

Renormalized bosonic interaction of excitons

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An effective bosonic Hamiltonian of $1s$ excitons with “spin” degrees of freedom in two dimensions is obtained through a projection procedure, starting from a conventional electron-hole Hamiltonian \mathcal{H}_{eh} . We first demonstrate that a straightforward transformation of \mathcal{H}_{eh} into a Hamiltonian of bosonic excitons does not give the two-body interaction between an “up-spin” exciton and a “down-spin” exciton, which are created by the left- and right-circularly polarized light beams, respectively. We then show that this interaction is generated through a projection procedure onto the subspace spanned by $1s$ excitons, as a renormalization effect coming from higher exciton states. The projection also renormalizes the interaction between $1s$ excitons with the same spins by a large amount. These renormalization effects are crucial for the polarization dependence of the optical responses from semiconductors. The present theory gives the microscopic foundation of the phenomenology that was successfully applied to the analysis of four-wave mixing experiments in GaAs quantum wells strongly coupled to the radiation field in a high- Q microcavity.

I. INTRODUCTION

Various theoretical methods have been developed to study optical properties of semiconductors.^{1–5} These methods can be divided into two groups: a “fermionic method” and a “bosonic method.” The fermionic method^{1,2} is formulated on the Hilbert space of two fermionic species, i.e., photoexcited electrons and holes in semiconductors. In this method, one solves the coupled equations of motions, the semiconductor Bloch equations (SBE),² for the particle densities of electrons and of holes, and for the expectation value of the polarization of the system. Since this method basically relies on the Hartree-Fock (HF) approximation for electrons and holes, it is suitable for higher excitation density, where Coulombic screening effects guarantee that exciton correlation effects become less crucial. In order to extend this method to lower excitation density, where exciton correlations become important, the three- and four-particle correlations should be taken into account and the truncation scheme should be improved.

On the other hand, the bosonic method^{3–5} is based on excitons, bound states of an electron and a hole. Regarding this elementary excitation as a bosonic particle, one constructs an effective Hamiltonian of bosonic excitons, from which physical quantities, such as linear and nonlinear response functions, can be calculated. Here, the effective Hamiltonian should be constructed very carefully, as we will show in this paper. The bosonic method is believed to be valid when the optical excitation is weak and when the photon energy is close to the exciton energy (see Sec. II A), because under these conditions main contributions to the optical properties should come from the excitons created in the system. With increasing the photoexcitation intensity, the Coulombic force becomes weaker by increased screening and/or increased Fermi energy, and the contribution from free carriers becomes more important. Hence the bosonic method is not valid at high excitation intensity.

A remarkable feature of the light field as a probing tool of

materials is that it has the polarization degrees of freedom. Recent experimental studies of semiconductor optics make the best use of this fact to reveal more detailed properties of excited states. The polarization degrees of freedom of photons induce “spin” degrees of freedom of excitations (see below). Experiments, including the four-wave mixing experiment in the time domain, have revealed the crucial roles of the interaction between an exciton (or an eh pair) created by the left-circularly polarized light and the one created by the right-circularly polarized light.^{6,7} However, most of the existing theories could not treat the polarization dependence correctly. For example, since the SBE (in its original form) were discussed within HF theory,² the excitations with different polarization degrees of freedom (e.g., left and right) are completely decoupled. Hence it is impossible to explain the polarization dependence of the optical response. It is also the case with bosonic theories: in Ref. 3, the polarization degrees of freedom were not included in the derivation of the interaction of excitons. References 8 and 9 treated the polarization degrees of freedom of excitons. However, since they essentially end up with the HF approximation of $1s$ excitons, the excitons with opposite spins are completely decoupled,¹⁰ as will be discussed in Sec. III C.

In this paper, we show that it is not at all trivial to derive the interaction of excitons created by opposite polarized light in a bosonic theory. One crucial result is that to go beyond the HF approximation of $1s$ excitons becomes of paramount importance. One of such investigations is seen in Ref. 11, where the modification of exciton binding energy is discussed.

Up to now, a strong objection against the bosonic method was that excitons are not bosonic particles. Moreover, until recently there were almost no experimental evidence which verifies an effective bosonic theory both qualitatively and quantitatively. However, important experimental evidence for the validity of the boson picture has been reported recently in a two-dimensional system.^{12,13} The experiment, discussed in detail below, is a nonlinear version of normal-

mode coupling in a high- Q microcavity. This experiment was stimulated by the fact that optical responses from semiconductors that are strongly coupled to the photon field have received much attention in recent years. The strong coupling is obtained by confining optically active regions in a high- Q microcavity, which is made possible by the development of nanostructure technology. For linear optical responses, the most noticeable phenomenon is a large Rabi splitting, which has been observed in both inorganic¹⁴ and organic¹⁵ semiconductors (insulators).

Kuwata-Gonokami and co-workers have performed four-wave mixing in a GaAs quantum well (QW) that is strongly coupled to the radiation field in a high- Q microcavity.^{12,13} They investigated several polarization configurations under the condition that the excitation density is very low, and measured the polariton-polariton scattering signals. The experimental results were successfully reproduced by a phenomenological model, which is called the weakly interacting boson model (WIBM).^{12,13,16} In the WIBM, excitons are treated as interacting bosons. The good agreement with the experimental results demonstrated that the bosonic picture is reliable in the lower-excitation regime. The Hamiltonian of the WIBM is

$$\begin{aligned}
\mathcal{H}_{\text{WIBM}} = & \sum_{\mathbf{k}, \sigma} [\omega_c a_{\mathbf{k}\sigma}^\dagger a_{\mathbf{k}\sigma} + \omega_e b_{\mathbf{k}\sigma}^\dagger b_{\mathbf{k}\sigma} \\
& + g(a_{\mathbf{k}\sigma}^\dagger b_{\mathbf{k}\sigma} + b_{\mathbf{k}\sigma}^\dagger a_{\mathbf{k}\sigma})] \\
& + W \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3} b_{\mathbf{k}_1}^\dagger + b_{\mathbf{k}_2}^\dagger + b_{\mathbf{k}_3}^\dagger - b_{\mathbf{k}_1 - \mathbf{k}_2 + \mathbf{k}_3} \\
& + R \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \sigma} b_{\mathbf{k}_1}^\dagger b_{\mathbf{k}_2}^\dagger b_{\mathbf{k}_3}^\dagger b_{\mathbf{k}_1 - \mathbf{k}_2 + \mathbf{k}_3, \sigma} \\
& - g\nu \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \sigma} (b_{\mathbf{k}_1}^\dagger b_{\mathbf{k}_2}^\dagger a_{\mathbf{k}_3}^\dagger b_{\mathbf{k}_1 - \mathbf{k}_2 + \mathbf{k}_3, \sigma} \\
& + b_{\mathbf{k}_1}^\dagger a_{\mathbf{k}_2} b_{\mathbf{k}_3}^\dagger b_{\mathbf{k}_1 - \mathbf{k}_2 + \mathbf{k}_3, \sigma}), \quad (1)
\end{aligned}$$

where photons and excitons are described by boson operators $a_{\mathbf{k}\sigma}$ and $b_{\mathbf{k}\sigma}$, respectively, with the spin index $\sigma = \pm$. This Hamiltonian has three parameters: W is the interaction strength of the excitons with opposite spin, R is the interaction strength of the excitons with the same spin, and $\nu (> 0)$ is the filling factor.¹⁷ The ratios of these three parameters were measured as $W/g\nu = -3.0$ and $R/g\nu = 0.2$. Once these three parameters are fixed, the experimental results for all polarization configurations are fitted very well. This indicates that a bosonic method is quite reliable in the lower excitation density. Nevertheless, existing bosonic theories³⁻⁵ could not give a *microscopic* foundation of the WIBM. In particular, they could not derive the “ W term,” the second term of the right-hand side of Eq. (1), see Sec. III. The term was introduced intuitively by an analogy with the discussion of a hydrogen molecule, when polarization dependence was discussed.^{5,12} Namely, from the analogy of a four-body problem corresponding to a hydrogen molecule, it was expected intuitively that the attractive interaction (the interaction term with a negative prefactor) should exist in a many-body Hamiltonian. It was also considered that the crucial interaction of exciton for biexciton formation has the form of

$b_+^\dagger b_-^\dagger b_- b_+$ with a negative prefactor, corresponding to the W term in Eq. (1), and that the binding energy of a biexciton is $|W|$. However, we will show that such a simple picture is wrong; it is not this term which is responsible for biexciton formation. The excitons which are not dipole active play an important role for the formation, as shown in Sec. V E.

The purpose of our paper is to derive an effective boson Hamiltonian of $1s$ excitons with “spin” degrees of freedom under the three conditions cited in Sec. II A. Through this derivation, a microscopic foundation of the WIBM, especially the interaction terms of excitons with the scattering amplitude W and R in the WIBM, is obtained. We show that effects of exciton states higher than $1s$ are crucial when deriving the effective Hamiltonian of $1s$ excitons:¹⁸ these effects yield the two-body interaction term corresponding to W in Eq. (1), and largely modify the strength of the interaction term corresponding to R in Eq. (1). The higher exciton states are taken into account through a projection procedure, and, as shown in Sec. II B, it is crucial to project the whole exciton space onto the $1s$ excitons subspace and to reconstruct of the interaction of $1s$ excitons. In a more general sense, the bosonic description of fermionic systems in two-dimension is one of the most attractive fields in condensed-matter theory.^{19,20} If the bosonic method is useful in two-dimensional semiconductors, it would be one of the examples of the success of a two-dimensional “bosonization.” We also discuss the relation of the exciton-exciton interaction and the formation of a biexciton.

