Study of resonant inelastic light scattering in Keldysh–Schwinger functional integral formalism

H C Lee
Department of Physics and Basic Science Research Institute, Sogang University, Seoul 121-742, Korea
E-mail: hyunlee@sogang.ac.kr

New Journal of Physics 15 (2013) 093039 (21pp)
Received 21 August 2013
Published 26 September 2013
Online at http://www.njp.org/
doi:10.1088/1367-2630/15/9/093039

Abstract. The scattering cross-section of the resonant inelastic light scattering is represented as a correlation function in the Keldysh–Schwinger functional integral formalism. The functional integral approach enables us to compute the cross-section in the Feynman diagram perturbation theory where many-body effects can be fully incorporated. This approach is applied to the one G-phonon Raman scattering of graphene, and the result is shown to agree with the one previously obtained by the conventional Fermi golden rule formula. Also, this approach is generalized to the systems in non-equilibrium conditions.
1. Introduction

The inelastic light scattering (ILS) is a very important experimental method for the investigation of the physical properties of condensed matter. In particular, the resonant inelastic x-ray scattering (often called RIXS) has developed very rapidly recently, mainly owing to the advent of the high power x-ray light sources [1]. RIXS has proven to be a versatile tool for the study of elementary excitations of strongly correlated electron materials, such as magnons [2] and orbitons, where many-body interactions play crucial roles [1]. Thus it is very desirable to have a theoretical scheme for the interpretation of the experimental ILS data which can incorporate many-body effects precisely.

In general, the transition rate \( w_{\text{ILS}} \) for ILS can be obtained by the Fermi golden rule which is applied up to the second order with respect to the interaction Hamiltonian \( \hat{H}' \) between light and matter:

\[
\begin{align*}
    w_{\text{ILS}} &= \frac{2\pi}{\hbar} \sum_i p_i \sum_F \left| \langle F|\hat{H}'_{\text{NR}}|I\rangle \right|^2 + \sum_n \frac{\langle F|\hat{H}'_{\text{R}}|n\rangle \langle n|\hat{H}'_{\text{R}}|I\rangle}{E_i - E_n} \right| \right|^2,
\end{align*}
\]

where \( |I\rangle, |n\rangle \) and \( |F\rangle \) denote the initial states, the (many-body) intermediate eigenstates and the final states of the light–matter system, respectively. \( E_{i,n} \) are the energy eigenvalues of the states and \( p_i \) is the probability for the initial states (\( \sum_i p_i = 1 \)). \( \hat{H}'_{\text{NR}} \) is the part of the Hamiltonian \( \hat{H}' \) quadratic in the photon vector potential operator \( \vec{A} \), which contributes to the non-resonant ILS [3]. \( \hat{H}'_{\text{R}} \) is linear in \( \vec{A} \), and it is responsible for the resonant ILS [3] (the second term of (1)).

The non-resonant transition amplitude often dominates over the resonant one, but in special cases where \( E_i \) is close to \( E_n \), the resonant transition amplitude can be significant. After some manipulations involving the expectation value of the photon operators, (1) can be shown to be essentially the Kramers–Heisenberg formula of ILS. In this paper, for simplicity, we will consider only the case \( \hat{H}'_{\text{R}} = -\frac{1}{c} \int d\vec{r} \vec{A}(\vec{r}) \cdot \vec{J}_e(\vec{r}) \), where \( \vec{J}_e(\vec{r}) \) is the electric current operator.

It is well known that the scattering cross-section of non-resonant contribution can be expressed in the form of correlation function of stress tensor operators [3, 4] by using the van Hove–Placzek procedure [5, 6]. By employing the fluctuation–dissipation theorem the correlation function can be directly linked to the retarded Green’s function (e.g. see [4, p 151]).
Then the retarded Green’s function can be computed systematically (including full many-body effects) in the imaginary time Matsubara formalism [4, 7] by using an analytic continuation technique. Thus as far as the non-resonant contribution to ILS is concerned we have a firmly established a theoretical scheme where full many-body physics can be incorporated precisely. Henceforth we will assume that the resonance condition is satisfied and will focus on the resonant contributions only.

The first goal of this paper is to express the resonant contribution to ILS in the form of correlation function just like the case of the non-resonant contribution mentioned above. To achieve this goal, in principle, we have to carry out the sum over the intermediate states $|n\rangle$ of the second term of (1). In general, very little is known about the states $|n\rangle$, and we have to employ some kinds of assumptions or approximations about the states. Correlation functions are defined without reference to intermediate states, so in our approach the explicit sum over the intermediate states $|n\rangle$ should be avoided. Then we have to go back to the time-dependent perturbation theory involving time evolution operator. Recall that the time-dependent perturbation theory requires time-ordering of operators [8], namely

$$\hat{T}(\hat{X}(t)\hat{Y}(t')) = \theta(t-t')\hat{X}(t)\hat{Y}(t') \pm \theta(t'-t)\hat{Y}(t')\hat{X}(t),$$

where minus sign applies when both $\hat{X}(t)$ and $\hat{Y}(t')$ are fermion operators and $\hat{T}$ is the time-ordering symbol$^1$. $\theta(t)$ is the unit step function.

Now as can be seen in (1), we have to take the complex conjugate of transition amplitude to obtain the transition rate. In quantum mechanics the complex conjugate of a transition amplitude of a process is related to a transition amplitude of the time-reversed process, and this observation necessitates the introduction of anti-time-ordering

$$\hat{T}(\hat{X}(t)\hat{Y}(t')) = \theta(t'-t)\hat{X}(t)\hat{Y}(t') \pm \theta(t-t')\hat{Y}(t')\hat{X}(t),$$

where $\hat{T}$ is the anti-time-ordering symbol.

From the above discussion it is evident that the scattering cross-section of ILS (generically all transition probability) requires not only forward time evolution but also backward time evolution in the time representation of the time-dependent perturbation theory. Therefore, we are naturally led to the concept of closed time contour introduced by Keldysh and Schwinger (KS) [9, 10].

The KS formulation of time-dependent perturbation theory has been employed in the earlier works on the x-ray spectra of metals [11–13]. However, these works did not fully disentangle the time orderings of involved operators, so that they were limited to the time-representation of Feynman diagrams or the time-ordered Feynman diagrams (see, e.g. [12, figure 2]; [13, equation (11)]). If the time orderings are fully disentangled then all time-integrals can be done from $-\infty$ to $\infty$, and all Feynman diagrams can be calculated in an energy–momentum representation.

The second goal of this paper is to elaborate on the formalism developed in [11–13], so that the correlation function obtained in the first goal can be recast in the KS functional integral formalism, which will disentangle the time orderings completely. The correlation functions in KS functional integral formalism can be computed in the standard Feynman diagram method of quantum field theory [14] without being limited to time-ordered diagrams, etc.

The third goal of this paper is to generalize the KS functional integral approach to the systems in non-equilibrium conditions. The KS methods were originally developed for the study

$^1$ It can be easily checked that $\hat{T}(\hat{X}(t)\hat{Y}(t')) = \pm \hat{T}(\hat{Y}(t')\hat{X}(t))$. 

New Journal of Physics 15 (2013) 093039 (http://www.njp.org/)
of non-equilibrium phenomena. As such, the KS functional integral formalism can be readily generalized for the study of ILS in non-equilibrium situation.

The main results of this paper are (21), (23), (47), (74) and (75). This paper is organized as follows. In section 2 we derive the correlation function corresponding to the scattering cross-section of the resonant ILS. In section 3 the correlation function is represented in the KS functional integral formalism. The key step is the disentanglement of the time orderings of electric current operators. In section 4 we compare our approach with the Kramers–Heisenberg formula and point out differences. In section 5 we apply our method to the case of G-phonon Raman scattering of graphene and compare the results with the one obtained by the conventional Fermi golden rule method. In section 6 we generalize the KS functional integral formalism of ILS to the non-equilibrium situations. We close this paper with discussions and summary in section 7.

2. Scattering cross-section of inelastic light scattering (ILS) and correlation function

First we recall that the scattering cross-section of ILS is defined by the transition rate (induced by light–matter interaction) divided by the flux of incoming photons

$$d\sigma = \sum_{l} \sum_{p} \frac{|M_{FI}(t_F, t_i)|^2}{(t_F - t_i)} / \left( \frac{C}{\mathcal{V}} \right),$$

(4)

where $M_{FI}$ is the transition amplitude from an initial state $|l\rangle$ to a final state $|F\rangle$ in time interval $[t_i, t_F]$ and $\mathcal{V}$ is the volume of space for the box quantization of photons. Our goal in this section is to express (4) in the form of correlation function. The key quantity is the transition amplitude $M_{FI}(t_F, t_i)$ which is most naturally presented in the framework of time-dependent perturbation theory [8].

