(ii) resonance-enhancement factors, and (iii) line broadening[4,27] in the case of impurity scattering, which is expected to be discernible in an experiment.

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[*] Present address: Xerox Palo Alto Research Center, 3180 Porter Drive, Palo Alto, Calif. 94304.

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Effect of Electronic Polarization on States of Localized Electrons in Insulators[*]

Shao-fu Wang, Harbans L. Arora, and Mitsuru Matsuura
Department of Physics, University of Waterloo, Waterloo, Ontario, Canada

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Starting from the electronic-polaron theory we derive an expression for the interaction between a localized electron and a massive hole via the polarization field for all electron-hole separations. The form of this interaction remains unchanged even when both electron and hole masses are effectively infinite. The expression obtained is compared with the corresponding expression in the Haken-Schottky (HS) theory. Also expressions for polaron effects such as self-energy, mass correction, and Lamb-shift-type corrections are derived in second-order perturbation theory. Owing to the complete analogy between the electron and lattice polarons, our results are also applicable to the case of a bound lattice polaron. Our results are then compared with those of other authors for a bound lattice polaron.

I. INTRODUCTION

From recent work such as those of Refs. 1–3, it is seen that in studying quantum states of a localized electron in insulators, the electronic polarization plays a significant role and considerably affects not only the positions of the electronic energy levels but also the transition energies. In the above references, the treatment of electronic-polarization effects on the system under consideration is based on the electronic-polaron theory of Toyozawa[4] and Haken and Schottky[5] (T-HS). This theory is an analog of the usual lattice-polaron theory and can be used to study not only the effective potential of a source particle but also the polaron effects (such as self-energy, mass correction, and Lamb-shift-type corrections) due to the electronic-polarization field.

On the one hand, starting from the T-HS theory, the effective Hamiltonian, which includes the electronic-polarization field, is obtained in Ref. 2 by the Hartree-type self-consistent-field method. This method is appropriate for deeply localized electronic states but is not adequate for intermediate electron-hole separations. On the other hand, in Refs. 1 and 3, the expression for the electronic-polarization potential is obtained by adopting the Haken-Schottky (HS) result,[5] which is derived for large-radius Wannier excitons, i.e., for states of large electron-hole separations, from the electronic-polaron theory of T-HS.

In this paper, we adopt the T-HS theory for the electronic-polarization field and employ a different approach to derive the effective interaction between a localized electron and a massive hole (effectively of infinite mass; for simplicity, we shall call it just "hole" in this work) for all electron-hole separations. We examine the consequences of this interaction and the resultant polaron effects.

In Sec. II, we derive an analytic expression which takes into account the effect of the electronic polarization on the interaction between a localized electron and a hole. This result is then used to obtain expressions for the polaron effects on the localized electron. Owing to the complete analogy between the electronic- and lattice-polaron theories these results can also be applied to the case of a bound lattice polaron, by making formal substitutions for the appropriate physical quantities involved. However, the results for polaron effects can be applied only to the case when a single phonon energy is greater than the binding energy of the localized electron. In Sec. III, the expressions obtained in Sec. II are further studied in both nonband-mass theory and the band-mass approximation. Then the obtained results are compared with those of HS and other authors.[6,7] Finally, in Sec. IV a summary of the work is given.

II. THEORY

A. Formulation

In the absence of ionic polarization, the Hamiltonian of a system consisting of an electron and a massive hole located at an ion site (such as a localized exciton or an F center) in a perfect insulator may be written as

$$H = H_e - V(|\vec{r} - \vec{R}|) + \mathcal{H}(\vec{r}, \vec{R}, [b_{\vec{w}}]) , \qquad (1)$$

with

$$H_e = \frac{p^2}{2m} + \sum_\alpha V(\vec{r} - \vec{R}_{\alpha 0}) , \qquad (2)$$

$$\mathcal{H} = (\vec{r}, \vec{R}, [b_{\vec{w}}]) = \sum_{\vec{w}} b^\dagger_{\vec{w}} b_{\vec{w}} E_{ex} + \sum_{\vec{w}} (V_w b_{\vec{w}} e^{i\vec{w}\cdot\vec{r}} + \text{c. c.})$$
$$- \sum_{\vec{w}} (V_w b_{\vec{w}} e^{i\vec{w}\cdot\vec{R}} + \text{c. c.}) , \qquad (3)$$

where the $\vec{p}$, m, and $\vec{r}$ are, respectively, the momentum, the free-electron mass, and the position vector of the (localized) electron. $V(\vec{r} - \vec{R}_{\alpha 0})$ is the interaction of the electron with the αth ion (or atom) at $\vec{R}_{\alpha 0}$ when the αth ion is at its equilibrium position and its core and valence electrons are in the ground state. Thus the last term of Eq. (2) is the interaction of the electron with all ions making up the ideal insulator. The term $-V(|\vec{r} - \vec{R}|)$ in Eq. (1) is the attractive interaction between the bare electron and the hole at R, which is a Coulombic potential for large electron-hole separations. The Hamiltonian given by Eq. (3) was first derived by Toyozawa,[4] by considering the virtual excitation of crystalline valence electrons (these excitations are believed to be responsible for the electronic polarization) as a virtual exciton (approximately a boson), to describe the electronic-polarization field in the crystal and the interaction of this field with the source particles, i.e., the electron and the hole. Later, the same Hamiltonian [Eq. (3)] was also derived and justified by Haken and Schottky[5] from many-body considerations of the excitons. In this context $[b_{\vec{w}}]$ represents all coordinates of the virtual excitons, E_{ex} is the energy of a longitudinal exciton and is approximated to be independent of $\vec{w}$ as in Ref. 4, $b_{\vec{w}}$ and $b^\dagger_{\vec{w}}$ are, respectively, annihilation and creation operators for a longitudinal virtual exciton of wave vector $\vec{w}$, and V_w is given by the standard expression

$$V_w = -i \left[\frac{2\pi e^2 E_{ex}}{\Omega w^2} \left(1 - \frac{1}{\epsilon_\infty} \right) \right]^{1/2} , \qquad (4)$$

where Ω is the volume of the crystal under consideration.

