Infrared absorption in high-density electron-hole systems: The role of quantum fluctuations

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We study the infrared absorption of the macroscopic quantum state in highly excited semiconductors. The calculated spectrum clearly shows the BCS-like energy-gap (BEG) formation. We incorporate the large quantum fluctuation with the quasistatic Eliashberg equation for *e-h* systems that allows us to calculate the renormalized band energy, the BEG, and the wave function renormalization factor. We find that the collective phase fluctuation significantly modifies the spectra, and that the strong visible-light excitation distinctly stabilizes the *e-h* BCS state.

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I. INTRODUCTION

The issue of the Bose-Einstein condensation (BEC) of excitons has extensively been studied both experimentally and theoretically.[1,2] This is because the bound electron-hole pairs, or excitons, behave like bosons in low densities. Among a lot of experimental efforts to create the excitonic BEC, the fast and coherent propagations of excitons in Cu_2O (Ref. 3) and BiI_3 (Ref. 4) are expected to be the exciton superfluidity because the fast exciton transport is more pronounced with increasing the exciton density. However, the current understanding of these phenomena still remains tentative and controversial because of the complicated experimental situations, such as phonon effects,[5,6] spatial inhomogeneity of exciton density, band anisotropy, finite lifetime of excitons, and so forth.

Recently, Johnsen and Kavoulakis have calculated the infrared absorption spectrum of the excitonic BEC and shown that the spectral component associated with the $1s$-$2p$ transition of excitons strongly depends on the quantum degeneracy of the exciton gas.[7] This result is particularly interesting because either the phonon effect or the inhomogeneity of the exciton density is not sensitive to the absorption process accompanied by the $1s$-$2p$ transition of excitons.[8]

In order to clearly observe the macroscopic quantum phenomena in highly excited semiconductors, we need to increase the exciton density, which is sufficiently larger than the critical density for BEC. When the exciton density is so increased that the excitons are deeply overlapping with each other, the exciton concept is no longer appropriate, and the Fermion nature of electrons and holes plays a significant role. As a result, we should consider the state-filling effect, the band-gap renormalization, and the screening effect. The macroscopic quantum state is no longer regarded as the excitonic BEC but should be interpreted as the *e-h* BCS state, where the bound *e-h* pairs are similar to the Cooper pairs in superconductors. The deviation from the Boson picture is known to be significant when the mean interparticle distance is less than $10a_{\mathrm{B}}$ (a_{B} is the exciton Bohr radius).[24]

The *e-h* pair correlation in semiconductors has initially been discussed by Keldysh and co-workers.[9] They have analyzed the stability of the *e-h* BCS state and the dispersion relation of the elementary excitations within the BCS-like mean-field theory. The analysis has been further extended to incorporate the band anisotropy,[10–13] the spin effect,[14,15] and the spatial confinement effects.[16]

We should remark that the *e-h* BCS state is essentially different from the excitonic BEC. In the *e-h* BCS state, the relative motion of *e-h* pairs determines the macroscopic quantum coherence and the order parameter is the BCS-like energy gap (BEG) at the quasi-Fermi level. In the excitonic BEC, on the other hand, the macroscopic quantum state is governed by the center-of-mass motion of excitons and the order parameter is the density of the condensed excitons.

When we analyze the macroscopic quantum state in semiconductors, conventional approaches based either on the BCS-like mean-field theory or on the interacting Boson model are not appropriate. In many practical experiments of highly excited semiconductors, the deviation from the BCS-like mean-field theory predominates mainly because the carrier density cannot become high enough to justify the mean-field approximation. The system is often in the crossover regime between the *e-h* BCS state and the excitonic BEC, where the collective phase fluctuation associated with the center-of-mass motion of *e-h* pairs predominates.

The BCS-BEC crossover problem has been discussed in a variety of physical contexts including superconductivity,[17–19] nuclear matter,[20] and superfluid ^{3}He (Ref. 21). Much attention has been attracted to the BCS-BEC crossover in connection with the high-T_{c} cuprate superconductors, where the pseudogap structure is observed in the normal state density-of states for underdoped cuprates almost up to the room temperature.[22] In the cuprate superconductors, the coherence length is known to be the same order as the mean interparticle distance,[23] and this property is in contrast with the conventional superconductors where the Cooper pairs are strongly overlapping in real space. The optically excited semiconductors have a marked advantage to investigate the BCS-BEC crossover because the macroscopic quantum state can easily be controlled without changing the composition of materials.

Recently, we have analyzed the BCS-BEC crossover in high-density *e-h* systems by calculating the luminescence spectra.[24] We have shown that the broad spectral component arising from the pair recombination in the *e-h* BCS state splits into the P and P_2 lines with decreasing the particle density, where the P (P_2) line originates from the radiative recombination of an exciton associated with the excitation of another exciton from $1s$ to continuum $(2s)$ state. In addition, we have shown that the coherent emission line at the quasi-Fermi level continuously changes to the exciton line in the exciton condensate with decreasing the particle density. The analysis is based on the Bethe-Salpeter equation combined with the generalized random-phase approximation (GRPA);[25] this approach allows us to consider the state-filling effect, the band-gap renormalization, and the strong (exciton formation) and weak (*e-h* Cooper pair formation) pair correlation on the same basis.

In the present paper, we analyze the infrared or THz light response of the high-density *e-h* systems. The analysis is based on the quasistatic Eliashberg equation (QSEE) for *e-h* systems, which considers the collective phase fluctuation associated with the center-of-mass motion of *e-h* pairs. We numerically solve the QSEE and quantitatively evaluate the renormalized band energy, the BEG, and the wave function renormalization factor. We employ the GRPA to consider the leading contribution of the collective phase fluctuation. The QSEE is obtained by a variational calculation with respect to the Bogoliubov parameters, where the expectation value of the model Hamiltonian is calculated with the GRPA.

We use the GRPA for $r_s \leqslant 1$, where r_s is the mean interparticle distance scaled by the exciton Bohr radius. The GRPA is valid in this density regime because the high-density effects predominate for $r_s \lesssim 2$. Namely, it is known that the quasi-Fermi surface is generated for $r_s \lesssim 5$ (Ref. 24), and the ground state energy per *e-h* pair considerably deviates from the exciton binding energy for $r_s \lesssim 2$ (Ref. 26). In addition, we have shown in Ref. 24 that the GRPA analysis gives the correct density dependence of the band-gap renormalization for CuCl (Ref. 27) and ZnO (Refs. 28 and 29). These results are in marked contrast with those for electron gases where the RPA analysis is valid only in very high densities.[30] The difference between these systems arises because, in highly excited semiconductors, the single *e-h* pair excitation energy has a gap.

We investigate the quasiequilibrium state where the *e-h* pair density is given by the quasi-chemical-potential, and the stationary state driven by a monochromatic pump light. In both states, we assume that the effective carrier temperature is low enough to exhibit the macroscopic quantum state. In practical experiments, the high-density *e-h* systems at low effective temperature are difficult to generate. This is because the large excess energy originating from the large band-gap shift prevents from thermalization during the carrier lifetime. However, the generation of the cold degenerate *e-h* plasma has recently been reported using the resonant excitation of the exciton level of CuCl (Ref. 31). According to this experiment, the quasi-Fermi level is found to build up within 0.3 ps; this time scale is much shorter than the carrier lifetime that is the order of a few nanoseconds. This result indicates that the present assumption can be used at least approximately to practical systems.