The organization of this paper is as follows: In Sec. II, after explaining the necessity of the projection, exciton bosonic operators are introduced and exciton spins are defined. In Sec. III, the exciton interaction is discussed without the projection, which corresponds to the two-dimensional case of the existing bosonic theory.³ It is shown that the exciton interaction corresponding to the W term in WIBM is *not* obtained in this approximation. In Sec. IV, which is the main part of this paper, the projection procedure is introduced,¹⁸ resulting in appearance of the W term and modifications of the interaction strength of excitons with the same spins. Discussions and remarks are presented in Sec. V. In Sec. VI, the results obtained in this paper are summarized. Throughout this paper, the units $\hbar = 1, e = 1, \epsilon_0 = 1$ are used.

II. MODEL AND STRATEGY

A. Conditions for the validity of the effective theory

The purpose of the present paper is to derive an *effective* Hamiltonian of $1s$ excitons in a QW. Here, the “effective Hamiltonian” means that it (approximately) describes the optical responses of the QW correctly, although it is a function of $1s$ exciton operators only.

Since any effective theory is valid only in some specific physical situations, we first clarify the physical situations or conditions under which we construct the effective theory. In compensation for this limitation, the effective theory describes the physics simply. In contrast, such insights are hardly obtainable from straightforward calculations using the electron-hole Hamiltonian.

We construct an effective Hamiltonian that describes optical responses of semiconductor QW’s under the following conditions: (i) The excitation is weak (weak excitation re-

gime) so that the mean distance l_{ex} of photocreated (virtual and/or real) excitons is much larger than the Bohr radius a_0 of the $1s$ exciton;

$$l_{ex} \gg a_0, \quad (2)$$

and (ii) all the photon energies (pump, probe, and signal) $\hbar \omega_i$ are close to the energy E_{1s} of the $1s$ exciton;

$$|E_{1s} - \hbar \omega_i| \ll |E_{2p} - \hbar \omega_i| \quad (i = \text{pump, probe, signal}), \quad (3)$$

where E_{2p} is the energy of $2p$ excitons, and (iii) the linewidth Γ_{1s} of $1s$ exciton is smaller than the detuning energies;

$$\Gamma_{1s} < |E_{1s} - \hbar \omega_i| \quad (i = \text{pump, probe, signal}). \quad (4)$$

Physical meaning of these conditions will be explained in Sec. V D.

Under these conditions, nonlinear optical signals would not be strong in general. One must therefore devise experimental methods for detecting the signals with a high sensitivity. For this purpose, a well-prepared method was proposed by Kuwata-Gonokami *et al.*,¹² in which an optical cavity with a high- Q value is utilized. This point will be discussed later in Sec. V D.

B. Necessity of projection and renormalization

The Hamiltonian of an eh system is defined on the eh Hilbert space \mathbf{H}_{eh} that is a fermionic Hilbert space spanned by e and h states. As long as photo excitations are concerned, all excited states are charge neutral. We can thus limit ourselves in the charge neutral sector of \mathbf{H}_{eh} . The effective Hamiltonian of excitons, which is defined on a bosonic Hilbert space spanned by the exciton states, should describe the dynamics of the eh system in this charge neutral sector of \mathbf{H}_{eh} . Note that there are two (or more) choices for the bosonic Hilbert space: one is the whole exciton space \mathbf{H}_{ex} that is spanned by all exciton states, whereas the other is its subspace \mathbf{H}_{1s} that is spanned by the $1s$ states only. The effective Hamiltonian depends on the choice of the bosonic Hilbert space.

We will first consider in Sec. III the effective Hamiltonian defined on \mathbf{H}_{ex} . Its interaction part takes the following form:

$$\begin{aligned} \mathcal{H}^{full-int} = & \frac{1}{2\Omega} \sum_{\{S\}} \sum_{\{\nu\}} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} V_{ex}(\mathbf{q}; \{\nu\}; \{S\}) \\ & \times b_{\mathbf{k}+\mathbf{q}\nu_1}^\dagger s_1 b_{\mathbf{k}'-\mathbf{q}\nu_2}^\dagger s_2 b_{\mathbf{k}'\nu_4} s_4 b_{\mathbf{k}\nu_3} s_3 + \dots, \end{aligned} \quad (5)$$

where $b_{\mathbf{k}\nu S}$ denotes the excitonic boson operators (defined later), and Ω is the area of the QW. The first term denotes the two-body interactions between excitons of various states, and \dots denotes three- and more-body interactions. Since *exact* calculations, which take account of *all* terms of Eq. (5) to infinite order, are impossible, *one has to make approximations*. Because of conditions (i)–(iii) in Sec. II A, it is tempting to take only the two-body interactions of $1s$ excitons, \mathcal{H}_{1s}^{int} , among many terms of Eq. (5);

$$\begin{aligned} \mathcal{H}_{1s}^{int} = & \frac{1}{2\Omega} \sum_{\{S\}} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} V_{ex} \\ & \times (\mathbf{q}; \{\nu=1s\}; \{S\}) b_{\mathbf{k}+\mathbf{q}1s}^\dagger s_1 b_{\mathbf{k}'-\mathbf{q}2s}^\dagger s_2 b_{\mathbf{k}'4s} s_4 b_{\mathbf{k}3s} s_3, \end{aligned} \quad (6)$$

where $b_{\mathbf{k}S}$ denotes $b_{\mathbf{k}\nu S}$ with $\nu=1s$.²¹ Unfortunately, however, we will show later that the replacement $\mathcal{H}^{full-int} \rightarrow \mathcal{H}_{1s}^{int}$ is a very poor approximation, which cannot explain the experimental results *even qualitatively*. This originates from the complete neglect of effects of higher exciton states $\nu=2p, 3d, \dots$, which, however, play important roles as intermediate states.

To resolve this difficulty, we will then consider in Sec. IV the effective Hamiltonian defined on \mathbf{H}_{1s} . It is obtained by the projection procedure, by which the dynamics in \mathbf{H}_{ex} is projected onto the subspace \mathbf{H}_{1s} that is spanned by $1s$ excitons only. In general, a projection procedure generates dissipative terms in the projected dynamics in the subspace. Under conditions (i)–(iii) of Sec. II A, however, we may neglect the dissipative terms. Namely, the dynamics in \mathbf{H}_{1s} can be described by an Hamiltonian dynamics, whose Hamiltonian (effective Hamiltonian) is a function of the boson operators $b_{\mathbf{k}S}$ for $1s$ excitons only. Its interaction part, $\tilde{\mathcal{H}}_{1s}^{full-int}$, consists of two- and more-body interactions among $1s$ excitons. For example, the two-body interaction $\tilde{\mathcal{H}}_{1s}^{int}$ takes the following form:

$$\tilde{\mathcal{H}}_{1s}^{int} = \frac{1}{2\Omega} \sum_{\{S\}} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} \tilde{V}_{ex}(\mathbf{q}; \{S\}) b_{\mathbf{k}+\mathbf{q}1s}^\dagger s_1 b_{\mathbf{k}'-\mathbf{q}2s}^\dagger s_2 b_{\mathbf{k}'4s} s_4 b_{\mathbf{k}3s} s_3. \quad (7)$$

In this two-body interaction, effects of higher exciton states have been (partly) included as ‘‘renormalization effects,’’ which have modified (renormalized) the forms and the strengths of $\tilde{\mathcal{H}}_{1s}^{int}$. Therefore, in contrast to the case of $\mathcal{H}_{1s}^{full-int}$, it is reasonable to take $\tilde{\mathcal{H}}_{1s}^{int}$ as an approximation to $\tilde{\mathcal{H}}_{1s}^{full-int}$. In fact, we will show that the replacement $\tilde{\mathcal{H}}_{1s}^{full-int} \rightarrow \tilde{\mathcal{H}}_{1s}^{int}$ is a good approximation, which agrees with the WIBM and experimental results.

In short, one must perform the projection onto the subspace \mathbf{H}_{1s} to get a correct effective interaction of $1s$ excitons. The projection procedure modifies both the form and strengths of the effective interaction. This is essential to justify the WIBM. In what follows, we will derive the effective interaction $\tilde{\mathcal{H}}_{1s}^{int}$ of $1s$ excitons in a QW from the conventional interacting electron-hole Hamiltonian.

