

# A Scaling Relation of Two-Photon Absorption in Antiferromagnetic Insulators

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We discuss some properties of nonlinear absorption spectrum in insulators with the gap originating from the electron correlation. We formulate a method of evaluating the nonlinear susceptibility and apply this to the antiferromagnetic insulator. A scaling relation for the two-photon absorption is derived and applied to a quantitative estimation, which gives values comparable with experiments on Mott insulators. The direct transition term is found to be dominant in many-body systems. It is also shown that our formulation naturally introduces the final-states interaction (the charge fluctuation and the excitonic effect), which gives sufficient contribution to two-photon absorption spectrum.

KEYWORDS: nonlinear optics, antiferromagnetic insulator, two-photon absorption, electron correlation, Mott insulator

## 1. Introduction

It has been found that several kinds of nonlinear optical responses in low-dimensional strongly correlated electron systems (SCES) are large compared with those of band insulators.<sup>1-3)</sup> The following becomes a subject of discussion. It is known that the scaling relation of the two-photon absorption (TPA) exists in semiconductors.<sup>4,5)</sup> As compared with this scaling relation, TPA in SCES is large.<sup>1)</sup> In these systems, however, how a scaling relation behaves has not been known and there has been no quantitative estimation. In this paper we consider a scaling relation in insulators with the gap originating from the electron correlation. We evaluate TPA in antiferromagnetic (AF) insulators and then make relation to Mott insulators.

This paper includes presentation of a new method of evaluating the nonlinear optical susceptibility. The reason why we require this method is as follows. So far methods of the transition probability<sup>6)</sup> and the spectral representation with discrete energy levels and dipole moments between them<sup>7)</sup> are known. These are useful in the case where energy levels and wave functions are determined or small systems which can be diagonalized numerically. Here Green's function method is presented. Although this is equivalent to the above two methods in principle in the case we can calculate exactly, this method is appropriate to include the many-body effect systematically in the bulk system. The calculation of the small system with an

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artificial broadening of discrete spectrum is not considered to give quantitatively appropriate information.

We use the Hartree-Fock method with the conserving approximation,<sup>8)</sup> however, it is shown that AF insulators have different features of absorption spectrum compared with the Peierls insulator. The direct transition term is found to be dominant in SCES because the gap results from the electron correlation. This term is obtained as a result of the systematic derivation of the formula for the nonlinear optical susceptibility. The other result obtained by this formulation is the final-states interaction in the nonlinear response. This interaction effect is naturally introduced by Green's function method with the conserving approximation. There are two main effects; the charge fluctuation and the excitonic effect. We finally obtain a scaling relation which is applied to a quantitative estimation and gives results comparable with experiments in order of magnitude. (Here we do not mean a verification of the functional form because of the absence of systematic variation of the energy gap in experiments. This is rather a problem of consistency between  $\beta$  and  $1/U$  (§3.3).) This relation also poses a question about the interpretation based on the spin-charge separation in previous works.

## 2. Formalism

We consider the nonlinear optical susceptibility ( $\chi^{(3)}$ ) defined as follows with the nonlinear polarization ( $P^{(3)}$ ) and the external field ( $E(\omega)$ ) (see Appendix for definitions of  $\chi^{(3)}$  and  $K^{(3)}$ ).

$$P^{(3)}(\omega) = \iiint d\omega_1 d\omega_2 d\omega_3 \chi^{(3)}(\omega, \omega', \omega'') E(\omega_3) E(\omega_2) E(\omega_1) \quad (1)$$

with  $\omega = \omega_1 + \omega_2 + \omega_3$ ,  $\omega' = \omega_2 + \omega_3$  and  $\omega'' = \omega_3$ , and

$$\chi^{(3)}(\omega, \omega', \omega'') = \frac{K^{(3)}(\omega, \omega', \omega'')}{\omega\omega_1\omega_2\omega_3} \quad (2)$$

Here the correlation function  $K^{(3)}$  is written with the current operator. The diagrammatic calculation to include the many-body effect is usually performed with use of the Matsubara frequency, and therefore the analytic continuation is required as follows.

$$K^{(3)}(i\omega_p, i\omega_q, i\omega_r) \xrightarrow{\text{a.c.}} K^{(3)}(\omega, \omega', \omega'') \quad (3)$$

Here  $\omega_l = 2\pi Tl$  ( $T$  is temperature and  $l$  is integer) and  $\xrightarrow{\text{a.c.}}$  indicates that  $i\omega_p \rightarrow \omega + i\delta_p$ ,  $i\omega_q \rightarrow \omega' + i\delta_q$  and  $i\omega_r \rightarrow \omega'' + i\delta_r$ . The relation between the response function and the correlation function is not trivial especially in the nonlinear response. The correct analytic continuation to obtain the causal response from the correlation function  $K^{(3)}$  is given by putting  $\delta_{p,q,r} \rightarrow +0$  on the condition that  $\delta_p > \delta_q > \delta_r$ . This formalism is equivalent to Keldysh's time dependent Green's function technique<sup>9)</sup> if the conserving approximation is used. The susceptibility with the polarization operator is equivalent to that of the current operator in the long-wavelength limit (see Appendix). In this paper we consider the current-current cor-

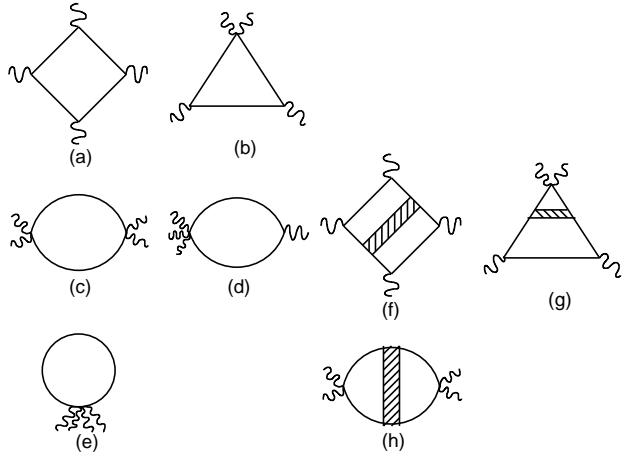


Fig. 1. The diagrammatic representation of  $K^{(3)}$  in our approximation. (The full diagrammatic representation is presented in ref.<sup>10</sup>) The solid lines express the one-particle Green function. The vertex with  $n$ -wavy lines implies  $\partial^{(n-1)}\hat{v}_k/\partial k^{(n-1)}$ . The shaded rectangle denotes the reducible four-point vertex  $\hat{\Gamma}$  (the generalized final-states interaction).

relation function because we take the kinetic energy as the unperturbed Hamiltonian. The expression without vertex corrections is written as follows, with the Green function  $\hat{G}$  and the velocity  $\hat{v}$  defined in specific models. (We put  $e = c = \hbar = 1$  except in the quantitative evaluation.)

