Direct Determination of Exciton Wave Function Amplitudes by the Momentum-Resolved Photo-Electron Emission Experiment

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We study conceptional problems of a photo-electron emission (PEE) process from a free exciton in insulating crystals. In this PEE process, only the electron constituting the exciton is suddenly emitted out of the crystal, while the hole constituting the exciton is still left inside and forced to be recoiled back to its original valence band. This recoil on the hole is surely reflected in the spectrum of the PEE with a statistical distribution along the momentum-energy curve of the valence band. This distribution is nothing but the square of the exciton wave function amplitude, since it shows how the electron and the hole are originally bound together. Thus, the momentum-resolved PEE can directly determine the exciton wave function. These problems are clarified, taking the Γ and the saddle point excitons in GaAs, as typical examples. New PEE experiments are also suggested.

I. INTRODUCTION

The momentum- or the angle-resolved photo-electron emission (PEE) spectroscopy is a powerful method to elucidate the energy band dispersion of solids. Nowadays, by combining this method with the time-resolved technique, we can see the temporal evolution of the energy distribution function in the momentum space, being able to detect the ultrafast transient dynamics of electrons directly [1]. Then, the basic experimental technique and theoretical understanding of this method have already been established, with a great contribution to the materials science [2, 3]. Here, however, we explore a new functionality of this momentum-resolved PEE spectroscopy.

In most cases of this PEE, we can tacitly assume that the initial one-electron energy level, from which the electron starts to be photo-excited and emitted, is definite, at any rate. If the starting state is an ordinary exciton, freely moving in an insulating crystal, however, such an initial one-electron energy level is indefinite in principle, even though the exciton itself has a well-defined energy level and a momentum. The free exciton is a bound state between an electron and a hole only in their relative space, being a superposition of various electron-hole pair excitations, extending over the wide energy region of the conduction and valence bands [4] . Thus, the starting one-electron energy level of this PEE is, in principle, indefinite.

By the PEE from this free exciton, only the electron is suddenly emitted out of the insulating crystal, while the hole is still left inside, and recoiled back to its original valence band. This recoil on hole has a statistical distribution along the momentum-energy curve of the valence band and surely appears in the spectrum of the PEE. This distribution is nothing but the square of the exciton wave function amplitude in the momentum space, since it shows how the electron and the hole are bound together. Thus, the momentum-resolved PEE can determine the exciton wave function amplitude, directly. Incidentally, it is already well known that the PEE can never determine the phase of the emitted electron waves.

At present, we have no other method to determine the exciton wave function directly by an experimental observation, although we can theoretically calculate it, by assuming an effective coulombic attraction between the electron and the hole, as well as their energy band dispersions [6]. Thus, this direct observation will contribute to open new aspects of the real electron-hole correlation in insulating solids, and will provide a novel functionality of the momentum-resolved PEE.

In order to perform this novel experiment, the following quite high level optical measurement technique is required. At first, a free exciton in an insulating crystal should be resonantly excited by an intense and temporary short laser pulse. A little after this laser pulse excitation having been completed, another intense and temporary short laser pulse should be shone to make the electron (constituting this exciton) photo-emitted out of the crystal. Finally, the energy and the momentum of this photo-emitted electron should be determined quite precisely. Thus, the present problem inevitably becomes a quite novel momentum- and time-resolved PEE.

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Very recently, such an experiment has been performed by Tanimura et al. in GaAs [7]. In this PEE, in addition to the ordinary Γ exciton in the band gap [8], the saddle point exciton just below the L valley has also been quite clearly detected. Only by the ordinary optical measurements, the existence of the saddle point exciton in GaAs has been suggested in Refs. [9] and [10]. Since this saddle point exciton is the bound state immersed in the continuum of the electron-hole pair excitations, it is known to be the Fano resonance state [11].

