Pump Built-in Hamiltonian Method for Pump-Probe Spectroscopy

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(March 13, 2018)

We propose a new method of calculating nonlinear optical responses of interacting electronic systems. In this method, the total Hamiltonian (system + system-pump interaction) is transformed into a different form that (apparently) does not have a system-pump interaction. The transformed Hamiltonian, which we call the pump built-in Hamiltonian, has parameters that depend on the strength of the pump beam. Using the pump built-in Hamiltonian, we can calculate nonlinear responses (responses to probe beams as a function of the pump beam) by applying the linear response theory. We demonstrate the basic idea of this new method by applying it to a one-dimensional, two-band model, in the case the pump excitation is virtual (coherent excitation). We find that the exponent of the Fermi edge singularity varies with the pump intensity.

Nonlinear optics for semiconductors is one of active fields in solid state physics. Recently, research interests of nonlinear optics are extended to Mott-Hubbard and charge-transfer insulators, for which electron-electron interactions play central roles. The nonlinear optical responses, especially pump-probe spectroscopy, from these systems are experimentally studied, and quite interesting phenomena are reported. However, it is practically impossible to apply the standard method of calculation of $\chi^{(3)}$ to such systems, because it is quite difficult to obtain the wavefunctions of the ground states and the excited state.

In this letter, we propose a new approach, in which the total Hamiltonian $H(t)$ (system Hamiltonian $H_{\text{sys}} +$ system-pump interaction $H_{\text{sys-pump}}(t)$) is transformed into a different form that (apparently) does not have a system-pump interaction. The transformed Hamiltonian is called a “pump built-in Hamiltonian”. If the pump built-in Hamiltonian is tractable, one can calculate nonlinear responses (responses to probe beams as a function of the pump beam) by applying the linear response theory to the pump built-in Hamiltonian. We here present the main idea by explicitly constructing the pump built-in Hamiltonian in a two-band model when the pump excitation is virtual (in the sense we explain below). It is shown that we can include effects of the pump beam as modification of the parameters of the original Hamiltonian in some cases. Namely, the system with a pump beam is equivalent to the system without the pump beam with different values of parameters. Using the pump built-in Hamiltonian, we successfully evaluate the Fermi edge singularity of the absorption of the probe beam. It is shown that the exponent of the singularity varies as a function of the pump intensity. This success indicates that the pump built-in Hamiltonian method could have potential to analyze nonlinear optical responses of various systems in which electron-electron interactions play important roles.

We assume that the pump excitation is “virtual”, i.e., the quantum-mechanical coherence is preserved when the pump beam is shining the sample. The virtual excitation is realized when the spectrum of the pump beam does not overlap with the absorption spectrum of the system. If the state vector $|\Psi(t)\rangle$ of the system is initially the ground state $|G\rangle$ of $H_{\text{sys}}$ before the pump beam is on, it evolves adiabatically the “instantaneous ground state” of $H(t) = H_{\text{sys}} + H_{\text{sys-pump}}(t)$ in the rotating frame. Namely,

$$|\Psi(t)\rangle = |G[E(t)]\rangle,$$

where $|G[E(t)]\rangle$ denotes the ground state of $H(t)$ in the rotating frame when $t$ is regarded as a parameter. It is a function of the instantaneous value of the envelope function $E(t)$ of the pump beam at time $t$. Therefore, it is sufficient to calculate the linear response of $|G[E]\rangle$ to a probe beam for each fixed value of $E$; it gives the nonlinear response of the system at a time $t$ that satisfies $E = \bar{E}(t)$. We denote $H(t)$ for $E = \bar{E}(t)$ simply as $H$.

To demonstrate the basic idea of the pump built-in Hamiltonian method, we here consider a simple model; a one-dimensional, two-band model which has a finite band gap $E_g$. The one-dimensional system may be realized, e.g., in a quantum wire made of semiconductors. The two bands, which we call “electron” and “hole” bands, can be the lowest conduction subband and highest valence subband, or the second and first subbands of the conduction band. The latter case may be more suitable to realize the virtual excitation. We assume that the pump beam is polarized in the direction normal to the quantum wire direction, whereas the probe beam is polarized parallel to the wire. Let $c_{k,\sigma}$ ($c_{k,\sigma}^\dagger$) and $d_{-k,\sigma}$ ($d_{-k,\sigma}^\dagger$) be the Fermi annihilation (creation) operators for an electron and a hole, respectively. We take the model Hamiltonian used in Refs. and , which has the following form in the rotating frame:

$$H =$$
\[ H = H_1 + H_2, \]
\[ H_1 = \sum_{k, \sigma} \left( E_c(k) c_{k\sigma}^\dagger c_{k\sigma} + E_d(k) d_{-k' - \sigma}^\dagger d_{-k' - \sigma} \right) \]
\[ + \sum_{k, \sigma} \lambda(k) (c_{k\sigma} d_{-k' - \sigma} + d_{-k' - \sigma}^\dagger c_{k\sigma}), \]
\[ H_2 = \frac{1}{2} \sum_{\sigma, \sigma', k, k', q} U(q) \left( c_{k+q+\sigma}^\dagger c_{k' - q + \sigma'}^\dagger c_{k' \sigma'} c_{k \sigma} 
\right. \]
\[ + d_{-k' - q - \sigma}^\dagger d_{-k' + q - \sigma'}^\dagger d_{-k' - \sigma'} d_{-k - \sigma} \]
\[ - 2c_{k+q+\sigma}^\dagger c_{k\sigma} d_{-k' + q - \sigma'}^\dagger d_{-k' - \sigma'} \right). \]

Here, the electron and hole energies in the effective-mass approximation are taken as
\[ E_c(k) = \frac{k^2}{2m_c} + E_g - \omega_p, \]
\[ E_d(k) = \frac{k^2}{2m_d}, \]
respectively, where \( m_c \) and \( m_d \) are the electron (\( c \)) and hole (\( d \)) effective masses, and \( \omega_p \) is the central frequency of the pump beam. The \( \lambda(k) \) is the product of \( \mathcal{E} \) and the interband transition dipole. The \( U(q) > 0 \) represents the interaction strength between particles. The detuning energy \( \Delta = E_g - \omega_p \) is assumed positive and large enough to satisfy the condition of the virtual excitation. To simplify the calculations, we assume that the initial state \( |G \rangle \) is two Fermi seas of electrons and holes with the same Fermi wavenumber \( k_F \). This may be realized, e.g., by creating electrons and holes by another optical beam; the pump beam is shined after the carriers cool down. To further simplify the calculations, we consider the case where
\[ E_g \gg |E_c(k_F) - E_d(0)| \gg |U|. \]

Under these conditions, spontaneous band mixing, such as the instability toward an excitonic insulator, are unfavorable. Moreover, many-body scatterings occur only among electrons and holes near their Fermi points. Furthermore, we can neglect interband scatterings (hence such terms have not been included in \( H \)).

Our purpose is to construct Hamiltonian in which the pump term, the second term in Eq. (3), is eliminated. To this end, we first diagonalize \( H_1 \) by the Bogoliubov transformation
\[ \tilde{c}_{k\sigma} = \cos \theta_k c_{k\sigma} + \sin \theta_k d_{-k' - \sigma}, \]
\[ \tilde{d}_{-k\sigma} = \cos \theta_k d_{-k' - \sigma} - \sin \theta_k c_{k\sigma}, \]
as
\[ H_1 = \sum_{k, \sigma} \tilde{E}_+(k) \tilde{c}_{k\sigma}^\dagger \tilde{c}_{k\sigma} + \tilde{E}_-(k) \tilde{d}_{-k' - \sigma}^\dagger \tilde{d}_{-k' - \sigma} \equiv \tilde{H}_1^0, \]

where
\[ \tilde{E}_\pm(k) = \sqrt{E_+(k)^2 + \lambda(k)^2} \pm E_-(k), \]
\[ \cos 2\theta_k = \frac{E_+(k)}{\sqrt{E_+(k)^2 + \lambda(k)^2}}, \]
\[ \sin 2\theta_k = -\lambda(k)/\sqrt{E_+(k)^2 + \lambda(k)^2}, \]
\[ \tilde{E}_\pm(k) \equiv \pm |E_c(k) + E_d(k)|/2. \]

Note that \( \tilde{E}_+ - E_c \) and \( \tilde{E}_- - E_d \) represent the optical Stark shifts of the (non-interacting) electron and hole, respectively.