C. Model

We consider the conduction and the heavy-hole bands in a GaAs QW, which has a direct band gap. We start from the following conventional form of the electron-hole Hamiltonian \mathcal{H}_{eh} :

$$\begin{aligned} \mathcal{H}_{eh} = & \sum_i \int dx \hat{\psi}_i^\dagger(x) \left(-\frac{\nabla^2}{2m_i} + E_i \right) \hat{\psi}_i(x) \\ & + \sum_{i,i'} \frac{z_i z_{i'}}{2} \int dx dx' \hat{\psi}_i^\dagger(x) \hat{\psi}_{i'}^\dagger(x') \\ & \times V(\mathbf{r}_i - \mathbf{r}_{i'}) \hat{\psi}_{i'}(x') \hat{\psi}_i(x). \end{aligned} \quad (8)$$

$$\begin{pmatrix} |+\rangle \\ |-\rangle \\ |\alpha\rangle \\ |\beta\rangle \end{pmatrix} = \begin{pmatrix} |-1/2, +3/2\rangle \\ |+1/2, -3/2\rangle \\ |+1/2, +3/2\rangle \\ |-1/2, -3/2\rangle \end{pmatrix}. \quad (10)$$

Here, $V(\mathbf{r})$ denotes the Coulomb potential, which behaves in a QW of width L as $V(\mathbf{r}) \approx e^2/\epsilon r$ for $|\mathbf{r}| \geq L$, where ϵ is the static dielectric constant, and $V(\mathbf{r}) \approx \text{constant}$ for $|\mathbf{r}| \leq L$. The calculation is simplified by taking the limit $L \rightarrow 0$ whenever the singularity at $\mathbf{r} = \mathbf{0}$ is irrelevant. In Eq. (8), $\hat{\psi}_{e(h)}(x)$ is the field operator of an electron (hole), $i = \{e, h\}$, $z_{e(h)} = 1 (-1)$, $x \equiv (\mathbf{r}_{e(h)}, J_{e(h)}^z)$, $\int dx \equiv \sum_{J_i^z} \int d^2 r_i$, and similarly for i' and x' . The index $J_{e(h)}^z$ denotes the z component of the total angular momentum, which is a good quantum number, when the z axis is taken in the direction normal to the QW layers. The J_h^z is defined as -1 times J^z of the corresponding valence band electron. In a GaAs QW, $J_h^z = \pm 3/2$ for a heavy hole, and $J_e^z = \pm 1/2$.¹ A photon with $J_{ph}^z = +1 (-1)$ creates an electron-hole pair with $J_e^z = -1/2 (+1/2)$ and $J_h^z = +3/2 (-3/2)$ to conserve the total angular momentum.

D. Strategy

Since all states that are excited by photons are electrically neutral, the discussion is confined to the charge-neutral sector. Then the following exciton operator can be defined:²²

$$\begin{aligned} b_{\mathbf{q}\nu S} \equiv & \sum_{J_e^z, J_h^z} \int d^2 r_e d^2 r_h \frac{1}{\sqrt{\Omega}} \exp\left(i\mathbf{q} \cdot \frac{m_e \mathbf{r}_e + m_h \mathbf{r}_h}{M}\right) \\ & \times \varphi_\nu(\mathbf{r}_e - \mathbf{r}_h) \langle S | J_e^z, J_h^z \rangle \hat{\psi}_e(\mathbf{r}_e, J_e^z) \hat{\psi}_h(\mathbf{r}_h, J_h^z). \end{aligned} \quad (9)$$

Here, the plane wave corresponds to the center-of-mass motion of an electron-hole pair, $\varphi_\nu(\mathbf{r})$ is a wave function for the e - h relative motion, $\langle S | J_e^z, J_h^z \rangle$ the Clebsch-Gordan (CG) coefficient, Ω the QW area, and $M \equiv m_e + m_h$. In the following, the heavy hole condition $0 < m_e \ll m_h$ is assumed, and $\mu(\mu') \equiv m_{e(h)}/M$.

From the explicit calculation of the commutation relation for these operators, they can be treated as bosonic operators when the particle density is very low. This is satisfied under condition (i) in Sec. II A. When $\varphi_\nu(\mathbf{r})$ is the wave function with the quantum number $\nu (= 1s, 2p_+, 2p_-, \dots)$ of a hydrogen atom in two dimensions,¹ the operator $b_{\mathbf{q}\nu S}$ is identified with the bosonic operator for an exciton with the relative motion index ν . Then, the exciton states are labeled by indices \mathbf{q} , ν and S , where \mathbf{q} is the momentum of the center-of-mass motion, ν denotes the set of quantum numbers for the relative motion ν , and S denotes combinations of J_e^z and J_h^z , as shown in Eq. (10). Since S is related to the total angular momentum, it is referred to as ‘‘spin’’ index in the following. For transitions from the heavy-hole band to the electron band in a GaAs QW, possible changes of the total angular momentum are $\Delta J^z = \pm 1$ and ± 2 . We here take the final states corresponding to $\Delta J^z = +1, -1, +2, -2$ as $S = +, -, \alpha, \beta$, respectively. They are related with $|J_e^z, J_h^z\rangle$ as

Since the dipole transition is associated with $\Delta J^z = \pm 1$, $|+\rangle$ and $|-\rangle$ are dipole active, coupling to circularly polarized light with $J_{ph}^z = \pm 1$, whereas $|\alpha\rangle$ and $|\beta\rangle$ are dipole inactive. The general form of Eq. (10) for elliptically polarized light is discussed in Ref. 8.

III. INTERACTION OF EXCITONS BEFORE PROJECTION

In this section, the interaction Hamiltonian corresponding to Eq. (6) is obtained in order to clarify the difference from the two-body interaction obtained through the projection procedure which is discussed in the next section. For this purpose, we calculate the scattering amplitude of excitons without any intermediate states.

A. 1s exciton scattering amplitude

In this subsection, scattering processes which involve only 1s excitons are considered without the projection procedure, resulting to the interaction Hamiltonian of 1s excitons through a straightforward transformation.

Such scattering processes are schematically shown in Fig. 1, where the index ‘‘ex. i ’’ should be read as the set of indices $\{\mathbf{k}_i, \nu_i, S_i\}$. In this subsection, $\nu_i = 1s$ for any i and the index is dropped if any confusion is not expected. These processes are composed of two parts: one is a direct process, Figs. 1(a)–1(d), and the other is a fermionic exchange process, Figs. 1(e)–1(h). The form of the interaction Hamiltonian of 1s excitons is

$$\mathcal{H}_{1s}^{int} = \frac{1}{2\Omega} \sum U(\mathbf{q}; \{S\}) b_{\mathbf{k}+\mathbf{q}, S_1}^\dagger b_{\mathbf{k}'-\mathbf{q}, S_2}^\dagger b_{\mathbf{k}', S_3} b_{\mathbf{k}, S_4}, \quad (11)$$

where the scattering amplitude is written as

$$\begin{aligned} U(\mathbf{q}; \{S\}) = & U_D^o(\mathbf{q}) U_D^s(S_1, S_2; S_3, S_4) \\ & + U_{Ex}^o(\mathbf{q}) U_{Ex}^s(S_1, S_2; S_3, S_4). \end{aligned} \quad (12)$$

The $U_D^o(\mathbf{q}) U_D^s(S_1, S_2; S_3, S_4)$ and $U_{Ex}^o(\mathbf{q}) U_{Ex}^s(S_1, S_2; S_3, S_4)$ are the direct and the exchange scattering amplitudes, respectively. The expressions of each component in Eq. (12) are

$$\begin{aligned} U_D^o(\mathbf{q}) = & \Omega \int d\mathbf{r}_e d\mathbf{r}'_e d\mathbf{r}_h d\mathbf{r}'_h \phi_{\mathbf{k}+\mathbf{q}}(\mathbf{r}_e, \mathbf{r}_h) \phi_{\mathbf{k}'-\mathbf{q}}(\mathbf{r}'_e, \mathbf{r}'_h) \\ & \times \{V(\mathbf{r}_e - \mathbf{r}'_e) + V(\mathbf{r}_h - \mathbf{r}'_h) - V(\mathbf{r}_e - \mathbf{r}'_h) \\ & - V(\mathbf{r}'_e - \mathbf{r}_h)\} \phi_{\mathbf{k}}(\mathbf{r}_e, \mathbf{r}_h) \phi_{\mathbf{k}'}(\mathbf{r}'_e, \mathbf{r}'_h), \end{aligned} \quad (13)$$

$$\begin{aligned} U_D^s(S_1, S_2; S_3, S_4) = & \sum_{J_e^z, J_e'^z, J_h^z, J_h'^z} \langle S_1 | J_e^z, J_h^z \rangle \langle S_2 | J_e'^z, J_h'^z \rangle \\ & \times \langle S_3 | J_e^z, J_h^z \rangle \langle S_4 | J_e'^z, J_h'^z \rangle, \end{aligned} \quad (14)$$