In this paper, we study the recoil effect of the hole in the PEE from the free exciton. Our goal is to conceptionally show that the momentum-resolved PEE spectrum directly reflects the exciton wave function. The momentum-resolved PEE can evaluate the real electron-hole correlation in the exciton, basically without any theoretical assistance.

For the explanation of this recoil effect, we need to evaluate the exciton wave function. Here we do it only by a simple method, since it may be enough to make our limited and conceptional purpose of the present paper clearly understood by readers. Then, prior to the argument of this recoil effect, we introduce a phenomenological model for exciton states in GaAs. We reproduce the known experimental results as simple as possible. We will be concerned only with resonant excitations to the Γ exciton or the saddle point one. Even in the cases of various non-resonant excitations, a conventional two-color two-photon PEE will also occur. However, their intensities are very weak as compared with those of resonant cases, and hence the non-resonant effects would not affect the essential aspect of the present argument.

This paper is organized as follows: In Sec. II, we introduce a phenomenological model for exciton states in GaAs. In Sec. III, we study the recoil effect of the hole in PEE from excitons, and show the momentum-resolved PEE can evaluate the exciton wave function amplitude directly. Finally, in Sec. IV, we give a summary.

II. A PHENOMENOLOGICAL MODEL FOR EXCITON IN GAAS

To evaluate the exciton wave functions for both the Γ exciton and the saddle point one in GaAs, we introduce a phenomenological model, wherein model parameters are adjusted so as to reproduce the experimental results of the excitons in GaAs. For this purpose, we start from the following Hamiltonian ($\equiv H$),

$$H = H_0 + H_{eh},$$  \hspace{1cm} (1)

$$H_0 \equiv \sum_{k} E_c(k) a_k^\dagger a_k + \sum_{k} E_h(k) b_k^\dagger b_k,$$  \hspace{1cm} (2)

$$H_{eh} \equiv -\sum_{l, \Delta l} U_{\text{eff}}(\Delta l) n_{e,l} n_{h,l+\Delta l},$$  \hspace{1cm} (3)

where $a_k^{(1)}$ and $b_k^{(1)}$ are annihilation (creation) operators for an electron and a hole with wave vector $k$, respectively. Spin index is neglected as we focus on a singlet exciton. The electron number and the hole one at a lattice site $l$ are defined as $n_{e,l} \equiv a_l^\dagger a_l$ and $n_{h,l} \equiv b_l b_l$, respectively, where $a_l (b_l) = N^{-1/2} \sum_k e^{ikl} a_k (b_k)$. $N$ is the total number of the lattice sites. $E_c(k)$ and $E_h(k)$ are the band dispersions for the conduction electron and for the valence hole, respectively. This energy is parametrized into an analytic function, whose details are given in Appendix A, by referring the result in Ref. [10]. The modeled band energy along the Γ-L line is shown in Fig. 1. In our model, all the energies are referenced from the valence band maximum. $-U_{\text{eff}}(\Delta l)$ is the effective attraction between an electron at a site $l$ and a hole at a site $l+\Delta l$, and is an adjustable parameter to be determined so as to reproduce the experimentally observed energies of excitons. The unit of length is the spatial periodicity of the crystal along the Γ-L direction.

The excitation spectrum ($\equiv S(E)$) by a light with the photon energy ($\equiv E$) is given as

$$S(E) = \sum_j | \langle j | P | 0 \rangle |^2 \delta (E - E_j),$$  \hspace{1cm} (4)

where $P$ is the polarization or photo-excitation operator, and is given as

$$P = N^{-\frac{1}{2}} \sum_l (a_l^\dagger b_l^\dagger + \text{h.c.}) = N^{-\frac{1}{2}} \sum_k (a_k^\dagger b_{-k}^\dagger + \text{h.c.}).$$  \hspace{1cm} (5)

