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MASSES AND OSCILLATOR STRENGTHS OF WANNIER EXCITONS IN POLAR SEMICONDUCTORS

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Polaron effects on total masses and oscillator strengths of Wannier excitons are discussed by the variational calculation. The dependence of these effects on the internal motion of the exciton is shown to be quite important.

I. Introduction

In polar semiconductors the Wannier exciton interacts strongly with the longitudinal optical (LO) phonon and then its optical properties may deviate much from those of the simple hydrogenic model [1,2]. In the present work, using a simple two band model and the Fröhlich electron-phonon interaction, we show that polaron effects on exciton masses and oscillator strengths of the wannier exciton depend strongly on the internal (relative) motion and the nature of the exciton-phonon interaction is quite different between the ground(1s) state and excited states.

II. Calculation and Discussion

We consider an s-type Wannier exciton interacting with the LO phonon. The variational method of [1-3], which may incorporate the state dependency of the exciton-LO phonon interaction well, is used to treat polaron effects on the exciton. We set

$$\zeta_i \vec{Q} = \sum_{\vec{k}} |v_{\vec{k}}|^2 \vec{k}\{(f^i_{\vec{k}})^2 - f^i_{\vec{k}} f^j_{\vec{k}} \langle\psi_n|\cos(\vec{k}\cdot\vec{r})|\psi_n\rangle\},\quad i\neq j=1,2\ , \tag{1}$$

where $f^i_{\vec{k}}$'s, which appear as variational parameters in the unitary transformation in the calculation and physically play a role as governing the amount of the shift of the phonon coordinate by the interaction with the exciton, have been determined variationally by the minimamization of the energy. $\hbar\omega v_{\vec{k}}$ ($\hbar\omega$, being the LO phonon energy) is the electron-phonon coupling constant and ψ_n is the wave function of the internal state n. For small $\vec{Q}$, after solving eq.(1) for ζ_i, the energy of an exciton with the total momentum $\vec{Q}$ and the internal state n is given by

$$E_{\vec{Q},n} = E_n(\vec{Q}=0) + \hbar^2 Q^2/2M^*_n. \tag{2}$$

The energy $E_n(\vec{Q}=0)$ is the energy of an internal exciton state n, involved in the optical absorption. In general the exciton mass M^*_n depends on the internal state n and is neither equal to the bare exciton mass $M=m_1+m_2$, (where m_1 and m_2 are the electron and hole masses, respectively) nor to the fully renormalized exciton polaron mass $M^*_p = m^*_1+m^*_2$, (where m^*_i is the polaron mass of an electron (i=1) or a hole (i=2)). The oscillator strengths of the zero phonon line and one phonon side band of an exciton state n in the allowed case of the optical absorption are given by

$$f_n^{(0)} = A|\psi_n(0)|^2 \exp(-g_n) \quad \text{and} \quad f_n^{(1)} = g_n f_n^{(0)}, \tag{3}$$

respectively. Here A is a constant and $\psi_n(0)$ is the amplitude of the wave function of the internal state at the origin. The quantity g_n, which describes the transfer of the oscillator strength from the zero phonon line to phonon side bands, is given by $g_n = \Sigma_{\vec{K}} |v_{\vec{K}}|^2 \,|f^1_{\vec{K}}(\vec{Q}=0) - f^2_{\vec{K}}(\vec{Q}=0)|^2$ and is sensitive to the difference of the electron and hole masses as appearing in $g_n=0$ for the equal masses. In order to characterize the present exciton-LO phonon system, it is convenient to use the following four physical parameters; the reduced mass μ/m (m being the free electron mass), the electron-hole mass ratio $\sigma=m_1/m_2$, the strength of the coulomb binding $R_y^\infty/\hbar\omega$ $(=\mu e^4/2\varepsilon_\infty^2\hbar^3\omega$, where ε_∞ is the high frequency dielectric constant) and the Fröhlich electron-phonon coupling constant α. These physical parameters are changed in the physically important region of $0.2 \leq \mu/m \leq 0.5$, $0.1 \leq \sigma \leq 1$, $2.5 \leq R_y^\infty/\hbar\omega \leq 10$, and $0 \leq \alpha \leq 4$ to see general properties of exciton masses and oscillator strengths. Typical results for these properties are shown and discussed in the following.