$$\begin{aligned}
K^{(3)}(i\omega_p, i\omega_q, i\omega_r) = & \frac{1}{3!} \left\{ \sum_{\{i_1, j_1\}} T \sum_{k, n} \text{Tr}[\hat{v}\hat{G}_1 \hat{v}\hat{G}_{i_1} \hat{v}\hat{G}_{j_1} \hat{v}\hat{G}_8] \right. \\
& + \sum_{\{j_2\}} T \sum_{k, n} \text{Tr}[\hat{v}\hat{G}_1 \frac{\partial \hat{v}}{\partial k} \hat{G}_{j_2} \hat{v}\hat{G}_8] + \sum_{\{i_2\}} T \sum_{k, n} \text{Tr}[\hat{v}\hat{G}_1 \hat{v}\hat{G}_{i_2} \frac{\partial \hat{v}}{\partial k} \hat{G}_8] \\
& + \sum_{\{i_1, j_1\}} T \sum_{k, n} \text{Tr}[\frac{\partial \hat{v}}{\partial k} \hat{G}_{i_1} \hat{v}\hat{G}_{j_1} \hat{v}\hat{G}_8] + \sum_{\{i_2\}} T \sum_{k, n} \text{Tr}[\frac{\partial \hat{v}}{\partial k} \hat{G}_{i_2} \frac{\partial \hat{v}}{\partial k} \hat{G}_8] \\
& \left. + \sum_{\{j_2\}} T \sum_{k, n} \text{Tr}[\frac{\partial^2 \hat{v}}{\partial k^2} \hat{G}_{j_2} \hat{v}\hat{G}_8] + T \sum_{k, n} \text{Tr}[\hat{v}\hat{G}_1 \frac{\partial^2 \hat{v}}{\partial k^2} \hat{G}_8] + T \sum_{k, n} \text{Tr}[\frac{\partial^3 \hat{v}}{\partial k^3} \hat{G}_8] \right\}
\end{aligned} \tag{4}$$

The suffices take the following combinations,  $\{i_1, j_1\} = \{57, 56, 37, 34, 26, 24\}$ ,  $\{i_2\} = \{5, 3, 2\}$ ,  $\{j_2\} = \{7, 6, 4\}$ . Here  $1 \sim 8$  indicate  $i\epsilon_n + i\omega_p$ ,  $i\epsilon_n + i\omega_p - i\omega_r$ ,  $i\epsilon_n + i\omega_p - i\omega_q + i\omega_r$ ,  $i\epsilon_n + i\omega_p - i\omega_q$ ,  $i\epsilon_n + i\omega_q$ ,  $i\epsilon_n + i\omega_q - i\omega_r$ ,  $i\epsilon_n + i\omega_r$ ,  $i\epsilon_n$ , respectively ( $\epsilon_n = \pi T(2n + 1)$ ,  $n$  is integer). The diagrammatic representation is given in (a ~ e) of Fig. 1. All these terms are necessary for the correct behavior at  $\omega \rightarrow 0$ ,  $\omega' \rightarrow 0$  and  $\omega'' \rightarrow 0$ . (In the case of many-body systems the problem of the zero frequency divergence<sup>11</sup>) is solved with use of the conserving approximation.) Usually only the first term (a) is effective and other terms has been neglected, however, we see that this does not necessarily hold in insulators with the gap originating from the electron correlation.

The model and approximation for AF insulator and the Peierls insulator are as follows.

In the case of AF insulator, the Hamiltonian is the Hubbard model with the on-site Coulomb interaction  $U$ ,

$$\mathcal{H} = \mathcal{H}_t + \mathcal{H}_U.$$

Here  $\mathcal{H}_t = \sum_{\langle ij \rangle \sigma} t_{ij} (c_{i\sigma}^\dagger c_{j\sigma} + c_{j\sigma}^\dagger c_{i\sigma})$  and  $\mathcal{H}_U = \frac{U}{2} \sum_{i\sigma} n_{i\sigma} n_{i-\sigma}$  ( $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$ ,  $\langle ij \rangle$  indicates the pair of the nearest neighbor sites and we put the transfer integral  $t_{ij} = -t$ ). The one-particle Green function in the Hartree-Fock approximation is

$$\hat{G}_k(i\epsilon_n) = \frac{1}{(i\epsilon_n)^2 - (\xi_k^2 + \Delta^2)} \begin{pmatrix} i\epsilon_n + \Delta & \xi_k \\ \xi_k & i\epsilon_n - \Delta \end{pmatrix}.$$

Here  $\Delta = \frac{U}{2} | \langle n_\uparrow \rangle - \langle n_\downarrow \rangle |$  (We consider  $U \geq 5t$  in the numerical calculation and then we can put  $\Delta = U/2$ .) and the velocity is  $\hat{v}_k = \begin{pmatrix} 0 & v_k \\ v_k & 0 \end{pmatrix}$ . ( $\xi_k = -2t\cos k$ ,  $v_k = \partial \xi_k / \partial k$ . We put the lattice constant  $a = 1$  hereafter except in the quantitative evaluation.) We also consider the two-dimensional AF insulator. In this case  $\xi_k = -2t(\cos k_x + \cos k_y)$ ,  $v_{k_x} = \partial \xi_k / \partial k_x$ .

In the case of the Peierls insulator,

$$\mathcal{H} = - \sum_{i\sigma} \frac{1}{2} (t + (-1)^i \Delta) (c_{i+1\sigma}^\dagger c_{i\sigma} + c_{i\sigma}^\dagger c_{i+1\sigma}),$$

$$\hat{G}_k(i\epsilon_n) = \frac{1}{(i\epsilon_n)^2 - |\xi_k|^2} \begin{pmatrix} i\epsilon_n & \xi_k^* \\ \xi_k & i\epsilon_n \end{pmatrix}$$

and  $\hat{v}_k = \begin{pmatrix} 0 & v_k^* \\ v_k & 0 \end{pmatrix}$ . ( $\xi_k = -t\cos k - i\Delta \sin k$ ,  $v_k = \partial \xi_k / \partial k$ ).

### 3. Results

#### 3.1 Analytic results without vertex corrections

Briefly we see linear absorption spectrum. The correlation function  $K^{(1)}$  is written as in eq. (4);

$$K^{(1)}(i\omega_l) = T \sum_{n,k} \text{Tr}[\hat{v}_k \hat{G}_k(i\epsilon_n + i\omega_l) \hat{v}_k \hat{G}_k(i\epsilon_n)] + T \sum_{k,n} \text{Tr}[\frac{\partial \hat{v}_k}{\partial k} \hat{G}_k(i\epsilon_n)] e^{i\epsilon_n 0^+}. \quad (5)$$

Then linear absorption spectrum ( $\text{Im}\chi^{(1)}(\omega) = -\text{Im}K^{(1)}(\omega)/\omega^2$ ) is as follows.

$$\text{Im}\chi^{(1)}(\omega) = \frac{-1}{\omega^2} \sum_k \int \frac{d\epsilon}{2\pi} \left( \tanh \frac{\epsilon}{2T} - \tanh \frac{\epsilon + \omega}{2T} \right) \text{Tr} \left[ \hat{v}_k \text{Im}\hat{G}_k^R(\epsilon + \omega) \hat{v}_k \text{Im}\hat{G}_k^R(\epsilon) \right] \quad (6)$$