$| 0 \rangle$ represents the electron-hole vacuum and $| j \rangle$ represents the exact excited state with the energy $E_j$, and hence

$$H | j \rangle = E_j | j \rangle.$$  \hspace{1cm} (6)
By using the relation
\[ \delta(E - E_j) = -\frac{1}{\pi} \text{Im} \left( \frac{1}{(E + i\epsilon) - E_j} \right), \]  
(7)
(where \( \epsilon \) is a positive infinitesimal,) Eq. (1) is written as
\[ S(E) = -\frac{1}{\pi} \text{Im} \langle 0 \mid P \frac{1}{(E + i\epsilon) - H} P \mid 0 \rangle. \]  
(8)
In the above equation, the right hand side can be expanded with respect to \( H_{\text{eh}} \) as
\[ \frac{1}{(E + i\epsilon) - H} = \frac{1}{(E + i\epsilon) - H_0} + \frac{1}{(E + i\epsilon) - H_0} H_{\text{eh}} \frac{1}{(E + i\epsilon) - H_0} + \cdots. \]  
(9)
This is the Green’s function of an excitonic state with an energy \( E \), which is given as
\[ G(E + i\epsilon) \equiv \frac{1}{(E + i\epsilon) - H} = \frac{1}{(E + i\epsilon) - H_0 - H_{\text{eh}}}. \]  
(10)
This can be also expanded with respect to \( H_{\text{eh}} \) as
\[ G = G_0 + G_0 H_{\text{eh}} G_0 + G_0 H_{\text{eh}} G_0 H_{\text{eh}} G_0 + \cdots, \]  
(11)
where \( G_0 \) is the Green’s function for the non-interacting system, and is given as
\[ G_0(E + i\epsilon) \equiv \frac{1}{(E + i\epsilon) - H_0}. \]  
(12)
By using this Green’s function, the excitation spectrum, Eq.(8), is expressed as
\[ S(E) = -\frac{1}{\pi} \text{Im} \langle 0 \mid PG(E + i\epsilon)P \mid 0 \rangle. \]  
(13)
If we take only the zeroth order term of \( G \), \( S(E) \) becomes the combined excitation density of states (DOS) of the non-interacting electron-hole pair (\( \equiv D(E) \)) as
\[ D(E) = -\frac{1}{\pi} \text{Im} \langle 0 \mid PG_0(E + i\epsilon)P \mid 0 \rangle. \]  
(14)
Then, as a scalar, we can define
\[ G_0(E + i\epsilon) \equiv \langle 0 \mid PG_0(E + i\epsilon)P \mid 0 \rangle. \]  
(15)
FIG. 2: (Color online) The Combined DOS $D(E)$ and $R(E)$ are shown. The dashed lines are represent $-U_{\Gamma}^{-1}$ and $-U_{S}^{-1}$, which are determined to so as to reproduce the experimental energy of excitons. For more details, see the text.

In the present model, it is explicitly written as

$$G_0(E + i\epsilon) = \frac{1}{N} \sum_k \frac{1}{E + i\epsilon - E_e(k) - E_h(-k)},$$

(16)

and

$$D(E) = \frac{1}{N} \sum_k \delta(E - E_e(k) - E_h(-k)),$$

(17)

with the condition $\int D(E) dE = 1$. We can also define

$$R(E) - i \pi D(E) \equiv G_0(E + i\epsilon),$$

(18)

where the real part $R(E)$ is connected with $D(E)$ by the the Kramers-Kronig relation as,

$$R(E) = \int dE' \frac{D(E')}{E - E'},$$

(19)

To determine the excitonic state so as to reproduce the experimental result, we consider only the effective on-site coulombic interaction between an electron and a hole, $U_{\text{eff}}(\Delta l) \rightarrow U_m = U_{\text{eq}}(0)$. In order to take original long range nature of the coulombic interaction into account, we assume this $U_m$ depends on the resultant exciton. Hence, we assign $m = \Gamma$ for the $\Gamma$ exciton and $m = S$ for the saddle point one, respectively. Then, the Green’s function for exciton is now represented as

$$G(E) \equiv \langle 0 \mid PG(E)P \mid 0 \rangle = \frac{G_0(E)}{1 + G_0(E)U_m}.$$