We now introduce a new operator given by

$$B_{\vec{w}} = b_{\vec{w}} - V^*_{\vec{w}} \, e^{-i\vec{w}\cdot\vec{R}} / E_{ex} . \qquad (5)$$

For the system considered here, since $\vec{R}$ is constant, the commutation relations $[b_{\vec{w}}, \vec{R}] = 0$ and $[b^\dagger_{\vec{w}}, \vec{R}] = 0$ hold. Accordingly the new operator $B_{\vec{w}}$ and its complex conjugate, i.e., $B^\dagger_{\vec{w}}$, satisfy the commutation relations for boson operators. Thus $B_{\vec{w}}$ can be defined as an annihilation operator for bosons of wave vector $\vec{w}$ by $B_{\vec{w}} | 0 \rangle = 0$, where $| 0 \rangle$ is the corresponding vacuum state. With the aid of the operators $B_{\vec{w}}$ and $B^\dagger_{\vec{w}}$, the above H can be transformed to

$$H = H_e - V(|\vec{r} - \vec{R}|) + \sum_{\vec{w}} \frac{|V_w|^2}{E_{ex}} (e^{i\vec{w}\cdot(\vec{r} - \vec{R})} + \text{c. c.})$$
$$- \sum_{\vec{w}} \frac{|V_w|^2}{E_{ex}} + \sum_{\vec{w}} B^\dagger_{\vec{w}} B_{\vec{w}} E_{ex}$$
$$+ \sum_{\vec{w}} (V_w B_{\vec{w}} e^{i\vec{w}\cdot\vec{r}} + \text{c. c.}) , \qquad (6)$$

without a change in the eigenvalues. The fifth term in (6) is the Hamiltonian of the virtual excitons characterized by $B_{\vec{w}}$, i.e., the modified excitons, and the last term is the interaction between these virtual excitons and the trapped electron. The meaning of the third and fourth terms will be discussed later.

Taking the last term in (6) as a perturbation, we obtain (from the Schrödinger equation for the unperturbed system)

$$H_0 \psi_n(\vec{r}) = \epsilon_n \psi_n(\vec{r}) \tag{7a}$$

and

$$\sum_{\vec{w}} B^\dagger_{\vec{w}} B_{\vec{w}} E_{\mathrm{ex}} |n_w\rangle = \epsilon_{\mathrm{ex}} |n_w\rangle , \tag{7b}$$

where $|n_w\rangle \equiv |n_1, n_2, \cdots\rangle$ is an eigenfunction of the exciton system and ϵ_{ex} is the corresponding eigenvalue. The ψ_n's and ϵ_n's are, respectively, eigenfunctions and eigenvalues of the electronic Hamiltonian H_0, where H_0 is given by

$$H_0 = H_e - V(|\vec{r} - \vec{R}|) + \sum_{\vec{w}} \frac{|V_w|^2}{E_{\mathrm{ex}}} (e^{i\vec{w}\cdot(\vec{r}-\vec{R})} + \mathrm{c.\,c.}) . \tag{8}$$

The fourth term of Eq. (6), which will be shown to be the self-energy of the hole due to its own polarization field, is a constant and is omitted hereafter. Thus the energy of the unperturbed state $\psi_n(\vec{r})|n_w\rangle$ is $\epsilon_n + \sum_{\vec{w}} n_{\vec{w}} E_{\mathrm{ex}}$, where n_w is the number of modified virtual excitons with energy E_{ex}. Let us start with the unperturbed state $\psi_n(\vec{r})|0\rangle$ in which there is a trapped electron and no excitons are present. The perturbation operating on this unperturbed state produces all states with one exciton present in the field. Therefore, the matrix element involved in first-order energy vanishes, and there is no first-order perturbation of the energy level of the electron. In second order, however, the matrix elements, for which the final state has one exciton, are nonvanishing and we get

$$\Delta E_n = - \sum_{\vec{w}, m} \frac{|V_w|^2 |\rho_{nm}|^2}{\epsilon_m + E_{\mathrm{ex}} - \epsilon_n} , \tag{9}$$

where

$$\rho_{nm} = \int \psi^*_n(\vec{r})\, \psi_m(\vec{r})\, e^{i\vec{w}\cdot\vec{r}}\, d\vec{r}$$

and m is summed over all electronic states. Equation (9) is analogous to that derived by Bethe for the self-energy of an electron in a stationary state of hydrogen, due to its interaction with virtual photons.[8] Hence the ΔE_n [Eq. (9)] is the electron self-energy due to its interaction with virtual excitons.

B. Further Physical Interpretations

To understand the physical meaning of the fourth term in Eq. (6), consider first Eq. (9) and the higher-order perturbation corrections to the total energy for the case in which the electronic transition energy $|\epsilon_m - \epsilon_n|$ (for which the corresponding $|\rho_{nm}|$ is significantly different from zero) is very much less than the exciton energy E_{ex} (order of the first exciton peak). Thus $|\epsilon_m - \epsilon_n|$ can be neglected compared to E_{ex}. In this case, since $\sum_m |\rho_{nm}|^2 = 1$, the above ΔE_n reduces to

$$E_s(e) = -\sum_{\vec{w}} \frac{|V_w|^2}{E_{\mathrm{ex}}} , \tag{10}$$

the electron self-energy. Further all the higher odd-order corrections are zero as in the case of the first-order correction. As for the higher even-order corrections first consider the fourth-order correction. It follows from the conclusion drawn in Ref. 9 that if $|\epsilon_m - \epsilon_n|$, for which $|\rho_{nm}|$ is far from zero, is very much less than E_{ex}, the fourth term is identically zero. We then expect, in this limit, the sixth and higher even orders to contribute zero, and thus $E_s(e)$ given by Eq. (10) is exact in this limit. Now compare the fourth term of Eq. (6) with $E_s(e)$. Both have the same form, and hence the former is the self-energy of the hole due to its own electronic-polarization field. Note that this self-energy of the hole is obtained by employing an exact canonical transformation, considering it as a source particle of infinite mass, i.e., effectively static with respect to virtual excitons. Thus an expression like the fourth term of Eq. (6) is exactly the self-energy of a source particle in the static approximation (this approximation is the opposite extreme to the adiabatic approximation used in the literature for the electron-phonon interaction). It follows then that $E_s(e)$ is also the self-energy of the electron in the static approximation and that a criterion for the validity of the static approximation is that the $|\epsilon_n - \epsilon_m|$, for which the corresponding $|\rho_{nm}|$ is large, should be much less than E_{ex}. This is consistent with what one has expected.