We also suppose that the effective masses of electrons and holes are equal and isotropic. The effects of the band anisotropy and the different electron and hole effective masses have been discussed by Kopaev,[11] Zittartz,[12] and Conti *et al.*[13] They found that the *e-h* pair correlation is apt to destroy either by the band anisotropy or by the effective mass difference between electrons and holes. Recently, we have numerically solved the QSEE for various temperatures with neglecting the wave function renormalization effect, and studied the effective mass difference between electrons and holes.[41] We find that the effective mass difference is not sensitive to the stability of the *e-h* BCS state even though the BEG for different effective mass cases becomes smaller than that for the equal mass case. We have used in that analysis the quasistatic single-plasmon-pole approximation (QSSPPA) to incorporate the screening effect; the calculated density dependence of the renormalized band-gap excellently agrees with experiments for CuCl (Ref. 27) and ZnO (Ref. 29). This point is in marked contrast with conventional discussions based either on the Thomas-Fermi screening[11] or on the constant potential,[13] because it is difficult with their approaches to quantitatively calculate the BCS-like gap, the band-gap renormalization, and the quasi-chemical-potential of *e-h* pairs.

In the present paper, we show that the BEG is clearly found in the calculated infrared response even though the strong quantum fluctuation considerably reduces the *e-h* pair correlation. This result suggests that the infrared response is a good candidate for a decisive observation of the *e-h* BCS state. This result is also important in the coherent nonlinear response in semiconductors because the BEG generated by the Coulomb interaction gives the local field effect that is the many-body modification of the optical field inside materials.[32]

The infrared response in highly photoexcited semiconductors has first been discussed by Galitskii *et al.*[33] In their analysis, the Coulomb interaction between carriers is neglected and the visible pump light is rigorously treated with the Bogoliubov transformation combined with the rotating-frame representation. Their model is therefore essentially the same as the assembly of the two-level systems driven by a oscillating field, and the energy gap at the quasi-Fermi level is identical with the Rabi frequency. Later, the analysis has been extended to consider the relaxation processes with the nonequilibrium Green function formalism[34,35] and the interparticle Coulomb interaction.[35–39] However, these results are not conclusive because the BCS-like mean-field theory overestimates the BEG.

We also show that the *e-h* pair correlation is distinctly enhanced by the intense visible pump light which causes the interband transition. As a consequence, the anomalous spectral line shape originating from the BEG is drastically pronounced with increasing the pump-light intensity. Whereas the similar behavior has been suggested in Ref. 37, the result is tentative because the BCS-like mean-field approximation overestimates the *e-h* pair correlation. Considering the fact that the BEG is still much larger than the Rabi frequency even considering the strong quantum fluctuation, the macroscopic quantum phenomenon is expected to be observed under strong photoexcitations.

The paper is organized as follows. In Sec. II, we derive the QSEE within the GRPA. We introduce the Bogoliubov quasiparticle operators to take into account the *e-h* pair correlation. The collective phase fluctuation from the *e-h* BCS state is considered by solving the linearized equation of motion of the pair operators with respect to the Bogoliubov quasiparticles. We numerically solve the QSEE and calculate the imaginary part of the intraband dielectric function in Sec. III. The discussions on the several assumptions employed in the present paper is presented in Sec. IV. In Appendix, we show that the GRPA dielectric function exactly satisfies the longitudinal f-sum rule in quasiequilibrium states where the e-h density is given by the quasi-chemical-potential. In the present analysis, we approximate the RPA dielectric function to the QSSPPA dielectric function to numerically solve the QSEE. The result suggests that this approximation does not lead to significant contradictions because the QSSPPA dielectric function also satisfies the longitudinal f-sum rule.[40]

II. FORMULATION

We consider a three-dimensional e-h system in a direct-gap semiconductor at zero temperature, which consists of the isotropic nondegenerate parabolic conduction and valence bands with identical electron and hole masses; the effect of the different effective mass between electrons and holes will be discussed in a forthcoming paper.[41] We derive the QSEE under the intense monochromatic laser light with visible frequency ω_L which causes the interband transition; the pump-light is treated classically, and the Rabi frequency λ is approximated to be a constant. The QSEE for quasi-equilibrium case (in the absence of the interaction between pump-light and particles) is obtained by replacing $\mu=\omega_L-E_g$ and $\lambda=0$, where μ is the quasi-chemical-potential of e-h pairs and E_g is the band-gap energy. We consider the e-h attractive Coulomb interaction together with the e-e and h-h repulsive Coulomb interactions. The spin degrees of freedom are disregarded to focus our attentions to the collective quantum fluctuation in the e-h BCS state. The interaction between the pump light and an e-h system is considered in the rotating-wave approximation. The finite temperature effect will be discussed in a forthcoming paper.[41]

The model Hamiltonian is written as

$$H=\sum_{\boldsymbol{k}}\{\mathcal{E}^e_{\boldsymbol{k}}c^\dagger_{\boldsymbol{k}}c_{\boldsymbol{k}}+\mathcal{E}^h_{\boldsymbol{k}}d^\dagger_{\boldsymbol{k}}d_{\boldsymbol{k}}\}+\lambda\sum_{\boldsymbol{k}}\{c^\dagger_{\boldsymbol{k}}d^\dagger_{-\boldsymbol{k}}\exp(-i\omega_L t)+\text{H.c.}\}+\frac{1}{2}\sum_{\boldsymbol{k},\boldsymbol{p},\boldsymbol{q}}V_{\boldsymbol{q}}\{c^\dagger_{\boldsymbol{k}+\boldsymbol{q}}c^\dagger_{\boldsymbol{p}-\boldsymbol{q}}c_{\boldsymbol{p}}c_{\boldsymbol{k}}+d^\dagger_{\boldsymbol{k}+\boldsymbol{q}}d^\dagger_{\boldsymbol{p}-\boldsymbol{q}}d_{\boldsymbol{p}}d_{\boldsymbol{k}}-2c^\dagger_{\boldsymbol{k}+\boldsymbol{q}}c_{\boldsymbol{k}}d^\dagger_{\boldsymbol{p}-\boldsymbol{q}}d_{\boldsymbol{p}}\},\tag{1}$$

where $c_{\boldsymbol{k}}$ ($d_{\boldsymbol{k}}$) is the annihilation operator for electrons (holes) whose single-particle energy is $\mathcal{E}^e_{\boldsymbol{k}}=k^2/(2m)+E_g$ $[\mathcal{E}^h_{\boldsymbol{k}}=k^2/(2m)]$, $V_{\boldsymbol{q}}=4\pi e^2/(\epsilon_0 q^2)$ is the Coulomb interaction with the unexcited background dielectric constant ϵ_0.