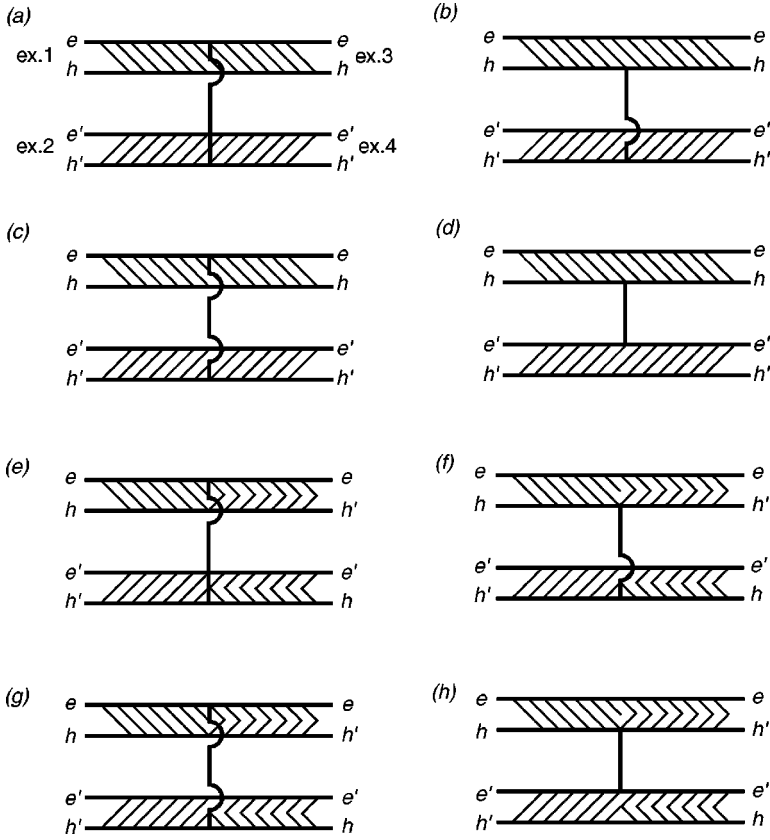


FIG. 1. Diagrammatically expression of direct (a)–(d) and fermionic exchange interaction (e)–(h) of 1s excitons. The horizontal lines represent an electron and a hole and the vertical line Coulomb interaction of two particles connected by the line.

$$U_{Ex}^o(\mathbf{q}) = -\Omega \int d\mathbf{r}_e d\mathbf{r}'_e d\mathbf{r}_h d\mathbf{r}'_h \phi_{\mathbf{k}+\mathbf{q}}(\mathbf{r}_e, \mathbf{r}_h) \phi_{\mathbf{k}'-\mathbf{q}}(\mathbf{r}'_e, \mathbf{r}'_h) \\ \times \{V(\mathbf{r}_e - \mathbf{r}'_e) + V(\mathbf{r}_h - \mathbf{r}'_h) - V(\mathbf{r}_e - \mathbf{r}'_h) \\ - V(\mathbf{r}'_e - \mathbf{r}_h)\} \phi_{\mathbf{k}}(\mathbf{r}'_e, \mathbf{r}_h) \phi_{\mathbf{k}'}(\mathbf{r}_e, \mathbf{r}'_h), \quad (15)$$

and

$$U_{Ex}^s(S_1, S_2; S_3, S_4) = \sum_{J_e^z, J_e^{z'}, J_h^z, J_h^{z'}} \langle S_1 | J_e^z, J_h^z \rangle \langle S_2 | J_e^{z'}, J_h^{z'} \rangle \\ \times \langle S_3 | J_e^{z'}, J_h^z \rangle \langle S_4 | J_e^z, J_h^{z'} \rangle, \quad (16)$$

where $\phi_{\mathbf{q}}(\mathbf{r}_e, \mathbf{r}_h)$ is the product of the wave functions of the center of mass and the relative motion of excitons,

$$\phi_{\mathbf{q}}(\mathbf{r}_e, \mathbf{r}_h) \equiv \frac{1}{\sqrt{\Omega}} e^{i\mathbf{q} \cdot (\mu\mathbf{r}_e + \mu'\mathbf{r}_h)} \varphi(\mathbf{r}_e - \mathbf{r}_h). \quad (17)$$

The wave function of the relative motion φ is the 1s wave function of a hydrogen atom in two dimensions:

$$\varphi(\mathbf{r}) = \frac{2\sqrt{2}}{\sqrt{\pi}a_0} e^{-2|\mathbf{r}|/a_0}, \quad (18)$$

where a_0 is the exciton Bohr radius. In the following, $U_{D(Ex)}^o$ and $U_{D(Ex)}^s$ are referred to as ‘‘orbital’’ and ‘‘spin’’ parts, respectively.

First, the orbital parts are calculated. Fourier representations of orbital parts in direct and exchange scattering amplitude are

$$U_D(\mathbf{q}) = \sum_{\mathbf{p}_1, \mathbf{p}_2} \tilde{V}(\mathbf{q}) [|\tilde{\varphi}(\mathbf{p}_1)|^2 |\tilde{\varphi}(\mathbf{p}_2)|^2 + \tilde{\varphi}(\mathbf{p}_1) \tilde{\varphi}^* \\ \times (\mathbf{p}_1 + \mathbf{q}) \tilde{\varphi}(\mathbf{p}_2) \tilde{\varphi}^*(\mathbf{p}_2 - \mathbf{q}) - 2\tilde{\varphi}(\mathbf{p}_1) |\tilde{\varphi}(\mathbf{p}_2)|^2 \tilde{\varphi}^* \\ \times (\mathbf{p}_1 + \mathbf{q})], \quad (19)$$

$$U_{Ex}(\mathbf{q}) = - \sum_{\mathbf{p}_1, \mathbf{p}_2} \tilde{V}(\mathbf{q} + \mathbf{p}_1 - \mathbf{p}_2) [|\tilde{\varphi}(\mathbf{p}_1)|^2 |\tilde{\varphi}(\mathbf{p}_2)|^2 \\ + \tilde{\varphi}(\mathbf{p}_1) \tilde{\varphi}^*(\mathbf{p}_1 + \mathbf{q}) \tilde{\varphi}(\mathbf{p}_2) \tilde{\varphi}^*(\mathbf{p}_2 - \mathbf{q}) \\ - 2\tilde{\varphi}(\mathbf{p}_1) |\tilde{\varphi}(\mathbf{p}_2)|^2 \tilde{\varphi}^*(\mathbf{p}_2 - \mathbf{q})]. \quad (20)$$

Here, the notations with tilde are defined as

$$\tilde{V}(\mathbf{p}) = \int d^2\mathbf{r} e^{i\mathbf{p} \cdot \mathbf{r}} \frac{1}{|\mathbf{r}|} = \frac{2\pi}{|\mathbf{p}|}, \quad (21)$$

$$\tilde{\varphi}(\mathbf{p}) = \frac{1}{\sqrt{\Omega}} \int d^2\mathbf{r} e^{i\mathbf{p} \cdot \mathbf{r}} \varphi(\mathbf{r}) \\ = \frac{1}{\sqrt{\Omega}} \frac{\sqrt{2\pi}a_0}{[1 + (|\mathbf{p}|a_0/2)^2]^{3/2}}. \quad (22)$$

Since the transferred momentum in the exciton scattering processes is fairly small, which is of the order of the photon momentum, the direct and the double fermionic exchange interactions are negligible⁸ and the momentum dependence of the exchange interaction can be omitted. This allows the approximation $U_D^o(\mathbf{q}) \approx U_D^o(\mathbf{q}=0) \equiv U_D^o$ and $U_{Ex}^o(\mathbf{q}) \approx U_{Ex}^o(\mathbf{q}=0) \equiv U_{Ex}^o$. The \mathbf{q} dependence of the interaction strength is discussed in Ref. 8. Under these conditions, U_D^o and U_{Ex}^o can be directly obtained from Eqs. (19) and (20) for

$\mathbf{q} \rightarrow 0$:

$$U_D^o = 0, \quad (23)$$

$$U_{Ex}^o = 2 \sum_{\mathbf{p}, \mathbf{p}'} \tilde{V}(\mathbf{p} - \mathbf{p}') [|\tilde{\varphi}(\mathbf{p})|^2 \tilde{\varphi}(\mathbf{p})^* \tilde{\varphi}(\mathbf{p}') - |\tilde{\varphi}(\mathbf{p}')|^2 \tilde{\varphi}(\mathbf{p}')^* \tilde{\varphi}(\mathbf{p})]. \quad (24)$$

Equation (23) reflects the charge neutrality of the system.