It is seen from the derivation of Eq. (10) that when $|\epsilon_n - \epsilon_m|$, for which $|\rho_{nm}|$ is significantly different from zero, is considerably less than the exciton energy E_{ex}, the higher-order perturbation corrections are very small compared to the second-order energy correction given by Eq. (9). Therefore the second-order perturbation theory is sufficient in calculating a correction to the total energy of a localized electron, described by a Hamiltonian like that given by Eq. (6), when its ground state is closer to the conduction band than to the valence band. In the following we shall limit ourselves to this case.

The interpretation of the fourth term of Eq. (6) as the self-energy of a massive hole dressed with virtual excitons has its analogy in the meson theory of nuclear forces. Then, following the interpretation given in the meson theory for a term analogous to the third term in Eq. (6), we interpret this term, i.e.,

$$V_e=\sum_{\vec{w}} \frac{|V_w|^2}{E_{\text{ex}}}\left(e^{i\mathbf{w}\cdot(\vec{r}-\vec{R})}+\text{c. c.}\right) , \tag{11a}$$

as the interaction between the electron and the hole via the electronic-polarization field. This interaction is due to the exchange of virtual excitons between the electron and the hole. Hereafter let us call this interaction the exchange interaction.

Replacing the summation over the wave vector $\vec{w}$ in Eq. (11a) by an integration with respect to this vector over the first Brillouin zone, as is the usual procedure, we obtain

$$V_e=\left(1-\frac{1}{\epsilon_\infty}\right)\frac{e^2}{|\vec{r}-\vec{R}|}\,\frac{2}{\pi}\int_0^{w_m|\vec{r}-\vec{R}|}\frac{\sin\mu}{\mu}\,d\mu , \tag{11b}$$

where $\mu=w|\vec{r}-\vec{R}|$ and w_m is the maximum value of $|\vec{w}|$. w_m is approximately π/d for alkali halides with NaCl structure, where d is the nearest-neighbor distance. When the electron-hole separation $|\vec{r}-\vec{R}|$ is very much larger than d, V_e reduces to $(1-1/\epsilon_\infty)e^2/|\vec{r}-\vec{R}|$ because of the decreasing nature of the integrand with increasing $|\vec{r}-\vec{R}|$. In this case the second term in Eq. (8) becomes a Coulombic potential and hence Eq. (8) reduces to

$$H_0=H_e-e^2/\epsilon_\infty|\vec{r}-\vec{R}| .$$

This means that the exchange interaction is nothing but the dielectric screening interaction. This interaction reduces the electron-hole interaction "$-V(|\vec{r}-\vec{R}|)$" in Eq. (8) and, thereby, the magnitudes of ϵ_n and ϵ_m so that $|\epsilon_m-\epsilon_n|$ becomes much less than E_{ex}. Therefore, as noted above on the use of perturbation theory for this problem, the perturbation series will be much more rapidly convergent using the transformed Hamiltonian rather than the original Hamiltonian.

We note that the exchange interaction obtained here can be applied to an electron-hole pair of arbitrary separation so long as $\mathcal{H}$ expressed by Eq. (3) exists. This is because no limitation for $|\vec{r}-\vec{R}|$ is included in making the canonical transformation. For the case of large electron-hole separations, the exchange interaction merely introduces the usual high-frequency dielectric constant ϵ_∞ into the electron-hole interaction in Eq. (8) as discussed above. Thus the eigenfunctions of H_0 [Eq. (8)] and those of the Hamiltonian equal to the sum of the first two terms in this equation are functionally the same, differing only in the eigenvalues. Whereas for intermediate and small separations, the exchange interaction introduces the $\vec{r}$-dependent dielectric constant into the electron-hole interaction. Consequently, the unperturbed states become complicated compared to those in the case of large separations. Now, however, the transformation to H_0 [Eq. (8)] places us predominantly in the region of large separations thus simplifying the problem (see Sec. II C for details).

The exchange interaction will be further discussed and compared with the corresponding HS expression in Sec. III.

C. Evaluation of ΔE_n

In the theory of a bound lattice polaron, attempts have been made to evaluate an equation like Eq. (9).[6,7] In this work we shall use a different approach so that ΔE_n can be evaluated more accurately than by the methods used in these references.

We can rewrite Eq. (9) in the form

$$\Delta E_n=-\sum_{\vec{w}}|V_w|^2\left\langle n\left|e^{i\vec{w}\cdot\vec{r}}\,\frac{1}{E_{\text{ex}}+H_0-\epsilon_n}\,e^{-i\vec{w}\cdot\vec{r}}\right|n\right\rangle \tag{12}$$

for the state $|n\rangle$ under consideration. If we use the formal expansion, this can be written as

$$\Delta E_n=-\sum_{k=0}^{\infty}(-1)^k\sum_{\vec{w}}\frac{|V_w|^2}{E_{\text{ex}}}\times\left\langle n\left|e^{i\vec{w}\cdot\vec{r}}\left(\frac{H_0-\epsilon_n}{E_{\text{ex}}}\right)^k e^{-i\vec{w}\cdot\vec{r}}\right|n\right\rangle . \tag{13}$$

Using the relation

$$e^{i\vec{w}\cdot\vec{r}}\,p^2\,e^{-i\vec{w}\cdot\vec{r}}=(\vec{p}-\hbar\vec{w})^2 ,$$

we get

$$\Delta E_n=-\sum_{k=0}^{\infty}(-1)^k\sum_{\vec{w}}\frac{|V_w|^2}{E_{\text{ex}}}\times\left\langle n\left|\left(\frac{\hbar^2w^2/2m-\hbar\vec{w}\cdot\vec{p}/m+H_0-\epsilon_n}{E_{\text{ex}}}\right)^k\right|n\right\rangle \tag{14}$$

or

$$\Delta E_n=-\sum_{\vec{w}}|V_w|^2\times\left\langle n\left|\left(E_{\text{ex}}+\frac{\hbar^2w^2}{2m}-\frac{\hbar\vec{w}\cdot\vec{p}}{m}+H_0-\epsilon_n\right)^{-1}\right|n\right\rangle . \tag{15}$$