The transition amplitude $M_{FI}(t_F, t_i)$ is defined by

$$M_{FI}(t_F, t_i) = \langle F | \hat{U}_{\text{tot}}(t_F, t_i) | l \rangle,$$

(5)

where $\hat{U}_{\text{tot}}(t_F, t_i)$ is the time evolution operator governed by the total Hamiltonian $\hat{H}_{\text{tot}}$ of light–matter system from time $t_i$ to $t_F$:

$$\hat{U}_{\text{tot}}(t_F, t_i) = \exp \left[ -\frac{i}{\hbar} \int_{t_i}^{t_F} d\tilde{r} \hat{H}_{\text{tot}}(\tilde{r}) \right].$$

(6)

The total Hamiltonian consists of the Hamiltonian of matter $\hat{H}_m$, the Hamiltonian of photons $\hat{H}_p$ (see appendix (A.1)) and the interaction Hamiltonian between light and matter $\hat{H}^I$:

$$\hat{H}_{\text{tot}} = (\hat{H}_m + \hat{H}_p) + \hat{H}^I \equiv \hat{H}_0 + \hat{H}^I.$$

(7)

Next we employ the interaction picture and treat $\hat{H}^I$ perturbatively. In the interaction picture the time evolution operator $\hat{U}_{\text{tot}}(t_F, t_i)$ becomes (see, e.g. [15, pp 722–724])

$$\hat{U}_{\text{tot}}(t_F, t_i) = \hat{U}_{\hat{H}_0}(t_F, t_i) \hat{V}(t_F, t_i),$$

(8)

where $\hat{U}_{\hat{H}_0}(t_F, t_i)$ is the time evolution operator governed by the Hamiltonian $\hat{H}_0$ (see (7)), and

$$\hat{V}(t_F, t_i) = \exp \left[ -\frac{i}{\hbar} \int_{t_i}^{t_F} d\tilde{r} \hat{H}^I_{\hat{H}_0}(\tilde{r}) \right].$$

(9)
where $\hat{H}_{\text{ph}}^\prime$ denotes the Hamiltonian $\hat{H}^\prime$ in the Heisenberg picture with respect to $\hat{H}_0$. Now plugging (8) into (5) and expanding (9) up to the second order in $\hat{H}_{\text{ph}}^\prime$, we obtain the second order contribution which is responsible for the resonant ILS:

$$\mathcal{M}^{(2)}_{\text{FI}} = \frac{1}{2!} \left( -\frac{i}{\hbar} \right)^2 \left\langle I \right| \hat{U}_{\hat{H}_0} \int_{\hat{h}}^{\hat{H}_{\text{ph}}} \hat{d}_1 \int_{\hat{h}}^{\hat{H}_{\text{ph}}} \hat{d}_2 \mathcal{T}[\hat{H}_{\text{ph}}^\prime(\hat{t}_1) \hat{H}_{\text{ph}}^\prime(\hat{t}_2)] \right| I \rangle. \tag{10}$$

Then the transition probability can be expressed as

$$|\mathcal{M}^{(2)}_{\text{FI}}|^2 = \frac{1}{(2!)^2 \hbar^4} \left\langle I \right| \int_{\hat{h}}^{\hat{H}_{\text{ph}}} \hat{d}_3 \int_{\hat{h}}^{\hat{H}_{\text{ph}}} \hat{d}_4 \mathcal{T}[\hat{H}_{\text{ph}}^\prime(\hat{t}_4) \hat{H}_{\text{ph}}^\prime(\hat{t}_3)] \hat{U}_{\hat{H}_0} | F \rangle \times \left\langle F \right| \hat{U}_{\hat{H}_0} \int_{\hat{h}}^{\hat{H}_{\text{ph}}} \hat{d}_1 \int_{\hat{h}}^{\hat{H}_{\text{ph}}} \hat{d}_2 \mathcal{T}[\hat{H}_{\text{ph}}^\prime(\hat{t}_1) \hat{H}_{\text{ph}}^\prime(\hat{t}_2)] | I \rangle. \tag{11}$$

Note the appearance of the anti-time-ordering $\mathcal{T}$ in (11).

The next key step is to compute the matrix elements of (11) with respect to the photon states (only). For that purpose, the initial $|I\rangle$ and the final $|F\rangle$ states are represented by the tensor product of photon Hilbert space $|k_{i,f}, \vec{e}_{i,f}\rangle$ and matter Hilbert space $|i/f\rangle$

$$|I\rangle = |k_i, \vec{e}_i\rangle \otimes |i\rangle, \quad |F\rangle = |k_f, \vec{e}_f\rangle \otimes |f\rangle, \tag{12}$$

where $k_{i,f}$ and $\vec{e}_{i,f}$ are the wavenumber vector and the polarization vector of incoming and outgoing photon, respectively. Since the matter Hamiltonian $\hat{H}_m$ commutes with the photon Hamiltonian $\hat{H}_\text{ph}$ which is non-interacting (see appendix (A.1)), the action of $\hat{H}_\text{ph}$ and its time evolution operator $U_{\hat{H}_\text{ph}}$ in (11) on photon Hilbert space can be evaluated exactly. After evaluating the action of $U_{\hat{H}_\text{ph}}$, we can define an operator $\hat{D}_{\vec{k},\vec{e}}(t_1, t_2)$ and its Hermitian conjugate $\hat{D}_{\vec{k},\vec{e}}^\dagger(t_3, t_4)$ which act only on the matter Hilbert space:

$$\hat{D}_{\vec{k},\vec{e}}(t_1, t_2) = \langle k_f, \vec{e}_f | \mathcal{T}[\hat{H}_{\text{ph}}^\prime(\hat{t}_1) \hat{H}_{\text{ph}}^\prime(\hat{t}_2)] | k_i, \vec{e}_i \rangle, \tag{13}$$

$$\hat{D}_{\vec{k},\vec{e}}^\dagger(t_3, t_4) = \langle k_i, \vec{e}_i | \mathcal{T}[\hat{H}_{\text{ph}}^\prime(\hat{t}_3) \hat{H}_{\text{ph}}^\prime(\hat{t}_4)] | k_f, \vec{e}_f \rangle, \tag{14}$$

where $\vec{k}_{i,f}$ and $\vec{e}_{i,f}$ collectively.

Then the transition probability (11) can be recast as

$$|\mathcal{M}^{(2)}_{\text{FI}}|^2 = \frac{1}{(2!)^2 \hbar^4} \prod_{i=1}^{4} \int_{\hat{h}}^{\hat{H}_{\text{ph}}} \hat{d}_i \langle i | \hat{D}_{\vec{k}_i,\vec{e}_i}^\dagger(\hat{t}_3, \hat{t}_4) \hat{U}_{\hat{H}_m} | f \rangle \langle f | \hat{U}_{\hat{H}_m} \hat{D}_{\vec{k}_i,\vec{e}_i}(\hat{t}_1, \hat{t}_2) | i \rangle. \tag{15}$$

We note that the operator $\hat{D}_{\vec{k}_i,\vec{e}_i}(t_1, t_2)$ is time-ordered, while the operator $\hat{D}_{\vec{k}_i,\vec{e}_i}^\dagger(t_3, t_4)$ is anti-time-ordered.

The initial photon state $|k_i, \vec{e}_i\rangle$ can be taken to a pure state, so that the probability distribution for the initial state $p_i$ pertains only to the initial matter Hilbert space. The probability distribution $p_i$ for the initial matter Hilbert space can be specified by a density matrix $\hat{\rho}_m$ (Tr$_m$ indicates trace over matter Hilbert space)

$$p_i = \langle i | \hat{\rho}_m | i \rangle, \quad \text{Tr}_m \hat{\rho}_m = 1. \tag{16}$$
The scattering cross-section of resonant ILS now takes the form
\[ d\sigma = \frac{1}{c/\sqrt{t_F - t_i}} \sum_{k, \vec{e}_t} \sum_{i} \langle i | \hat{\rho}_m | i \rangle \sum_{f} | \mathcal{M}_{fi}^{(2)} |^2. \]  
(17)

The sum over the final matter Hilbert space of (17) can be done using the closure relation of matter Hilbert space \( \sum_i | i \rangle \langle i | = I \) (\( I \) is an identity operator). From (15) we obtain
\[ \sum_{f} | \mathcal{M}_{fi}^{(2)} |^2 = \frac{1}{(2\pi)^2 \hbar^4} \int_{t_i}^{t_F} dt_i \langle i | \hat{D}_{k_i, \vec{e}_t} (\vec{t}_3, \vec{t}_4) \hat{D}_{k_i, \vec{e}_t} (\vec{t}_1, \vec{t}_2) | i \rangle. \]  
(18)

The sum over the initial matter Hilbert space \( | i \rangle \) of (17) can be done by using (16):
\[ d\sigma = \frac{1}{(2\pi)^2 \hbar^4} \frac{1}{c/\sqrt{t_F - t_i}} \sum_{k, \vec{e}_t} \int_{t_i}^{t_F} dt_i \text{Tr}_m \langle \hat{\rho}_m \hat{D}_{k_i, \vec{e}_t} (\vec{t}_3, \vec{t}_4) \hat{D}_{k_i, \vec{e}_t} (\vec{t}_1, \vec{t}_2) \rangle. \]  
(19)

Equation (19) represents the scattering cross-section in the form of correlation function of operators \( \hat{D}_{k_i, \vec{e}_t} \) and \( \hat{D}_{k_i, \vec{e}_t}^\dagger \) which act only on the matter Hilbert space. The detailed form of the operator \( \hat{D}_{k_i, \vec{e}_t} \) is worked out in the appendix.