In Fig.1 the exciton mass M_n^* for 1s and 2s states is shown as a function of α. Values of M_n^* deviate from those of the bare exciton mass $M=m_1+m_2$ (values at $\alpha=0$ in Fig.1), when α becomes large. However it is clear that the exciton mass for 1s state M_{1s}^* receives much less polaron effects than that for 2s state M_{2s}^*. In reality values of M_{2s}^* are very close to those of the fully renormalized exciton polaron mass $M_p^*=m_1^*+m_2^*$, where, in the present case, the polaron mass of an electron or a hole is given by $m_i^* = m_i(1+\alpha_i/6)$ $(\alpha_i=\alpha(m_i/m)^{1/2})$. This is also true for 3s state and thus exciton masses for excited states are considered to be nearly equal to the fully renormalized exciton polaron mass M_p^*. On the other hand the polaron effects on the exciton mass M_{1s}^* are rather small and M_{1s}^* may be closer to the

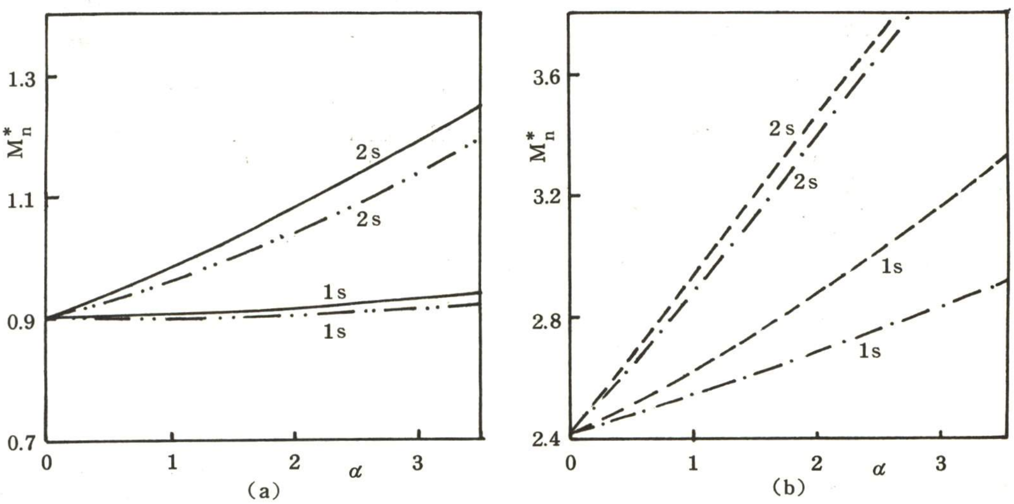

Fig.1 Exciton mass M_n^* (in units of the free electron mass) vs α; $\mu/m=0.2$ (a) $\sigma=0.5$, $R_y^\infty/\hbar\omega=5$ (solid) and 10 (broken with two dots) and (b) $\sigma=0.1$, $R_y^\infty/\hbar\omega=5$ (broken) and 10 (broken with a dot)

bare exciton mass, as in the previous works of the perturbational calculation [4,5] and the variational calculation [3]. As seen in Fig.1 differences of M^*_{1s} and M^*_{2s} are about 10% even for $\alpha \approx 1$ and may be larger than 20% for $\alpha > 2$. Therefore, we think, the differences of the exciton masses for the ground and excited states should be observed for some materials in experiments such as the resonant Brillouin scattering and the resonant Raman scattering.

Next we discuss the oscillator strengths of an s-type Wannier exciton in the optical absorption. In Fig.2 the oscillator strength ratios of the zero phonon lines, $f^{(0)}_n / f^{(0)}_{1s}$, are shown as a function of α. It is seen that these ratios may be considerably reduced compared to the hydrogenic values (values at α=0 in Fig.2) when α becomes large. There are two sources for the reduction as expected from eq.(3); (i) the large extension of the wave functions for excited states compared to that for the ground state and (ii) the larger transfer of the oscillator strengths from zero phonon lines to phonon side bands for excited states than for the ground state. The former is the main reason for the reduction of the oscillator strength ratio for $\sigma > 0.5$. However, the latter also has a large effect for $\sigma \lesssim 0.4$, i.e., for the large difference of the electron and hole masses. To see this point, values of g_n as a function of α are plotted in Fig.3. Figure 3 indicates that for the large difference of the electron and hole masses g_n for excited state may become an order of unity when the electron-phonon coupling α becomes large such as $\alpha \gtrsim 3$. This means, as seen in eq.(3), that for excited states the one-phonon side band may be as strong as the zero phonon line and the large transfer of the oscillator strength from the zero phonon line to the phonon side bands may occur. On the other hand the corresponding quantity for the ground state, g_{1s}, is much smaller than g_n's for excited states and the large transfer of the oscillator strength as for excited states does not occur. Then the oscillator strength ratio $f^{(0)}_n / f^{(0)}_{1s}$ may