(20)

The estimated $D(E)$ and $R(E)$ are shown in Fig. 2 wherein each exciton state corresponds to the crossing point of the $R(E)$ curve and $-U_m^{-1}(1 + R(E)U_m = 0)$. To determine the adequate $U_m$'s, we use the energy of excitons obtained by the experiment.: (1) The $\Gamma$ exciton is just 5 meV below the conduction band bottom, and the band gap energy is set at 1.45 eV in our model. (2) For the saddle point exciton, its excitation energy is around 3 eV from Ref. \[7\]. From these conditions, we obtain $U_{\Gamma} = 2.1$ eV for the $\Gamma$ point exciton, and $U_{S} = 0.88$ eV for saddle point one, respectively.

In our model, anti-resonance states corresponding to the $\Gamma$ exciton and the saddle point one may also exist. However, they are not relevant to the present purpose.

The evaluated $S(E)$ is shown in Fig. 3. Both the $\Gamma$ exciton and the saddle point one basically give Lorentz shapes. However, Fano type anti-symmetric tails in each low intensity regions can also be seen clearly. Since the saddle point exciton is a transient state, the obtained spectral width reflects the decay rate ($\equiv \Gamma_S$). One can evaluate this $\Gamma_S$, by assuming the Lorentz form of spectral shape as

$$S(E) \propto \frac{1}{(E_{x,S} - E)^2 + \Gamma_S^2},$$

(21)
where \( E_{x,S} \) is the energy of the saddle point exciton and is set at 3.0 eV in the present model. By the curve fitting, we can estimate that \( \Gamma_S \approx 0.1 \) eV. Thus, the present model well reproduces the basic characteristics of the experiments \[ \text{[7, 10]}. \]

III. PEE FROM EXCITONS

In this section, we discuss the PEE from the excitons introduced in the preceding section. Since we focus on the resonant excitation to each exciton in the two-photon process as mentioned before, we start from the condition that the \( \Gamma \) exciton or the saddle point one has already been created. When this exciton is shone by the light with an energy (\( \equiv E_p \)), its electron is excited up to the high energy continuum state within the crystal. This high energy continuum is the literal “high energy continuum”, in the sense that the electron with any crystal momentum can find corresponding state at “any high energy”, and can go out of the crystal, whereafter it is sorted according to the initial crystal momentum and the energy. This is nothing but the function of the momentum-resolved PEE apparatus.

In the PEE from the exciton, only the electron is emitted out from the crystal and the hole is still left inside, as mentioned before. Since the electron and the hole are bound, the energy of the emitted electron is shifted by recoil effect of the hole. Then, the energy of emitted electron (\( \equiv E_{em}(k) \)), sorted in the crystal momentum \( k \), is given from the energy conservation law as

\[
E_{x,m} + E_p = E_{em}(k) + W + E_r(k),
\]

where \( W \) is the energy to remove an electron from the crystal, and \( E_{x,m} \) is the energy of exciton with \( m = \Gamma \) for the \( \Gamma \) exciton and \( m = S \) for the saddle point one, as mentioned before. \( E_r(k)(= E_h(-k) > 0) \) is the recoil energy of the hole. \( E_{em}(k) \) depends on \( E_{x,m}, E_p \) and \( W \), as well as \( E_r(k) \). However, these former three quantities can be fixed, once we fix a target exciton state. Then, by observing \( E_{em}(k) \) experimentally, we can determine \( E_r(k) \). Since the hole is recoiled back to its original valence band, this recoil on hole has a statistical distribution along the momentum-energy curve of the valence band. The intensity of this statistical distribution corresponds to the existing probability of the electron-hole pair constituting the exciton. Thus, the statistical distribution obtained by the recoil on hole is nothing but the square of the exciton wave function amplitude in the momentum space, since it shows how the electron and hole are bound together.

