpha_1)$$

$$+ W_-(\alpha_1, \lambda)\} + \big|C_{\lambda,\delta_1}(q)\big|^2 G^{(+)}_{\lambda\delta_2}(\bar{\omega}_{123})\{W_+(\lambda, \alpha_1) - W_-(\alpha_1, \lambda) - W_-(\lambda, \gamma_1)$$

$$+ W_-(\gamma_1, \lambda)\} - \big|C_{\lambda_1,\delta_1}(q)\big|^2 G^{(-)}_{\lambda\delta_2}(\bar{\omega}_{123})\{W_+(\alpha_1, \lambda) - W_-(\lambda, \alpha_1)$$

$$- W_+(\gamma_1, \lambda) + W_-(\lambda, \gamma_1)\}\big], \tag{6.22}$$

$$\Gamma^{(24)}_{\beta_1\delta_1\delta_2}(\bar{\omega}_{123})(f_{\delta_2} - f_{\beta_1})$$

$$= \sum_{q}\sum_{\lambda}\big[\,\big|C_{\delta_2,\lambda}(q)\big|^2 G^{(+)}_{\beta_1\lambda}(\bar{\omega}_1)\{W_+(\beta_1, \lambda) - W_-(\lambda, \beta_1)\}$$

$$- \big|C_{\delta_2,\lambda}(q)\big|^2 G^{(-)}_{\beta_1\lambda}(\bar{\omega}_1)\{W_+(\lambda, \beta_1) - W_-(\beta_1, \lambda)\}$$

$$+ \big|C_{\lambda,\delta_1}(q)\big|^2 G^{(+)}_{\lambda\delta_2}(\bar{\omega}_{123})\{W_+(\lambda, \gamma_1) - W_-(\gamma_1, \lambda) - W_+(\lambda, \beta_1) + W_-(\beta_1, \lambda)\}$$

$$- \big|C_{\lambda_1,\delta_1}(q)\big|^2 G^{(-)}_{\lambda\delta_2}(\bar{\omega}_{123})\{W_+(\gamma_1, \lambda) - W_-(\lambda, \gamma_1) - W_+(\beta_1, \lambda) + W_-(\lambda, \beta_1)\}\big], \tag{6.23}$$

where we have used the following relations:

$$L_v a^+_\alpha a_\beta = \sum_{q,\lambda} \big(b_q + b^+_{-q}\big)\{C_{\lambda,\alpha}(q)a^+_\lambda a_\beta - C_{\beta,\lambda}(q)a^+_\alpha a_\lambda\},$$

$$L_d b^\pm_q a^+_\alpha a_\beta = (E_\alpha - E_\beta \pm \hbar\omega_q)b^\pm_q a^+_\alpha a_\beta,$$

$$L_d a^+_\alpha a_\beta a^+_\gamma a_\delta = (E_\alpha + E_\gamma - E_\beta - E_\delta)a^+_\alpha a_\beta a^+_\gamma a_\delta,$$

$$L_d b^+_{q_1} b_{q_2} a^+_\alpha a_\beta = (E_\alpha - E_\beta + \hbar\omega_{q_1} - \hbar\omega_{q_2})b^+_{q_1} b_{q_2} a^+_\alpha a_\beta.$$

In equations (6.20)–(6.23), the solutions near the resonance points $\hbar\omega_{123} \approx E_{\delta_1\delta_2}$, $\hbar\bar{\omega}_{12} \approx E_{\beta_2\beta_1} + E_{\gamma_2\gamma_1}$ and $\hbar\bar{\omega}_1 \approx E_{\gamma_2\gamma_1}$ were considered, and we have ignored the terms including $C_{\alpha\alpha}(q)$ and $(\hbar\omega \pm \hbar\omega_q)^{-1}$ by the same reason as in equations (5.7) and (5.8). Note that the result given in [23] has the same form as the present, but does not include the terms corresponding to equations (6.20)–(6.23). This suggests that the present result provides a more specific and significant interpretation of the phonon emissions and absorptions in all electron transition processes.

## 7. Concluding remarks

The linear and nonlinear optical conductivity tensors for a system of electrons interacting with phonons were derived using the reduction identity and the state-dependent projection

technique introduced by the authors. The linewidth terms appearing in the conductivity contain the electron and phonon distribution functions more phenomenologically. Therefore, all possible electron transitions, as well as phonon emission and absorption, can be explained properly.

As far as we know, there is no explicit similarity between the present projection technique and the conventional diagram method. The term-by-term manipulations are different in the two methods, in general. Note that the method used here is different from the standard perturbation technique usually utilized in the standard diagram method. However, the two methods are sure to lead to same acceptable results if correct manipulations are made. We should remember the old saying 'All roads lead to Rome'. However, we expect that the present technique is simpler and requires less amount of calculation in dealing with the problem chosen in the present paper.

The difference between the present theory and the previous one [33] lies in the use of different projection operators. The present result is more general than the previous one. The higher order terms of the nonlinear conductivity can be obtained in a similar manner using higher order terms in equation (2.5) and (3.7) with the method used in this paper. Although the theory for an electron–phonon system was formulated, electron–impurity scattering on damping can also be included in this theory. It is expected that the result can be applied to nonlinear optical phenomena such as the sum-frequency generation [37] and second-harmonic generation [38, 39]. All of these will be studied in the future.