Substituting (A.6) and (A.7) into (19) we arrive at
\[ d\sigma = \frac{1}{t_F - t_i} \frac{(2\pi/\hbar)^2}{c \omega_l \omega_F} \int_{k, \vec{e}_t} \left( d\vec{t}_i \right) e^{-i\omega_{t_1} + i\omega_{t_2} + i\omega_{t_3} - i\omega_{t_4}} C_{k_i, \vec{e}_t, k_i, \vec{e}_t} (t_3, t_4, t_1, t_2), \]  
(20)

where the correlation function \( C_{k_i, \vec{e}_t, k_i, \vec{e}_t} (t_3, t_4, t_1, t_2) \) is given by \( (\vec{J}_e (\vec{k}, t) \) is the electric current operator in momentum space, see (A.8))
\[ C_{k_i, \vec{e}_t, k_i, \vec{e}_t} (t_3, t_4, t_1, t_2) = \text{Tr}_m \langle \hat{\rho}_m \hat{\tilde{T}} [\vec{J}_e (\vec{k}_i, t_3) \cdot \hat{\tilde{e}}_t \vec{J}_e (-\vec{k}_i, t_4) \cdot \hat{\tilde{e}}_t] \rangle \int [\vec{J}_e (-\vec{k}_i, t_1) \cdot \hat{\tilde{e}}_t \vec{J}_e (\vec{k}_i, t_2) \cdot \hat{\tilde{e}}_t] \rangle. \]  
(21)

For matter systems at equilibrium the correlation function \( C_{k_i, \vec{e}_t} (t_3, t_4, t_1, t_2) \) possesses the time translation invariance which is manifest in the following form:
\[ C_{k_i, \vec{e}_t, k_i, \vec{e}_t} (\{ t_i \}) = \int_{t_1}^{t_2} \prod_{j=1}^{4} d\omega_j \frac{2\pi}{2\pi} e^{-i\sum_j \omega_j t_j} (2\pi)^3 \delta \left( \sum_j \omega_j \right) \tilde{C}_{k_i, \vec{e}_t, k_i, \vec{e}_t} (\{ \omega_j \}). \]  
(22)

Plugging (22) into (20), changing variable \( t_i \rightarrow t_i + (t_F + t_i)/2 \), and carrying out the time integrals in the limit \( t_F - t_i \rightarrow \infty \), we obtain \( (\sum k_i = V \int d^3 \vec{k}_i / (2\pi)^3) \)
\[ d\sigma = \frac{(2\pi/\hbar)^2}{c \omega_l \omega_F} \int \frac{d^3 \vec{k}_i}{(2\pi)^3} \sum_{\vec{e}_t} \tilde{C}_{k_i, \vec{e}_t, k_i, \vec{e}_t} (\omega_t, -\omega_t, -\omega_t, \omega_t), \]  
(23)

where the energy conserving delta function is substituted by
\[ 2\pi \delta(0) = \int_{-(n-\hbar/2)}^{(n-\hbar/2)} dt e^{i\omega t} \rightarrow (t_F - t_i). \]  
(24)

Equations (21) and (23) constitute the first goal of this paper.
3. Keldysh–Schwinger functional integral formulation of correlation function

As we have seen in section 2 the computation of scattering cross-section has been reduced to the computation of the correlation function (21), which we rephrase as

$$\Pi(\tau_1, \tau_2, \tau_3, \tau_4) = \text{Tr}_m(\hat{t}[\hat{A}(\tau_1)\hat{B}(\tau_2)]\text{Tr}[\hat{C}(\tau_3)\hat{D}(\tau_4)]\hat{\rho}_m)$$

for the sake of more convenient notations. From now on we will drop the subscript ‘m’ denoting the matter sector. In this section the correlation function (25) will be represented in the KS functional integral formulation, and in this representation all time-orderings will be disentangled completely.

The literature on the method of KS closed time contour are very vast, and we cite only [16, 17] which appear to be more useful for condensed matter physicists in the author’s opinion. We will also need the method of coherent state path integral, and the detailed treatments can be found in [18]. In the following many well-known detailed steps (for which readers are referred to the references cited above) will be omitted and we will focus on disentangling the time orderings of (25).

The key concept of the KS method is the time evolution along closed time contour. A characteristic property of this closed time contour $C = [t_1 \rightarrow t_F \rightarrow t_1]$ (see figure 1) is (where we put $\hat{\rho}_m = \hat{\rho}(t_1)$)

$$Z \equiv \text{Tr}[\hat{U}(t_1, t_F)\hat{U}(t_F, t_1)\hat{\rho}(t_1)] = \text{Tr}[\hat{U}\hat{C}\hat{\rho}(t_1)] = 1,$$

which follows from the unitarity of time evolution operator $\hat{U}$. We start by resolving the time orderings of (25) explicitly:

$$\hat{T}[\hat{A}(\tau_1)\hat{B}(\tau_2)] = \theta(\tau_2 - \tau_1)\hat{A}(\tau_1)\hat{B}(\tau_2) + \theta(\tau_1 - \tau_2)\hat{B}(\tau_2)\hat{A}(\tau_1),$$

$$\text{Tr}[\hat{C}(\tau_3)\hat{D}(\tau_4)] = \theta(\tau_3 - \tau_4)\hat{C}(\tau_3)\hat{D}(\tau_4) + \theta(\tau_4 - \tau_3)\hat{D}(\tau_4)\hat{C}(\tau_3).$$

Plugging (27) into (25) we can write

$$\Pi(\{\tau_j\}) = \theta(\tau_2 - \tau_1)\theta(\tau_3 - \tau_4)\Pi^{(1)}(\{\tau_j\}) + \theta(\tau_2 - \tau_1)\theta(\tau_4 - \tau_3)\Pi^{(2)}(\{\tau_j\}) + \theta(\tau_1 - \tau_2)\theta(\tau_3 - \tau_4)\Pi^{(3)}(\{\tau_j\}) + \theta(\tau_1 - \tau_2)\theta(\tau_4 - \tau_3)\Pi^{(4)}(\{\tau_j\}),$$

where the correlation functions $\Pi^{(1,2,3,4)}(\{\tau_j\})$ are given by

$$\Pi^{(1)}(\{\tau_j\}) = \text{Tr}[\hat{A}(\tau_1)\hat{B}(\tau_2)\hat{C}(\tau_3)\hat{D}(\tau_4)\hat{\rho}(t_1)], \quad \Pi^{(2)}(\{\tau_j\}) = \text{Tr}[\hat{A}(\tau_1)\hat{B}(\tau_2)\hat{D}(\tau_4)\hat{C}(\tau_3)\hat{\rho}(t_1)],$$

$$\Pi^{(3)}(\{\tau_j\}) = \text{Tr}[\hat{B}(\tau_2)\hat{A}(\tau_1)\hat{C}(\tau_3)\hat{D}(\tau_4)\hat{\rho}(t_1)], \quad \Pi^{(4)}(\{\tau_j\}) = \text{Tr}[\hat{B}(\tau_2)\hat{A}(\tau_1)\hat{D}(\tau_4)\hat{C}(\tau_3)\hat{\rho}(t_1)].$$

New Journal of Physics 15 (2013) 093039 (http://www.njp.org/)
Next we represent each correlation function \( \Pi^{(1,2,3,4)}(\{\tau_j\}) \) in the KS functional integral formalism. Working out the case for \( \Pi^{(1)}(\{\tau_j\}) \) will be enough. The operators \( \hat{A}(\tau_j), \hat{B}(\tau_j), \hat{C}(\tau_j), \hat{D}(\tau_j) \) in the Heisenberg picture can be expressed in terms of the time evolution operator. For \( \hat{A}(\tau_j) \),

\[
\hat{A}(\tau_j) = \hat{U}^\dagger(\tau_j, t_1) \hat{A} \hat{U}(\tau_j, t_1) = \hat{U}(t_1, \tau_j) \hat{A} \hat{U}(\tau_j, t_1),
\]

and similarly for other operators. Substituting (30) into \( \Pi^{(1)} \) of (29) and employing the composition property of time evolution operator \( \hat{U}(t_1, t_3) = \hat{U}(t_1, t_2) \hat{U}(t_2, t_3) \), we obtain

\[
\Pi^{(1)}(\{\tau_j\}) = \text{Tr}[\hat{U}(t_1, \tau_1) \hat{A} \hat{U}(\tau_1, t_2) \hat{B} \hat{U}(t_2, t_3) \hat{C} \hat{U}(t_3, t_4) \hat{D} \hat{U}(t_4, t_1) \hat{\rho}(t_1)],
\]

where the turning point of the closed time contour is marked with a vertical bar. From now on we will take a limit \( t_\ell \to \infty, t_i \to -\infty \). The time evolution to the left of the bar of (31) is backward in time, while that to the right of the bar is forward in time. Thus in (31) we have time evolution along the closed time contour, and the operators are inserted along the contour.