Here  $G_k^R(\epsilon) = G_k(i\epsilon_n \rightarrow \epsilon + i\delta)$  is the retarded Green function. We consider the case of  $\omega \gg T$  and then  $(\tanh \frac{\epsilon}{2T} - \tanh \frac{\epsilon + \omega}{2T}) \neq 0$  only in the case of  $\epsilon \times (\epsilon + \omega) < 0$ . This simplifies the matrix calculation when we put the above  $\hat{G}_k(i\epsilon_n)$  into this equation. In AF insulator,

$$\text{Im}\chi^{(1)}(\omega) = \frac{1}{\omega} \sum_k \delta(\omega - 2\sqrt{\xi_k^2 + \Delta^2}) \frac{\Delta^2}{\xi_k^2 + \Delta^2} v_k^2 \quad (7)$$

The summation over  $k$  is replaced by the integration as usual and the result is as follows.

$$\text{Im}\chi^{(1)}(\omega) = \frac{2\Delta^2 \sqrt{16t^2 + 4\Delta^2 - \omega^2}}{\omega^3 \sqrt{\omega^2 - 4\Delta^2}}. \quad (8)$$

In the two dimensional case,

$$\text{Im}\chi^{(1)}(\omega) = \frac{\Delta^2}{\pi\omega^3} \frac{1}{\eta} \int_{\eta-1}^1 dx \frac{\sqrt{1-x^2}}{\sqrt{1-(\eta-x)^2}}. \quad (9)$$

Here  $\eta = \frac{\sqrt{(\omega/2)^2 - \Delta^2}}{2t} \leq 2$ . The integral is also written with the elliptic function. In the Peierls insulator,

$$\text{Im}\chi^{(1)}(\omega) = \frac{4t^2 \Delta^2}{\omega^3 \sqrt{\omega^2 - 4\Delta^2} \sqrt{4t^2 + 4\Delta^2 - \omega^2}}. \quad (10)$$

We see that one-photon absorption (OPA) spectrum shows the allowed band-edge behavior in both of AF insulator and the Peierls insulator. In the two dimensional case there is also the lower band-edge divergence, contrary to the continuum model. However it is shown that the width of the divergent region is narrow for the two dimensional case than the one dimensional case.

Next we see how the above behavior changes in TPA. The vertex correction exists in many body systems but we discuss it later because the numerical calculation is required. We consider the case of  $\omega_1 = -\omega_2 = \omega_3 = \omega$  in eqs. (2) and (4). Then two-photon absorption spectrum is written as follows. (We write only the relevant terms; the corresponding combinations of the frequency indices in eq. (4) are {1568, 1378, 1348, 1268}, {168}, {138}, {378, 348}, {38} for the first  $\sim$  fifth term, respectively.)

$$\begin{aligned} \text{Im}\chi^{(3)}(\omega, 0, \omega) = & \frac{2}{3! \omega^4} \int \frac{d\epsilon}{2\pi} \{ 4\text{Tr}[\hat{v}_k \text{Re}\hat{G}_k^R(\epsilon + \omega) \hat{v}_k \text{Im}\hat{G}_k^R(\epsilon + 2\omega) \hat{v}_k \text{Re}\hat{G}_k^R(\epsilon + \omega) \hat{v}_k \text{Im}\hat{G}_k^R(\epsilon)] \\ & + 4\text{Tr}[\hat{v}_k \text{Re}\hat{G}_k^R(\epsilon + \omega) \hat{v}_k \text{Im}\hat{G}_k^R(\epsilon + 2\omega) \frac{\partial \hat{v}_k}{\partial k} \text{Im}\hat{G}_k^R(\epsilon)] \\ & + \text{Tr}[\frac{\partial \hat{v}_k}{\partial k} \text{Im}\hat{G}_k^R(\epsilon + 2\omega) \frac{\partial \hat{v}_k}{\partial k} \text{Im}\hat{G}_k^R(\epsilon)] \} \end{aligned} \quad (11)$$

Here we replace the thermal factor,  $\tanh \frac{\epsilon}{2T} - \tanh \frac{\epsilon+2\omega}{2T}$ , by  $-2$  with the same reason as in the linear absorption. We also shift the frequency in the integral for some terms, which simplifies the result. In AF insulator (we write the result in the following functional form to compare with that of ref.<sup>4)</sup>)

$$\text{Im}\chi^{(3)}(\omega, 0, \omega) = \frac{1}{\Delta^3} \left[ \frac{4}{3} F_{\text{af}}^{(a)} \left( \frac{\omega}{\Delta} \right) + F_{\text{af}}^{(b)} \left( \frac{\omega}{\Delta} \right) + \frac{1}{12} F_{\text{af}}^{(c)} \left( \frac{\omega}{\Delta} \right) \right]. \quad (12)$$

Here,

$$F_{\text{af}}^{(a)} \left( \frac{\omega}{\Delta} \right) = \frac{\sqrt{(\omega/\Delta)^2 - 1} ((2t/\Delta)^2 + 1 - (\omega/\Delta)^2)^{3/2}}{(\omega/\Delta)^9},$$

$$F_{\text{af}}^{(b)}\left(\frac{\omega}{\Delta}\right) = \frac{\sqrt{(\omega/\Delta)^2 - 1}\sqrt{(2t/\Delta)^2 + 1 - (\omega/\Delta)^2}}{(\omega/\Delta)^7},$$

and

$$F_{\text{af}}^{(c)}\left(\frac{\omega}{\Delta}\right) = \frac{\sqrt{(\omega/\Delta)^2 - 1}}{(\omega/\Delta)^5\sqrt{(2t/\Delta)^2 + 1 - (\omega/\Delta)^2}}.$$

Here  $(a, b, c)$  correspond to those of Fig. 1 and also to the first, the second and the third term in eq. (11), respectively.  $\text{Im}K^{(3b,c)}$  take finite values and give large contribution to  $\chi^{(3)}$ , especially in the case of large  $\Delta$ , compared with  $\text{Im}K^{(3a)}$  (see Fig. 7). This is contrary to the case of the band insulator as noted below and ref.<sup>12)</sup> The two dimensional case is similarly written as in the case of  $\text{Im}\chi^{(1)}$  with  $\eta = \frac{\sqrt{\omega^2 - \Delta^2}}{2t} \leq 2$ . In the Peierls insulator,

$$\text{Im}\chi^{(3)}(\omega, 0, \omega) = \frac{F_{\text{pr}}(\omega/\Delta)}{6\Delta^3} \left(\frac{t}{2\Delta}\right)^2. \quad (13)$$

Here

$$F_{\text{pr}}\left(\frac{\omega}{\Delta}\right) = \frac{\sqrt{(\omega/\Delta)^2 - 1}\sqrt{(t/\Delta)^2 - (\omega/\Delta)^2}}{(\omega/\Delta)^9}.$$

The band-edge behavior at low energy is same as in ref.<sup>14)</sup> where the linearized dispersion is used.