To see this, we calculate the wave function for the exciton. The Green’s function itself in Eq. \[ \text{[10]} \] is already a kind of wave function \[ \text{[12]} \]. However, here we derive it more straightly. The eigenstate for the \( \Gamma \) exciton with the total momentum zero can be written as

\[
| \Gamma \rangle = N^{-\frac{1}{2}} \sum_k f_\Gamma(k) a_k^\dagger b_{-k}^\dagger | 0 \rangle,
\]

where \( f_\Gamma(k) \) is the wave function of the \( \Gamma \) exciton, and we determined it variationally. The relation between the present \( k \) space representation for the exciton and the real space one is shown in the Appendix B.

Let us define the Lagrangian (\( \equiv \mathcal{L} \)) as

\[
\mathcal{L}(E_{x,\Gamma}) = \langle \Gamma | (H_0 + H_{eh} - E_{x,\Gamma}) | \Gamma \rangle,
\]

FIG. 3: (Color online) The excitation spectrums for (a) \( \Gamma \) exciton and (b) Saddle point exciton are shown.
which satisfies
\[ \frac{\partial \mathcal{L}(E_{x, \Gamma})}{\partial f^*_r(k)} = 0. \]  
(25)

Since \( U_{\text{eff}}(\Delta l) \) in Eq. (3) is now \( U_\Gamma \), from Eqs. (24) and (25), we obtain
\[ f_r(k) \propto \frac{1}{E_{x, \Gamma} - E_e(k) - E_h(-k)}. \]  
(26)

In case of the Fano type saddle point exciton, due to its transient nature, the above variational method cannot be used. Here, however, we use a simple extrapolation method to calculate the wave function for the saddle point exciton. At first, we tentatively or formally assume that the saddle point exciton has an infinite life time, \( \varepsilon^{-1} \). In this case, this exciton becomes a good eigenstate of the exciton Hamiltonian, Eq. (4), being locally stable (or an extremal) state, although it is only around the saddle point region with the energy \( \sim E_{x,S} + i\varepsilon \). The eigenstate for the saddle point exciton with the total momentum zero can be written as
\[ | S \rangle = N^{-1/2} \sum_k f_S(k) a_k^\dagger b_{-k}^\dagger | 0 \rangle, \]  
(27)

where \( f_S(k) \) is the wave function of the saddle point exciton. Hence, we can determine the wave function \( f_S(k) \) from the locally stable or the extremal condition of the Lagrangian \( \mathcal{L}(E_{x,S}) \), which is given by
\[ \mathcal{L}(E_{x,S}) = \langle S | \{ H_0 + H_{eh} - (E_{x,S} + i\varepsilon) \} | S \rangle. \]  
(28)

The locally stable or the extremal condition becomes as
\[ \frac{\partial \mathcal{L}(E_{x,S})}{\partial f_S^*(k)} = 0. \]  
(29)

By replacing \( U_S(\Delta l) \) in Eq. (3) by \( U_S \), we obtain
\[ f_S(k) \propto \frac{1}{E_{x,S} + i\varepsilon - E_e(k) - E_h(-k)}. \]  
(30)

from Eqs. (28) and (29).

In the next, only within this context, we introduce its finite life time, by the extrapolation, \( \varepsilon \rightarrow \Gamma_S \), from the previous Green’s function calculation result, and we finally get
\[ f_S(k) \propto \frac{1}{E_{x,S} + i\Gamma_S - E_e(k) - E_h(-k)}. \]  
(31)

The calculated \(|f_m(k)|^2\)'s are shown in Figs. 4 and 5. One can see the one-to-one correspondence between \(|f_m(k)|^2\) and \(E_r(k)\). For example, the peak of \(|f_S(k)|^2\) in Fig. 4 is around the saddle point between the \(\Gamma\) valley and the \(L\) one, with \(E_r(k) \sim 0.5\) eV. Thus, the measured \(E_m(k)\) in the momentum-resolved PEE reflects the wave function of the exciton through \(E_r(k)\).