B. Scattering amplitude of higher exciton states than $1s$

The calculation of the exciton scattering amplitude which includes excitons with $\nu > 1s$ is quite similar to the previous calculation involving excitons with only $\nu = 1s$. For such scattering processes that satisfy the conservation laws, the amplitude for scattering from initial state (ν_1, ν_2) to the final state (ν_3, ν_4) is obtained as

$$U_{D\{\nu\}}^o = 0, \quad (25)$$

$$U_{Ex\{\nu\}}^o = 2 \sum_{\mathbf{p}, \mathbf{p}'} \tilde{V}(\mathbf{p} - \mathbf{p}') [\tilde{\varphi}_{\nu_1}(\mathbf{p})^* \tilde{\varphi}_{\nu_2}(\mathbf{p}) \tilde{\varphi}_{\nu_3}(\mathbf{p}') \tilde{\varphi}_{\nu_4}(\mathbf{p}') - \tilde{\varphi}_{\nu_1}(\mathbf{p}')^* \tilde{\varphi}_{\nu_2}(\mathbf{p}') \tilde{\varphi}_{\nu_3}(\mathbf{p}) \tilde{\varphi}_{\nu_4}(\mathbf{p})], \quad (26)$$

under the same assumptions as in the previous calculation. Here, $\tilde{\varphi}_\nu(\mathbf{p})$ is the Fourier transform of the corresponding wave function in the real space:

$$\tilde{\varphi}_\nu(\mathbf{p}) = \frac{1}{\sqrt{\Omega}} \int d^2\mathbf{r} e^{i\mathbf{p} \cdot \mathbf{r}} \varphi_\nu(\mathbf{r}). \quad (27)$$

C. Interaction Hamiltonian of excitons before projection

From the results in previous two subsections, the bosonic Hamiltonian of excitons $\mathcal{H} \equiv \mathcal{H}^0 + \mathcal{H}^{int}$ is obtained when it is assumed that the exciton density is low, where \mathcal{H}^0 is the free part of excitons. The general form of \mathcal{H}^{int} through a straightforward transformation is

$$\mathcal{H}^{int} = \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}\{\nu\}\{S\}} \frac{U_{Ex\{\nu\}}^o U_{Ex\{S\}}^s}{2\Omega} b_{\mathbf{k}+\mathbf{q}\nu_1 S_1}^\dagger \times b_{\mathbf{k}'-\mathbf{q}\nu_2 S_2}^\dagger b_{\mathbf{k}'\nu_4 S_4} b_{\mathbf{k}\nu_3 S_3}. \quad (28)$$

Using this formula, we express \mathcal{H} as

$$\mathcal{H} = \mathcal{H}^0 + \mathcal{H}_{1s}^\pm + \mathcal{H}'_{1s} + \mathcal{H}_{others}, \quad (29)$$

where $\mathcal{H}_{1s}^\pm + \mathcal{H}'_{1s} \equiv \mathcal{H}_{1s}^{int}$, [see Eq. (6)], includes the $\nu = 1s$ operators only, and \mathcal{H}_{others} denotes such remaining terms as the interaction between $1s$ and $2p$ excitons, the interaction between $2p$ and $2p$ excitons and other similar interactions. The \mathcal{H}_{1s}^\pm consists only of the operators with $S = \pm$, corresponding to dipole active excitons, whereas \mathcal{H}'_{1s} consists of terms of $S = \alpha$ and β operators, including cross terms with $S = \pm$ operators.

From the explicit calculation of the spin part of the exchange scattering amplitude, $U_{Ex\{S\}}^s$, for all combinations of $\{S_1, S_2, S_3, S_4\} = \{+, -, \alpha, \beta\}$, \mathcal{H}_{1s}^\pm and \mathcal{H}'_{1s} are obtained as

$$\mathcal{H}_{1s}^\pm = \frac{U}{2\Omega} \sum_{S=\pm} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} b_{\mathbf{k}+\mathbf{q}S}^\dagger b_{\mathbf{k}'-\mathbf{q}S}^\dagger b_{\mathbf{k}'S} b_{\mathbf{k}S}, \quad (30)$$

$$\mathcal{H}'_{1s} = \frac{U}{\Omega} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} \left[\sum_{S=\alpha, \beta} \left(\frac{1}{2} b_{\mathbf{k}+\mathbf{q}S}^\dagger b_{\mathbf{k}'-\mathbf{q}S}^\dagger b_{\mathbf{k}'S} b_{\mathbf{k}S} + b_{\mathbf{k}+\mathbf{q}S}^\dagger b_{\mathbf{k}'-\mathbf{q}S}^\dagger b_{\mathbf{k}'S} b_{\mathbf{k}S} + b_{\mathbf{k}+\mathbf{q}S}^\dagger b_{\mathbf{k}'-\mathbf{q}S}^\dagger b_{\mathbf{k}'S} b_{\mathbf{k}S} \right) + (b_{\mathbf{k}+\mathbf{q}S}^\dagger + b_{\mathbf{k}'-\mathbf{q}S}^\dagger - b_{\mathbf{k}'S} - b_{\mathbf{k}S}) \right], \quad (31)$$

where $b_{\mathbf{q}S} \equiv b_{\mathbf{q}1sS}$, and the effective interaction strength $U \equiv U_{Ex\{\nu\}=\{1s\}}^o$.

For $d=2$, and in the limit of $L \rightarrow 0$, Eq. (24) is evaluated as

$$U = 2\pi a_0^2 \left(1 - \frac{315\pi^2}{4096} \right) E_{ex}^b \approx 1.52 a_0^2 E_{ex}^{2D}, \quad (32)$$

where a_0 is the exciton Bohr radius and E_{ex}^{2D} is the binding energy of the two-dimensional exciton.¹⁷ Note that the estimation of Eq. (24) in the three-dimensional case, using three-dimensional $1s$ wave function, gives the hard-core scattering strength obtained in Ref. 3. This fact shows that the theory presented in this section corresponds to the result obtained by the Usui transformation.²³

The point which should be emphasized in the dipole active part is that there are no interaction terms between the exciton with $S = +$ and the exciton with $S = -$ (opposite spin exciton interaction), which are represented in the form of $\propto b_+^\dagger b_-^\dagger b_- b_+$, and that there are only the interaction terms between the excitons with $S = + (-)$ (equal spin exciton interaction). This is due to vanishing of $U_{Ex\{S\}}^s$ for the corresponding combination of $\{S\}$. The absence of the interaction between $S = +$ and $S = -$ excitons agrees with the result of Ref. 8, in which the case of elliptic polarization with an elliptic polarization angle α is discussed. See the fifth row of Table I in Ref. 8. The case of $\alpha = 0$ in Ref. 8 corresponds to the case of circular polarization discussed in this section. The existence of the interaction in the case of $\alpha \neq 0$ in Ref. 8 is due to the finite inner product of $S = +$ and $-$, and that of $S = -$ and $-$.

Here two ways of thinking are possible. One is that the excitons with $S = +$ and the excitons with $S = -$ will not interact with each other. The other is that the absence of such an interaction term is due to the fact that so far we calculated to lowest order only. A large number of experiments show that excitons with the opposite spins do interact and that such an interaction is crucial, for instance, for four-wave mixing in the time domain. So it is not appropriate to interpret \mathcal{H}_{1s}^\pm as the effective Hamiltonian for dipole active $1s$ excitons. The straightforward transformation presented in this section is not appropriate for deriving the effective Hamiltonian of $1s$ excitons and the projection procedure discussed in the next section is indispensable.

IV. PROJECTION PROCEDURE

In this section, the interaction Hamiltonian of $1s$ excitons, which corresponds to Eq. (7), is obtained through the projec-

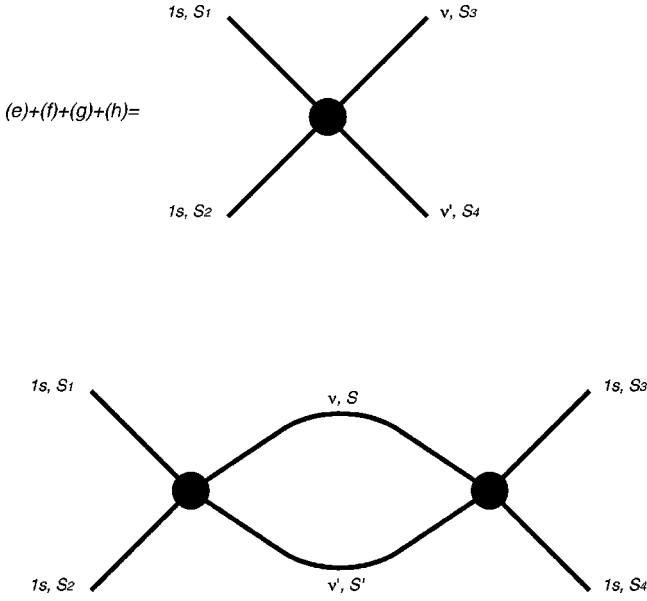


FIG. 2. Successive exchange scattering processes of excitons taken into the projection procedure. The bullet shows the sum of the single exchange scattering processes between the $1s$ and ν excitons.

tion procedure.¹⁸ This procedure yields the correct two-body interactions of $1s$ excitons in the subspace of $1s$ excitons. In particular, the interaction term of excitons with the opposite spins is obtained through the projection, whereas such an interaction was not obtained in the previous section. In other words, such an interaction is obtained when going beyond the HF approximation of $1s$ excitons alone. Moreover, the projection yields a large renormalization of the interaction strength of excitons with the same spins. Among works which have the similar motivation of going beyond HF, modification of the exciton binding energy beyond HF is discussed by considering screening effects in Ref. 11.

In the previous section, the effects of higher exciton states have been completely excluded in deriving the interaction Hamiltonian of $1s$ excitons, Eqs. (30) and (31). In this section, the theory is discussed in the subspace spanned by $1s$ excitons, where the effects of higher exciton states are renormalized. Since this scenario is quite similar to the derivation of an effective Hamiltonian from the Hubbard model with large on-site Coulomb repulsion through a projection procedure,²⁴ the method used here is referred to as projection. In other words, this is nothing but the real part of the second-order vertex correction in the field theory, yielding the energy shift.