In both of the one-dimensional case and the Peierls insulator TPA shows the forbidden band-edge behavior ( $\propto \sqrt{(\omega/\Delta)^2 - 1}$ ), contrary to OPA. However there are two different points; the existence of the direct transition term ((b,c) terms) and vertex corrections. The latter is discussed in the next subsection and we discuss the former here. (The direct transition means that the coupling to the external field by  $\partial v/\partial k$  makes possible the transition to the upper band by frequency  $2\omega$ . On the other hand the coupling by  $v$  indicates  $\omega$ -absorption and then the two-photon absorption is made possible only by way of the virtual states.)

In the Peierls insulator  $\text{Im}K^{(3b,c)}(\omega, 0, \omega) = 0$  and only the self-transition terms are effective. This is same as in semiconductors.<sup>13)</sup> In the band insulator the energy gap results from the difference between the transfer integrals. Therefore the upper and lower bands do not couple with each other by  $\partial\hat{v}/\partial k$ . (Mathematically, the matrix  $\hat{G}$  and  $\partial\hat{v}/\partial k$  can be diagonalized at the same time.) On the other hand the gap in AF insulator results from the electron correlation, then the coupling between the upper and lower bands by  $\partial\hat{v}/\partial k$  remains. With the notation in Appendix,  $j^d$  is a conserved quantity in the absence of the electron correlation (i.e.  $[\mathcal{H}_t, j^d] = 0$ ), and then the absorption by  $j^d$ -term at the finite energy vanishes. On the other hand  $[\mathcal{H}_U, j^d] \neq 0$  and this makes the absorption by  $j^d$ -term possible. (Strictly speaking this kind of absorption also exists in the Peierls insulator because the electron correlation would not vanish. However the origin of the gap indicates that the transfer term is dominant and then our conclusion is considered to hold in this case.)

The functional form in the semiconductor<sup>4)</sup> is different from both of the above two. These differences result from the dimensionality and the origin of the gap. Therefore the absolute

value of TPA in SCES cannot be measured by the relation derived in semiconductors.

### 3.2 Vertex corrections

Next we consider the vertex correction on the basis of the conserving approximation. (The diagrammatic representation is given in  $(f, g, h)$  of Fig. 1. These are relevant terms to TPA. We omit the vertex correction which is attached to the single external field line and irrelevant to TPA.) This effect is interpreted as the generalization of the final-states interaction. For an example the excitonic effect arises from this type of interaction. Therefore we consider the extended Hubbard model with the nearest neighbor interaction,

$$\mathcal{H} = \mathcal{H}_t + \mathcal{H}_U + \mathcal{H}_V.$$

Here  $\mathcal{H}_V = V \sum_{i\sigma\sigma'} n_{i\sigma} n_{i+1\sigma'}$ . With use of the Hartree-Fock approximation, the vertex correction to the correlation function is written as follows.

$$K_{\text{vc}}^{(3)}(i\omega_p, i\omega_q, i\omega_r) = K_{\text{vc},f}^{(3)}(i\omega_p, i\omega_q, i\omega_r) + K_{\text{vc},g}^{(3)}(i\omega_p, i\omega_q, i\omega_r) + K_{\text{vc},h}^{(3)}(i\omega_p, i\omega_q, i\omega_r).$$

Here,

$$K_{\text{vc},f}^{(3)}(i\omega_p, i\omega_q, i\omega_r) = \sum_{\{i,j,l,l'\}} \frac{-1}{3} \sum_{k,k'} (T \sum_n \hat{G}_j \hat{v} \hat{G}_l \hat{v} \hat{G}_i)_{\vec{k}} \hat{\Gamma}_{k,k'}(i-j) (T \sum_n \hat{G}_i \hat{v} \hat{G}_{l'} \hat{v} \hat{G}_j)_{k'},$$

$$K_{\text{vc},g}^{(3)}(i\omega_p, i\omega_q, i\omega_r) = \sum_{\{i,j,l\}} \frac{-1}{3} \sum_{k,k'} (T \sum_n \hat{G}_j \frac{\partial \hat{v}}{\partial k} \hat{G}_i)_{\vec{k}} \hat{\Gamma}_{k,k'}(i-j) (T \sum_n \hat{G}_i \hat{v} \hat{G}_l \hat{v} \hat{G}_j)_{k'}$$

and

$$K_{\text{vc},h}^{(3)}(i\omega_p, i\omega_q, i\omega_r) = \sum_{\{i,j\}} \frac{-1}{3} \sum_{k,k'} (T \sum_n \hat{G}_j \frac{\partial \hat{v}}{\partial k} \hat{G}_i)_{\vec{k}} \hat{\Gamma}_{k,k'}(i-j) (T \sum_n \hat{G}_i \frac{\partial \hat{v}}{\partial k} \hat{G}_j)_{k'}.$$

Here  $\vec{(\hat{A})}$  ( $(\hat{A})$ ) indicates the column (row) vector made by four components of  $2 \times 2$  matrix  $\hat{A}$ ,  $(\hat{A}) = (A_{11}, A_{22}, A_{12}, A_{21})$ .  $i, j, l, l'$  take the appropriate combinations of  $1 \sim 8$  as in eq. (4). Each of  $\sum_{\{i,j,l,l'\}}$  and  $\sum_{\{i,j,l\}}$  contains 12 terms, and  $\sum_{\{i,j\}}$  3 terms. In this two-photon absorption calculation,  $\{i, j, l, l'\} = \{3, 8, 1, 7\}, \{3, 8, 1, 4\}, \{1, 6, 8, 2\}, \{1, 6, 8, 5\}$  in  $\sum_{\{i,j,l,l'\}}$ ,  $\{i, j, l\} = \{1, 6, 8\}, \{3, 8, 1\}, \{3, 8, 4\}, \{3, 8, 7\}$  in  $\sum_{\{i,j,l\}}$  and  $\{i, j\} = \{2, 8\}$  in  $\sum_{\{i,j\}}$ . The reducible four-point vertex  $\hat{\Gamma}$  is defined as

$$\hat{\Gamma}_{k,k'}(i\omega_l) = \hat{I}_{k,k'} + \sum_{k_1} \hat{I}_{k,k_1} \hat{g}_{k_1}(i\omega_l) \hat{\Gamma}_{k_1,k'}(i\omega_l). \quad (14)$$

The diagrammatic representation of  $\Gamma$  and  $I$  are given in Fig. 2. Here  $\hat{I}$  is the irreducible four-point vertex which is the functional derivative of the self-energy by the one-particle Green function,<sup>8)</sup>  $\hat{I}_{k,k'} = \begin{pmatrix} \hat{I}^u & \hat{0} \\ \hat{0} & \hat{I}_{k,k'}^v \end{pmatrix}$ . Here,  $\hat{I}^u = \frac{U}{2} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$ ,  $\hat{I}_{k,k'}^v = \frac{-V_{k-k'}}{2} \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$  and

$$g_k^{\epsilon\xi,\eta\zeta}(i\omega_l) = T \sum_n G_k^{\xi\eta}(i\epsilon_n + i\omega_l) G_k^{\zeta\epsilon}(i\epsilon_n).$$

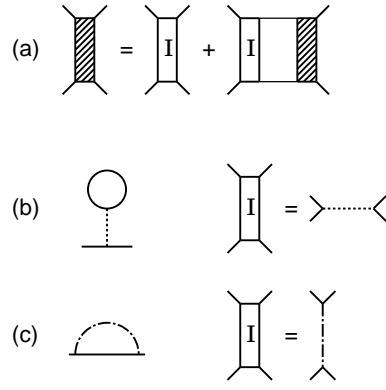


Fig. 2. (a) The diagrammatic representation of the integral equation (14). The shaded rectangle denotes  $\hat{\Gamma}$  as in Fig. 1. The blank rectangle denotes the irreducible four-point vertex  $\hat{I}$ . (b) The self-energy including  $U$ -term and its vertex correction  $\hat{I}^u$ . (c) The self-energy including  $V$ -term and its vertex correction  $\hat{I}^v$ .