We can proceed further by an expansion to get

$$\Delta E_n=-\sum_{\vec{w}}\frac{|V_w|^2}{E_{\text{ex}}+\hbar^2w^2/2m}\times\left\langle n\left|\left[1-\left(\frac{\hbar\vec{w}\cdot\vec{p}/m+\epsilon_n-H_0}{E_{\text{ex}}+\hbar^2w^2/2m}\right)\right]^{-1}\right|n\right\rangle \tag{16}$$

$$=\sum_{j=0}^{\infty}J_j , \tag{17}$$

where

$$J_j = -\sum_{\vec{w}} \frac{|V_w|^2}{E_{ex}+\hbar^2 w^2/2m} \left\langle n \left| \left(\frac{\hbar \vec{w}\cdot\vec{p}/m + \epsilon_n - H_0}{E_{ex}+\hbar^2 w^2/2m} \right)^j \right| n \right\rangle . \tag{18}$$

For the cases $j = 0,\ 1$,

$$J_0 = -\sum_{\vec{w}} \frac{|V_w|^2}{E_{ex}+\hbar^2 w^2/2m} , \tag{19}$$

$$J_1 = -\sum_{\vec{w}} \frac{|V_w|^2}{(E_{ex}+\hbar^2 w^2/2m)^2} \left\langle n \left| \frac{\hbar \vec{w}\cdot\vec{p}}{m} \right| n \right\rangle = 0 . \tag{20}$$

For the case of $j \geq 2$, we can use the expression

$$J_j = -\sum_{\vec{w}} \frac{|V_w|^2}{(E_{ex}+\hbar^2 w^2/2m)^{j+1}} \left\langle n \left| \frac{\hbar \vec{w}\cdot\vec{p}}{m} \times \left(\frac{\hbar \vec{w}\cdot\vec{p}}{m} + \epsilon_n - H_0 \right)^{j-2} \frac{\hbar \vec{w}\cdot\vec{p}}{m} \right| n \right\rangle . \tag{21}$$

Then using the eigenvalue equation for H_0, i.e., $H_0 |n\rangle = \epsilon_n |n\rangle$, we can get the expression for arbitrary $J_j\ (j \geq 2)$, in principle. For example,

$$J_2 = -\sum_{\vec{w}} \frac{|V_w|^2}{(E_{ex}+\hbar^2 w^2/2m)^3} \left\langle n \left| \left(\frac{\hbar \vec{w}\cdot\vec{p}}{m} \right)^2 \right| n \right\rangle , \tag{22}$$

$$\begin{aligned} J_3 &= -\sum_{\vec{w}} \frac{|V_w|^2}{(E_{ex}+\hbar^2 w^2/2m)^4} \left\langle n \left| \frac{\hbar \vec{w}\cdot\vec{p}}{m} \left(\frac{\hbar \vec{w}\cdot\vec{p}}{m} + \epsilon_n - H_0 \right) \frac{\hbar \vec{w}\cdot\vec{p}}{m} \right| n \right\rangle \\ &= -\sum_{\vec{w}} \frac{|V_w|^2}{(E_{ex}+\hbar^2 w^2/2m)^4} \left\langle n \left| \left[H_0 , \frac{\hbar \vec{w}\cdot\vec{p}}{m} \right] \frac{\hbar \vec{w}\cdot\vec{p}}{m} \right| n \right\rangle \\ &= -\sum_{\vec{w}} \frac{|V_w|^2}{(E_{ex}+\hbar^2 w^2/2m)^4} \left\langle n \left| \left[v(\vec{r}) \left(\frac{\hbar \vec{w}\cdot\vec{p}}{m} \right)^2 - \frac{\hbar \vec{w}\cdot\vec{p}}{m}\, v(\vec{r})\, \frac{\hbar \vec{w}\cdot\vec{p}}{m} \right] \right| n \right\rangle , \end{aligned} \tag{23}$$

$$\begin{aligned} J_4 &= -\sum_{\vec{w}} \frac{|V_w|^2}{(E_{ex}+\hbar^2 w^2/2m)^5} \left\langle n \left| \frac{\hbar \vec{w}\cdot\vec{p}}{m} \left(\frac{\hbar \vec{w}\cdot\vec{p}}{m} + \epsilon_n - H_0 \right)^2 \frac{\hbar \vec{w}\cdot\vec{p}}{m} \right| n \right\rangle \\ &= -\sum_{\vec{w}} \frac{|V_w|^2}{(E_{ex}+\hbar^2 w^2/2m)^5} \left\langle n \left| \left\{ \left(\frac{\hbar \vec{w}\cdot\vec{p}}{m} \right)^2 + \left[H_0 , \frac{\hbar \vec{w}\cdot\vec{p}}{m} \right] \right\} \left\{ \left(\frac{\hbar \vec{w}\cdot\vec{p}}{m} \right)^2 - \left[H_0 , \frac{\hbar \vec{w}\cdot\vec{p}}{m} \right] \right\} \right| n \right\rangle \\ &= -\sum_{\vec{w}} \frac{|V_w|^2}{(E_{ex}+\hbar^2 w^2/2m)^5} \left\langle n \left| \left\{ \left(\frac{\hbar \vec{w}\cdot\vec{p}}{m} \right)^4 - \left[\left(v(\vec{r}) , \frac{\hbar \vec{w}\cdot\vec{p}}{m} \right) \right]^2 \right\} \right| n \right\rangle , \end{aligned} \tag{24}$$

where $v(\vec{r})$ is the potential-energy term in H_0 and is given by

$$v(\vec{r}) = \sum_{\alpha} V(\vec{r} - \vec{R}_{\alpha 0}) - V(\vec{r}) + \sum_{\vec{w}} \frac{|V_w|^2}{E_{ex}} (e^{i\vec{w}\cdot\vec{r}} + \text{c.c.}) , \tag{25}$$

choosing the origin of all vectors to be at the center of the hole. The higher-order expressions have more complicated forms but can be obtained straightforwardly.