We analyze the time-independent Hamiltonian in the rotating-frame representation obtained by the Galitskii transformation[33,38,39] that is defined by the following unitary operator:

$$U(t)=\exp\left[-\frac{i\omega_L t}{2}\sum_{\boldsymbol{k}}(c^\dagger_{\boldsymbol{k}}c_{\boldsymbol{k}}+d^\dagger_{-\boldsymbol{k}}d_{-\boldsymbol{k}})\right].\tag{2}$$

The transformed Hamiltonian is written as follows:

$$H=\sum_{\boldsymbol{k}}\{\varepsilon^e_{\boldsymbol{k}}c^\dagger_{\boldsymbol{k}}c_{\boldsymbol{k}}+\varepsilon^h_{\boldsymbol{k}}d^\dagger_{\boldsymbol{k}}d_{\boldsymbol{k}}\}+\lambda\sum_{\boldsymbol{k}}\{c^\dagger_{\boldsymbol{k}}d^\dagger_{-\boldsymbol{k}}+d_{-\boldsymbol{k}}c_{\boldsymbol{k}}\}+\frac{1}{2}\sum_{\boldsymbol{k},\boldsymbol{p},\boldsymbol{q}}V_{\boldsymbol{q}}\{c^\dagger_{\boldsymbol{k}+\boldsymbol{q}}c^\dagger_{\boldsymbol{p}-\boldsymbol{q}}c_{\boldsymbol{p}}c_{\boldsymbol{k}}+d^\dagger_{\boldsymbol{k}+\boldsymbol{q}}d^\dagger_{\boldsymbol{p}-\boldsymbol{q}}d_{\boldsymbol{p}}d_{\boldsymbol{k}}-2c^\dagger_{\boldsymbol{k}+\boldsymbol{q}}c_{\boldsymbol{k}}d^\dagger_{\boldsymbol{p}-\boldsymbol{q}}d_{\boldsymbol{p}}\},\tag{3}$$

where $\varepsilon^{e(h)}_{\boldsymbol{k}}=\mathcal{E}^{e(h)}_{\boldsymbol{k}}-\omega_L/2$, is the single-particle energy of electrons (holes) in the new representation.

We consider the effect of collective phase fluctuation associate with the center-of-mass motion of e-h pairs with the GRPA. We calculate the linearized equation of motion for the Bogoliubov quasiparticle pair operators,

$$\begin{aligned}\Psi^{\boldsymbol{q}}_0(\boldsymbol{k})&=\alpha^\dagger_{\boldsymbol{k}}\alpha_{\boldsymbol{k}+\boldsymbol{q}},\\ \Psi^{\boldsymbol{q}}_1(\boldsymbol{k})&=\alpha^\dagger_{\boldsymbol{k}}\beta^\dagger_{-\boldsymbol{k}-\boldsymbol{q}},\\ \Psi^{\boldsymbol{q}}_2(\boldsymbol{k})&=\beta_{-\boldsymbol{k}}\alpha_{\boldsymbol{k}+\boldsymbol{q}},\\ \Psi^{\boldsymbol{q}}_3(\boldsymbol{k})&=\beta_{-\boldsymbol{k}}\beta^\dagger_{-\boldsymbol{k}-\boldsymbol{q}}.\end{aligned}\tag{4}$$

Here $\alpha_{\boldsymbol{k}}$ and $\beta_{\boldsymbol{k}}$ are the annihilation operators of Bogoliubov quasiparticles defined by the Bogoliubov transformation,

$$\begin{aligned}c_{\boldsymbol{k}}&=u_{\boldsymbol{k}}\alpha_{\boldsymbol{k}}+v_{\boldsymbol{k}}\beta^\dagger_{-\boldsymbol{k}},\\ d_{-\boldsymbol{k}}&=u_{\boldsymbol{k}}\beta_{-\boldsymbol{k}}-v_{\boldsymbol{k}}\alpha^\dagger_{\boldsymbol{k}}.\end{aligned}\tag{5}$$

The Bogoliubov parameters, $u_{\boldsymbol{k}}$ and $v_{\boldsymbol{k}}$, satisfy the constraint $u^2_{\boldsymbol{k}}+v^2_{\boldsymbol{k}}=1$ to ensure the unitarity condition. In the present analysis, we consider the most divergent terms which give the dominant contribution in the limit of small center-of-mass momentum $\boldsymbol{q}$ of Bogoliubov quasiparticle pairs.

The linearized equations of motion for Ψ's are given by a straightforward calculation of the following commutators:

$$[\Psi^{\boldsymbol{q}\dagger}_0(\boldsymbol{k}),H]=-(E_{\boldsymbol{k}+\boldsymbol{q}}-E_{\boldsymbol{k}})\Psi^{\boldsymbol{q}\dagger}_0(\boldsymbol{k}),\tag{6a}$$

$$[\Psi^{\boldsymbol{q}\dagger}_1(\boldsymbol{k}),H]=(E_{\boldsymbol{k}+\boldsymbol{q}}+E_{\boldsymbol{k}})\Psi^{\boldsymbol{q}\dagger}_1(\boldsymbol{k})+C^{(3)}_{\boldsymbol{k},\boldsymbol{k}+\boldsymbol{q}}V_{\boldsymbol{q}}\sum_{\boldsymbol{p}}C^{(3)}_{\boldsymbol{p},\boldsymbol{p}+\boldsymbol{q}}[\Psi^{\boldsymbol{q}\dagger}_1(\boldsymbol{p})-\Psi^{\boldsymbol{q}\dagger}_2(\boldsymbol{p})],\tag{6b}$$

$$[\Psi^{\boldsymbol{q}\dagger}_2(\boldsymbol{k}),H]=-(E_{\boldsymbol{k}+\boldsymbol{q}}+E_{\boldsymbol{k}})\Psi^{\boldsymbol{q}\dagger}_2(\boldsymbol{k})+C^{(3)}_{\boldsymbol{k},\boldsymbol{k}+\boldsymbol{q}}V_{\boldsymbol{q}}\sum_{\boldsymbol{p}}C^{(3)}_{\boldsymbol{p},\boldsymbol{p}+\boldsymbol{q}}[\Psi^{\boldsymbol{q}\dagger}_1(\boldsymbol{p})-\Psi^{\boldsymbol{q}\dagger}_2(\boldsymbol{p})],\tag{6c}$$

$$[\Psi_3^{q\dagger}(\boldsymbol{k}),H]=(E_{\boldsymbol{k}+\boldsymbol{q}}-E_{\boldsymbol{k}})\Psi_3^{\boldsymbol{q}}(\boldsymbol{k}). \tag{6d}$$