Here  $G^{ij}$  is  $(ij)$ -component of  $\hat{G}$ . In the extended Hubbard model  $V_{k-k'} = V(\text{cos}k\text{cos}k' + \text{sin}k\text{sin}k')$ . The first and the second term are relevant to TPA and OPA, respectively. In the case of TPA the solution to the integral equation of  $\hat{\Gamma}$  is written as follows.

$$\hat{\Gamma}_{k,k'}(i\omega_l) = \begin{pmatrix} \hat{\Gamma}_u(i\omega_l) & \hat{\Gamma}_a(i\omega_l)\text{cos}k' \\ \text{cos}k\hat{\Gamma}_a^t(i\omega_l) & \text{cos}k\hat{\Gamma}_v(i\omega_l)\text{cos}k' \end{pmatrix}. \quad (15)$$

The components of this matrix are,  $\Gamma_v^{11} = -(V/2)[(1 + Ug_u)(1 + Vg_v^{22}/2) - UV(g_a^{22})^2/2]/D$ ,  $\Gamma_v^{22} = -(V/2)[(1 + Ug_u)(1 + Vg_v^{11}/2) - UV(g_a^{11})^2/2]/D$ ,  $\Gamma_v^{12} = \Gamma_v^{21} = (V/2)[(1 + Ug_u)Vg_v^{12}/2 + UVg_a^{11}g_a^{22}/2]/D$ ,  $\Gamma_a^{11} = -\Gamma_a^{21} = (UV/4)[(1 + Vg_v^{22}/2)g_a^{11} + Vg_v^{12}g_a^{22}/2]/D$ ,  $\Gamma_a^{22} = -\Gamma_a^{12} = (UV/4)[(1 + Vg_v^{11}/2)g_a^{22} + Vg_v^{12}g_a^{11}/2]/D$ ,  $\Gamma_u^{11} = \Gamma_u^{22} = U/4 - (U/4)[(1 + Vg_v^{11}/2)(1 + Vg_v^{22}/2) - (Vg_v^{12}/2)^2]/D$ ,  $\Gamma_u^{12} = \Gamma_u^{21} = U/2 - \Gamma_u^{11}$ . Here

$$D(i\omega_l) = (1 + Ug_u)[(1 + Vg_v^{11}/2)(1 + Vg_v^{22}/2) - (Vg_v^{12}/2)^2] - UV[(1 + Vg_v^{11}/2)g_a^{22} + (1 + Vg_v^{22}/2)g_a^{11} + Vg_v^{12}g_a^{11}g_a^{22}], \quad (16)$$

and

$$\hat{g}(i\omega_l) = \begin{pmatrix} \hat{g}_u & \hat{g}_a \\ \hat{g}_a^t & \hat{g}_v \end{pmatrix} = \sum_k \begin{pmatrix} \hat{1} & \hat{0} \\ \hat{0} & \text{cos}k\hat{1} \end{pmatrix} \hat{g}_k(i\omega_l) \begin{pmatrix} \hat{1} & \hat{0} \\ \hat{0} & \text{cos}k\hat{1} \end{pmatrix}. \quad (17)$$

Physically,  $\hat{\Gamma}^u$  mainly indicates the charge fluctuation. The charge fluctuation is also studied on the basis of the time-dependent Hartree-Fock simulation.<sup>15)</sup> In the linear response with the Hartree-Fock approximation,  $\hat{\Gamma}^u$  and  $\hat{\Gamma}^a$  are absent and are peculiar to the excited states. Firstly we evaluate the contribution to TPA from the vertex correction in the case of  $V = 0$ . The result is in Fig. 3. It is seen that the vertex correction removes the high-energy spectrum and enhances the spectrum at lower energy. The band-edge divergence at high-energy is removed by the vertex correction in the one dimensional system. This is the cancellation



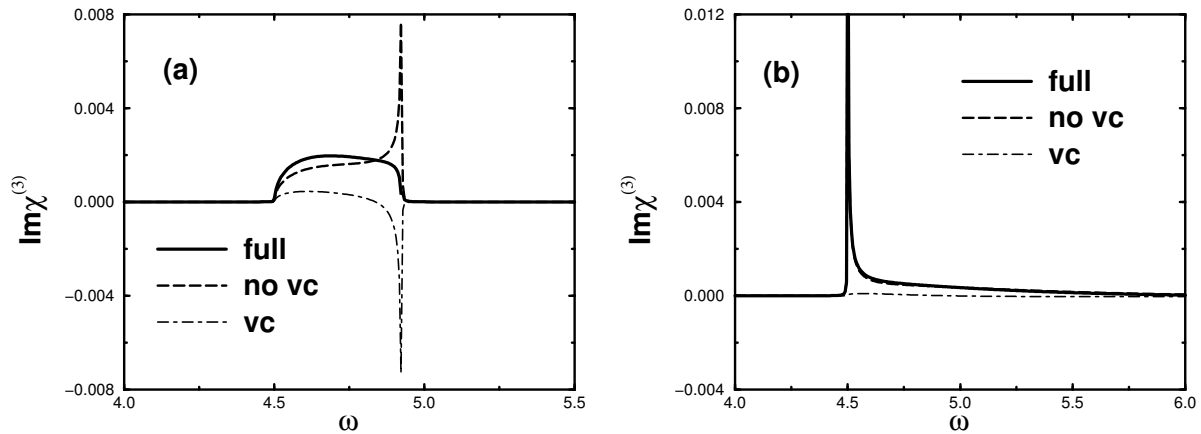


Fig. 3. The spectrum of  $\text{Im}\chi^{(3)}$  for the one dimensional (a) and the two dimensional case (b) in AF insulator based on the Hubbard model with the dispersion relation shown in the text. ‘no vc’ and ‘vc’ indicate the calculation without the vertex correction and with only the vertex correction, respectively. ‘full’ indicates the sum of these two.  $U = 9$ ,  $V = 0$  and  $t = 1$ .