If we retain terms up to the order $(E_n^b / E_{ex})^2$ where E_n^b is the binding energy of the nth state, we get from the above results

$$\begin{aligned} \Delta E_n = &-\sum_{\vec{w}} \frac{|V_w|^2}{E_{ex}+\hbar^2 w^2/2m} - \sum_{\vec{w}} \frac{|V_w|^2}{(E_{ex}+\hbar^2 w^2/2m)^3} \left\langle n \left| \left(\frac{\hbar \vec{w}\cdot\vec{p}}{m} \right)^2 \right| n \right\rangle - \sum_{\vec{w}} \frac{|V_w|^2}{(E_{ex}+\hbar^2 w^2/2m)^4} \\ &\times \left\langle n \left| \left[v(\vec{r}) \left(\frac{\hbar \vec{w}\cdot\vec{p}}{m} \right)^2 - \frac{\hbar \vec{w}\cdot\vec{p}}{m}\, v(\vec{r}) \frac{\hbar \vec{w}\cdot\vec{p}}{m} \right] \right| n \right\rangle - \sum_{\vec{w}} \frac{|V_w|^2}{(E_{ex}+\hbar^2 w^2/2m)^5} \left\langle n \left| \left(\frac{\hbar \vec{w}\cdot\vec{p}}{m} \right)^4 \right| n \right\rangle . \end{aligned} \tag{26}$$

Replacing the summation over $\vec{w}$ by an integration with respect to this vector in the first Brillouin zone, we obtain

$$\Delta E_n = -\alpha_1 E_{ex} - \tfrac{1}{6} \alpha_2 E_{ex} A_n + \tfrac{1}{12} \alpha_3 E_{ex} B_n - \tfrac{3}{40} \alpha_4 E_{ex} C_n , \tag{27}$$

where

$$\begin{aligned} A_n &= \frac{1}{2mE_{ex}} \langle n | p^2 | n \rangle , \\ B_n &= \frac{1}{2mE_{ex}{}^2} \langle n | (\vec{p} v(\vec{r})) \cdot \vec{p} | n \rangle , \\ C_n &= \frac{1}{(2mE_{ex})^2} \langle n | p_4 | n \rangle , \end{aligned} \tag{28}$$

$$\alpha_1 = \frac{2\alpha}{\pi} \tan^{-1} u \,, \tag{29a}$$

$$\alpha_2 = \frac{2\alpha}{\pi} \left(\frac{-2u}{(1+u^2)^2} + \frac{u}{1+u^2} + \tan^{-1} u \right) , \tag{29b}$$

$$\alpha_3 = \frac{2\alpha}{\pi} \left(\frac{-8u}{3(1+u^2)^3} + \frac{2u}{3(1+u^2)^2} + \frac{u}{1+u^2} + \tan^{-1} u \right) , \tag{29c}$$

and

$$\alpha_4 = -\frac{32\alpha}{3\pi} \frac{u^3}{(1+u^2)^4} + \alpha_3 \,, \tag{29d}$$

u being

$$u = (\hbar^2/2mE_{\text{ex}})^{1/2} w_m \,. \tag{30}$$

w_m is the maximum value of $|\vec{w}|$ in the first Brillouin zone and α is

$$\alpha = \frac{e^2}{2E_{\text{ex}}} \left(1 - \frac{1}{\epsilon_\infty} \right) \left(\frac{2mE_{\text{ex}}}{\hbar^2} \right)^{1/2} . \tag{31}$$

The α_j's in Eqs. (29) reduce to α when w_m is replaced by infinity. This approximation is reasonable only when the integrand of the $\vec{w}$ integral decreases very rapidly as $|\vec{w}|$ increases for given E_{ex} and m.

In Eq. (27), the first term represents the *usual* self-energy of the electron due to electronic polarization, the second term is nothing but the mass correction, and the third and fourth terms are the so-called Lamb-shift-type corrections. The magnitude of these polaron effects depends on the coupling constant α, the electronic mass m, the exciton energy E_{ex}, and the form of the potential $v(\vec{r})$ (see Sec. III for details).

We note that in the above calculation the $|n\rangle$ was treated as an eigenfunction of H_0 only and thus the above results are not only applicable in the non-band-mass theory but also in the band-mass approximation [i.e., $p^2/2m + \sum_\alpha V(\vec{r} - \vec{R}_{\alpha 0}) \to p^2/2m_b$, where m_b is the electronic band mass] if the free electron mass m is replaced by m_b. When the band-mass approximation holds for a trapped electron, the electron-hole separation is usually large and the exchange interaction given by Eq. (11) becomes $(1 - 1/\epsilon_\infty) e^2/|\vec{r} - \vec{R}|$. Consequently, H_0 [expressed by Eq. (8)] reduces to

$$H_0 = p^2/2m_b - e^2/\epsilon_\infty |\vec{r} - \vec{R}| \,. \tag{32}$$

This is a hydrogenic Hamiltonian and its eigenfunctions $|n\rangle$ are hydrogenlike.

When the band-mass approximation is not applicable, however, the electron-hole separation is usually not so large that the H_0 becomes hydrogenic. The corresponding eigenfunctions are thus complicated even if one ignores the exchange interaction in H_0. In this case, as in the semicontinuum calculation of the F center,[2] the hydrogenic wave functions may be chosen as the trial wave functions in a variational calculation to obtain numerical values of the total energy including the correction ΔE_n.

As a further comment we see that, because of the complete analogy between the electronic polaron and the lattice polaron, the exchange interaction and ΔE_n for a bound lattice polaron are given by Eqs. (11) and (27), respectively, with $E_{\text{ex}} \to \hbar\omega$ and $(1 - 1/\epsilon_\infty) \to (1/\epsilon_\infty - 1/\epsilon_s)$, where ϵ_s is the static dielectric constant and ω is the frequency of a longitudinal optical phonon of wave vector $\vec{w}$. The exchange interaction thus obtained is applicable in both cases of $\omega_e > \omega$ and $\omega_e < \omega$, where ω_e is the frequency of the trapped electron. However, note that ΔE_n obtained in this way is valid only for $\omega_e < \omega$, i.e., the weak coupling case.

III. DISCUSSION OF RESULTS AND COMPARISON WITH OTHER CALCULATIONS

In this section we proceed to examine and compare the results of Sec. II, i.e., polaron effects and the exchange interaction in relation to other calculations in the literature. We discuss polaron effects, given by Eq. (27), in Sec. III A and the exchange interaction, given by Eq. (11), in Sec. III B.

