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CONCERNING THE THEORY OF THE EXCITON STATE IN SEMICONDUCTORS

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Exciton states are treated by the Green's function method. The spectra of Frenkel and Mott excitons are determined for an arbitrary temperature.

INTRODUCTION

THE investigation of exciton states in semiconductors can provide certain information concerning the band spectrum of electrons. The presence of exciton states to some degree provides a measure of the deviation of the energy spectrum of a system of electrons from the spectrum described by the band approximation, which in the majority of cases is sufficient to explain the available experimental data. The existence of excitons is related to correlation effects in a system of electrons which are located in the crystal lattice.

There are many experimental works confirming the exciton structure of the electron energy spectra near the bottom of the conduction band.[1-9] The majority of them are concerned with investigations of polar crystals, and only two investigations[8, 9] relate to the atomic semiconductor Ge. In the latter case the exciton level lies very close to the bottom of the conduction band and is thus quite difficult to detect experimentally. Investigations of the internal[10, 11] and external[12, 13] photoeffects in semiconductors also point to the existence of excitons. Of the theoretical works, which have appeared in recent years, we may mention references 14–19. There is considerable interest in works concerning diffusion of excitons,[20-22] and concerning their contribution to the dielectric constant[23-25] and to the coefficient of heat conduction, etc.

This wide variety of questions concerning excitons might usefully be considered from a single viewpoint. We wish to show that this is apparently possible by using the Green's function method. The superiority of this method over the others usually employed is that the treatment is carried out for an arbitrary temperature from the very outset. On the other hand the exciton energy spectrum (including the problem of damping), the absorption of light, the dielectric constant, the diffusion coefficient, and the thermal conductivity can be obtained by investigating the two-particle Green's function.[26]

In the present article we consider only the problem of excitons in an undeformed lattice.

EXCITON STATES IN AN UNDEFORMED LATTICE

Let us consider a system of valence electrons placed in the crystal lattice. The lattice sites are assumed stationary. In the general case the system of valence electrons is described by the Hamiltonian[27]

$$H = \sum_{f_1 f_2} L(f_1 f_2)\, a_{f_1}^{+} a_{f_2}^{+} + \frac{1}{2} \sum_{f_1 f_2 f_3 f_4} F(f_1 f_2 f_4 f_3)\, a_{f_1}^{+} a_{f_2}^{+} a_{f_3} a_{f_4}, \qquad (1)$$

which takes into account the interaction between the electrons and the lattice site and between the electrons themselves. The index f defines the coordinate of the site and the state of the electron at this site. In particular, if the electron state is described by means of the Wannier function, then f defines the number of the site and of the band.

The band spectrum of the electrons can be obtained from the Hamiltonian (1) in the self-consistent field approximation. The problem is to calculate the correlation effects in the system of electrons; these effects have a considerable influence on the character of the energy spectrum close to the bottom of the conduction band. We shall investigate these correlation effects by making use of the two-particle (and double-time) retarded and advanced Green's functions determined by Bogolyubov and Tyablikov[28] (for convenience these functions are multiplied by the factor −i, cf. reference 26):

$$\langle\langle a_{f'}^{+}(\tau)\, a_{f''}(\tau) | a_{g'}^{+}(\tau')\, a_{g''}(\tau') \rangle\rangle^{ret}$$

$$= -i\theta(\tau-\tau') \langle [a_{f'}^{+}(\tau)\, a_{f''}(\tau),$$

$$a_{g'}^{+}(\tau') a_{g''}(\tau')] \rangle, \qquad (2)$$

$$\langle\langle a^+_{f'}(\tau)\, a_{f''}(\tau) | a^+_{g'}(\tau')\, a_{g''}(\tau')\rangle\rangle^{adv}$$
$$= i\theta(\tau'-\tau)\,\langle[a^+_{f'}(\tau)\, a_{f''}(\tau),\ a^+_{g'}(\tau')\, a_{g''}(\tau')]\rangle, \quad (3)$$

where we denote

$$\langle\ldots\rangle = Q^{-1}\,\mathrm{Sp}\,(\ldots e^{-H/\theta}), \qquad Q = \mathrm{Sp}\, e^{-H/\theta}.$$

The Fourier transforms of the functions (2) and (3) can be analytically continued in the upper and lower half-planes respectively and are to all intents and purposes a single analytic function throughout the whole of the E plane, which has a linear cut along the real axis.[28]

This situation enables us to determine the double-time correlation function $\langle a^+_{f'}(\tau)\, a_{f''}(\tau)\, a^+_{g'}(\tau')\, a_{g''}(\tau')\rangle$, by means of which we calculate the majority of the physical quantities relating to the electron system.[26]

The equations for the Fourier transforms of (2) and (3) coincide. They are easily obtained by using the equation of motion for the operators in the Heisenberg representation. By simple calculations we obtain

$$\begin{aligned}
E\langle\langle a^+_{f'}a_{f''} | a^+_{g'}a_{g''}\rangle\rangle &= \langle a^+_{g'}a_{f''}\rangle\,\delta_{g''f'} - \langle a^+_{f'}a_{g''}\rangle\,\delta_{g'f''}\\
&+ \sum_f L_{eff}(ff')\langle\langle a^+_f a_{f''} | a^+_{g'}a_{g''}\rangle\rangle - \sum_f L_{eff}(f''f)\langle\langle a^+_{f'}a_f | a^+_{g'}a_{g''}\rangle\rangle\\
&+ \sum_{f_1f_2f} (F(ff_1f'f_2) - F(ff_1f_2f'))\,\langle a^+_f a_{f''}\rangle\langle\langle a^+_{f_1}a_{f_2} | a^+_{g'}a_{g''}\rangle\rangle\\
&- \sum_{f_1f_2f} (F(f''f_1ff_2) - F(f''f_1f_2f))\langle a^+_{f'}a_f\rangle\langle\langle a^+_{f_1}a_{f_2} | a^+_{g'}a_{g''}\rangle\rangle;
\end{aligned}$$

$$L_{\mathbf{eff}}(f_1f_2) = L(f_1f_2) + \sum_{f'f''}\{F(f_1f'f_2f'') - F(f_1f'f''f_2)\}\,\langle a^+_{f'}a_{f''}\rangle. \quad (4)$$