## Appendix A. Derivation of the second-order reduction identity

Using $[H, \rho(H)] = 0$ and $T_R(ABC) = T_R(BCA)$, the following are obtained:

$$
\begin{aligned}
T_R\{\rho[[[LX, A], B], C]\} &= T_R\{\rho[[[HX - XH, A], B], C]\} \\
&= T_R\{\rho(XABCH - CHXAB - BHXAC + CBHXA \\
&\quad - AHXBC + CAHXB + BAHXC - CBAHX)\} \\
&\quad + T_R\{\rho(-XHABC + CXHAB + BXHAC - CBXHA \\
&\quad + AXHBC - CAXHB - BAXHC + HCBAX)\} \quad\quad \text{(A.1)}
\end{aligned}
$$

and

$$
\begin{aligned}
T_R\{\rho[[[X, LA], B], C]\} &+ T_R\{\rho[[[X, A], LB], C]\} + T_R\{\rho[[[X, A], B], LC]\} \\
&= T_R\{\rho[[[X, HA - AH], B], C]\} + T_R\{\rho[[[X, A], HB - BH], C]\} \\
&\quad + T_R\{\rho[[[X, A], B], HC - CH]\} \\
&= T_R\{\rho(XHABC - CXHAB - BXHAC + CBXHA - CBHAX)\} \\
&\quad + T_R\{\rho(AHXBC - CAHXB - BAHXC + CBAHX)\} \\
&\quad + T_R\{\rho(-AXHBC + CAXHB)\} + T_R\{\rho(BHXAC)\} \\
&\quad + T_R\{\rho(BAXHC - HCBAX)\} + T_R\{\rho(-XABCH + CHXAB)\}. \quad \text{(A.2)}
\end{aligned}
$$

So, we have derived the second-order reduction identity, i.e. (A.1) $= -$(A.2). By similar procedures, the higher order terms can be derived.

## Appendix B. Derivation of equation (6.12)

Applying equation (4.6) to $(\hbar\bar{\omega}_{12} - L_{\text{eq}})^{-1}$ in equation (6.10) with $A \equiv \hbar\bar{\omega}_{12}$ and $B \equiv L_{\text{eq}}$ gives

$$D_2(\bar{\omega}_{12})\hbar\bar{\omega}_{12} = T_R\big\{\rho\big[K(\bar{\omega}_1)\big[\big[L_v a_{\delta_1}^+ a_{\delta_2}, a_{\alpha_1}^+ a_{\alpha_2}\big], a_{\beta_1}^+ a_{\beta_2}\big], a_{\gamma_1}^+ a_{\gamma_2}\big]\big\}$$
$$+ T_R\big\{\rho\big[K(\bar{\omega}_1)\big[L_{eq}K(\bar{\omega}_{12})\big[L_v a_{\delta_1}^+ a_{\delta_2}, a_{\alpha_1}^+ a_{\alpha_2}\big], a_{\beta_1}^+ a_{\beta_2}\big], a_{\gamma_1}^+ a_{\gamma_2}\big]\big\}$$
$$\equiv D_{21}(\bar{\omega}_1) + D_{22}(\bar{\omega}_{12}). \tag{B.1}$$

$D_{21}(\bar{\omega}_1)$ and $D_{22}(\bar{\omega}_{12})$ are calculated by applying the zeroth- and first-order reduction identity, respectively, as

$$D_{21}(\bar{\omega}_1) = -V_{21}\big/\big(\hbar\bar{\omega}_1 + E_{\gamma_1\gamma_2}\big) \tag{B.2}$$

$$D_{22}(\bar{\omega}_{12}) = -\big(E_{\beta_1\beta_2} + E_{\gamma_1\gamma_2}\big)D_2(\bar{\omega}_{12}) - V_{22} - V_{23}, \tag{B.3}$$

in the second-order approximation of $L_v$. Therefore, inserting equations (B.2) and (B.3) into equation (B.1), we obtain

$$D_2(\bar{\omega}_{12}) = \frac{-V_{21}\big/\big(\hbar\bar{\omega}_1 + E_{\gamma_1\gamma_2}\big) - V_{22} - V_{23}}{\hbar\bar{\omega}_{12} + E_{\beta_1\beta_2} + E_{\gamma_1\gamma_2}}. \tag{B.4}$$

$E_2(\bar{\omega}_{123})$ given in equation (6.11) is calculated by applying the second-order reduction identity as

$$E_2(\bar{\omega}_{123}) = -V_{24} - V_{25} - V_{26} \tag{B.5}$$

since $T_R\big\{\rho\big[K(\bar{\omega}_1)\big[K(\bar{\omega}_{12})[Q_2 K(\bar{\omega}_{123})L_v a_{\delta_1}^+ a_{\delta_2}, a_{\alpha_1}^+ a_{\alpha_2}], a_{\beta_1}^+ a_{\beta_2}\big], a_{\gamma_1}^+ a_{\gamma_2}\big]\big\} = 0$. Finally, inserting equations (B.4) and (B.5) into equation (6.9), equation (6.12) is obtained.

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