Equation (6) is the semiconductor version of the Anderson-Rickayzen equation (SCARE) for superconductors,[25] and it gives the leading contribution of the collective phase fluctuation. For the notational convenience, we introduce the coherence factors that are expressed in terms of the Bogoliubov parameters,

$$C_{\boldsymbol{k},\boldsymbol{p}}^{(0)}=u_{\boldsymbol{k}}u_{\boldsymbol{p}}+v_{\boldsymbol{k}}v_{\boldsymbol{p}},$$

$$C_{\boldsymbol{k},\boldsymbol{p}}^{(1)}=u_{\boldsymbol{k}}v_{\boldsymbol{p}}+v_{\boldsymbol{k}}u_{\boldsymbol{p}},$$

$$C_{\boldsymbol{k},\boldsymbol{p}}^{(2)}=u_{\boldsymbol{k}}u_{\boldsymbol{p}}-v_{\boldsymbol{k}}v_{\boldsymbol{p}},$$

$$C_{\boldsymbol{k},\boldsymbol{p}}^{(3)}=u_{\boldsymbol{k}}v_{\boldsymbol{p}}-v_{\boldsymbol{k}}u_{\boldsymbol{p}}. \tag{7}$$

As shown below, the Bogoliubov parameters, $u_{\boldsymbol{k}}$ and $v_{\boldsymbol{k}}$, are determined with the variational principle. The energy of a pair of Bogoliubov quasiparticles (the excitation energy from the e-h BCS state) is written as $2E_{\boldsymbol{k}}=\xi_{\boldsymbol{k}}^R C_{\boldsymbol{k},\boldsymbol{k}}^{(2)}-\lambda_{\boldsymbol{k}}^R C_{\boldsymbol{k},\boldsymbol{k}}^{(1)}$, where $2\xi_{\boldsymbol{k}}^R=(\varepsilon_{\boldsymbol{k}}^e+\varepsilon_{\boldsymbol{k}}^h)-\Sigma_{\boldsymbol{p}}V_{\boldsymbol{k}-\boldsymbol{p}}\{1-C_{\boldsymbol{p},\boldsymbol{p}}^{(2)}\}$ is the renormalized e-h pair energy. The renormalized Rabi frequency is written by $\lambda_{\boldsymbol{k}}^R=\lambda-\frac{1}{2}\Sigma_{\boldsymbol{p}}V_{\boldsymbol{k}-\boldsymbol{p}}C_{\boldsymbol{p},\boldsymbol{p}}^{(1)}$, where the second term on the right-hand side arises from the macroscopic quantum coherence generated by the e-h Coulomb interaction.[32] We find from Eq. (6) that $\Psi_j^{\boldsymbol{q}}(\boldsymbol{k})$ $(j=0,3)$ is the energy eigenoperator with eigenvalue $\mp(E_{\boldsymbol{k}+\boldsymbol{q}}-E_{\boldsymbol{k}})$, and they describe the Bogoliubov quasiparticles that are already present in the initial state. These operators can be neglected in the subsequent analysis because we calculate the expectation value of the model Hamiltonian at zero temperature.[25]

In the following analysis, we calculate the expectation value of the model Hamiltonian with $\langle H\rangle=\langle H\rangle_0+\int_0^1 dg\langle H_{\rm int}\rangle_g$, where $H_{\rm int}$ is the third term on the right-hand side of Eq. (3); $\langle\cdots\rangle_0$ and $\langle\cdots\rangle_g$ represent the ground-state expectation value of the system with $V_{\boldsymbol{q}}=0$ and $V_{\boldsymbol{q}}=gV_{\boldsymbol{q}}$, respectively. In order to calculate $\langle H_{\rm int}\rangle_g$, we introduce the 2×2 matrix Green function defined by

$$i[\boldsymbol{G}_{\boldsymbol{k},\boldsymbol{p}}^{\boldsymbol{q}}(t)]_{\alpha,\beta}=\Theta(t)\langle[\Psi_\alpha^{\boldsymbol{q}\dagger}(\boldsymbol{k},t),\Psi_\beta^{\boldsymbol{q}}(\boldsymbol{p},0)]\rangle, \tag{8}$$

$(\alpha,\beta=1,2)$; this Green function is analytically evaluated with Eq. (6). The expectation value of the interaction Hamiltonian $\langle H_{\rm int}\rangle$ is expressed in terms of $\boldsymbol{G}_{\boldsymbol{k},\boldsymbol{p}}^{\boldsymbol{q}}(t)$ as follows:

$$\langle H_{\rm int}\rangle=-\sum_{\boldsymbol{k},\boldsymbol{p},\boldsymbol{q}}\int_0^\infty\frac{d\omega}{4\pi}V_{\boldsymbol{q}}C_{\boldsymbol{k},\boldsymbol{k}+\boldsymbol{q}}^{(3)}C_{\boldsymbol{p},\boldsymbol{p}+\boldsymbol{q}}^{(3)}\times\mathrm{Im}[\mathrm{tr}\{(\boldsymbol{\tau}_0-\boldsymbol{\tau}_1)\boldsymbol{G}_{\boldsymbol{k},\boldsymbol{p}}^{\boldsymbol{q}}(\omega)\}], \tag{9}$$

where $\boldsymbol{G}_{\boldsymbol{k},\boldsymbol{p}}^{\boldsymbol{q}}(\omega)$ is the Fourier transform of $\boldsymbol{G}_{\boldsymbol{k},\boldsymbol{p}}^{\boldsymbol{q}}(t)$; $\boldsymbol{\tau}_0$ and $\boldsymbol{\tau}_j$ $(j=1,2,3)$ are the 2×2 unit matrix and the Pauli matrices, respectively. The expectation value of the model Hamiltonian with respect to the GRPA ground state takes the following form:

$$\langle H\rangle=-\sum_{\boldsymbol{q}}\{(1-C_{\boldsymbol{q},\boldsymbol{q}}^{(2)})\xi_{\boldsymbol{q}}^0-C_{\boldsymbol{q},\boldsymbol{q}}^{(1)}\lambda\}+\sum_{\boldsymbol{q}}\int_{-\infty}^{+\infty}\frac{d\omega}{4\pi i}\ln[1+V_{\boldsymbol{q}}\Pi_{\boldsymbol{q}}(\omega+i\gamma)], \tag{10}$$

where $2\xi_{\boldsymbol{k}}^0=(\varepsilon_{\boldsymbol{k}}^e+\varepsilon_{\boldsymbol{k}}^h)-\Sigma_{\boldsymbol{q}}V_{\boldsymbol{q}}$ is the bare e-h pair energy, and $\Pi_{\boldsymbol{q}}(\omega)$ is the polarization function given by

$$\Pi_{\boldsymbol{q}}(\omega)=-2\sum_{\boldsymbol{k}}\frac{(E_{\boldsymbol{k}+\boldsymbol{q}}+E_{\boldsymbol{k}})}{\omega^2-(E_{\boldsymbol{k}+\boldsymbol{q}}+E_{\boldsymbol{k}})^2}C_{\boldsymbol{k},\boldsymbol{k}+\boldsymbol{q}}^{(3)2}. \tag{11}$$

We introduce, in Eq. (10), the e-h pair recombination rate γ. In the present analysis, we only consider the radiative recombination; the nonradiative recombination due to phonons is neglected. In this case, the radiation field plays the role of reserver and the pair recombination processes is regarded as Markovian, i.e., independent of frequency, because of the broad spectrum of the vacuum fluctuation of the radiation field.[43] This point is in contrast with the strong phonon-carrier coupling case where the nonradiative recombination rate is non-Markovian.