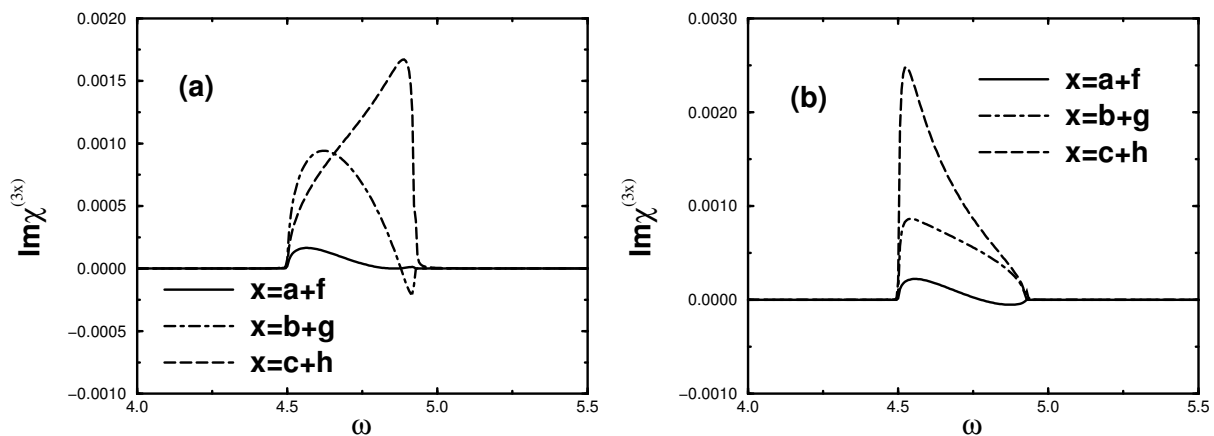


Fig. 4. The decomposition of TPA spectrum to several parts in the one-dimensional system ; (a)+(f), (b)+(g) and (c)+(h) of Fig. 1. (a)  $U = 9$ ,  $V = 0$ , (b)  $U = 9$ ,  $V = 2$

between (c)-term and (h)-term, which is not dependent on the value of  $U$ . It is also shown that the width of the divergent region is narrow for the two dimensional system as in the case of the one-photon absorption spectrum. The damping effect is not included in our calculation and this effect broadens the spectral width and makes the absorption below  $\omega < \Delta$  possible as the strong coupling approach (the perturbation by  $t$ ) will show.

The decomposition of TPA spectrum, classified by the way of coupling with the external field, is shown in (a) and (b) of Fig. 4, in the case of  $V = 0$  and  $V = 2$ , respectively. The

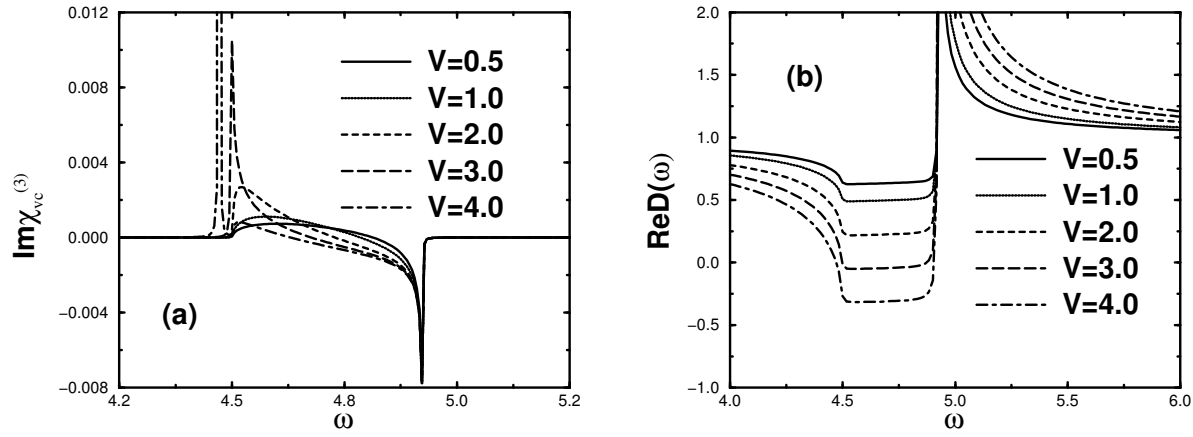


Fig. 5. (a) The vertex correction to TPA spectrum ((f)+(g)+(h) of Fig. 1) with various values of  $V$  and  $U = 9$  in the one-dimensional system. (b)  $D(\omega)$  (the denominator of the four-point vertex  $\Gamma(\omega)$ ) with various values of  $V$  and  $U = 9$ .

direct transition terms,  $(b + g)$  and  $(c + h)$ , are dominant, independent of the value of  $V$ . In the Peierls insulator it is shown that  $(T \sum_{k,n} \hat{G}_j \frac{\partial \hat{v}}{\partial k} \hat{G}_i) = 0$  in  $(g, h)$  of Fig. 1 except for the self-transition term. (This is similar to the result that  $(b, c)$  in Fig. 1 have no contribution to  $K^{(3)}$  for TPA.) The  $V$ -term shifts the spectrum to lower energy by the excitonic effect. The vertex correction to TPA spectrum with various values of  $V$  is shown in Fig. 5 (a). The high-energy band edge is independent of  $V$ , which cancels with the no-vertex correction term ((c) of Fig. 1). On the other hand, spectrum grows around lower energy with increasing  $V$ . The poles of  $\hat{\Gamma}$  indicate some kinds of excitons. The denominator  $D(\omega)$  of  $\Gamma(\omega)$  is shown in Fig. 5 (b). With increasing  $V$ ,  $\text{Re}D(\omega)$  decreases and this enhances spectrum at lower energy. The sharp isolated peak exists at the frequency in which  $\text{Re}D(\omega) = 0$  and  $\omega < \Delta$ .

The excitonic effect in OPA is described by the diagram which is similar to (h) of Fig. 1 but with one external field line at each side of the vertex. This means that the factor  $\cos k$  is replaced by  $\sin k$  in eqs. (15) and (17). This difference of the parity on  $k \rightarrow -k$  corresponds to that of even- and odd-states in TPA and OPA, respectively. Then the relation between TPA and OPA about the excitonic effect is not trivial. In general the factor  $\sin k$  enhances the lower energy part because the excitation around the lower band-edge arises from  $k \simeq \pi/2$ . Therefore  $\text{Re}D(\omega)$  in the linear response is smaller than that of TPA around the lower band-edge.