To develop the functional integral representation, the time evolution operators of (31) are to be expressed by the Trotter product formula [18] in accordance with the closed time contour of figure 1. For \( t > t' \) (the time evolution in forward branch)

\[
\hat{U}(t, t') = \lim_{N \to \infty} \prod^{N-\text{times}} e^{-i\Delta t \hat{H}/\hbar} = \prod^{N-\text{times}} e^{-i\Delta t \hat{H}/\hbar}, \quad \Delta t = \frac{t - t'}{N}, \quad t > t',
\]

and for \( t < t' \) (the time evolution in backward branch) we note \( \hat{U}(t, t') = (\hat{U}(t', t))^{-1} \), so that

\[
\hat{U}(t, t') = \lim_{N \to \infty} \prod^{N-\text{times}} e^{+i\Delta t \hat{H}/\hbar} = \prod^{N-\text{times}} e^{+i\Delta t \hat{H}/\hbar}, \quad \Delta t = \frac{t' - t}{N}, \quad t' > t.
\]

Then between each infinitesimal time evolution operator \( e^{\pm i\Delta t \hat{H}/\hbar} \), we insert the following closure relation of coherent states (\( I \) is an identity operator):

\[
I = \int D[\{\phi_{\pm, k}\}] e^{-\phi^*_k \hat{\phi}^\pm_k} \langle \{\phi_{\pm, k}\} | \{\phi_{\pm, k}\} \rangle,
\]

where \( \{\phi_{\pm, k}\} \) and \( \{\phi^*_k\} \) denote the collective coherent state variables at the \( k \)th time slice along the forward (+) and the backward (−) branch. Recall that the ket coherent state is an eigenstate of annihilation operator, while the bra coherent state is an eigenstate of creation operator:

\[
\hat{c}|\phi_{\pm, k}\rangle = \phi_{\pm, k}|\phi_{\pm, k}\rangle, \quad \langle \phi_{\pm, k}|\hat{c}^\dagger = \langle \phi_{\pm, k}|\phi^*_k\rangle.
\]

This property makes the computation of matrix elements of any normal-ordered operators very easy:

\[
\langle \phi | \hat{A}_{\text{normal ordered}}(\hat{c}^\dagger, \hat{c}) | \phi' \rangle = e^{\hat{c}^* \phi'} A(\hat{c}^\dagger \to \phi^*, \hat{c} \to \phi').
\]

Note that the symbol \( A \) in the right hand side of (36) is not an operator but a function of coherent state variables \( \phi^*, \phi' \). All operators appearing in correlation function (31) are assumed to be normal-ordered. Employing (34–36) it is straightforward to develop the functional integral representation of the correlation function (31). Below we exhibit a few details of the computations near operator insertion points. Consider, for example, the insertion of the operator \( \hat{C} \) in (31). Clearly the time slicing can be done in such a way that the set of time slices include
the time slice at \( \tau_3 \) (or arbitrarily close to it). Let \( |\phi_{+k}\rangle \) be the ket of the time slice corresponding to the time \( \tau_3 \) (recall that the operator \( \hat{C} \) is located in the forward branch). Then the relevant matrix elements are

\[
\cdots |\phi_{+,k+1}\rangle \langle \phi_{+,k+1}^*| \hat{C} e^{-i\Delta t\hat{H}/\hbar} |\phi_{+,k}\rangle e^{-\phi_{+,k}^*\phi_{+,k}^\dagger} |\phi_{+,k+1}\rangle \cdots,
\]

Since the time interval \( \Delta t \) is infinitesimally small, we can approximate \( e^{-i\Delta t\hat{H}/\hbar} \approx 1 - i\Delta t\hat{H}/\hbar \) and obtain

\[
\langle \phi_{+,k+1}| \hat{C} e^{-i\Delta t\hat{H}/\hbar} |\phi_{+,k}\rangle \approx \left( \phi_{+,k+1}^* \phi_{+,k} + 1 \right) \hat{C} \left[ 1 - i\Delta t\frac{\hat{H}}{\hbar} \right] |\phi_{+,k}\rangle.
\]

Inserting a closure relation (34) into (38), we obtain

\[
\langle \phi_{+,k+1}| \hat{C} e^{-i\Delta t\hat{H}/\hbar} |\phi_{+,k}\rangle = \int D[\xi^*, \xi] e^{-\xi^*\xi} \langle \phi_{+,k+1}| \hat{C} |\xi\rangle \langle \xi| \left( \hat{C} \left[ 1 - i\Delta t\frac{\hat{H}}{\hbar} \right] \right) |\phi_{+,k}\rangle.
\]

The operator \( \hat{C} = \hat{C}(\hat{\xi}^*, \hat{\xi}) \) and the Hamiltonian \( \hat{H} \) can be assumed to be bounded polynomials of the creation \( \hat{\xi}^\dagger \) and the annihilation operator \( \hat{\xi} \). Now \( \xi^*, \xi \) integrals of (39) can be done exactly. This is because the operator \( \hat{C} \) and \( \hat{H} \) are assumed to be polynomials of \( \hat{\xi} \) and \( \hat{\xi}^\dagger \), so that the most general integrals involving \( \xi \) and \( \xi^* \) are of the following form \( (n, m) \) are non-negative integers:

\[
\int D[\xi^*, \xi] e^{-\xi^*\xi} \phi_{+k+1}^* \xi \xi^n = \phi_{+k+1}^* \phi_{+k}^* \xi^n.
\]

From (40), it follows that (since polynomial form is preserved through integral)

\[
\langle \phi_{+,k+1}| \hat{C} e^{-i\Delta t\hat{H}/\hbar} |\phi_{+,k}\rangle \approx C(\phi_{+,k+1}^*, \phi_{+,k}) \langle e^{\phi_{+,k+1}^*\phi_{+,k}} \rangle \hat{C} e^{-i\Delta t\hat{H}/\hbar} \phi_{+,k+1}^* \phi_{+,k}^* \phi_{+,k}.
\]

In the continuum limit \( N \to \infty \), the difference between \( \phi_{+,k+1}^* \) and \( \phi_{+,k}^* \) in \( C(\phi_{+,k+1}^*, \phi_{+,k}) \) can be ignored. Then (41) proves that the operator \( \hat{C}(\tau) \) which is inserted in the forward branch maps to \( C(\phi_{+,\tau}^*(\tau), \phi_{+,\tau}(\tau)) \) in the KS coherent functional integral formulation

\[
\hat{C}(\hat{\xi}^\dagger(\tau), \hat{\xi}(\tau))|\text{forward branch} \to C(\phi_{+,\tau}^*(\tau), \phi_{+,\tau}(\tau)).
\]

In an entirely similar way it can be shown that

\[
\hat{A}(\hat{\xi}^\dagger(\tau), \hat{\xi}(\tau))|\text{backward branch} \to A(\phi_{-,\tau}^*(\tau), \phi_{-,\tau}(\tau)).
\]

Combining the above results we arrive at the following KS functional integral representation of the correlation function (31):

\[
\Pi^{(1)}(\{\tau_j\}) = \int D[\phi^*, \phi] A(\phi_{+,\tau_1}^*(\tau_1), \phi_{-,\tau_1}(\tau_1)) B(\phi_{+,\tau_2}^*(\tau_2), \phi_{-,\tau_2}(\tau_2)) C(\phi_{+,\tau_3}^*(\tau_3), \phi_{+,\tau_3}(\tau_3))
\]

\[
\times D(\phi_{+,\tau_4}^*(\tau_4), \phi_{+,\tau_4}(\tau_4)) e^{i\Delta \phi_{+,\tau_4}/\hbar} e^{-i\Delta \phi_{-,\tau_4}/\hbar + \delta \Delta S_{\pm}},
\]

where \( S_{\pm} \) is the well-known classical action of functional integral defined on the forward (+) and backward (−) branch. \( \delta \Delta S_{\pm} \) is infinitesimally small terms which couple the forward and the backward branch functional integral variables. For the subtle roles played by \( \delta S_{\pm} \), see [16] and [19]. We emphasize that the \( A, B, C, D \) of (44) are functions of ordinary variables (or anti-commuting Grassman numbers for fermion operators) and that they are not operators, so that the relative order between them does not matter.

*New Journal of Physics* **15** (2013) 093039 (http://www.njp.org/)
Now recalling the definitions of the correlation functions \( \Pi^{(1,2,3,4)}(\{\tau_i\}) \) from (29), we find that all of \( \Pi^{(1,2,3,4)}(\{\tau_i\}) \) are given by the same expression, namely (44), in the KS functional integral. But they are defined in the separate regions of time variables \( \{\tau_i\} \) as signified by the products of step functions in (28). Noting a trivial identity
\[
[\theta(\tau_1 - \tau_2) + \theta(\tau_2 - \tau_1)] \times [\theta(\tau_3 - \tau_4) + \theta(\tau_4 - \tau_3)] = 1 \cdot 1 = 1,
\]
we finally obtain
\[
\Pi(\{\tau_i\}) = \text{Tr}_m(\hat{T}[\hat{A}(\tau_1)\hat{B}(\tau_2)]\hat{T}[\hat{C}(\tau_3)\hat{D}(\tau_4)]\hat{\rho}_m)
\]
\[
= \int D[\phi^+, \phi] A(\phi^+ + 1(\tau_1), \phi + 1(\tau_1)) B(\phi^+ + 1(\tau_2), \phi + 1(\tau_2))
\times C(\phi^+ + 1(\tau_3), \phi + 1(\tau_3)) D(\phi^+ + 1(\tau_4), \phi + 1(\tau_4)) e^{iS_\text{F}/\hbar - iS_\text{H}/\hbar + \delta S_{\text{S}}}. \tag{46}
\]
Applying the result (46) to the correlation function of ILS (21) we arrive at
\[
C_{\ell_1,\ell_2,\ell_3,\ell_4}(t_1, t_2, t_3, t_4) = \int D[\phi^+, \phi] e^{iS_\text{F}/\hbar - iS_\text{H}/\hbar + \delta S_{\text{S}}}
[\hat{J}_e(\phi^+ + 1(\ell_1), \phi + 1(\ell_1)) \cdot \hat{\ell}_1^+]
\times [\hat{J}_e(\phi^+ + 1(\ell_2), \phi + 1(\ell_2)) \cdot \hat{\ell}_2^+]
[\hat{J}_e(\phi^+ + 1(\ell_3), \phi + 1(\ell_3)) \cdot \hat{\ell}_3^+]
\times [\hat{J}_e(\phi^+ + 1(\ell_4), \phi + 1(\ell_4)) \cdot \hat{\ell}_4^+],
\tag{47}
\]
which is the second main result of this paper. (47) is the four-current correlation function which can be computed directly by using the standard functional integral methods such as Feynman diagram perturbation theory or semi-classical approximations.