The QSEE for e-h systems is obtained with a variational calculation with respect to the Bogoliubov parameter under the constraint $u_{\boldsymbol{k}}^2+v_{\boldsymbol{k}}^2=1$: $\delta\langle H\rangle/\delta u_{\boldsymbol{k}}=0$ with $(\delta u_{\boldsymbol{k}}/\delta v_{\boldsymbol{k}})=-v_{\boldsymbol{k}}/u_{\boldsymbol{k}}$. A straightforward calculation gives the following expression for the QSEE,

$$\zeta_{\boldsymbol{k}}=\frac{1}{2}(\varepsilon_{\boldsymbol{k}}^e+\varepsilon_{\boldsymbol{k}}^h)-\frac{1}{2}\sum_{\boldsymbol{p}}V_{\boldsymbol{p}}-\frac{1}{2}Z_{\boldsymbol{k}}\sum_{\boldsymbol{p}}V_{\boldsymbol{k}-\boldsymbol{p}}C_{\boldsymbol{k},\boldsymbol{p}}^{(2)}[1+2\chi_{\boldsymbol{k}-\boldsymbol{p}}(-E_{\boldsymbol{k}}-E_{\boldsymbol{p}})], \tag{12a}$$

$$\Delta_{\boldsymbol{k}}=\lambda+Z_{\boldsymbol{k}}\sum_{\boldsymbol{p}}V_{\boldsymbol{k}-\boldsymbol{p}}C_{\boldsymbol{k},\boldsymbol{p}}^{(1)}[1+2\chi_{\boldsymbol{k}-\boldsymbol{p}}(-E_{\boldsymbol{k}}-E_{\boldsymbol{p}})], \tag{12b}$$

$$Z_{\boldsymbol{k}}^{-1}=1+2\sum_{\boldsymbol{p}}V_{\boldsymbol{k}-\boldsymbol{p}}C_{\boldsymbol{k},\boldsymbol{p}}^{(3)2}\left[\frac{\partial\chi_{\boldsymbol{k}-\boldsymbol{p}}(\omega)}{\partial\omega}\right]_{\omega=-E_{\boldsymbol{k}}-E_{\boldsymbol{p}}}, \tag{12c}$$

where $2\zeta_{\boldsymbol{k}}$ and $\Delta_{\boldsymbol{k}}$ are the renormalized single e-h pair excitation energy and the BEG, respectively. The Bogoliubov parameters are given by

$$u_{\boldsymbol{k}}^2=\frac{1}{2}\left(1-\frac{\zeta_{\boldsymbol{k}}}{\sqrt{\zeta_{\boldsymbol{k}}^2+\Delta_{\boldsymbol{k}}^2}}\right),\quad v_{\boldsymbol{k}}^2=\frac{1}{2}\left(1+\frac{\zeta_{\boldsymbol{k}}}{\sqrt{\zeta_{\boldsymbol{k}}^2+\Delta_{\boldsymbol{k}}^2}}\right). \tag{13}$$

The dynamic screening effect is considered in the present theory through the partial screening function[26] defined by

$$\chi_{\boldsymbol{k}}(\omega)=\frac{1}{\pi}\int_0^\infty dz\frac{\epsilon_0\mathrm{Im}[\epsilon_{\boldsymbol{k}}^{-1}(z)]}{(z-\omega)}, \tag{14}$$

where $\epsilon_{\boldsymbol{k}}(\omega)=\epsilon_0[1+V_{\boldsymbol{k}}\Pi_{\boldsymbol{k}}(\omega)]$ is the dielectric function in GRPA. We show in Appendix that the GRPA dielectric function exactly satisfies the longitudinal f-sum rule for $\lambda=0$, The wave function renormalization factor $Z_{\boldsymbol{k}}$ reflects the collective phase fluctuation from the e-h BCS state. This effect arises from the retardation effect in the screened Coulomb interaction, and is absent in the BCS-like mean-field analysis. In contrast to the Eliashberg theory for superconductors,[42] the BEG and the wave function renormalization factor are independent of ω in the present analysis. The QSEE is not taken into account the effect of collision broadening of the e-h pair energy arising from the imaginary part of the self-energy. However, this effect does not contribute significantly in the present case because the collision broadening quadratically vanishes at the quasi-Fermi level at zero temperature.[44,45]

In the following, we simplify the QSEE using the partial screening function in the QSSPPA;[26,46] this approximation is known to produce the relatively good self-energy corrections in three-dimensional systems.[24,47] The dielectric function in QSSPPA is given by

$$\epsilon_{\boldsymbol{k}}^{-1}(z)=\epsilon_0^{-1}\left(1+\frac{\omega_{\rm pl}^2}{z^2-\omega_{\boldsymbol{k}}^2}\right), \tag{15a}$$

where $\omega_{\rm pl}=[4\pi ne^2/(\epsilon_0 M)]^{1/2}$ is the plasma frequency (n is the e-h pair density, and M is the reduced mass of e-h pairs). The dispersion of the effective plasmon mode is chosen as

$$\omega_{\boldsymbol{k}}^2=\omega_{\rm pl}^2\left(1+\frac{k^2}{k_{\rm TF}^2}\right)+G_{\rm eff}^2, \tag{15b}$$

where $k_{\rm TF}=[16Me^2/(\pi\epsilon_0)]^{1/2}(6\pi^2 n)^{1/6}$ is the Thomas-Fermi wave number and $G_{\rm eff}=2\min_{\boldsymbol{k}}E_{\boldsymbol{k}}$ is the effective gap. The partial screening function and its derivative take the following simple forms:

$$\chi_{\boldsymbol{k}}(\omega)=\frac{\omega_{\rm pl}^2}{2\omega_{\boldsymbol{k}}}\left(\frac{1}{\omega-\omega_{\boldsymbol{k}}}\right),$$

$$\frac{\partial\chi_{\boldsymbol{k}}(\omega)}{\partial\omega}=-\frac{\omega_{\rm pl}^2}{2\omega_{\boldsymbol{k}}}\left(\frac{1}{\omega-\omega_{\boldsymbol{k}}}\right)^2. \tag{15c}$$

III. NUMERICAL ANALYSIS

In the numerical analysis, we use the units where the exciton binding energy and the exciton Bohr radius are taken as unity. We iteratively solve the QSEE Eq. (12), and the infrared absorption spectra are calculated with

$$\alpha(\omega)=\frac{\omega}{n(\omega)}\mathrm{Im}\,\epsilon(\omega), \tag{16}$$

where $n(\omega)$ is the refractive index. The dielectric function $\epsilon(\omega)$ is given by

$$\epsilon(\omega)=\epsilon_0[1+\lim_{\boldsymbol{k}\to\boldsymbol{0}}V_{\boldsymbol{k}}\Pi_{\boldsymbol{k}}(\omega)], \tag{17}$$

where the polarization function $\Pi_{\boldsymbol{k}}(\omega)$ is defined by Eq. (11). In the following analysis, γ is set to 0.03.