We then rewrite \( H_2 \) using the transformed operators, \( \tilde{c}, \tilde{c}^\dagger, \tilde{d}, \) and \( \tilde{d}^\dagger \). Substituting these operators in \( H_2 \), and ordering them in their normal order, we obtain \( H_2 = H'_1 + H'_2 + \tilde{H}_2' \). Here \( H'_1 \) is the one-body part, whereas \( H'_2 \) and \( \tilde{H}_2' \) are many-body interactions. The \( H'_2 \) consists of terms of the forms of \( \tilde{c}^\dagger \tilde{c}^\dagger \tilde{c} \tilde{c}, \tilde{d}^\dagger \tilde{d}^\dagger \tilde{d} \tilde{d}, \) and \( \tilde{c}^\dagger \tilde{d}^\dagger \tilde{c} \tilde{d} \). On the other hand, \( \tilde{H}_2' \) consists of other combinations of \( \tilde{c}, \tilde{c}^\dagger, \tilde{d}, \) and \( \tilde{d}^\dagger \). Needless to say, only gauge invariant combinations appear.

Collecting all the one-body terms, we obtain the one-body part of the transformed Hamiltonian, \( \tilde{H}_1 \equiv \tilde{H}_1^0 + \tilde{H}_1' \), in the following form:
These recursion relations are solved numerically. On the other hand, the form of \( \tilde{\lambda} \) is

\[
\tilde{\lambda}(k)(\tilde{c}_{k\sigma}\tilde{d}_{k-\sigma} + \tilde{d}_{-k-\sigma}^{\dagger}\tilde{c}_{k\sigma}^{\dagger}).
\]  

(15)

We consider two simplified cases: (a) \( U(0) \gg U(2k_{F}) \) and (b) \( U(0) \simeq U(2k_{F}) \).

Firstly, we consider case (a), for which backward scattering is negligible. In this case, under the condition (7), we can take \( k = \pm k_{F} \), \( k' = \pm k_{F} \), and \( q \simeq 0 \). Therefore \( \tilde{U}(q, k, k') \) is represented by a single parameter \( \tilde{U}(0, \pm k_{F}, \pm k_{F}) \equiv \tilde{U} \).

We then find that \( \tilde{H}_{2} \) is negligible. Comparing the explicit forms of \( H_{2} \) and \( \tilde{H}_{2} \), we also find that \( \tilde{H}_{2} \) has exactly the same form as \( H_{2} \), and that the parameters are changed as

\[
\tilde{E}_{c/d}(k) = \tilde{E}_{c/d}(k) + \frac{U}{2} \sin^{2}\theta_{k}
\]

\[
-\frac{3U}{2} \sin^{2}\theta_{k} \cos 2\theta_{k} + \frac{U}{2} \sin^{2} 2\theta_{k},
\]  

(17)

\[
\tilde{\lambda}(k) = -\frac{U}{2} \sin 2\theta_{k},
\]  

(18)

and \( \tilde{U} = U \). Furthermore, the Fermi wavenumber of the transformed system, \( \tilde{k}_{F} \), is equal to \( k_{F} \) because we have assumed that \( |E(k_{F}) - E(0)| \gg |U| \) and that the excitation is virtual; in the virtual excitation, the densities of “dressed particles” (\( \tilde{c} \) and \( \tilde{d} \)) are conserved.

Although our purpose is to eliminate the pump term, a new pump term has been generated through the many-body interactions, with the coefficient \( \tilde{\lambda}(k) \). To eliminate this term, we perform Bogoliubov transformations successively: since the form of \( \tilde{H}_{2} \) is the same as \( H_{2} \), we can perform the same procedure successively. The mapping functions from \( \{E_{c}, E_{d}, \lambda\} \to \{\tilde{E}_{c}, \tilde{E}_{d}, \tilde{\lambda}\} \), Eqs. (17) and (18), are denoted by \( f_{j}(E_{c}, E_{d}, \lambda) \) \((j = 1, 2, 3)\). Then the recursion relations between steps \( n \) and \( n + 1 \) are written as follows:

\[
E_{c}^{(n+1)} = f_{1}(E_{c}^{(n)}, E_{d}^{(n)}, \lambda^{(n)}),
\]  

(19)

\[
E_{d}^{(n+1)} = f_{2}(E_{c}^{(n)}, E_{d}^{(n)}, \lambda^{(n)}),
\]  

(20)

\[
\lambda^{(n+1)} = f_{3}(E_{c}^{(n)}, E_{d}^{(n)}, \lambda^{(n)}).
\]  

(21)

These recursion relations are solved numerically.