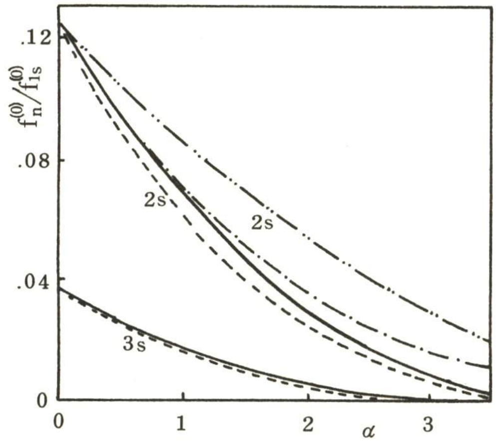

Fig.2 Ratio of the oscillator strengths of zero phonon lines $f^{(0)}_n / f^{(0)}_{1s}$ vs α; μ/m=0.2, $R^{\infty}_y/\hbar\omega$=5 and σ=0.5 (solid) and 0.1 (broken); μ/m=0.2, $R^{\infty}_y/\hbar\omega$=10 and σ=0.5 (broken with two dots) and 0.1 (broken with a dot)

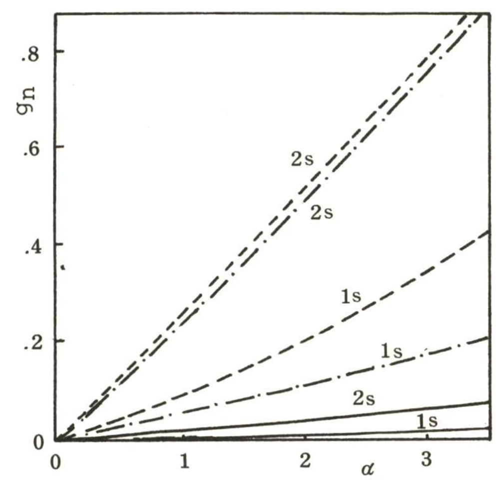

Fig.3 $g_n = \Sigma_{\vec{k}} |v_{\vec{k}}|^2 (f^1_{\vec{k}} - f^2_{\vec{k}})^2$ vs α; μ/m = 0.2, $R^{\infty}_y/\hbar\omega$=5 and σ=0.5 (solid) and 0.1 (broken); μ/m=0.2, $R^{\infty}_y/\hbar\omega$=10 and σ=0.1 (broken with a dot)

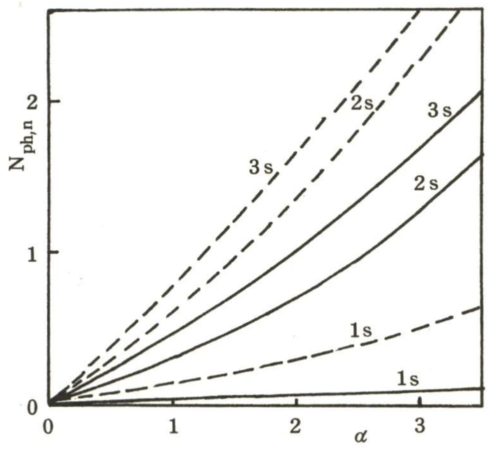

Fig.4 Average virtual-phonon number $N_{ph,n}$ vs α; $\mu/m=0.2$, $R_y^\infty/\hbar\omega=5$, and $\sigma=0.5$ (solid) and 0.1 (broken)

be considerably reduced by the transfer of the oscillator strengths of zero phonon lines to phonon side bands for small σ such as $\sigma \leq 0.4$.

All the above results indicate that polaron effects for excited states are much stronger than those for the ground state. In order to see this point more clearly, in Fig.4 the virtual phonon number involved in the state n, $N_{ph,n} = \langle\Psi_{n,0}|\Sigma_{\vec{k}} a^+_{\vec{k}} a_{\vec{k}}|\Psi_{n,0}\rangle$, (where $\Psi_{n,0}$ is the zero phonon exciton wave function and $a^+_{\vec{k}}(a_{\vec{k}})$ denotes the creation (annihilation) operator of the phonon with the wave vector $\vec{k}$) is plotted as a function of α. It is seen that the virtual phonon number for the ground state is smaller by one order of magnitude than those for excited states and thus the exciton-phonon coupling for the ground state is much weaker than for excited states. This result may be understood as a consequence of the exciton consisting of two particles with the opposite charges, i.e., an electron and a hole. In the physically important region the exciton radius for the ground state is smaller than the sum of the electron and hole polaron radii, $R_1 + R_2$. Therefore the cancellation of the phonon clouds around an electron and a hole occurs to a large extent and thus the exciton-phonon coupling for the ground state is not strong even for large α such as $\alpha \approx 3$-4. On the other hand for excited states the exciton radii are much larger than $R_1 + R_2$ and the cancellation of the phonon clouds is very small. Then the exciton-phonon coupling for excited states is strong when α becomes large. This difference of the strengths of the exciton-phonon interaction, arising from the difference of the exciton radii, causes the strong dependence of the exciton mass on the internal motion and the large reduction of the oscillator strength ratios of the zero phonon lines $f_n^{(0)}/f_{1s}^{(0)}$ as seen above.

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