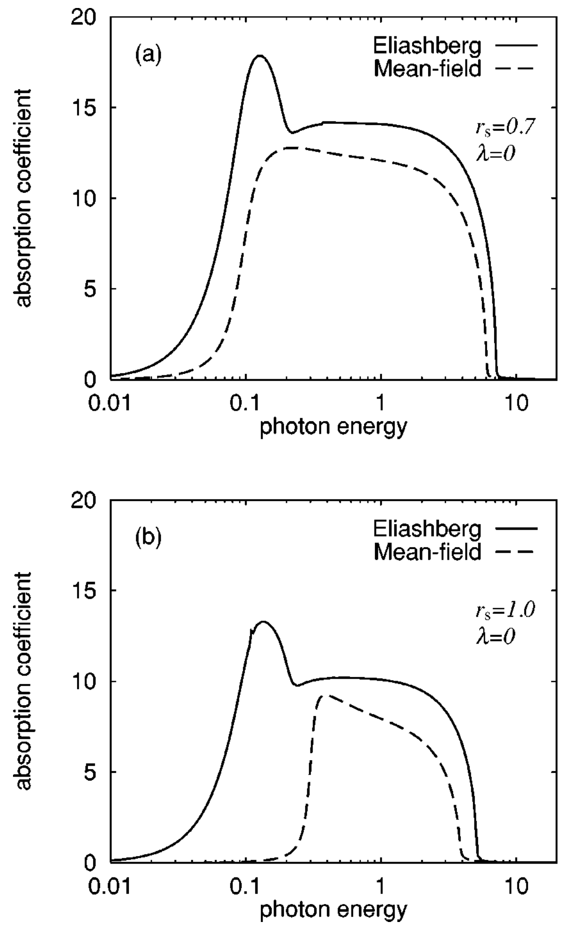

FIG. 1. (a) The infrared absorption spectra in the absence of the pump light ($\lambda=0$). (a) is spectra for $r_s=0.7$ and (b) is that for $r_s=1.0$. The solid line is given by the present theory and the dashed line is given by the BCS-like mean-field theory.

We first discuss the absorption spectra in the absence of the pump light ($\lambda=0$); the effect of the pump light will be shown later. In this case, the particle density is determined by the quasi-chemical-potential of e-h pairs. Figure 1 depicts the calculated absorption spectra for $r_s=0.7$ and $r_s=1.0$, where $r_s=(4\pi n/3)^{-1/3}$ (n is the e-h pair density) is the dimensionless mean interparticle distance. As a reference, we also show the absorption spectra calculated with the BCS-like mean-field analysis (QSEE with $Z_{\boldsymbol{k}}=1$ approximation) for the same densities. The present analysis gives the strong infrared absorption below the plasma frequency $\omega_{\rm pl}$ ($\omega_{\rm pl}=6.0$ for $r_s=0.7$, and $\omega_{\rm pl}=3.5$ for $r_s=1.0$), as expected.

We find that the present theory gives the stronger absorption spectra with larger plasma frequency than that of the BCS-like mean-field theory. This behavior arises from the mass renormalization effect. As shown in Fig. 2, the numerical solution of QSEE gives $m^*/m=0.86$ for $r_s=0.7$ and $m^*/m=1.2$ for $r_s=1.0$, while the BCS-like mean-field analysis gives $m^*/m=0.79$ for $r_s=0.7$ and $m^*/m=0.76$, where $m^*=[(\partial^2\zeta_{\boldsymbol{k}}/\partial k^2)_{\boldsymbol{k}=\boldsymbol{0}}]^{-1}$ and m are the renormalized and bare masses of particles, respectively.

In the BCS-like mean-field analysis, we find a transparent region in $\omega\lesssim0.15$ for $r_s=0.7$ and $\omega\lesssim0.30$ for $r_s=1.0$ that

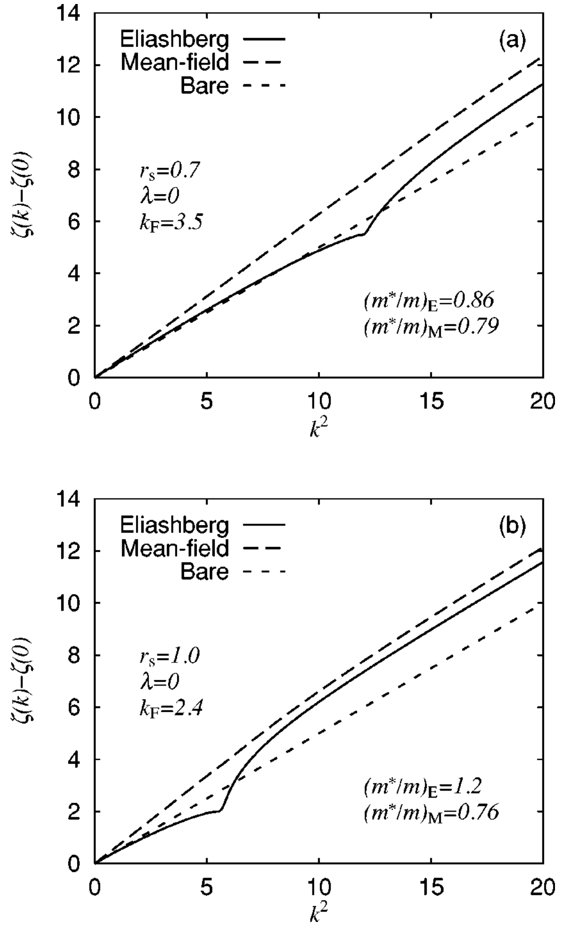

FIG. 2. The momentum dependence of the renormalized band energy in the absence of the pump-light ($\lambda=0$). (a) is the result for $r_s=0.7$ and (b) is that for $r_s=1.0$. The Fermi momentum for $r_s=0.7$ is $k_F=3.5$, and that for $r_s=1.0$ is $k_F=2.4$.

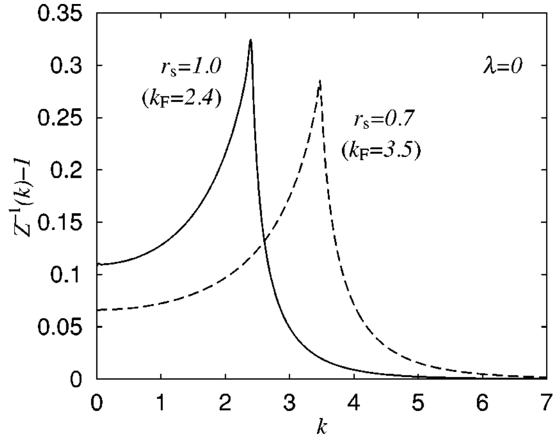

FIG. 3. The momentum dependence of $Z^{-1}(k)-1$ in the absence of the pump light ($\lambda=0$). The solid line represents the result for $r_s=0.7$, and the dashed line is the result for $r_s=1.0$.

originates from the BEG formation at the quasi-Fermi level. In the present analysis, on the other hand, the quantum fluctuation effect considerably reduces the transparent region. Furthermore, it gives a peaked structure at $\omega \simeq 0.12$ for $r_s=0.7$ and $\omega \simeq 0.1$ for $r_s=1.0$. This peaked structure originates from the collective excitation mode associated with the center-of-mass motion of e-h pairs that corresponds to the Anderson mode in superconductors.[25] With increasing the e-h density, the quantum fluctuation effect reduces, and the transparent region given by the present analysis is almost the same as that given by the BCS-like mean-field analysis, as expected.