A. Polaron Effects

First consider the case of shallow electronic states within the band-mass approximation. In this case, as discussed in Sec. II C, the unperturbed Hamiltonian H_0 is hydrogenic and the unperturbed states are hydrogenlike so that the second-order energy corrections to the $1s$-, $2s$-, and $2p$-like hydrogenic states, according to Eq. (27), are

$$\Delta E_{1s} = -\alpha_{1b} E_{\text{ex}} - \tfrac{1}{6} \alpha_{2b} E_{\text{ex}} (E_{1s}^b / E_{\text{ex}}) + (\tfrac{1}{3} \alpha_{3b} - \tfrac{3}{8} \alpha_{4b}) E_{\text{ex}} (E_{1s}^b / E_{\text{ex}})^2 \,, \tag{33a}$$

$$\Delta E_{2s} = -\alpha_{1b} E_{\text{ex}} - \tfrac{1}{6} \alpha_{2b} E_{\text{ex}} (E_{2s}^b / E_{\text{ex}}) + (\tfrac{16}{24} \alpha_{3b} - \tfrac{39}{40} \alpha_{4b}) E_{\text{ex}} (E_{2s}^b / E_{\text{ex}})^2 \,, \tag{33b}$$

$$\Delta E_{2p} = -\alpha_{1b} E_{\text{ex}} - \tfrac{1}{6} \alpha_{2b} E_{\text{ex}} (E_{2p}^b / E_{\text{ex}}) + (0 - \tfrac{7}{40} \alpha_{4b}) E_{\text{ex}} (E_{2p}^b / E_{\text{ex}})^2 \,, \tag{33c}$$

where E_n^b is the magnitude of the energy of the usual hydrogenic levels measured from the bottom of the conduction band, that is,

$$E_{1s}^b = m_b e^4 / 2\epsilon_\infty^2 \hbar^2 \,, \quad E_{2s}^b = E_{2p}^b = m_b e^4 / 8\epsilon_\infty^2 \hbar^2 \,, \tag{34}$$

and the α_{jb}'s are those given by Eq. (29) with the electronic mass m replaced by the band mass m_b.

The first term of Eqs. (33) is the constant energy shift due to electronic polarization and is the same for all electronic states. This corresponds to the energy shift in the usual lattice polaron theory. However, the magnitude of this shift is different because of the different values of α_b and E_{ex}. The second term arising from $-\frac{1}{6}\alpha_2 E_{ex} A_n$ in Eq. (27) gives the mass correction and has a form similar to that in the weak-coupling theory of lattice polaron. The last term in each of the Eqs. (33) is the Lamb-shift-type correction, which is a function of the state in question. The first and second terms in the bracket of this last term arise from $\frac{1}{12}\alpha_3 \times E_{ex} B_n$ and $-\frac{3}{40}\alpha_4 E_{ex} C_n$ in Eq. (27), respectively.

The corresponding expression for a bound lattice polaron, obtained from the present results by making formal replacements, differs from those given in Refs. 6 and 7. In Ref. 6, only the term corresponding to $\frac{1}{12}\alpha_{3b} E_{ex} B_n$ of Eq. (27) exists and the term corresponding to $-\frac{3}{40}\alpha_{4b} E_{ex} C_n$ does not appear. This is because in Ref. 6, the expansion of the second-order energy correction was truncated excluding the terms of the order $(p^2/2m)^2$. The net result of this is that Lamb-shift-type correction is opposite in sign to that in Ref. 6 and smaller than it in magnitude.

On the other hand, Ref. 7 includes terms like $-\frac{3}{40}\alpha_{4b} E_{ex} C_n$, but excludes $(\frac{1}{12}\alpha_{3b} E_{ex} B_n)$-like terms, which include the potential $v(\vec{r})$. This is due to an approximation used in this reference, i.e., setting $[F, H_0] = [F, p^2/2m_b]$, where $H_0 = p^2/2m_b - e^2/\epsilon_s r$, and F is an operator satysfying the following equation:

$$([F, H_0] - \hbar\omega F)|n\rangle = e^{-i\vec{w}\cdot\vec{r}}|n\rangle .$$

If we make an equivalent approximation, our results reduce to those of Ref. 7, and thus lead to an overestimation of the Lamb-shift correction for s states.

It is instructive to compare our Lamb-shift correction with that due to the photon field in the hydrogen atom.[8] It is seen that only the term $\frac{1}{12}\alpha_3 \times E_{ex} B_n$ is analogous to the Lamb shift in the hydrogen atom. In the problem of Lamb shift in hydrogen, as the momentum of the photon is small compared with the electron momentum, the contribution to the kinetic-energy term by the photon momentum can be neglected. Thus there is no correction like $-\frac{3}{40}\alpha_4 E_{ex} C_n$ term. In our problem, this contribution is not negligible. In the case of $\alpha_{3b} \simeq \alpha_{4b}$, this contribution is larger than the usual Lamb-shift-type corrections for $1s$, $2s$, and $2p$ states, and the energy of the $2s$ state is lower than that of the $2p$ state contrary to the result of the Lamb-shift in hydrogen atom [cf. Eqs. (33b) and (33c)].

Second, we estimate the corrections represented by Eq. (27) for a deeper state in the nonband-mass theory and compare these with those discussed above in the band-mass approximation. The evaluation of B_n for any given electronic state is very complicated. For the sake of simplicity, we consider an F-center-like electron and approximate its ground state by a hydrogenlike $1s$ wave function. Then, for the potential $v(\vec{r})$ given by Eq. (25), the ΔE_{1s} in the nonband-mass theory is worked out to be

$$\Delta E_{1s} = -\alpha_1 E_{ex} - \frac{1}{6}\alpha_2 \frac{\hbar^2\lambda^2}{2m} + \frac{\alpha_3}{6}\frac{\hbar^2\lambda^2}{2m}\frac{e^2\lambda}{E_{ex}}\sum_i{}' z_i e^{-2\lambda d_i} - \frac{\alpha_3}{12\pi}\left(1 - \frac{1}{\epsilon_\infty}\right)\frac{\hbar^2\lambda^2}{2m}\frac{e^2\lambda}{E_{ex}}\left(\tan^{-1}x - \frac{x}{1+x^2}\right) - \frac{\frac{3}{8}\alpha_4(\hbar^2\lambda^2/2m)^2}{E_{ex}} . \quad (35)$$