![FIG. 1. The semi-log plot of \( \lambda^{(n)} \) versus the number of iterations in case (a) \( U(0) \gg U(2k_{F}) \), for several values of \( 4U/\Delta = 6, 4, 2, \) and \( 1 \) (from the top to the bottom). The \( \lambda \) of the original system is fixed.](image-url)
Figure 1 shows the semi-log plot of $\lambda^{(n)}$ for several values of $U/\Delta$, where the pump-system coupling strength of the original system, $\lambda$, is fixed. It is found that $\lambda^{(n)}$ decreases exponentially with increasing the number of iterations, converging to zero for all values of $U/\Delta$. If we define the decay exponent $\mu$ by $\lambda^{(n)} \propto \exp[-\mu n]$, the plot shows that the larger the forward scattering strength is, the smaller the decay exponent is.

![Figure 1](image)

**FIG. 2.** The semi-log plot of $\lambda^{(n)}$ versus the number of iterations in case (a) $U(0) \gg U(2k_F)$, for several values of $4\lambda/\Delta=6$, 4, 2, and 1 (from the top to the bottom on the left vertical axis). The value $4U/\Delta = 1$ is fixed.

Figure 2 shows the semi-log plot of $\lambda^{(n)}$ for several values of $\lambda/\Delta$, where $4U/\Delta = 1$ is fixed. It shows that $\lambda^{(n)}$ converges to zero in the limit of $n \to \infty$ for all values of $\lambda/\Delta$. The plot also shows that the decay exponent is almost independent of $\lambda$ of the original system. We thus find that $\lambda^{(n)}$ converges to zero after iterations for any values of $U > 0$ and $\Delta > 0$, and that the decay exponent depends only on the strength of the forward scattering.

Next the recursion relations for the kinetic energies are considered. Figure 3 shows that the kinetic energies of the electron and the hole in the limit of $n \to \infty$, for $4U/\Delta = 1$ and $4\lambda/\Delta = 1$. The curvatures of the dispersions become slightly smaller than the original ones (dotted lines). Qualitatively similar results are obtained for other values of $U/\Delta > 0$. We thus conclude that the elimination of the pump term makes the masses of electron and hole heavier.

Therefore the pump term is eliminated in the limit of $n \to \infty$, and we obtain the pump built-in Hamiltonian in case (a).

![Figure 3](image)

**FIG. 3.** The dispersion curves of the electron and the hole (black lines) in the limit of $n \to \infty$ in case (a) $U(0) \gg U(2k_F)$. The dotted lines show the initial dispersions.

Secondly, we consider case (b), where $U(0) \simeq U(2k_F)$. This situation may be realized, e.g., for a screened interaction if the screening length is larger than $1/k_F$. By dropping the wave number dependence of the interaction strength, we find

$$\hat{H}_2 = \frac{U}{2} \sum_{\sigma,\sigma',k,k',q} \left( \hat{c}_{k+q,\sigma} \hat{c}_{k',-q,\sigma'} \hat{c}_{k',\sigma'} \hat{c}_{k,\sigma} \right)$$
The built-in Hamiltonian is diagonalized in terms of the charge \( \eta \) of the pump beam. The explicit form of the pump beam in the presence of a pump beam is given by the real part of

\[
\begin{align*}
&+ d_{k-q}^\dagger \sigma d_{k-k'}^\dagger \sigma' d_{k'-\sigma}\sigma \\
&- 2 \tilde{c}^\dagger_{k+q} \tilde{c}_{\sigma} d_{-k-q} \tilde{c}^\dagger_{\sigma'} d_{k'-\sigma'}
\end{align*}
\]

(22)

and \( \tilde{H}_2 = 0 \). As in case (a), we obtain

\[
\tilde{E}_{c/d}(k) = \tilde{E}_{c/d}(k) + \frac{U}{2} I_2
\]

\[
= -\frac{3U}{2} I_2 \cos 2\theta_k + UI_1 \sin 2\theta_k,
\]

(23)

\[
\tilde{\lambda}(k) = -UI_2 \sin 2\theta_k - UI_1 \cos 2\theta_k,
\]

(24)

and \( \tilde{U} = U \). The \( I_{1(2)} \) are defined by \( I_1 = (1/2) \sum_k \sin 2\theta_k \) and \( I_2 = \sum_k \sin^2 \theta_k \). From successive transformations, the recursion relations corresponding to Eqs. (19)–(21) are obtained. Figure 4 is the semi-log plot of \( \lambda^{(n)} \) as a function of the number of iterations. One can see that \( \lambda \) converges to zero exponentially as in case (a), although the decay exponent is smaller than that of case (a). The plot corresponding to Fig. 3 is also obtained, which yields the same conclusion that the elimination of the pump beam makes the effective masses of electron and hole slightly heavier. We thus obtain the pump built-in Hamiltonian in the limit of \( n \to \infty \) also in case (b).