4. Comparison with Kramers–Heisenberg formula

Let us compare the results (21) and (23) of this paper with the Kramers–Heisenberg formula which is essentially the Fermi golden rule (1). At this point we emphasize that in deriving (21) and (23) no approximations have been made regarding any intermediate states.

Below we will point out that the Kramers–Heisenberg formula was obtained by ignoring certain terms which are attributed to the sudden turning on of perturbation [8]. This implies that our results (21) and (23) contain more contributions than the Kramers–Heisenberg formula, so that we have to select a subset of Feynman diagrams contributing to (21) and (23) which indeed correspond to the Kramers–Heisenberg formula.

We start by evaluating the time integrals of (10) by introducing intermediate states \(|n\rangle\). For simplicity put \(t_1 = 0\), and resolve time-ordering explicitly.
\[
\mathcal{M}^{(2)}_{\text{F}} = \left(\frac{-i}{\hbar}\right)^2 \int_0^{t_F} d\bar{t}_1 \int_0^{\bar{t}_1} d\bar{t}_2 \langle F|\hat{U}\hat{H}_{\text{R}_0}\hat{H}_{\text{F}}(\bar{t}_1)|n\rangle|n\rangle_{\hat{H}_{\text{R}_0}(\bar{t}_2)}|n\rangle
\]
\[
= \frac{i}{\hbar} e^{-i\bar{t}_F E_{\text{F}}/\hbar} \sum_n \frac{\langle F|\hat{H}|n\rangle\langle n|\hat{H}'|n\rangle\langle n|\hat{H}'|n\rangle\langle n|\hat{H}'|n\rangle}{E_n - E_1} \int_0^{t_F} d\bar{t}_1 \left( e^{\frac{\bar{t}_F}{t_F}} - \frac{e^{\frac{\bar{t}_F}{t_F}}}{\text{Kramers–Heisenberg}} - \frac{e^{\frac{\bar{t}_F}{t_F}}}{\text{Kramers–Heisenberg ignored}} \right). \tag{48}
\]
In the limit \(t_F - t_1 \to \infty\), only the first term in the parentheses of (48) yields a contribution which is proportional to \(t_F - t_1\) and it implements the energy conservation \(\delta(E_{\text{F}} - E_n)\). This contribution leads to the Fermi golden rule (1) and to the Kramers–Heisenberg formula. In the same limit the
second term does not give substantial contribution unless \( E_f \sim E_n \), and this constraint greatly suppresses the number of possible intermediate states \(|n\rangle\), resulting in negligible contribution compared to the first one \[8\]. The constraint \( E_f \sim E_n \) can be enforced by introducing \( \delta(E_f - E_n) \) in the sum over intermediate states. This delta function can be naturally understood as a spectral function which is the imaginary part of retarded Green’s function. Therefore, in our formalism, we have to ignore the Feynman diagrams which have more spectral functions than those which correspond to the Kramers–Heisenberg formula. This assertion will be explicitly demonstrated in section 5. In transition probability, the ignored terms (which is to be squared) will allow processes which are not constrained by the energy conservation \( E_f = E_i \). These processes will turn out to be strongly suppressed by more factors of spectral function. An explicit example will be given in section 5.

So this is the price we have to pay: we have obtained a correlation function which does not depend on intermediate states, but we have to select appropriate Feynman diagrams. Fortunately, the selection of diagrams will turn out to be more or less obvious and straightforward.

We also note that the ignored terms can be eliminated by adiabatic turning on of perturbations \[8\], but in our formalism this method turns out to be very cumbersome, so it is not pursued further.

5. Application: single G-phonon Raman intensity of graphene

In this section we apply the KS functional integral formalism to the case of the single G-phonon Raman intensity of graphene and compare the results with those obtained by conventional approach \[21\] based on the Fermi golden rule.

For explicit calculations we need more elaborations in KS formalism. More details can be found in \[16, 19\]. For fermions let us use the notation \( (\psi_\pm, \psi^*_\pm) \) for coherent state variables instead of \( (\phi_\pm, \phi^*_\pm) \), and reserve the notation \( (\phi_\pm, \phi^*_\pm) \) for bosons. It turns out that in practical calculations it is more convenient to take a linear combination of coherent state variables (often called Keldysh basis) in the following way \[16, 19\]. For bosons, the Keldysh basis is given by \( (\text{‘cl’} = \text{classical}, \text{‘q’} = \text{quantum, see} [16]) \)

\[
\begin{align*}
\phi_{\text{cl}}(k, t) &= \frac{1}{\sqrt{2}}[\phi_+(k, t) + \phi_-(k, t)], & \phi_{\text{q}}(k, t) &= \frac{1}{\sqrt{2}}[\phi_+(k, t) - \phi_-(k, t)], \\
\phi^*_{\text{cl}}(k, t) &= \frac{1}{\sqrt{2}}[\phi^*_+(k, t) + \phi^*_-(k, t)], & \phi^*_{\text{q}}(k, t) &= \frac{1}{\sqrt{2}}[\phi^*_+(k, t) - \phi^*_-(k, t)].
\end{align*}
\]  

(49)

For fermions, a different combination is chosen:

\[
\begin{align*}
\psi_1(k, t) &= \frac{1}{\sqrt{2}}[\psi_+(k, t) + \psi_-(k, t)], & \psi_2(k, t) &= \frac{1}{\sqrt{2}}[\psi_+(k, t) - \psi_-(k, t)], \\
\tilde{\psi}_1(k, t) &= \frac{1}{\sqrt{2}}[\psi^*_+(k, t) - \psi^*_-(k, t)], & \tilde{\psi}_2(k, t) &= \frac{1}{\sqrt{2}}[\psi^*_+(k, t) + \psi^*_-(k, t)].
\end{align*}
\]  

(50)

Then Green’s functions of bosons (such as phonon) are given by \((a, b = \text{cl, q})\)

\[
-i\langle \phi_a(k, t) | \phi^*_b(k, t') \rangle = \hat{D}^{ab}_{k}(t, t') = \begin{pmatrix} D^K_{k}(t, t') & D^R_{k}(t, t') \\ D^K_{k}(t, t') & 0 \end{pmatrix},
\]

(51)

where the superscripts ‘K,R,A’ indicate Keldysh, retarded and advanced Green’s functions, respectively. On the other hand, Green’s functions of fermions are given by \((a, b = 1, 2)\)

\[
-i\langle \psi_a(k, t) | \psi^*_b(k, t') \rangle = \hat{G}^{ab}_{k}(t, t') = \begin{pmatrix} G^K_{k}(t, t') & G^R_{k}(t, t') \\ 0 & G^A_{k}(t, t') \end{pmatrix}.
\]

(52)
At equilibrium, the Keldysh Green’s function is determined by retarded and advanced Green’s functions owing to the fluctuation–dissipation theorem [16, 17]

\[
D^K(\omega) = \coth \frac{\omega}{2k_B T} \left[ D^R(\omega) - D^A(\omega) \right] : \text{bosons,}
\]

\[
G^K(\epsilon) = \tanh \frac{\epsilon}{2k_B T} \left[ G^R(\epsilon) - G^A(\epsilon) \right] : \text{fermions.}
\]

Since the retarded and the advanced Green’s functions in frequency space are complex conjugate of each other, (53) implies that the Keldysh Green’s function is purely imaginary (or anti-Hermitian). We recall the difference between the retarded and the advanced Green’s function in energy space is \(2i\) times the spectral function, which means that in equilibrium the Keldysh Green’s function is always proportional to the spectral function.

Now we turn to the case of graphene (for a review, see [20]). The graphene has honeycomb lattice structure with two sublattices (denoted by A, B in figure 2), so that all the electron Green’s functions become \(2 \times 2\) matrix in sublattice space (the spin will be neglected for simplicity). More concretely,

\[
G_k^R(\epsilon) = \left[ (\epsilon + i0^+)I_2 - \begin{bmatrix} 0 & t_k \\ t_k^* & 0 \end{bmatrix} \right]^{-1}, \quad G_k^A(\epsilon) = \left[ G_k^R(\epsilon) \right]^\dagger, \tag{54}
\]
Figure 3. The lowest order Feynman diagram for one-phonon Raman scattering. The solid lines and the way line represent the electron KS Green’s function and the phonon KS Green’s function, respectively. The black boxes indicate the insertion of current operators of the correlation function (47).

where $t_k = -t_0 \sum_{i=1,2,3} e^{-i k \cdot d_i}$ with $d_i$ being vectors connecting nearest neighbors and $t_0$ is a hopping amplitude. $I_2$ is the identity matrix in sublattice space. The retarded Green’s function of G-phonon ($\vec{u}$ in figure 2) is given by ($M$ is the mass of carbon atom)

$$D^R_q(\omega) = \frac{1}{M (\omega + i 0^+)^2 - \omega_q^2}, \quad D^A_q(\omega) = [D^R_q(\omega)]^*, \quad (55)$$

where $\omega_q$ is the G-phonon dispersion. The Einstein phonon approximation $\omega_q \approx \omega_{\text{ph}}$ will be assumed.