Here λ is the parameter involved in $1s$ wave function $\psi_{1s} = (\lambda^3/\pi)^{1/2} e^{-\lambda r}$, d_i is the distance of the ith ion from the center of the trap, z_i is the charge on the ith ion, and the prime on the sum in the third term implies summation over all ions except the trap. The x involved in the fourth term is equal to $w_m/2\lambda$. We note that α_j's in the above equation are those given by Eqs. (29) in the nonband-mass theory and that the mass m involved in the expression for α, Eq. (31), is the free-electron mass. This value of α is different from that of α_b and for a given crystal $\alpha = (m/m_b)^{1/2}\alpha_b$. Thus the first two terms, which give, respectively, the energy shift and the mass correction in the nonband-mass theory, are α_1/α_{1b} and α_2/α_{2b} times the corresponding values in the band-mass approximation, respectively. Combining the second term with the kinetic energy

$$\langle 1s|p^2/2m|1s\rangle = \hbar^2\lambda^2/2m$$

gives the electronic-polaron mass

$$m^* = m/(1 - \frac{1}{6}\alpha_2) . \quad (36)$$

For typical alkali halide crystals such as NaCl and KCl, $m \simeq 2m_b$, $E_{ex} \simeq 8$ eV, $\epsilon_\infty \simeq 2$, $w_m \simeq \pi/d \simeq 1$ (d is the nearest-neighbor distance), and $\alpha_2 \simeq 0.84\alpha_{2b}$, where α_{2b} is of the order of 0.3. Thus the effect of the electronic polarization on the electronic mass, and thereby on the kinetic energy, is small. The third and fourth terms in Eq. (35) arise from the third term, and the last term comes from the last term in the expansion of ΔE_n, Eq. (27). These three terms contribute to the Lamb-shift correction. For typical alkali halide crystals, $\alpha_3 \simeq 0.5\alpha$, $\alpha_4 \simeq 0.25\alpha$, and the sum of these corrections is about "-0.02 eV" for $\lambda = 0.7$ Å^{-1} and "0.002 eV" for $\lambda = 0.4$ Å^{-1}. These values are small as compared with the optical absorption and emission energies due to point defects.

B. Exchange Interaction

It is interesting to note that the exchange interaction obtained in Sec. II, i.e., Eq. (11), is exactly the same as that obtained when both the electron and hole masses are effectively infinite (equivalently, the frequencies of the electron and hole are much less than the frequency of the polarization field, i.e., the static approximation). Indeed, in the static limit, Eq. (1) becomes

$$H_s = \sum\nolimits_\alpha V(\vec{r} - \vec{R}_{\alpha 0}) - V(|\vec{r} - \vec{R}|) + \mathcal{H} . \tag{37}$$

We introduce the operator

$$\beta_{\vec{w}} = b_{\vec{w}} - \frac{V_w^* e^{-i\vec{w}\cdot\vec{R}}}{E_{ex}} + \frac{V_w^* e^{-i\vec{w}\cdot\vec{r}}}{E_{ex}} . \tag{38}$$

This operator and its Hermitian conjugate $\beta_{\vec{w}}^\dagger$ satisfy the commutation relations for bosons in this limit and can be defined, respectively, as an annihilation operator and a creation operator for bosons of wave vector $\vec{w}$ with respect to a vacuum state $\| 0\rangle$, defined by $\beta_{\vec{w}} \| 0\rangle = 0$. With the aid of these new operators, Eq. (37) can be transformed into

$$H_s = \sum\nolimits_\alpha V(\vec{r} - \vec{R}_{\alpha 0}) - V(|\vec{r} - \vec{R}|) + \sum_{\vec{w}} \beta_{\vec{w}}^\dagger \beta_{\vec{w}} E_{ex} + \sum_{\vec{w}} \frac{|V_w|^2}{E_{ex}} (e^{i\vec{w}\cdot(\vec{r}-\vec{R})} + \text{c. c.}) - 2\sum_{\vec{w}} \frac{|V_w|^2}{E_{ex}} . \tag{39}$$

The third term is the Hamiltonian of excitons characterized by these new operators, and the last term is the sum of the self-energies of the electron and the hole in this limit. The fourth term is thus the corresponding exchange interaction and is exactly that given by Eq. (11), obtained for an electron moving around a massive hole. The exchange interaction obtained in Ref. 1 has been generally interpreted as being valid only in the static approximation, whereas we see that, in fact, it has a more general validity when the hole is fixed.

Next let us compare the exchange interaction given in Eq. (11) with that of Haken and Schottly.[5] Since the HS theory for an exciton in a polarizable field starts with the band-mass approximation, we limit ourselves to this approximation in the following discussion. The exchange energy in this calculation is given by

$$E_e = \left\langle \Psi \middle| \sum_{\vec{w}} \frac{|V_w|^2}{E_{ex}} (e^{i\vec{w}\cdot(\vec{r}-\vec{R})} + \text{c. c.}) \middle| \Psi \right\rangle \tag{40}$$

for an eigenstate Ψ of H given by Eq. (6). If this Ψ is approximated by the product of the vacuum state of virtual excitons and an eigenstate of $\mathcal{H}_e$ is given by

$$\mathcal{H}_e = p^2/2m_b - e^2/|\vec{r} - \vec{R}| , \tag{41}$$

$|k\rangle$ say, then

$$E_e^0 = \left\langle k \middle| \sum_{\vec{w}} \frac{|V_w|^2}{E_{ex}} (e^{i\vec{w}\cdot(\vec{r}-\vec{R})} + \text{c. c.}) \middle| k \right\rangle . \tag{42}$$