### 3.3 Scaling relations

We evaluate this result quantitatively to compare with experiments. The experimentally observed values are  $\beta(\omega)$  and  $\alpha(\omega)$ , which are related to  $\text{Im}\chi^{(3)}$  and  $\text{Im}\chi^{(1)}$  as follows.

$$\beta(\omega) = \frac{24\pi^2\omega}{n_0c^2} \text{Im}\chi^{(3)}(\omega) \quad (18)$$

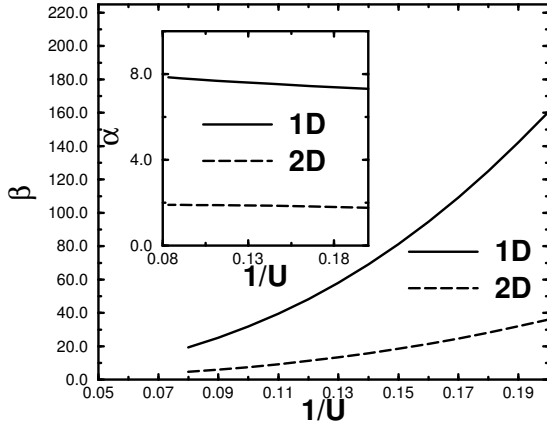


Fig. 6. The dependence of the average of TPA coefficient,  $\beta = \frac{1}{w_\beta} \int d\omega \beta(\omega)$ , on  $1/U$ . The dependence of the average of the OPA coefficient,  $\alpha = \frac{1}{w_\alpha} \int d\omega \alpha(\omega)$ , on  $1/U$  is shown in the inset.  $w_\beta$  and  $w_\alpha$  are the width of the spectrum for the two-photon and one-photon absorption, respectively.  $w_\beta = \sqrt{\Delta^2 + (2t(1+d))^2} - \Delta$  and  $w_\alpha = 2w_\beta$  for  $d$  dimensional system. We put  $t = 1$  and the unit is determined as in the text.

Here  $n_0 = \sqrt{1 + 4\pi\chi^{(1)}}$

$$\alpha^{(1)}(\omega) = \frac{4\pi\omega}{c} \text{Im}\chi^{(1)}(\omega) \quad (19)$$

The calculated results do not include the damping effect and therefore we consider the averaged value as in Fig. 6. (We put  $V = 0$ . The finite value of  $V$  does not change the average value because the positive part of the vertex correction cancels with the negative part as can be seen in Fig. 5 (a).) If we put the lattice constant  $a = 5[\text{\AA}]$  and the transfer integral  $t = 0.5[\text{eV}]$ , the values in this figure have the dimension  $\beta[\text{cm/GW}]$  and  $\alpha[10^5\text{cm}^{-1}]$ . It is known in the experiment<sup>16)</sup> that  $\beta \simeq 120, 12 [\text{cm/GW}]$  for the one and two dimensional system, respectively. In the same way  $\alpha \simeq 4, 1 [10^5\text{cm}^{-1}]$ . The calculation is comparable to the experiments in order of magnitude around  $U \simeq 6$  and  $U \simeq 8$  for the one and two dimensional system, respectively. This agrees with the following fact; the experimental values of  $J$  noted in ref.<sup>16)</sup> ( $J \simeq 2000 \sim 3000$  and  $1400 [\text{K}]$  for the one and two dimensional system, respectively) corresponds to  $U \simeq 4 \sim 6$  and  $U \simeq 8$ . This result is also consistent with the fact that the experiments are performed with the Mott insulator in which  $U$  is greater than the band width in general. This value of  $U$  is considered to be a realistic value even if the damping effect which broadens spectrum is included.

Another point to be emphasized is the strong dependence of  $\beta$  on  $1/U$ , which is contrary to that of  $\alpha$ . It is known that the exact solution in the one dimensional case gives  $\int_0^\infty \omega \text{Im}\chi^{(1)}(\omega) d\omega \propto t^2/U$  in the limit of  $t/U \rightarrow 0$ .<sup>17)</sup> In our calculation  $\int_0^\infty \omega \text{Im}\chi^{(1)}(\omega) d\omega$  is proportional to  $w_\alpha \alpha$ . The result that  $\alpha$  is almost independent of  $1/U$  is consistent with this

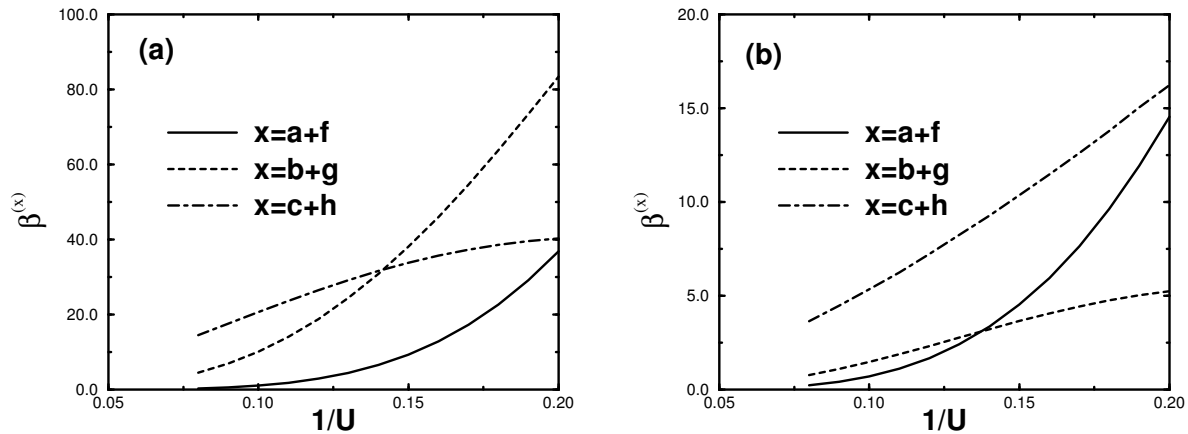


Fig. 7. The decomposition of  $\beta$  into several parts, classified as in Fig. 4, (a)+(f), (b)+(g) and (c)+(h) of Fig. 1. (a) the one dimensional system (b) the two dimensional system

sum rule because of  $w_\alpha \propto t^2/U$  for  $U \gg t$ . The strong dependence of  $\beta$  on  $1/U$  shows that the large nonlinear optical response is expected in the systems with large exchange interaction ( $J$ ).

The decomposition of  $\beta$  classified by the way of coupling to the external field is shown in Fig. 7. All of these parts increase with decreasing  $U$ . The difference of increasing rate originates from the difference of dependence of absorption spectra on frequency.  $\beta^x$  becomes large if it has a large portion of the spectrum at lower frequency. The dominant contribution comes from the direct transition term, which is peculiar to SCES.

#### 4. Summary and Discussion

In conclusion we formulate the nonlinear optical susceptibility of many-body systems on the basis of Green's function. We find that the direct transition term makes sufficient contribution to TPA in many-body systems, contrary to the band insulator. The vertex corrections which describe the charge fluctuation and the excitonic effect are obtained in our formulation. This effect shifts the spectrum to the lower frequency region. A scaling relation of TPA as a function of the electron-electron interaction is calculated. The calculated result agrees with the experimental value semi-quantitatively, which has not been clarified by previous works with small system size.