The electric currents of (47) of graphene [21] can be found in terms of coherent state variables using (50)

$$\vec{J}_\pm(q) = (-e) \frac{1}{2} \sum_k \tilde{\Psi}(k-q) E_\pm \otimes \tilde{V}_{k-q/2} \Psi(k), \quad (56)$$

where $\vec{v}_k = \frac{\partial}{\partial k}$, $\Psi = (\psi_{1A}, \psi_{1B}, \psi_{2A}, \psi_{2B})^t$, and

$$E_\pm = \begin{pmatrix} \pm 1 & 1 \\ 1 & \pm 1 \end{pmatrix}, \quad \tilde{V}_{k-q/2} = \begin{pmatrix} 0 & \vec{v}_{k-q/2} \\ \vec{v}_{k-q/2}^* & 0 \end{pmatrix}. \quad (57)$$

For the Raman scattering of graphene, the photon momentum can be neglected, so that $q \approx 0$. Finally, the (simplified) interaction between electron and G-phonon in action form is given by ($N$ is the number of lattice sites)

$$S_{e-ph} = -\lambda \frac{1}{\sqrt{N}} \int dt \sum_{q,k} \tilde{\Psi}(k+q)(u^d_{q}I_2 + u^a_{q}\sigma_z) \otimes \hat{f}_{k,q} \Psi(k), \quad (58)$$

where $\gamma^{cl} = I_2$ (identity matrix) and $\gamma^q = \sigma_z$ (Pauli matrix) act on the Keldysh space and $f_{k,q} = \begin{pmatrix} 0 & \hat{f}_{k,q} \\ \hat{f}_{k,q}^* & 0 \end{pmatrix}$, $u^{cl,q} = \frac{1}{\sqrt{2M_{\omega_q}}} [\phi^{cl,q}(q) + \phi^{cl,q*(-q)}]$ is the phonon coordinate in Keldysh basis and $\lambda$ is the electron–phonon coupling constant. The detailed forms of functions $v_k$ and $f_{k,q}$ do not concern us in this paper.

Now we are ready to compute the single G-phonon Raman intensity graphene in KS functional integral formalism. The single phonon Raman intensity in the lowest order perturbation theory is described by the Feynman diagram of figure 3. The direct application of Feynman rules yields the following result for the correlation function (47):

$$\tilde{C}_{\omega_i,\omega_f} = \lambda^2 M^a_- [iD_{q=0}^{ab}(\omega_i - \omega_f)] M^b_+, \quad a, b = \text{cl, q}, \quad (59)$$

New Journal of Physics 15 (2013) 093039 (http://www.njp.org/)
where \( (M_{a-}) \) is for the loop in the left side of figure 3, and \( M_{b+} \) is for the loop in the right)

\[
M_{a-} = \sum_k \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} \text{Tr}[\hat{G}_k(\epsilon)(E_- \otimes \hat{V}_k)\hat{G}_k(\epsilon + \omega_l)(\gamma^a \otimes \hat{j}_{k,q=0})\hat{G}_k(\epsilon + \omega_l)(E_- \otimes \hat{V}_k)]
\]

\[
+ \text{Tr}[\hat{G}_k(\epsilon)(E_- \otimes \hat{V}_k)\hat{G}_k(\epsilon - \omega_l)(\gamma^a \otimes \hat{j}_{k,q=0})\hat{G}_k(\epsilon - \omega_l)(E_- \otimes \hat{V}_k)]
\]

\[
M_{b+} = \sum_k \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} \text{Tr}[\hat{G}_k(\epsilon)(E_+ \otimes \hat{V}_k)\hat{G}_k(\epsilon - \omega_l)(\gamma^b \otimes \hat{j}_{k,q=0})\hat{G}_k(\epsilon - \omega_l)(E_+ \otimes \hat{V}_k)]
\]

\[
+ \text{Tr}[\hat{G}_k(\epsilon)(E_+ \otimes \hat{V}_k)\hat{G}_k(\epsilon + \omega_l)(\gamma^b \otimes \hat{j}_{k,q=0})\hat{G}_k(\epsilon + \omega_l)(E_+ \otimes \hat{V}_k)]
\]

The trace ‘Tr’ is over both Keldysh and sublattice spaces. The evaluation of the correlation function proceeds as follows: (i) take the trace over Keldysh space; (ii) then using the (anti) Hermiticity properties of matrices of Green’s functions it is easily shown that \( M_{a-}^\dagger = (M_{b+}^\dagger)^* \) and \( M_{a-}^\dagger = -(M_{b+})^* \); and (iii) sort out all terms with more than two Keldysh Green’s function \( G^R \) or spectral functions; these terms belong to the ignored terms discussed in section 4.

Direct integration indeed shows that there is no resonant behavior (roughly two delta functions eliminate all singular behaviors); (iv) after the sorting, a relation \( M_{a-}^\dagger = -M_{a-} \) can be shown by using the Hermiticity of matrices and the property \( \int d\epsilon G^R(\epsilon)G^K(\epsilon \pm \omega_{l/1})G^R(\epsilon \pm \omega_{l/1}) = 0 \) (also for the advanced Green’s function) which follows from the analyticity of the retarded Green’s functions (5) finally using the property \( [G^R]_1 = G^A \) and \( [G^K]_1 = -G^K \), \( M_{a-} \) can be shown to be purely imaginary. Then the correlation function boils down to

\[
\tilde{C}_{\omega_l\tilde{\alpha}_l\omega_l\tilde{\alpha}_l} = \lambda^2 [-(-M_{a-}^\dagger)^2] \left[ D^K(\omega_l - \omega_l) + D^R(\omega_l - \omega_l) - D^A(\omega_l - \omega_l) \right]
\]

\[
= \lambda^2 [-(-M_{a-}^\dagger)^2] \left[ \coth \left( \frac{\omega_l - \omega_l}{2k_BT} \right) + 1 \right] \left[ D^K(\omega_l - \omega_l) - D^A(\omega_l - \omega_l) \right]
\]

\[
= \frac{\lambda^2}{\pi M_{\omega_{ph}}} [-(-M_{a-}^\dagger)^2] \left[ \coth \left( \frac{\omega_l - \omega_l}{2k_BT} \right) + 1 \right] \left[ \delta(\omega_l - \omega_l - \omega_{ph}) - \delta(\omega_l - \omega_l + \omega_{ph}) \right]
\]

where (53) is employed in the second line and the explicit form of phonon Greens’ function (55) is used in the third line. The delta function \( \delta(\omega_l - \omega_l - \omega_{ph}) \) of (62) corresponds to the Stokes process of phonon emission which dominates over the anti-Stokes process of phonon absorption in the low temperature limit due to the thermal occupation number factor \( \coth([\omega_l - \omega_l]/2k_BT) + 1 \). Now the computation of \( M_{a-}^\dagger \) is rather straightforward (but tedious).

Substituting (62) into (23), the integral over the final photon momentum \( k_f \) is done by the delta function of (62), leaving behind an integral over the solid angle of \( k_f \), which defines a differential cross-section. The result obtained in this way is identical with equations (7), (12a) of [21] \( (F_0\alpha_3 \text{ part of (12b,c,d))} \). More precisely, Basko [21] also includes additional phonon–photon coupling specific to graphene which is irrelevant in our discussion.

The Feynman diagram of figure 4 depicts a process for the correlation function (47) which is allowed by Feynman rule but does not satisfy the correct energy conservation, as can be seen clearly in the phonon energy \( \omega_l + \omega_{ph} \). This Feynman diagram corresponds to the squared ignored terms discussed in section 4. Direct computation shows that all terms contributing to this Feynman diagrams contain at least two factors of spectral functions or Keldysh Green function, so they are strongly suppressed and do not contribute to the resonant scattering.
Figure 4. The Feynman diagram which does not satisfy correct energy conservation but is allowed by Feynman rule.

A simple criterion for this type of diagrams is that they have \(E_+\) and \(E_-\) vertices in the same loop (compare with figure 3).

6. ILS for systems in non-equilibrium conditions

The KS functional integral approach for ILS can be readily generalized to the systems in non-equilibrium conditions. In fact, the study of non-equilibrium phenomena was the original motivation for the introduction of KS method. We will assume that the non-equilibrium conditions are created by certain driving forces for the matter systems. Specifically we will consider the semiconductors of direct transition type such as GaAs, where the driving forces are pump pulse laser field which can be treated classically. This pump pulse field prepares the initial matter state \(|i]\) at time \(t_I\) for ILS, and then the probe photon of ILS is incident [22].