The \(\Gamma\) exciton is the ordinary one in the energy gap, and with a large electron-hole radius, or only in the small \(k\) region around the \(\Gamma\) point. On the other hand, the saddle point exciton arises mainly from the van Hove singularity of \(D(E)\) due to the \(L\) valley structure. Because of the electron-hole attraction, however, it has shifted to a little below this singular point. Its radius is smaller than that of the \(\Gamma\) exciton, or it is in the larger \(k\) region around the \(\Gamma_S\) point. It is resonating with the high energy region of the \(L\) valley as seen from \(D(E)\) in Fig. 2 and becomes the Fano state.

Here, we would like to suggest new PEE experiments not only for the excitons in GaAs, but also other ones in other materials. Especially, we would like to recommend the excitons with a large binding energy and (or) a short electron-hole radius. In this case, the wave function \(f(k)\) extends to a wide region in the Brillouin zone, as shown in Fig. 5. Hence, the energy-momentum resolution of the PEE experiment becomes rather easy. The direct experimental measurement of this \(f(k)\) in the large \(k\) region is very important, since it includes the true information of the real electron-hole attraction at a short distance of each material. Although various studies have already been devoted for this problem, true natures of electron-hole attraction at a short distance in each material are, even at present, still not so well clarified, since they are directly related to complicated “local” atomic, electronic and dielectric characteristics of each material, which is not in the ground state, but under a resonant optical “excitation”. By the experiments suggested here, research for this problem is expected to develop greatly.
IV. SUMMARY

In GaAs, we have, thus, shown that the PEE from the exciton can determine the square of the exciton wave function amplitude in the momentum space directly, reflecting the recoil effect of the hole left in valence band. Since the PEE spectrum itself is the exciton wave function, we can obtain the information of the real electron-hole correlation in insulating solids, basically without any theoretical assistance. When we look at this thing from an opposite standing point, the momentum-resolved PEE spectrum from the exciton may provide an opportunity to examine a validity of various theoretical and computational methods. These new aspects are due to the two-body nature of exciton, with the apparent break down of the one-body picture, in the initial state of the PEE. On this point, the PEE from the exciton is quite unique, and gives us an interesting insight into the many-body effect in quantum systems.
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Appendix A: Modeled band dispersion for GaAs

The band dispersion for GaAs is modeled without losing their basic characteristics, by referring the result in Ref. [10]. In our parametrization, the unit of length is the lattice constant along [111] direction of the GaAs crystal, and all the energies are referenced from the top of the valence band. In the reciprocal lattice space, the wave vector along the Γ-L direction is represented as $k_L$. Two wave vectors normal to $k_L$ are represented as $k_{L1}$ and $k_{L2}$, respectively. All the energies are given in eV. In our model, spin and orbital degeneracies are neglected, since there is no mixing among these degenerate states.

In conduction band, the wave vector at the saddle point between Γ valley and L one is set to $k_S = \alpha_S \pi$, where $k_L = 0$ for Γ point and $| k_L | = \pi$ for L one. The band dispersion for Γ valley ($0 \leq k_{L1}, k_{L1}, k_{L2} \leq k_S$) is parametrized with the following forms:

$$E_c(k) = E_g + c_g \left\{ \left( \frac{k_{L1}}{k_S} \right)^2 + \left( \frac{k_{L2}}{k_S} \right)^2 \right\},$$

where $E_g$ is the band gap. The band dispersion for L valley ($k_S \leq k_L \leq \pi$, $0 \leq k_{L1}, k_{L2} \leq k_S$) is parametrized as

$$E_c(k) = E_{g2} + c_{L1} \left\{ d_L \left( \frac{\pi - k_{L1}}{\alpha_L \pi} \right)^2 - \left( \frac{\pi - k_{L1}}{\alpha_L \pi} \right)^4 \right\} + c_{L2} \left\{ \left( \frac{k_{L1}}{k_S} \right)^2 + \left( \frac{k_{L2}}{k_S} \right)^2 \right\}.$$  \hspace{1cm} (A2)

The continuity of the conduction band at the saddle point is of no importance, since only the combined (joint) DOS between the conduction band and the valence one with the total momentum zero dominates the degree of the resonance of the photo-excited electron-hole pair.