The higher exciton states are taken as the intermediate states in the scattering processes of $1s$ excitons. Schematically, the scattering processes of excitons shown in Fig. 2 are considered. Since our purpose is to obtain the effective interaction of $1s$ excitons, the relative motion indices of four external lines must be $\nu = 1s$. As for the intermediate states, excitons which are connected with $1s$ excitons by dipole transitions are considered. Then, the lowest energy excitons for the intermediate states are $2p_{\pm}$ excitons. Note that in a two-dimensional system, the p states are doubly degenerated.

The Hamiltonian for the $1s - \nu$ interaction processes is

$$\begin{aligned} \mathcal{H}_{\text{out}} &\equiv \sum_{\nu} \mathcal{H}_{1s-\nu} \\ &= \sum_{\nu} \sum_{\{S\}} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} g_{\nu}(\mathbf{q}) b_{1s, \mathbf{k}+\mathbf{q}, S_1}^{\dagger} b_{1s, \mathbf{k}'-\mathbf{q}, S_2}^{\dagger} b_{\nu, \mathbf{k}', S_4} b_{\nu, \mathbf{k}, S_3} \\ &\quad + \text{H.c.} \end{aligned} \quad (33)$$

Let \mathcal{H}_{org} be the Hamiltonian which includes the kinetic and the interaction terms of $1s$ and ν excitons, and with eigenstates and eigenenergies defined by

$$\mathcal{H}_{\text{org}}|\Phi\rangle = E_{\Phi}|\Phi\rangle. \quad (34)$$

Consider the following Schrödinger equation:

$$(\mathcal{H}_{\text{org}} + \mathcal{H}_{\text{out}})|\Psi\rangle = E|\Psi\rangle. \quad (35)$$

Symbolically, the solution of this equation is shown as

$$\begin{aligned} |\Psi\rangle &= \frac{\mathcal{H}_{\text{out}}}{E - \mathcal{H}_{\text{org}}}|\Psi\rangle = \sum_{\Phi} |\Phi\rangle \frac{\langle\Phi|\mathcal{H}_{\text{out}}|\Psi\rangle}{E - E_{\Phi}} \\ &\quad + \frac{\mathcal{P}}{E - \mathcal{H}_{\text{org}}}\mathcal{H}_{\text{out}}|\Psi\rangle, \end{aligned} \quad (36)$$

where the projection operator \mathcal{P} is defined as

$$\mathcal{P} \equiv 1 - \sum_{\Phi} |\Phi\rangle\langle\Phi|. \quad (37)$$

The $|\Psi\rangle$ is rewritten as

$$|\Psi\rangle = \sum_{\Phi} a_{\Phi}|\Psi_{\Phi}\rangle, \quad (38)$$

where

$$a_{\Phi} \equiv \frac{\langle\Phi|\mathcal{H}_{\text{out}}|\Psi\rangle}{E - E_{\Phi}}, \quad (39)$$

and

$$|\Psi_{\Phi}\rangle \equiv |\Phi\rangle + \frac{\mathcal{P}}{E - \mathcal{H}_{\text{org}}}\mathcal{H}_{\text{out}}|\Psi_{\Phi}\rangle. \quad (40)$$

In lowest order, this wave function can be approximated as

$$|\Psi_{\Phi}\rangle \approx |\Phi\rangle + \frac{1}{E - \mathcal{H}_{\text{org}}}\mathcal{H}_{\text{out}}|\Phi\rangle. \quad (41)$$

From them, the Schrödinger equation for the projected out Hamiltonian, that is, the effective Hamiltonian is obtained as

$$(E - E_{\Phi})a_{\Phi} = \sum_{\Phi'} a_{\Phi'} \langle\Phi|\mathcal{H}_{\text{out}} \frac{1}{E - \mathcal{H}_{\text{org}}} \mathcal{H}_{\text{out}}|\Phi'\rangle. \quad (42)$$

Here, two successive exchange scattering processes are considered, whose individual scattering amplitudes are obtained from Eq. (26). The resolvent $1/(E - \mathcal{H}_{\text{org}})$ is replaced with the difference of the kinetic energy between two ν excitons and two $1s$ excitons, because the interaction energy of each exciton can be assumed to be small as compared to their kinetic energy. The orbital part of the renormalized scattering amplitude of $1s$ excitons, $U_{Ex}^{o'}$, which is calculated by second-order perturbative calculation, is

$$\begin{aligned}
U_{Ex}^{o'} &= \frac{1}{\Omega} \sum_{\mathbf{k}, \nu \neq 1s} \frac{|g_\nu(\mathbf{K})|^2}{2(E_\nu + \mathbf{K}^2/2M) - 2E_{1s}} \\
&= \frac{1}{\Omega} \sum_{\mathbf{k}, \nu \neq 1s} \frac{1}{2(E_\nu + \mathbf{K}^2/2M) - 2E_{1s}} \left| \sum_{\mathbf{p}, \mathbf{p}'} \tilde{V}(\mathbf{p} - \mathbf{p}' + \mathbf{K}) \right. \\
&\quad \times [-\tilde{\varphi}_{1s}^*(\mathbf{p}) \tilde{\varphi}_{1s}^*(\mathbf{p}') \tilde{\varphi}_\nu(\mathbf{p}) \tilde{\varphi}_\nu(\mathbf{p}') + 2\tilde{\varphi}_{1s}^*(\mathbf{p}) \tilde{\varphi}_{1s}^* \\
&\quad \times (\mathbf{p} - \mathbf{K}) \tilde{\varphi}_\nu(\mathbf{p}) \tilde{\varphi}_\nu(\mathbf{p}') - \tilde{\varphi}_{1s}^*(\mathbf{p}) \tilde{\varphi}_{1s}^*(\mathbf{p}') \tilde{\varphi}_\nu(\mathbf{p} - \mathbf{K}) \\
&\quad \left. \times \tilde{\varphi}_\nu(\mathbf{p}' + \mathbf{K}) \right|^2. \quad (43)
\end{aligned}$$

Note that the long-range part of Eq. (43) compared with the exciton Bohr radius describes the van der Waals force.

As for the spin part, $U_{Ex}^{s'}$, it is obtained by the simple product of two successive spin weights U_{Ex}^s ,

$$\begin{aligned}
U_{Ex}^{s'}(S_1, S_2; S_3, S_4) \\
= \sum_{S, S'} U_{Ex}^s(S_1, S_2; S, S') U_{Ex}^s(S, S'; S_3, S_4). \quad (44)
\end{aligned}$$

From the explicit representation of the renormalized scattering amplitude $U' \equiv U_{Ex}^{o'} U_{Ex}^{s'}$, the renormalized Hamiltonian of 1s excitons is written in the form

$$\tilde{\mathcal{H}}_{1s} = \tilde{\mathcal{H}}_{1s}^0 + \tilde{\mathcal{H}}_{1s}^\pm + \tilde{\mathcal{H}}_{1s}', \quad (45)$$

where $\tilde{\mathcal{H}}_{1s}^0$ is the Hamiltonian of free 1s excitons, and $\tilde{\mathcal{H}}_{1s}^\pm + \tilde{\mathcal{H}}_{1s}' \equiv \tilde{\mathcal{H}}_{1s}^{\text{int}}$, [see Eq. (7)], includes the $\nu=1s$ operators only. The $\tilde{\mathcal{H}}_{1s}^\pm$ consists only of operators with $S=\pm$, whereas $\tilde{\mathcal{H}}_{1s}'$ consists of terms of $S=\alpha$ and β operators, including cross terms with $S=\pm$ operators.

The interaction Hamiltonian which has only dipole active 1s excitons $\tilde{\mathcal{H}}_{1s}^\pm$, is

$$\begin{aligned}
\tilde{\mathcal{H}}_{1s}^\pm &= \frac{U-U'}{2\Omega} \sum_{S=\pm} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} b_{\mathbf{k}+\mathbf{q}}^\dagger b_{\mathbf{k}'-\mathbf{q}}^\dagger b_{\mathbf{k}'} b_{\mathbf{k}} \\
&\quad - \frac{U'}{\Omega} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} b_{\mathbf{k}+\mathbf{q}}^\dagger + b_{\mathbf{k}'-\mathbf{q}}^\dagger - b_{\mathbf{k}'} - b_{\mathbf{k}}, \quad (46)
\end{aligned}$$

where $U' = U_{Ex}^{o'}$ is a positive constant which arises from the renormalization of higher exciton states ($\nu = 2p_+, 2p_-, \dots$). Comparing the right-hand side of Eq. (46) with that of Eq. (30), we see that the coefficient of the first term is renormalized as $U \rightarrow U - U'$, and that the second term is generated through the projection procedure, which leads to the opposite spin exciton interaction. This is due to the fact that though the spin weight $U_{Ex}^s(+, -, +, -)$ vanishes, the spin weight $U_{Ex}^{s'}(+, -, +, -) \neq 0$ because the intermediate states can make use of the state with the $S = \{\alpha, \beta\}$, and $U_{Ex}^s(+, -, \alpha, \beta) \neq 0$. This shows that the renormalization of higher exciton states results in a renormalized Hamiltonian $\tilde{\mathcal{H}}_{1s}^\pm$, which differs, both quantitatively

and qualitatively, from the Hamiltonian \mathcal{H}_{1s}^\pm , where higher exciton states than 1s exciton are completely ignored.