In the presence of the pumping field, the time translation invariance is certainly violated, so that the definition of differential cross-section (4) is modified in the following way:

\[
d\sigma = \frac{d}{dt_F} \sum_F \sum_I p_I \frac{|\mathcal{M}_{FI}(t_F, t_I)|^2}{c/V}.
\]

The initial matter state \(|i]\) is prepared from the remote past (say \(-\infty\)) state (which is denoted as \(|i_{-\infty}\]\) by the action of the pumping field and the interactions in matter systems. The pumping field is assumed to be turned off before the probe photon comes in, \(\hat{H}_{\text{pump}}(t) = 0\) for \(t > t_I\). Thus we have

\[
|i\rangle = \hat{U}_{\hat{H}_m + \hat{H}_{\text{pump}}}(t_I, -\infty) |i_{-\infty}\rangle,
\]

where

\[
\hat{U}_{\hat{H}_m + \hat{H}_{\text{pump}}}(t_I, -\infty) = \text{T exp}\left[\frac{-i}{\hbar} \int_{-\infty}^{t_I} d\bar{t} (\hat{H}_m(\bar{t}) + \hat{H}_{\text{pump}}(\bar{t}))\right].
\]

The distribution of the remote past states are described by density matrix

\[
p_{i_{-\infty}} = \langle i_{-\infty}|\hat{\rho}_{m_{-\infty}}|i_{-\infty}\rangle.
\]

Now equation (18) is modified to

\[
\sum_F |\mathcal{M}_{Fij}^{(2)}|^2 = \frac{1}{(2!)^2 \hbar^4} \prod_{l=1}^{4} d\bar{t}_l \langle i_{-\infty}|\hat{U}^\dagger_{\hat{H}_m + \hat{H}_{\text{pump}}}(t_f, -\infty)\hat{D}_l^\dagger (\hat{t}_3, \hat{t}_1) \hat{D}_l (\hat{t}_2, \hat{t}_4)\rangle \hat{U}_{\hat{H}_m + \hat{H}_{\text{pump}}}(t_I, -\infty)|i_{-\infty}\rangle.
\]
Then the differential cross-section takes the form

\[
\frac{1}{(2\pi)^2} \frac{1}{\sqrt{\hbar \nu}} \frac{d}{d\Omega} \sum_{k_i, t_i} \int_{t_i}^{t_f} dt_i \text{Tr}_m \rho_{m,-\infty} \hat{U}_{\text{Rm}+\text{R}_{\text{pump}}}(-\infty, t_i) \times \hat{D}_{\bar{k}_i, \bar{t}_i}(-\hat{t}_i, -\hat{t}_f) \hat{D}_{\bar{k}_i, \bar{t}_i}(-\hat{t}_i, -\hat{t}_f) \hat{U}_{\text{Rm}+\text{R}_{\text{pump}}}(t_i, -\infty). \tag{68}
\]

The expressions for the operators \(\hat{D}_{\bar{k}, \bar{t}}\) and \(\hat{D}_{\bar{k}, \bar{t}}\) (equations (A.6) and (A.7)) remain unchanged since the pumping field is turned off after \(t_i\). Substituting (A.6) and (A.7) into (68) and defining a correlation function

\[
\tilde{C}_{k, \bar{k}, \bar{k}, \bar{t}}(t_3, t_4, t_1, t_2) = \text{Tr}_m \rho_{m,-\infty} \hat{U}_{\text{Rm}+\text{R}_{\text{pump}}}(-\infty, t_i) \tilde{T}[\hat{J}_b(k_i, t) \cdot \hat{e}_i \hat{e}^*_i \hat{J}_b(-k_f, t_i) \cdot \hat{e}_f] \times \hat{T}[\hat{J}_b(-k_i, t_1) \cdot \hat{e}_i \hat{e}^*_i \hat{J}_b(k_f, t_2) \cdot \hat{e}_f] \hat{U}_{\text{Rm}+\text{R}_{\text{pump}}}(t_i, -\infty), \tag{69}
\]

we arrive at

\[
\frac{d\sigma}{d\Omega} = \frac{(2\pi/\hbar)^2}{c \omega_i \omega_f} \frac{d}{d\Omega} \sum_{k_i, t_i} \int_{t_i}^{t_f} (dt_i) e^{-i\omega_i t_i + i\omega_f t_f + i\omega_i t + - i\omega_f t} \tilde{C}_{k, \bar{k}, \bar{k}, \bar{t}}(t_3, t_4, t_1, t_2). \tag{70}
\]

Equation (70) is the differential cross-section of ILS for systems in non-equilibrium conditions in the form of correlation function.

Next we disentangle the time ordering of the correlation function (69) by employing the KS functional integral formulation as in section 3. The relevant correlation function is of the form (in the same notations as those of the section 3)

\[
\tilde{\Pi}([\tau_i]) = \text{Tr}_m \hat{U}_{\text{Rm}+\text{R}_{\text{pump}}}(-\infty, t_i) \tilde{T}[\hat{A}(\tau_i) \hat{B}(\tau_2)] \tilde{T}[\hat{C}(\tau_3) \hat{D}(\tau_4)] \hat{U}_{\text{Rm}+\text{R}_{\text{pump}}}(t_i, -\infty) \rho_{m}. \tag{71}
\]

The difference with equilibrium case (25) lies in the presence of the pumping factor \(\hat{U}_{\text{Rm}+\text{R}_{\text{pump}}}\).

Next we follow the same steps as (27–29). Employing the composition property of the time evolution operator we obtain

\[
\tilde{\Pi}^{(1)}([\tau_j]) = \text{Tr}[\hat{U}_{\text{Rm}+\text{R}_{\text{pump}}}(-\infty, t_i) \hat{U}_{\text{Rm}}(t_i, \tau_1) \hat{A} \hat{U}_{\text{Rm}}(t_1, \tau_2) \hat{B} \hat{U}_{\text{Rm}}(t_2, \tau_f) \hat{U}_{\text{Rm}}(t_f, \tau_3) \times \hat{C} \hat{U}_{\text{Rm}}(t_3, \tau_4) \hat{D} \hat{U}_{\text{Rm}+\text{R}_{\text{pump}}}(t_i, -\infty) \rho(-\infty)], \tag{72}
\]

Since the pumping field is turned off in the time interval \([t_i, t_f]\), the subscripts denoting the Hamiltonian governing time evolution are, in fact, redundant. Namely we can simply take the total Hamiltonian to be \(\hat{H}_{\text{Rm}+\hat{H}_{\text{pump}}}\) with the understanding that the pumping field vanishes for \(t > t_i\). Then again by the composition property of evolution operator, equation (72) reduces to

\[
\tilde{\Pi}^{(1)}([\tau_j]) = \text{Tr}[\hat{U}(-\infty, \tau_1) \hat{A} \hat{U}(\tau_1, \tau_2) \hat{B} \hat{U}(\tau_2, \infty) \hat{U}(+\infty, \tau_3) \hat{C} \hat{U}(\tau_3, \tau_4) \hat{D} \hat{U}(\tau_4, -\infty) \rho(-\infty)], \tag{73}
\]

where we have inserted an identity \(\hat{U}(t_f, +\infty)\hat{U}(+\infty, t_f) = I\). Now equation (73) is of the same form as that of (31), so that they have the identical form of the following functional integral representation:

\[
\tilde{\Pi}([\tau_i]) = \int D[\phi^*, \phi] A(\phi^*_{m} (\tau_1), \phi_-(\tau_1)) B(\phi^*_{m} (\tau_2), \phi_-(\tau_2)) C(\phi^*_{m} (\tau_3), \phi_-(\tau_3)) \times D(\phi^*_{m} (\tau_4), \phi_+(\tau_4)) e^{+iS_1/h - iS_2/h + iS_{-}}, \tag{74}
\]
where the actions $S_{\pm}$ now include the driving pumping field term

$$S_{\pm}[\phi_{\pm}] = \int_{-\infty}^{\infty} dt [i\phi_{\pm}^* \hbar \partial_t \phi_{\pm} - H_m(\phi_{\pm}) - H_{\text{pump}}(\phi_{\pm})].$$

(75)

With equations (74) and (75), ILS in non-equilibrium situation can be computed by using the standard functional integral method.

Next let us apply the above results to the case of ILS of the semiconductor pumped by intense laser pulse. The pumping Hamiltonian in the dipole approximation of optical interband transition [23] is given by (for simplicity two band approximation (one conduction and one valence band) is made and spin degrees of freedom is ignored, see [23, chapter 5])

$$\hat{H}_{\text{pump}} = -E(t) \sum_k (d_{cv} \hat{c}_{c,k}^+ \hat{c}_{v,k} + d_{cv}^* \hat{c}_{v,k}^+ \hat{c}_{c,k}).$$

(76)

where $E(t)$ is the electric field of the pumping pulse laser and $d_{cv}$ is the dipole matrix element. $\hat{c}_{c,v}$ is the electron annihilation operator for conduction and valence band, respectively.

The electric current operator in dipole approximation is given by ($w_{k,q}$ is the optical transition matrix element)

$$\vec{J}_e(q) = \sum_k \left( w_{k,q} \hat{c}_{k+q}^+ \hat{c}_k + w_{k,q}^* \hat{c}_k^+ \hat{c}_{k+q} \right).$$

(77)

In terms of the coherent state variables in Keldysh basis (77) becomes

$$\vec{J}_{e,\pm}(q) = \frac{1}{2} \sum_k \hat{\Psi}(k - q) E_{\pm} \otimes \hat{V}_{k-q/2} \hat{\Psi}(k),$$

(78)

where $\Psi = (\psi_{c1}, \psi_{v1}, \psi_{c2}, \psi_{v2})^T$, and

$$E_{\pm} = \begin{pmatrix} \pm 1 & 1 \\ 1 & \pm 1 \end{pmatrix}, \quad \hat{V}_{k-q/2} = \begin{pmatrix} 0 & \bar{w}_{k-q/2} \\ \bar{w}_{k-q/2}* & 0 \end{pmatrix}.$$  

(79)

Then the action for the pumping term is given by

$$S_{\text{pump}} = \int_{-\infty}^{\infty} dt E(t) \sum_k \hat{\Psi}_k(t) I \otimes \hat{d} \hat{\Psi}_k(t),$$

(80)

where $\hat{d} = \begin{pmatrix} 0 & d_{cv} \\ d_{cv}^* & 0 \end{pmatrix}$.