Figure 2 depicts the renormalized band dispersions for $r_s=0.7$ and $r_s=1.0$ in the absence of the pump light ($\lambda=0$). The present analysis shows the noteworthy deviation from the parabolic momentum dependence especially near the Fermi momentum ($k_F=3.5$ for $r_s=0.7$, and $k_F=2.4$ for $r_s=1.0$). This deviation originates from the anomalous behavior of the wave function renormalization factor as shown in Fig. 3. This anomaly arises from the large quantum fluctuation close to the quasi-Fermi level. We should remark that the wave function renormalization factor associated with the retardation effect in the screened Coulomb interaction predominates near the quasi-Fermi level. In the large momentum limit, $k \gg k_F$, the wave function renormalization effect is negligible and the self-energy correction for the band dispersion is well described by the BCS-like mean-field theory. In the small momentum limit $k \ll k_F$, on the other hand, the renormalized band dispersion is close to the bare band dispersion, $\zeta_{\boldsymbol{k}}^{(0)}=(\varepsilon_{\boldsymbol{k}}^e+\varepsilon_{\boldsymbol{k}}^h)/2-\Sigma_{\boldsymbol{p}} V_{\boldsymbol{p}}/2$, because the wave function renormalization cancels the self-energy correction. With increasing the particle density, the collective phase fluctuation weakens, and the anomalous behavior of $\zeta_{\boldsymbol{k}}$ at $k=k_F$ becomes small and the momentum dependence of $\zeta_{\boldsymbol{k}}$ given by the present theory shows almost the same behavior as that of the BCS-like mean-field analysis. In addition, the wave function renormalization factor approaches unity, as shown in Fig. 3.

Finally, we quantitatively discuss the light-enhanced e-h pair correlation using the QSEE. Figure 4 depicts the infrared absorption spectra under strong visible pump-light excitations (large λ); the pump-light frequency ω_L is set to E_g. In the present analysis, we find that the e-h pair density is not a monotonic function of λ because the system exhibits the resonatorless optical bistability as in Ref. 39; this subject will be discussed elsewhere.[41] We find that the transparent region near $\omega \simeq 0$ distinctly grows with increasing pump light intensity. This result indicates that the e-h pair correlation is considerably enhanced by the strong pump light because this transparent region originates from the BEG formation. With increasing pump-light intensity, the present theory also shows that the overall absorption weakens and its spectral line shape becomes similar to that of the BCS-like mean-field analysis. These behaviors arise because the strong pump light enhances the e-h pair correlation and it significantly reduces the quantum fluctuation. The light-enhanced e-h pair correlation was discussed in Refs. 37 and 38 using the BCS-like mean-field theory. We should remark that these results are, however, tentative and controversial because the BCS-like mean-field theory overestimates the e-h pair correlation as shown in Fig. 4(b), and the present study provides quantitative and reliable results.

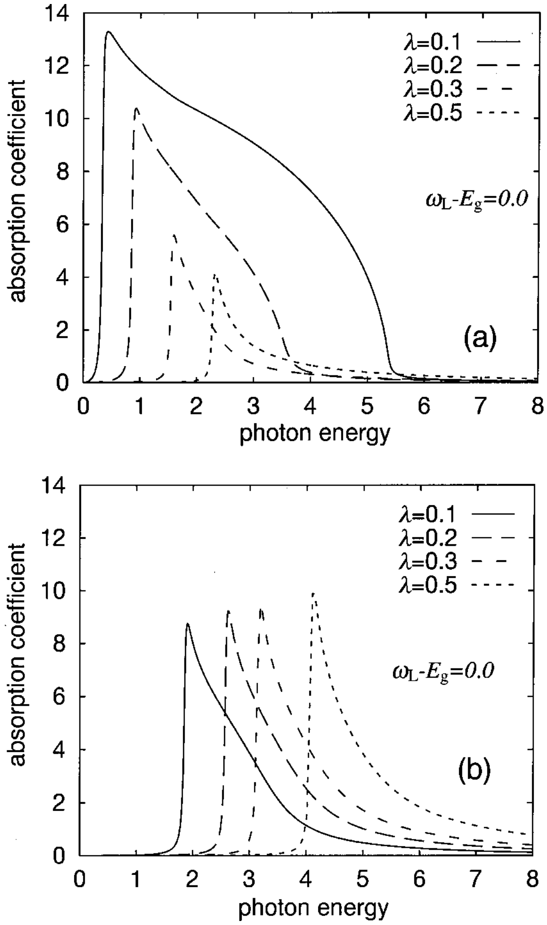

FIG. 4. The infrared absorption spectra under the intense pump light whose frequency is $\omega_{\rm L}=E_g$. (a) and (b) are given by the present theory and by the BCS-like mean-field theory, respectively. In (a) and (b), the mean interparticle distances (dimensionless densities) for $\lambda=0.1,0.2,0.3,0.5$ are $r_s=1.4,2.0,1.8,1.7$ ($na_{\rm B}^3=8.0\times10^{-2}$, 3.1×10^{-2}, 4.1×10^{-2},4.9×10^{-2}), and $r_s=1.4,1.3,1.3,1.2$ ($na_{\rm B}^3=9.6\times10^{-2}$, 1.0×10^{-2}, 1.1×10^{-2}, 1.4×10^{-2}), respectively.

IV. CONCLUSIONS AND DISCUSSIONS

We have analyzed the infrared or THz absorption spectrum of high-density e-h systems. The analysis is based on the QSEE for e-h systems that incorporates the wave function renormalization effect arising from the collective phase fluctuation associated with the center-of-mass motion of e-h pairs. It should be remarked that the quantum fluctuation effect often predominates in practical experiments for high-density e-h systems because the carrier density cannot become high enough to justify the BCS-like mean-field analysis. We show that the BEG is clearly found in the calculated spectra, even considering the strong quantum fluctuation. In the calculated spectra, we find the strong enhancement of the infrared absorption spectra originating from the strong mass renormalization effect. Furthermore, we find that the e-h pair correlation is distinctly enhanced by the intense visible pump light that causes the interband transition. These results suggest that the macroscopic quantum state in semiconductors is expected to be observed under intense photoexcitation.

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APPENDIX: A PROOF OF THE LONGITUDINAL *f*-SUM RULE FOR THE GRPA DIELECTRIC FUNCTION

In this appendix, we show that the GRPA dielectric function Eq. (17) rigorously satisfies the longitudinal f-sum rule when $\lambda=0$.