FIG. 4. The semi-log plot of \( \lambda^{(n)} \) against the number of iterations in case (b) \( U(0) = U(2k_F) \), for several values of \( 4U/\Delta = 6 \), 4, 2, and 1 (from the top to the bottom). The \( \lambda \) of the original system is fixed.

Since we have constructed the pump built-in Hamiltonian for both cases (a) and (b), we can discuss nonlinear optical responses by applying the linear response theory, the Kubo formula. For example, the absorption of the probe beam in the presence of a pump beam is given by the real part of

\[
\sigma(q, \omega) = \frac{1}{\omega} \int_{-\infty}^{t} dt' e^{i\omega(t-t')} \langle [j^{\dagger}(q, t), j(q, t')] \rangle,
\]

(25)

where \( j(q, t) \) is the current operator, and \( \langle \cdots \rangle \) denotes the expectation value in the ground state \( |G[\mathcal{E}] \rangle \) of the pump built-in Hamiltonian. For case (a), following Refs. (1) and (3), we evaluate the power-law singularity of the absorption of the probe beam: \( I(\omega) \sim |\omega - E_F|^{\eta(\mathcal{E})^{-1}} \). It is found that the exponent \( \eta(\mathcal{E}) \) varies as a function of the intensity of the pump beam. The explicit form of \( \eta(\mathcal{E}) \) is obtained by applying the bosonization technique, by which pump built-in Hamiltonian is diagonalized in terms of the charge \( (j = 1) \) and spin \( (j = 2) \) excitations. The result is

\[
\eta(\mathcal{E}) = \frac{\pi}{2} \sum_{i=1}^{2} \sum_{j=1}^{2} \left[ \frac{(u_i^j)^2 + (v_j^i)^2}{g_j^i} \sqrt{g_j^i} + 2u_i^j v_j^i \right],
\]

(26)

where \( [u_j^i, v_j^i] = R(\varphi_j) [(v_j^i)^{-1/2}, (v_j^i)^{-1/2}]^t \), \( [v_j^i, v_j^i] = R(\varphi_j) [(v_j^i)^{-1/2}, (v_j^i)^{-1/2}]^t \),
\[
\tilde{g}_j = \left[ \frac{1}{2} v_F^{c*} g_j^c + v_F^{d*} g_j^d \right. \\
\left. \pm \sqrt{(v_F^{c*} g_j^c - v_F^{d*} g_j^d)^2 + 4 \xi_j v_F^{c*} v_F^{d*}} \right],
\]

\( g_1^{c(d)} = v_F^{c(d)*} + U/\pi, g_2^{c(d)} = v_F^{c(d)*} - U/\pi, \xi_1 = -2U/\pi, \text{and} \xi_2 = 0. \) The \( R(\varphi_j) \) is the \( 2 \times 2 \) rotation matrix of angle \( \varphi_j \), where

\[
\varphi_c = \frac{1}{2} \tan^{-1} \left[ 2 \xi_1 \sqrt{v_F^{c*} v_F^{d*}/(v_F^{c*} g_1^c - v_F^{d*} g_1^d)} \right],
\]

and \( \varphi_2 = 0. \) The \( \eta(E) \) is determined by the Fermi velocity and the interaction strength in the pump built-in Hamiltonian. In the model under consideration, the Fermi velocity in the pump built-in Hamiltonian, \( v_F^c \), is smaller than the one without the pump beam, \( v_F \), whereas the interaction strength is unchanged. Hence, \( \eta(E) > \eta \), which should be observable in pump-probe experiments on quantum wires. Since \( |E(k_F) - E(0)| \gg |U| \) is assumed, the difference between \( v_F^c \) and \( v_F \) is small, hence the change of the exponent is also small. If we take other models, however, we expect a larger change.

Finally, we mention another significance of the pump built-in Hamiltonian method. Sato and coworkers recently observed a phase transition induced by a pump beam.\(^{15}\) Miyashita and coworkers recently suggested that this phenomenon can be explained if one assumes that parameters in the Hamiltonian varies as a function of the pump intensity.\(^{14}\) Since the pump built-in Hamiltonian has this property, the pump built-in Hamiltonian method may provide for the microscopic foundation of their assumption. However, we do not exclude other possibilities, such as a thermal effect, for the origin of the changes of the parameters in the experiment of Ref.\(^{15}\). Further studies are needed for settling it.

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