The renormalized interaction Hamiltonian which includes dipole inactive excitons is obtained similarly as

$$\begin{aligned}
\tilde{\mathcal{H}}_{1s}' &= \frac{U-U'}{\Omega} \sum_{S=\alpha, \beta} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} \left[\frac{1}{2} b_{\mathbf{k}+\mathbf{q}}^\dagger b_{\mathbf{k}'-\mathbf{q}}^\dagger b_{\mathbf{k}'} b_{\mathbf{k}} \right. \\
&\quad \left. + b_{\mathbf{k}+\mathbf{q}}^\dagger + b_{\mathbf{k}'-\mathbf{q}}^\dagger b_{\mathbf{k}'} b_{\mathbf{k}} + b_{\mathbf{k}+\mathbf{q}}^\dagger - b_{\mathbf{k}'-\mathbf{q}}^\dagger b_{\mathbf{k}'} b_{\mathbf{k}} \right] \\
&\quad + \frac{U}{\Omega} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} (b_{\mathbf{k}+\mathbf{q}}^\dagger + b_{\mathbf{k}'-\mathbf{q}}^\dagger - b_{\mathbf{k}'} b_{\mathbf{k}} + \text{H.c.}) \\
&\quad - \frac{U'}{\Omega} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} b_{\mathbf{k}+\mathbf{q}}^\dagger b_{\mathbf{k}'-\mathbf{q}}^\dagger b_{\mathbf{k}'} b_{\mathbf{k}}. \quad (47)
\end{aligned}$$

Since $\tilde{\mathcal{H}}_{1s}'$ includes dipole inactive 1s excitons with $S=\alpha, \beta$, it does not contribute to the optical response in its lowest order.^{25,26}

V. DISCUSSIONS AND REMARKS

A. Microscopic foundation of the WIBM

Kuwata-Gonokami *et al.*¹² introduced a phenomenological Hamiltonian, WIBM, which yields good agreement with the experimental four-wave mixing data. The WIBM has two kinds of interaction terms of excitons: One is a repulsive term R for the same spin excitons. The other is an attractive interaction W for the opposite spin excitons. As our first important result, we note that the phenomenological Hamiltonian¹² has the same form as our $\tilde{\mathcal{H}}_{1s}^\pm$, the dipole active part of $\tilde{\mathcal{H}}_{1s}$. This is quite reasonable because the other part $\tilde{\mathcal{H}}_{1s}'$, which is dipole inactive, should be invisible in low-order optical experiments.²⁶ We can therefore identify the parameters R and W of the phenomenological Hamiltonian¹² as

$$R = \frac{U-U'}{2\Omega}, \quad (48)$$

$$W = -\frac{U'}{\Omega}. \quad (49)$$

The value of U' , as given by Eq. (43), depends on the material parameters such as M and ϵ , and hence is different for different materials. It also depends on the QW parameter L . Moreover, when imperfections in the QW are non-negligible, the expressions of U' should be modified accordingly. Therefore even for the same material the values of R and W could vary from sample to sample, which seems to be consistent with recent experimental results.²⁷ Note, however, that the *existence* of both interaction terms of excitons $\tilde{\mathcal{H}}_{1s}^\pm$ is independent of such details.

Since the accurate evaluation of Eq. (43) is rather tedious, we here estimate the typical value of U' as follows. The \mathbf{K} summation in Eq. (43) is cut off for $K \geq C_L/L$ (through the cut off of \tilde{V}) and/or for $K \geq C_{a_0}/a_0$ (through $\tilde{\varphi}_\nu$), where C_L and C_{a_0} are cutoff parameters of the order of unity. For the

case of the QW sample of Ref. 12, $L \approx a_0$, hence we may cutoff the \mathbf{K} summation for $K \geq C/a_0$, where C is of the order of unity. For the ν summation, we may consider $\nu = 2p_{\pm}$ states only, because higher exciton states give much smaller overlap integrals. The summations for \mathbf{p} and \mathbf{p}' are replaced with integrals and U' is roughly evaluated, as $U' \approx 16.5a_0^2 C^2 E_{ex}^{2D}$.

Reference 12 reported the ratio $R:W$ as 1:−15. From Eqs. (48) and (49), we find that this ratio is reproduced by the present theory when the cutoff parameter $C \sim 0.3$, which is consistent with the requirement that C is of the order of unity. Considering that the values of R and W vary slightly from sample to sample,²⁷ the agreement seems satisfactory. Note that such a small value of R reported in Ref. 12 is due to the renormalization of $U \rightarrow U - U'$. Once the agreement of $\tilde{\mathcal{H}}_{1s}^{\pm}$ with the phenomenological Hamiltonian is established, the agreement with the experiment follows, as presented in Refs. 12, 13, and 28. That is, lowest-order perturbative calculations for the polariton-polariton scattering amplitudes agree with the experiment.^{12,25} From the above discussion, we conclude that the present theory yields the microscopic background of the two exciton interaction terms in the WIBM.

B. Correct effective Hamiltonian of 1s excitons

The correct form of the effective Hamiltonian of 1s excitons is the renormalized one instead of the Hamiltonian which is obtained without the projection procedure, i.e.,

$$\mathcal{H}_{1s}^{eff} = \tilde{\mathcal{H}}_{1s}^{\pm} + \tilde{\mathcal{H}}'_{1s}. \quad (50)$$

In fact, the opposite spin exciton interaction in $\tilde{\mathcal{H}}_{1s}^{\pm}$, which is absent in \mathcal{H}_{1s}^{\pm} , has been clearly observed experimentally in Refs. 6 and 12. We have used a low-order perturbation theory to derive $\tilde{\mathcal{H}}_{1s}$, where successive scatterings in the intermediate processes are not considered. However, this does not imply a total neglect of multiscattering processes, because we have calculated a Hamiltonian rather than observables. In fact, a systematic summing up of higher-order scattering processes is incorporated in our theory if one calculates higher-order perturbation terms from $\tilde{\mathcal{H}}_{1s}$ by writing down the Bethe-Salpeter equation.

Note that $\tilde{\mathcal{H}}_{1s}$ is not positive definite to the fourth-order in the exciton operators. The stability of the system should be preserved by higher-order terms. In general situations, properties of a system described by such a Hamiltonian should not be analyzed by a perturbation theory based on the vacuum of the free part. Nevertheless, we can use such a perturbation theory in our case, because our exciton theory has the built-in constraint that the ground state is the state with no excitons, i.e., the vacuum of $\tilde{\mathcal{H}}_{1s}^0$. The effective Hamiltonian $\tilde{\mathcal{H}}_{1s}$ together with this constraint constitutes a consistent theory, which justifies the low-order perturbation theory based on the given vacuum, if the optical excitation is sufficiently weak.

C. Comparison with existing bosonic theories

As shown in Sec. I, existing bosonic theories treated spinless excitons,³ or spinfull excitons without the renormaliza-

tion procedure.^{8,9} In the spinless theory,³ the result is that excitons interact repulsively. The attractive interaction of excitons with opposite spins is suggested from a variational calculation for the biexciton state. However, the existence of biexcitons is not directly related to the attractive interaction term of excitons with the opposite spins (the W term in WIBM), as will be discussed in Sec. V E. In the spinfull theories,^{8,9} on the other hand, an interaction Hamiltonian of 1s excitons with opposite spins was not derived. Since the theories essentially end up with the HF approximation of 1s excitons, the excitons with opposite spins are completely decoupled,¹⁰ as confirmed in many papers²⁹ (see Sec. III C). Such theories will not yield the interaction of excitons with opposite spins, as shown in Sec. III C.

The projection method discussed in this paper proves that one can obtain the explicit form of the interaction Hamiltonian of excitons with opposite spins beyond HF approximation of only 1s excitons. There are several papers where such an approach is motivated,¹¹ however, only the modification of the exciton binding energy is discussed. Using our interaction Hamiltonian, one can reproduce these results.²⁹

D. Validity of the theory

In deriving the effective Hamiltonian of 1s excitons, we have assumed three conditions listed in Sec. II A: (i) the excitation is weak so that the mean distance of photocreated excitons is much larger than the Bohr radius of the 1s exciton, (ii) all the photon energies (pump, probe, and signal) are close to the energy of the 1s exciton, and (iii) the linewidth of 1s exciton is smaller than the detuning energies.

The condition (i) allows us to use the boson representation of excitons. This condition may be confirmed experimentally by the fact that the signal intensity is precisely proportional to the square of the pump intensity, i.e., the optical response is well described by $\chi^{(3)}$. This suggests that the two-body scattering processes would be dominant. When l_{ex} is increased to $l_{ex} \sim a_0$, the deviation from the boson statistics of the operators $b_{\mathbf{q}\nu s}$ and $b_{\mathbf{q}\nu s}^{\dagger}$ will become non-negligible, which invalidates the boson representation.³⁰ The condition (ii) means that the 1s excitons give dominant contributions. This allows us to project out all states higher than 1s.