As usual,[30,48] we consider the retarded Green function given by

$$F^{\boldsymbol q}(t)=-i\Theta(t)\langle[\sigma_{\boldsymbol q}(t),\sigma_{\boldsymbol q}^\dagger(0)]\rangle, \tag{A1}$$

where $\sigma_{\boldsymbol q}=\sum_{\boldsymbol k}(c_{\boldsymbol{k+q}}^\dagger c_{\boldsymbol k}-d_{-\boldsymbol k}^\dagger d_{-\boldsymbol{k}-\boldsymbol{q}})$ is the charge-density operator. Introducing the spectral representation of $F^{\boldsymbol q}(t)$, we find that its Fourier transform fulfills the following identity:

$$V_{\boldsymbol q}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega\,{\rm Im}[F^{\boldsymbol q}(\omega)]=V_{\boldsymbol q}\langle[[H,\sigma_{\boldsymbol q}],\sigma_{\boldsymbol q}^\dagger]\rangle=-\omega_{\rm pl}^2. \tag{A2}$$

On the other hand, $F^{\boldsymbol q}(t)$ is expressed in terms of the Bogoliubov parameters as

$$F^{\boldsymbol q}(t)=-i\Theta(t)\sum_{\boldsymbol{k},\boldsymbol{p}}\{C_{\boldsymbol{k},\boldsymbol{k+q}}^{(0)}C_{\boldsymbol{p},\boldsymbol{p+q}}^{(0)}\langle[(\phi_{\boldsymbol{k+q}}^\dagger\boldsymbol{\tau}_0\phi_{\boldsymbol k})_t,(\phi_{\boldsymbol{p+q}}^\dagger\boldsymbol{\tau}_0\phi_{\boldsymbol p})_0^\dagger]\rangle-iC_{\boldsymbol{k},\boldsymbol{k+q}}^{(3)}C_{\boldsymbol{p},\boldsymbol{p+q}}^{(0)}\langle[(\phi_{\boldsymbol{k+q}}^\dagger\boldsymbol{\tau}_2\phi_{\boldsymbol k})_t,(\phi_{\boldsymbol{p+q}}^\dagger\boldsymbol{\tau}_0\phi_{\boldsymbol p})_0^\dagger]\rangle$$
$$+iC_{\boldsymbol{k},\boldsymbol{k+q}}^{(0)}C_{\boldsymbol{p},\boldsymbol{p+q}}^{(3)}\langle[(\phi_{\boldsymbol{k+q}}^\dagger\boldsymbol{\tau}_0\phi_{\boldsymbol k})_t,(\phi_{\boldsymbol{p+q}}^\dagger\boldsymbol{\tau}_2\phi_{\boldsymbol p})_0^\dagger]\rangle-C_{\boldsymbol{k},\boldsymbol{k+q}}^{(3)}C_{\boldsymbol{p},\boldsymbol{p+q}}^{(3)}\langle[(\phi_{\boldsymbol{k+q}}^\dagger\boldsymbol{\tau}_2\phi_{\boldsymbol k})_t,(\phi_{\boldsymbol{p+q}}^\dagger\boldsymbol{\tau}_2\phi_{\boldsymbol p})_0^\dagger]\rangle\}, \tag{A3}$$

where $\phi_{\boldsymbol k}=(\alpha_{\boldsymbol k},\beta_{-\boldsymbol k}^\dagger)$ and $(\phi_{\boldsymbol{k+q}}\boldsymbol{\tau}_0\phi_{\boldsymbol k})_t$ is the Heisenberg representation of $(\phi_{\boldsymbol{k+q}}\boldsymbol{\tau}_0\phi_{\boldsymbol k})$ at time t.

The SCARE indicates that $(\phi_{\boldsymbol{k+q}}\boldsymbol{\tau}_0\phi_{\boldsymbol k})_t$ is the linear combination of $(\phi_{\boldsymbol{k+q}}\boldsymbol{\tau}_0\phi_{\boldsymbol k})_0$ and $(\phi_{\boldsymbol{k+q}}\boldsymbol{\tau}_3\phi_{\boldsymbol k})_0$. Hence the first commutator on the right-hand side of Eq. (A3) vanishes. Similarly, we can show that the second and third terms vanish because these commutators are reduced to the linear combination of the annihilation and creation operators of the

Anderson mode. Therefore, $F^{\boldsymbol{q}}(t)$ is expressed in terms of $\boldsymbol{G}^{\boldsymbol{q}}_{\boldsymbol{k},\boldsymbol{p}}(t)$ defined by Eq. (8) as follows,

$$F^{\boldsymbol{q}}(t)=i\sum_{\boldsymbol{k},\boldsymbol{p}} C^{(3)}_{\boldsymbol{k},\boldsymbol{k}+\boldsymbol{q}}C^{(3)}_{\boldsymbol{p},\boldsymbol{p}+\boldsymbol{q}}\mathrm{tr}[(\boldsymbol{\tau}_0-\boldsymbol{\tau}_1)\boldsymbol{G}^{\boldsymbol{q}}_{\boldsymbol{k},\boldsymbol{p}}(t)]. \qquad (A4)$$

The Fourier transform of $\boldsymbol{G}^{\boldsymbol{q}}_{\boldsymbol{k},\boldsymbol{p}}(t)$ is directly obtained by solving its equation of motion by using the SCARE, and the result is written as follows:

$$\boldsymbol{G}^{\boldsymbol{q}}_{\boldsymbol{k},\boldsymbol{p}}(\omega)=\left(\frac{\omega\boldsymbol{\tau}_0-\Omega^{\boldsymbol{q}}_{\boldsymbol{k}}\boldsymbol{\tau}_3}{\omega^2-(\Omega^{\boldsymbol{q}}_{\boldsymbol{k}})^2}\right)\boldsymbol{\tau}_3\delta_{\boldsymbol{k},\boldsymbol{p}}-\left(\frac{V_{\boldsymbol{q}}C^{(3)}_{\boldsymbol{k},\boldsymbol{k}+\boldsymbol{q}}C^{(3)}_{\boldsymbol{p},\boldsymbol{p}+\boldsymbol{q}}}{\epsilon_0^{-1}\epsilon_{\boldsymbol{q}}(\omega)}\right)\times\left(\frac{\omega\boldsymbol{\tau}_0-\Omega^{\boldsymbol{q}}_{\boldsymbol{k}}\boldsymbol{\tau}_3}{\omega^2-(\Omega^{\boldsymbol{q}}_{\boldsymbol{k}})^2}\right)(\boldsymbol{\tau}_3-i\boldsymbol{\tau}_2)\left(\frac{\omega\boldsymbol{\tau}_0-\Omega^{\boldsymbol{q}}_{\boldsymbol{p}}\boldsymbol{\tau}_3}{\omega^2-(\Omega^{\boldsymbol{q}}_{\boldsymbol{p}})^2}\right)\boldsymbol{\tau}_3, \qquad (A5)$$

where $\Omega^{\boldsymbol{q}}_{\boldsymbol{k}}=E_{\boldsymbol{k}+\boldsymbol{q}}+E_{\boldsymbol{k}}$. Substituting Eq. (A4) into the left-hand side of Eq. (A2), we obtain the longitudinal *f*-sum rule,

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega\,\mathrm{Im}\left[\frac{1}{\epsilon_0^{-1}\epsilon_{\boldsymbol{q}}(\omega)}\right]=-\omega_{\mathrm{pl}}^2. \qquad (A6)$$

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