The experiments cited in this paper are performed in the Mott insulator, which remains insulating above the Néel temperature ( $T_N$ ). However the spectrum below  $T_N$  is similar to that above  $T_N$  at least in two dimensional case.<sup>16)</sup> (The experiment in quasi-one dimensional case below  $T_N$  is desired.) Our calculation is considered to be related to these systems below  $T_N$ . The finite couplings between chains are needed to obtain the finite  $T_N$ . This means that

the fluctuation is suppressed in real materials below  $T_N$ . Therefore our mean field analysis is considered to be applied in this ordered state. The difference of the effect of the fluctuation between the one and two dimensional systems is not treated in this approximation. However the fluctuation is considered to be small because of large value of  $U/t \gg 1$  used in this paper. (On the basis of the conserving approximation, the higher order correlation effect (the charge fluctuation) is included in the excited states even in the Hartree-Fock approximation.) The spin-charge separation as the mechanism of large nonlinear optical response is discussed in ref.<sup>18)</sup> This is contrary to our result because the spin-charge separation is obtained at large  $U$ . Our conclusion in this paper is that if we make a comparison between the systems with the same dimension, the opposite direction from the spin-charge separation is considered to be favorable for large  $\beta$ . The damping effect is not included in our calculation and is needed to obtain more precise behavior of the spectrum and the scaling relation. Especially the band-edge singularity should be broadened. This is a further problem which can be treated by Green's function method developed in this paper.

### Acknowledgements

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### Appendix: Derivation of the response function

In this appendix we show the general formula of the response function. We consider the uniform external field, which is included in the Hamiltonian as the Peierls factor as follows. (We put  $e = c = \hbar = 1$  and the indices indicating the space dimension are omitted.)

$$\mathcal{H}_A = \sum_{\langle ij \rangle \sigma} (t_{ij}^A c_{i\sigma}^\dagger c_{j\sigma} + t_{ji}^A c_{j\sigma}^\dagger c_{i\sigma}) + \mathcal{H}_U. \quad (\text{A}\cdot 1)$$

Here,  $t_{ij}^A = t_{ij} \exp[-iA \cdot (r_i - r_j)]$  and  $\mathcal{H}_U$  is the interaction term. In the linear response theory the current ( $J^{(1)}$ ) and the polarization ( $P^{(1)}$ ) are written as follows.

$$\begin{aligned} J^{(1)}(t) &= \frac{1}{i} \int_{-\infty}^t dt_1 \text{Tr}[[j^p(t_1), \rho_0] j^p(t)] A(t_1) + \text{Tr}[\rho_0 (-j^d)] A(t) \\ &=: - \int_{-\infty}^{\infty} dt_1 K^{(1)}(t - t_1) A(t_1) \end{aligned} \quad (\text{A}\cdot 2)$$

This equation also defines the correlation function  $K^{(1)}(t - t_1)$  used in this paper.

$$\begin{aligned} P^{(1)}(t) &= \frac{1}{i} \int_{-\infty}^t dt_1 \text{Tr}[[p(t_1), \rho_0] p(t)] E(t_1) \\ &=: \int_{-\infty}^{\infty} \chi^{(1)}(t - t_1) E(t_1) \end{aligned} \quad (\text{A}\cdot 3)$$

Here  $\rho_0$  is the density matrix in the equilibrium state and  $A$  is the vector potential.  $p := \sum_{i\sigma} r_i n_{i\sigma}$  is the polarization operator.  $j^p := i[\mathcal{H}_{A=0}, p]$  is the paramagnetic current operator.

$j^d := i[p, j^p]$  is the diamagnetic current operator. Similarly we can derive the nonlinear (the third order) response as follows. (These are derived with use of the nonequilibrium density matrix as usual.)

$$\begin{aligned}
J^{(3)}(t) &= \frac{1}{i^3} \int_{-\infty}^t dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \text{Tr}[[j^p(t_1), [j^p(t_2), [j^p(t_3), \rho_0]]]j^p(t)]A(t_3)A(t_2)A(t_1) \\
&\quad + \frac{1}{i^2} \int_{-\infty}^t dt_1 \int_{-\infty}^{t_1} dt_2 \text{Tr}[[j^p(t_1), [-\frac{1}{2}j^d(t_2), \rho_0]]j^p(t)]A(t_2)A(t_2)A(t_1) \\
&\quad + \frac{1}{i^2} \int_{-\infty}^t dt_1 \int_{-\infty}^{t_1} dt_2 \text{Tr}[[-\frac{1}{2}j^d(t_1), [j^p(t_2), \rho_0]]j^p(t)]A(t_2)A(t_1)A(t_1) \\
&\quad + \frac{1}{i^2} \int_{-\infty}^t dt_1 \int_{-\infty}^{t_1} dt_2 \text{Tr}[[j^p(t_1), [j^p(t_2), \rho_0]](-j^d(t))]A(t_2)A(t_1)A(t) \\
&\quad + \frac{1}{i} \int_{-\infty}^t dt_1 \text{Tr}[[\frac{-1}{2}j^d(t_1), \rho_0](-j^d(t))]A(t_1)A(t_1)A(t) \\
&\quad + \frac{1}{i} \int_{-\infty}^t dt_1 \text{Tr}[[\frac{1}{3!}j^f(t_1), \rho_0]j^p(t)]A(t_1)A(t_1)A(t_1) \\
&\quad + \frac{1}{i} \int_{-\infty}^t dt_1 \text{Tr}[[j^p(t_1), \rho_0]\frac{1}{2}j^f(t)]A(t_1)A(t)A(t) \\
&\quad + \text{Tr}[\rho_0 \frac{-1}{3!}j^g(t)]A(t)A(t)A(t) \\
&=: - \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \int_{-\infty}^{\infty} dt_3 K^{(3)}(t-t_1, t_1-t_2, t_2-t_3)A(t_3)A(t_2)A(t_1)
\end{aligned} \tag{A.4}$$

$$\begin{aligned}
P^{(3)}(t) &= \frac{1}{i^3} \int_{-\infty}^t dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \text{Tr}[[p(t_1), [p(t_2), [p(t_3), \rho_0]]]p(t)]E(t_3)E(t_2)E(t_1) \\
&=: \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \int_{-\infty}^{\infty} dt_3 \chi^{(3)}(t-t_1, t_1-t_2, t_2-t_3)E(t_3)E(t_2)E(t_1)
\end{aligned} \tag{A.5}$$

$j^f := i[p, j^d]$ ,  $j^g := i[p, j^f]$  are the generalized diamagnetic current operator, which are peculiar to the lattice model. These quantities satisfy the following relation,  $j := i[\mathcal{H}_A, p] = j^p - j^d A + \frac{1}{2}j^f A^2 - \frac{1}{3!}j^g A^3 + \dots$ . Relations,  $J^{(1,3)}(t) = \frac{\partial P^{(1,3)}(t)}{\partial t}$ , hold true in general, which can be proved with use of the relation  $E(t) = -\frac{\partial A(t)}{\partial t}$  and the above definition of operators. Therefore the formalism based on the current operator is equivalent to that based on the polarization operator. The key to this result is taking the (generalized) diamagnetic current into account. This should be done carefully so as to ensure the consistency between the current and the Hamiltonian. If we neglected this, we would reach incorrect conclusions. This is the reason why Xu and Sun erroneously stated that inclusion of the diamagnetic current cannot solve the problem of the zero-frequency divergence.<sup>19)</sup> The recent study also lacks the systematic derivation of the nonlinear response function and shows incorrect signs and the omission of several relevant terms.<sup>20)</sup>

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