We note that this E_e^0 is the exact exchange energy in second-order perturbation theory taking

$$H_e - V(|\vec{r} - \vec{R}|) + \sum\nolimits_{\vec{w}} b_{\vec{w}}^\dagger b_{\vec{w}} E_{ex} \equiv \mathcal{H}_e + \sum\nolimits_{\vec{w}} b_{\vec{w}}^\dagger b_{\vec{w}} E_{ex}$$

in Eq. (1) as the unperturbed Hamiltonian and the interactions of the electronic polarization field with the electron and hole as perturbations. Rewriting the above E_e^0 gives

$$\begin{aligned} E_e^0 &= \frac{1}{2}\left\langle k \middle| \sum_{\vec{w}} \frac{|V_w|^2}{E_{ex}} (e^{i\vec{w}\cdot(\vec{r}-\vec{R})} + \text{c. c.}) \middle| k \right\rangle \\ &\quad + \frac{1}{2}\left\langle k \middle| \sum_{\vec{w}} |V_w|^2 \left(e^{i\vec{w}\cdot\vec{r}} \frac{1}{E_{ex} + \mathcal{H}_e - \epsilon_k} e^{-i\vec{w}\cdot\vec{R}} + e^{i\vec{w}\cdot\vec{R}} \frac{1}{E_{ex} + \mathcal{H}_e - \epsilon_k} e^{-i\vec{w}\cdot\vec{r}} \right) \middle| k \right\rangle \\ &= \frac{1}{2}\sum_{\vec{w}} |V_w|^2 \Bigg[\left\langle k \middle| \frac{1}{E_{ex}} (e^{i\vec{w}\cdot(\vec{r}-\vec{R})} + \text{c. c.}) \middle| k \right\rangle \\ &\quad + \left\langle k \middle| \left(e^{i\vec{w}\cdot\vec{r}} \frac{1}{E_{ex} + \mathcal{H}_e - \epsilon_k} e^{-i\vec{w}\cdot\vec{r}} e^{i\vec{w}\cdot(\vec{r}-\vec{R})} + e^{-i\vec{w}\cdot(\vec{r}-\vec{R})} e^{+i\vec{w}\cdot\vec{r}} \frac{1}{E_{ex} + \mathcal{H}_e - \epsilon_k} e^{-i\vec{w}\cdot\vec{r}} \right) \middle| k \right\rangle \Bigg] , \end{aligned} \tag{43}$$

where we have used the fact that $\vec{R}$ is a constant and the relation $(E_{ex} + \mathcal{H}_e - \epsilon_k)^{-1} |k\rangle = E_{ex}^{-1} |k\rangle$, ϵ_k being the eigenenergy of $\mathcal{H}_e$. Following the method used in Sec. II to get an expansion for the second-order-energy correction, we obtain

$$E_e^0 = \left\langle k \middle| \left\{ \frac{1}{2} \sum_{\vec{w}} \frac{|V_w|^2}{E_{ex}} (e^{iw\cdot(\vec{r}-\vec{R})} + \text{c. c.}) + \frac{1}{2} \sum_{\vec{w}} \frac{|V_w|^2}{E_{ex} + \hbar^2 w^2/2m_b} (e^{i\vec{w}\cdot(\vec{r}-\vec{R})} + \text{c. c.}) \right. \right.$$

$$+\frac{1}{2}\sum_{\vec{w}}\frac{|V_w|^2}{(E_{ex}+\hbar^2w^2/2m_b)^2}\left(\frac{\hbar\vec{w}\cdot\vec{p}}{m_b}\,e^{i\vec{w}\cdot(\vec{r}-\vec{R})}+e^{-i\vec{w}\cdot(\vec{r}-\vec{R})}\,\frac{\hbar\vec{w}\cdot\vec{p}}{m_b}\right)$$

$$+\frac{1}{2}\sum_{\vec{w}}\frac{|V_w|^2}{(E_{ex}+\hbar^2w^2/2m_b)^3}\left[\left(\frac{\hbar\vec{w}\cdot\vec{p}}{m_b}\right)^2 e^{i\vec{w}\cdot(\vec{r}-\vec{R})}+e^{-i\vec{w}\cdot(\vec{r}-\vec{R})}\left(\frac{\hbar\vec{w}\cdot\vec{p}}{m_b}\right)^2\right]+\cdots\Bigg\}\,\Big|k\Big\rangle\,. \tag{44}$$

According to the HS theory, the effective Hamiltonian for the system of an electron localized around a massive hole is that for a Wannier exciton with the hole mass equal to infinity, i.e.,

$$H_{\rm HS}=\frac{p^2}{2m_b}-\frac{e^2}{|\vec{r}-\vec{R}|}+\frac{1}{2}\sum_{\vec{w}}\frac{|V_w|^2}{E_{ex}}\left(e^{i\vec{w}\cdot(\vec{r}-\vec{R})}+\text{c. c.}\right)+\frac{1}{2}\sum_{\vec{w}}\frac{|V_w|^2}{E_{ex}+\hbar^2w^2/2m_b}\left(e^{i\vec{w}\cdot(\vec{r}-\vec{R})}+\text{c. c.}\right)-\alpha_b\,E_{ex}-\sum_{\vec{w}}\frac{|V_w|^2}{E_{ex}}\,, \tag{45}$$

where m_b and α_b are, respectively, the polaron mass and the coupling constant in the HS theory, $-\alpha_b\,E_{ex}$ is the self-energy of the electron—a constant—and the last term is the self-energy of the massive hole. The sum of third and fourth terms in Eq. (45) is the exchange interaction in the HS work. This interaction is the same as the sum of the first two terms in the brackets $\{\ \}$ in Eq. (44). Consequently, the HS exchange interaction excludes terms such as third, fourth, and higher terms in Eq. (44). We anticipate from the remarks on the use of perturbation theory in Sec. II B that these terms are important for states whose binding energies are not very small compared to a single excitation energy (i.e., the exciton energy for the case of electronic polarizations and the phonon energy for the case of ionic polarization).

IV. SUMMARY

Within the polaron theory, an exact expression for the exchange interaction (i.e., the dielectric screening interaction) between an electron and a massive hole via a polarizable field was obtained for arbitrary electron-hole separation. The form of this interaction remains unchanged when the electron mass is also effectively infinite. Comparison of the obtained exchange interaction with that of Haken and Schottky showed that their exchange interaction is reliable only for the case of their interest, i.e., for very large electron-hole separations.

It has been shown that in the case of $\alpha_{3b}\simeq\alpha_{4b}$ (this is true for a bound lattice polaron in any polar crystal) the Lamb-shift-type correction due to a crystalline polarization field is opposite in sign to that due to the photon field in the hydrogen atom. This correction is small as compared with optical absorption and emission energies due to point defects in insulators.

It was also shown in the nonband-mass theory that the effect of the electronic polarization on the electronic mass and thereby on the binding energy is small.

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