For the valence band, we consider only the heavy hole band, since the light hole band is irrelevant for our present purpose. We parametrize the band dispersion for the Γ valley region ($0 \leq k_{L1}, k_{L1}, k_{L2} \leq k_S$) as

$$E_h(k) = c_h \left\{ \left( \frac{k_{L1}}{k_S} \right)^2 + \left( \frac{k_{L2}}{k_S} \right)^2 \right\},$$

while for the L valley region ($k_S \leq k_L \leq \pi$, $0 \leq k_{L1}, k_{L2} \leq k_S$) as

$$E_h(k) = E_{g3} - d_h \left( \frac{\pi - k_{L1}}{\alpha_L \pi} \right)^2 + c_h \left\{ \left( \frac{k_{L1}}{k_S} \right)^2 + \left( \frac{k_{L2}}{k_S} \right)^2 \right\}.$$  \hspace{1cm} (A4)

We set $\alpha_S = 0.3059$, $\alpha_L = 0.6941$, $E_g = 1.45$, $E_{g2} = 1.75$, $E_{g3} = 1.40$, $c_g = 1.1$, $c_{L1} = 0.80$, $c_{L2} = 1.1$, $c_h = 0.50$, $d_L = 2.0$, and $d_h = 0.90$. The resultant band dispersion along Γ-L line is shown in Fig. 1. It should be noted that there exist eight equivalent L points in the first Brillouin zone of GaAs, and this fact is taken into account in our calculation.

Appendix B: Exciton wave function in the Fourier space and the real space

Here, we explain the relation between the Fourier ($k$) space and the real (or relative) space of the photo-excited exciton wave function with no total momentum.

From Eq. (23), the Γ exciton is given as

$$| \Gamma \rangle = N^{-\frac{1}{2}} \sum_k f_\Gamma(k) a_{k}^\dagger b_{-k}^\dagger | 0 \rangle.$$  \hspace{1cm} (B1)
By using the lattice-site representations for the above electron and the hole operators, we can easily rewrite Eq. (B1) into a following localized excitation form, as

\[ | \Gamma \rangle = N^{-\frac{1}{2}} \sum_{k,l \Delta l} f_{\Gamma}(k)e^{-ik\cdot\Delta l}a_l b_{l+\Delta l}^\dagger | 0 \rangle, \]

(B2)

wherein the electron is at a lattice site \( l \), while the hole is at a site \( l + \Delta l \), keeping \( \Delta l \) distant from the electron. It is needless to say that this \( \Delta l \) is the relative space of the electron-hole pair. From this Eq. (B2), we can formally derive the photo-excited exciton wave function (\( \equiv w(\Delta l) \)) in the real (or relative) space \( \Delta l \) as

\[ w(\Delta l) \equiv N^{-1} \sum_k f_{\Gamma}(k)e^{-ik\cdot\Delta l}. \]

(B3)

Its normalization condition is as follows:

\[ \sum_{\Delta l} | w(\Delta l) |^2 = 1. \]

(B4)

Using this \( w(\Delta l) \) for Eq. (B2), we get the ordinary exciton in the real space as

\[ | \Gamma \rangle = N^{-\frac{1}{2}} \sum_{l \Delta l} w(\Delta l)a_l b_{l+\Delta l}^\dagger | 0 \rangle. \]

(B5)

Thus, we can see the Fourier component \( f_{\Gamma}(k) \) or its absolute value has a key information of the exciton. This is common to the other photo-excited exciton wave functions with no total momentum.

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