We can consider only the intraband electron–phonon interaction owing to band gap

$$\hat{H}_{\text{e-ph}} = \frac{\lambda}{\sqrt{N}} \sum_\alpha \sum_{k,q} \hat{c}_{\alpha k+q}^+ \sigma^\alpha_q \hat{c}_{\alpha k}, \quad \alpha = c, v.$$  

(81)

The action corresponding to (81) is

$$S_{\text{e-ph}} = -\frac{1}{\sqrt{N}} \int dt \sum_{q,k} \hat{\Psi}(k + q)(u_q^I I_4 + u_q^3 \sigma_x \otimes I_2) \hat{\Psi}(k),$$

(82)

Now everything boils down to the computation of the following correlation function from which the differential cross-section can be obtained via (70):

$$\tilde{C}_{\vec{k}, t_1, t_2, t_3, t_4} = \int D[\phi_{\pm}, \phi] e^{iS_4/\hbar} \left[ \tilde{J}_e(\phi_{\pm}(k_1, t_1), 0) \cdot \tilde{r}_e^* \right]$$

$$\times \left[ \tilde{J}_e(\phi^{\pm}_{-}(k_2, t_2), \phi_{-}(k_3, t_3)) \cdot \tilde{r}_e \right] \left[ \tilde{J}_e(\phi^{\pm}_{+}(k_4, t_4), \phi_{+}(k_4, t_4)) \cdot \tilde{r}_e^* \right].$$

(83)
The correlation function for the one-phonon Raman scattering (corresponding to (59) in equilibrium case with similar notations) is given by

$$\tilde{C}(t_1, t_2, t_3, t_4) = \lambda^2 \int_{-\infty}^{\infty} dt_5 \int_{-\infty}^{\infty} dt_6 M_{-+}^a(t_1, t_2, t_5)[iD_{q=0}(t_5, t_6)]M_{++}^b(t_3, t_4, t_6),$$

(84)

whose Feynman diagram is given in figure 5. The electron Green’s function satisfy the following Dyson equation which includes the pumping term ($\hat{G}_k^{(0)}(t-t')$ is the non-interacting Green’s function)

$$\hat{G}_k(t, t') = \hat{G}_k^{(0)}(t-t') + \int_{-\infty}^{\infty} dt'' \hat{G}_k^{(0)}(t-t'')\mathcal{E}(t'')I \otimes \hat{d}\hat{G}_k(t'', t'),$$

(85)

(85) follows from the standard Feynman rules. The phonon subsystem is also driven into non-equilibrium through the phonon self-energy. Its influence on ILS can be seen in higher order Feynman diagrams (see figure 6(b)). To best of our knowledge, the ILS in non-equilibrium condition has not been presented in the form of equations (84) and (85) before.

Figure 5. The lowest Feynman diagram for one-phonon Raman scattering in non-equilibrium condition. Note that the solid lines now represent the electron Green function in non-equilibrium (85).

Figure 6. Electron (a) and phonon (b) self-energy contributions to the Feynman diagram of ILS.
investigation of (84) and (85) is beyond the scope of this paper and is left for future studies.

7. Discussions

The KS functional integral approach to ILS possesses several advantages over the conventional approach based on the Fermi golden rule [21] even in the lowest order calculations. Firstly, the finite temperature effect is naturally incorporated as can be seen in (62), which then allows the discussion of both Stokes and anti-Stokes processes on equal footing. Secondly, the energy conservation for one-phonon process has been imposed by hand in [21], while in our approach it is a natural consequence of Feynman rule.

True advantages of KS functional approach lie in the study of higher order many-body effects. In terms of Feynman diagrams the many-body effects are often described by the self-energy corrections and the vertex corrections. For example, the Feynman diagram of the lowest order electron self-energy correction to ILS is given by the diagram (a) of figure 6, and the diagram (b) describes the lowest order phonon self-energy correction. A lowest order vertex correction to ILS is described in figure 7. With phonon self-energy corrections included, the phonon Green’s function in (59) can be taken to be fully interacting Green’s function, so that the phonon frequency shift and the phonon broadening effect (both stemming from phonon self-energy) for ILS can be precisely addressed.

Both self-energy and vertex correction are known to possess the logarithmic corrections [24], which implies that the inclusion of many-body effects are crucial for the proper interpretation of experimental ILS data. Our formalism provides a natural framework for the investigation of such higher order many effects. The study of the above many-body effects based on the Kramers–Heisenberg formula would be exceedingly difficult, if not impossible.

In summary, we have expressed the scattering cross-section of the resonant ILS in the form of correlation function both in equilibrium and non-equilibrium conditions, which then is recast in the framework of the KS functional integral. The correlation function in the KS functional integral can be computed by the Feynman diagram perturbation theory which permits the systematic study of many-body effects.

Acknowledgments

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government Mest (no. 2012-0008058).
Appendix. The calculations of the photon operator expectation values

The Hamiltonian of photon is given by ($\alpha$ is the photon polarization)

$$\hat{H}_p = \sum_{k, \alpha} \hbar \omega_k \left( a_{k\alpha}^{\dagger} a_{k\alpha} + \frac{1}{2} \right), \quad (A.1)$$

where $a_{k\alpha}$ and $a_{k\alpha}^{\dagger}$ are the photon annihilation operator and destruction operator, respectively. The Hamiltonian for the interaction between photon and matter is given by

$$\hat{H}_I(t) = -\frac{1}{c} \int d\vec{r} \hat{J}_c(\vec{r}, t) \cdot \hat{A}(\vec{r}, t), \quad (A.2)$$

where the operators $\hat{J}_c(\vec{r}, t)$ and $\hat{A}(\vec{r}, t)$ have acquired the time dependence via Heisenberg representation with respect to $\hat{H}_0$.

The second quantized vector potential operator $\hat{A}(\vec{r}, t)$ of photon is (in Gaussian unit)

$$\hat{A}(\vec{r}, t) = \frac{1}{\sqrt{\hbar}} \sum_{k, \alpha} \frac{2\pi \hbar c^2}{\omega_k} [\hat{\epsilon}_{k\alpha}^{\dagger} a_{k\alpha} e^{i\vec{k} \cdot \vec{r} - i\omega_k t} + \hat{\epsilon}_{k\alpha} a_{k\alpha}^{\dagger} e^{-i\vec{k} \cdot \vec{r} + i\omega_k t}], \quad (A.3)$$

where $\omega_k = c|\vec{k}|$. We recall the standard photon operator commutation relations

$$[a_{k\alpha}, a_{k'\alpha'}^{\dagger}] = \delta_{k, k'} \delta_{\alpha, \alpha'}, \quad [a_{k\alpha}, a_{k'\alpha'}] = [a_{k\alpha}^{\dagger}, a_{k'\alpha'}^{\dagger}] = 0. \quad (A.4)$$

Employing the commutation relations (A.4) it is straightforward to compute the following expectation values of photon operators:

$$\langle \hat{a}_{k\alpha}^{\dagger} \hat{a}_{k'\alpha'} | \hat{a}_{k'\alpha'}^{\dagger} \hat{a}_{k\alpha} \rangle = \delta_{k, k'} \delta_{\alpha, \alpha'}, \quad \delta_{k, k'} \delta_{\alpha, \alpha'}$$

In our case of ILS the initial and the final photon states are different, so that the second terms of (A.5) vanish.

Now we are ready to compute the operator $\hat{D}_{\vec{k}, \vec{k}'}$ (see equations (13) and (14) for definitions) explicitly. Plugging (A.3) into (A.2) and using the result (A.5) it is straightforward to obtain ($\omega_{i,t} = c|\vec{k}_{i,t}|$)

$$\hat{D}_{\vec{k}, \vec{k}'}(t_1, t_2) = \frac{4\pi \hbar}{\sqrt{\omega_{i,t} \omega_{t',t}}} \int d\vec{r}_1 d\vec{r}_2 T\{ [\hat{J}_c(x_1) \cdot \hat{\epsilon}_i e^{-i\vec{k} \cdot \vec{x}_1}] [\hat{J}_c(x_2) \cdot \hat{\epsilon}_i^{\dagger} e^{i\vec{k} \cdot \vec{x}_2}] \}, \quad (A.6)$$

where $x = (\vec{r}, t)$ and $k = (\vec{k}, \omega_k)$ are four-vectors, and $k \cdot x = \omega_k t - \vec{k} \cdot \vec{r}$. Similarly, we can obtain

$$\hat{D}_{\vec{k}, \vec{k}'}^+(t_3, t_4) = \frac{4\pi \hbar}{\sqrt{\omega_{i,t} \omega_{t',t}}} \int d\vec{r}_3 d\vec{r}_4 T\{ [\hat{J}_c(x_3) \cdot \hat{\epsilon}_i^{\dagger} e^{i\vec{k} \cdot \vec{x}_3}] [\hat{J}_c(x_4) \cdot \hat{\epsilon}_i e^{-i\vec{k} \cdot \vec{x}_4}] \}. \quad (A.7)$$

The spatial Fourier transform of the electric current operator is defined as

$$J^e_{\vec{k}}(t_j) = \int d\vec{r} e^{-i\vec{k} \cdot \vec{r}} J^e_{\vec{k}}(\vec{r}, t_j), \quad (A.8)$$
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