The final condition (iii) allows us to neglect relaxation process of 1s excitons. Namely, the equation of motion of the reduced density operator $\tilde{\rho}_{1s}$ in the 1s exciton subspace generally takes the following form:

$$\frac{\partial \tilde{\rho}_{1s}}{\partial t} = \frac{1}{i} [\tilde{\mathcal{H}}_{1s}, \tilde{\rho}_{1s}] + \tilde{\Gamma} \tilde{\rho}_{1s}. \quad (51)$$

Here, $\tilde{\mathcal{H}}_{1s}$ describes the unitary evolution of $\tilde{\rho}_{1s}$, and $\tilde{\Gamma}$ is the relaxation operator, which is described by the imaginary part of a vertex correction. Since the relaxation processes are less crucial under the condition (iii), one may disregard $\tilde{\Gamma}$, i.e., one may consider only the real part of the vertex correction. One might think that under these conditions the optical signals would not be strong enough to obtain experimental data. However, Kuwata-Gonokami *et al.*¹² proposed a well-prepared method to overcome this difficulty: they confined a GaAs QW in a high- Q microcavity. This results in a large

splitting of excitonic polariton spectrum. Some nonlinear optical signals are strongest at the polariton energies (upper and lower ones) because of the polariton resonance. On the other hand, the dissipation (which creates real excitons in the QW) is weak at these energies. [This may be understood by considering possible final states: When the initial state is a single photon state (coming from an external light source) that has the energy of the lower-polariton peak, the final state cannot be an exciton state (without a photon) because the energy is short to create a real exciton.] Therefore one can obtain strong signals without significant dissipation when the photon energies are close to the polariton energies in a high- Q optical cavity. Using this idea, Kuwata-Gonokami *et al.*¹² measured nonlinear optical responses under the conditions (i)–(iii), and showed the validity of the WIBM. Our theory is valid in such a case.

On the other hand, Shirane *et al.* recently demonstrated that the dissipation becomes important when the Q value of the optical cavity is lowered.¹³ In this case the relaxation processes of $1s$ exciton becomes important, which means that $\tilde{\Gamma}$ must be fully considered. These are closely related to the excitation induced dephasing (EID) discussed below. The relaxation effect is one of the future problems.

Another example to which the present theory is applicable may be the optical Stark effect in high quality samples at low temperature. In the past experiments of the optical Stark effect, one had to take the detuning rather large [hence condition (ii) is not satisfied] in order to avoid the absorption tail. The absorption tail would be reduced for samples with better quality and at lower temperatures.

E. Biexciton formation and attractive interaction of excitons

It has been conjectured¹² that a ‘‘biexciton effect’’ would be the origin of the ‘‘ W term,’’ i.e., the opposite spin exciton interaction. However, this argument is misleading. The biexciton state is analogous to a hydrogen molecule and is formed essentially through the mixing of two $1s$ states having different centers. The mixing of two hydrogen atoms yields the bonding and antibonding states, which are represented as $(1/\sqrt{2})(c_{1\uparrow}^\dagger c_{2\downarrow}^\dagger \pm c_{1\downarrow}^\dagger c_{2\uparrow}^\dagger)h_{1\sigma}^\dagger h_{2\sigma'}^\dagger |0\rangle$. Here, $c_{1(2)}^\dagger$ creates an electron in the $1s$ state located at nucleus 1(2), and $h_{1(2)}^\dagger$ creates the nucleus. The lower energy state is the bonding, that is, molecular state. In the case of excitons with $J_h^z = \pm 1/2$ and $J_h^z = \pm 3/2$, the corresponding states are $(1/\sqrt{2}) \times [b_+^\dagger b_-^\dagger \pm b_\alpha^\dagger b_\beta^\dagger] |0\rangle$, where the \mathbf{k} dependence is omitted in order to focus on the S dependence. The bonding state (– sign for a positive coupling constant) has a lower energy and is called biexciton. This energy splitting between the bonding and antibonding states is induced by an interaction of the form of $b_+^\dagger b_-^\dagger b_\alpha b_\beta + \text{H.c.}$, which is included in $\tilde{\mathcal{H}}'_{1s}$ [or, before the renormalization, in \mathcal{H}'_{1s} of Eq. (31)]. From a more general point of view, a typical bound state in an interacting boson model is the eigenstate

$$|\text{bound}\rangle = [a^\dagger b^\dagger - \text{sign}(g)c^\dagger d^\dagger] |0\rangle \quad (52)$$

of a boson Hamiltonian in the following form:

$$\mathcal{H} = \omega(a^\dagger a + b^\dagger b + c^\dagger c + d^\dagger d) + g(a^\dagger b^\dagger cd + d^\dagger c^\dagger ba). \quad (53)$$

The eigenenergy of $|\text{bound}\rangle$ is $2\omega - |g|$. Note that the existence of the bound state is independent of the sign of the interaction g .

On the other hand, the W term lowers the energies of *both* states by the same amount, hence does not play a central role in the formation of the biexciton state. The most important effect of the W term is to lower the energy of $b_+^\dagger b_-^\dagger |0\rangle$, relative to those of $b_+^\dagger b_+^\dagger |0\rangle$ and $b_-^\dagger b_-^\dagger |0\rangle$, and this effect was detected experimentally.¹² In the framework of the present bosonic theory, $\tilde{\mathcal{H}}'_{1s}$ lowers the energy of the bonding (biexciton) state relative to that of the antibonding state, and thus is crucial for the formation of the biexciton state, whereas the W term lowers the energy of both bonding and antibonding states. The point that should be emphasized is that the crucial interaction for biexciton formation is not $-U'b_+^\dagger b_-^\dagger b_- b_+$ but $U(b_+^\dagger b_-^\dagger b_\alpha b_\beta + \text{H.c.})$. The g in Eq. (53) corresponds to U in the present case.

F. Dipole decoupling and HF approximation

The original SBE are a theory within HF. There are two reasons that an extension beyond HF approximation becomes necessary. One reason is that it is necessary to take exciton-exciton correlation effects into account. When the excitation is low, the optical response from semiconductors are attributed to excitons. Similar features appear under high magnetic fields,³¹ where the most striking feature is that the signal of time-domain four-wave mixing does not decay exponentially, whereas the SBE predict a single-exponential decay. These typical two cases are beyond the scope of SBE.

As for the spin degrees of freedom, the excitations created by photons with the opposite circularly polarization are completely decoupled within HF. Furthermore, the experimental results of the polarization dependence of four-wave mixing signals and quantum beats are not treated within HF. The coupling of the excitation with the opposite spin is obtained beyond HF approximation.

Finally, we discuss the relation between the fermionic theories^{1,2,7,32,33} and the present bosonic theory. The HF factorization treatment of the SBE (Ref. 2) cannot produce the interaction between the excitation created by right-circularly polarized light and the excitation by left-circularly polarized light. The HF theory therefore corresponds to \mathcal{H}_{1s}^\pm , Eq. (30). It was argued in Refs. 7, 32, and 33 that the interactions of an exciton with higher states (including free carriers) are important, and that the interactions result in the energy shift, the EID, and the ‘‘biexcitonic correlations.’’ In the bosonic theory in the form of Eq. (29), these effects are included in \mathcal{H}'_{1s} and $\mathcal{H}_{\text{others}}$. After the projection is made, the relation is roughly as follows. The renormalized Hamiltonian $\tilde{\mathcal{H}}_{1s}^\pm$, Eq. (46), would include the HF term. The EID may be described by both $\tilde{\Gamma}$ and $\tilde{\mathcal{H}}'_{1s}$. The ‘‘biexcitonic correlation’’ would be included in $\tilde{\mathcal{H}}'_{1s}$. We believe that the present theory thus helps to bridge the gap between the bosonic theories^{3–5,12} and the fermionic theories^{1,2,7,32,33} of e - h systems. However, more detailed comparisons will be a subject of future studies.

As another future problem, the microscopic expression of the filling factor ν should be discussed on the same footing. Although such an expression was derived in Ref. 34, it cor-

responds to the “before projection” in our theory, so that the corresponding term “after projection” remains a subject of future research.

VI. CONCLUSIONS

In this paper, we have derived the effective Hamiltonian for $1s$ excitons with spin degree of freedom in two dimensions. This theory is valid when excitation density is weak and when the photon energy is close to the $1s$ exciton energy, because the boson operators of $1s$ excitons are used. Relaxation processes of excitons should be less important like, e.g., a QW in microcavity with high- Q value. What should be most emphasized is that the projection is crucial to obtain the effective Hamiltonian of excitons: the correct effective Hamiltonian of $1s$ excitons is not the one which is obtained by discarding the all exciton operators of $\nu \neq 1s$ among the full interaction of excitons [see Eqs. (5) and (6)], because it cannot explain the experimental results even qualitatively, as discussed in Sec. III. The higher exciton states $\nu=2p, 3d, \dots$ play important roles as intermediate

states. In order to include such effects, the projection is used. Through this procedure, the interactions of excitons with the opposite spins are obtained and the interaction strength of excitons with the same spins is drastically modified (renormalized) as shown in Sec. IV. In short, the procedure renormalizes both the form and strength of the effective interaction.

It is also shown that the effective Hamiltonian obtained through the projection provides the microscopic foundation of the phenomenological Hamiltonian $\mathcal{H}_{\text{WIBM}}$, proposed in Ref. 12. The agreement of the present theory with experiments supports the validity of a description of a fermionic system by bosonic fields in two dimensions, if the excitation is weak. This is a strong indication that bosonization can be a powerful tool also in higher than one dimension.

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