In the derivation of this equation we used the following approximation to the three-particle Green's function:

$$\begin{aligned}
\langle\langle a^+_{f_1}a^+_{f_2}a_{f_3}a_{f_4} | a^+_{g'}a_{g''}\rangle\rangle &= \langle a^+_{f_2}a_{f_3}\rangle\langle\langle a^+_{f_1}a_{f_4} | a^+_{g'}a_{g''}\rangle\rangle\\
&- \langle a^+_{f_2}a_{f_4}\rangle\langle\langle a^+_{f_1}a_{f_3} | a^+_{g'}a_{g''}\rangle\rangle + \langle a^+_{f_1}a_{f_4}\rangle\langle\langle a^+_{f_2}a_{f_3} | a^+_{g'}a_{g''}\rangle\rangle\\
&- \langle a^+_{f_1}a_{f_3}\rangle\langle\langle a^+_{f_2}a_{f_4} | a^+_{g'}a_{g''}\rangle\rangle. \quad (5)
\end{aligned}$$

Physically this approximation implies allowance for the interaction of the excited electron with the hole, which is formed in the background of filled single-electron states.

Equation (4) is easily solved by using the following subsidiary problem:

$$\begin{aligned}
E_\gamma u_\gamma(f'f'') &= \sum_f L_{\mathbf{eff}}(ff')\, u_\gamma(ff'') - \sum_f L_{eff}(f''f)\, u_\gamma(f'f)\\
&+ \sum_{f_1f_2f}\{(F(ff_1f'f_2) - F(ff_1f_2f'))\,\langle a_f^+a_{f''}\rangle\\
&-(F(f''f_1ff_2) - F(f''f_1f_2f))\,\langle a^+_{f'}a_f\rangle\}\, u_\gamma(f_1f_2), \quad (6)
\end{aligned}$$

with the normalizing condition

$$\sum_{f'f''} u_\gamma(f'f'')\, u^*_{\gamma'}(f'f'') = \delta_{\gamma\gamma'}.$$

The solution of (4) will have the form

$$\langle\langle a^+_{f'}a_{f''} | a^+_{g'}a_{g''}\rangle\rangle = \sum_{\alpha\alpha'} (E - E_\alpha)^{-1}\, c_{\alpha\alpha'} u_\alpha(f'f'')\, u^*_{\alpha'}(g'g''),$$

$$c_{\alpha\alpha'} = \sum_{g_1g_2f_1f_2}\{\langle \overset{+}{a}_{g_1}a_{f_2}\rangle\,\delta_{g_2f_1} - \langle\overset{+}{a}_{f_1}a_{g_2}\rangle\,\delta_{g_1f_2}\}\, u^*_\alpha(f_1f_2)\, u_{\alpha'}(g_1g_2). \quad (7)$$

From the result obtained we conclude that in the electron system there are possible collective oscillations (called excitons), the spectra of which are determined by the eigenvalues of (6). This deduction is based on the fact that the Green's function (7) has a pole on the real axis. We note that the eigenvalues of (6) depend on the temperature through the mean value $\langle a^+_{f_1} a_{f_2}\rangle$

Let us consider two concrete cases.

Frenkel Exciton. Let us suppose that the excited electron remains at that site, at which it existed in the ground state. Conditionally we will define: 0**g**-ground state of the electron at the site **g**; 1**g** excited state of the electron at the same site **g**. For simplicity we neglect the degeneracy of the excited state. In this case Eq. (6) takes the form

$$\begin{aligned}
E_\gamma u_\gamma(1g,\ 0g) &= (L_{\mathbf{eff}}(1g,\ 1g) - L_{eff}(0g,\ 0g))\, u_\gamma(1g,\ 0g)\\
&+ \sum_{g',\, g'',\, \nu=0,\,1}\{[F(\nu g'',\ 1g';\ 1g,\ 0g')\\
&- F(\nu g'',\ 1g';\ 0g',\ 1g)]\,\langle a^+_{\nu g''}a_{0g}\rangle - [F(0g,\ 1g';\ \nu g'',\ 0g')\\
&- F(0g,\ 1g';\ 0g',\ \nu g'')]\,\langle a^+_{1g}a_{\nu g''}\rangle\}\, u_\gamma(1g',\ 0g')\\
&+ \sum_{g',\, g'',\, \nu=0,\,1}\{[F(\nu g'',\ 0g';\ 1g,\ 1g')\\
&- F(\nu g'',\ 0g';\ 1g',\ 1g)]\,\langle a^+_{\nu g''}a_{0g}\rangle - [F(0g,\ 0g';\ \nu g'',\ 1g')\\
&- F(0g,\ 0g';\ 1g';\ \nu g'')]\,\langle a^+_{1g}a_{\nu g''}\rangle\}\, u_\gamma(0g',\ 1g'). \quad (8)
\end{aligned}$$

By analogy we can also write the equation for $u_\gamma(0\mathbf{g},\ 1\mathbf{g})$. From the translational invariance of the crystal lattice we find that $u_\gamma(1\mathbf{g}, 0\mathbf{g})$ and $u_\gamma(0\mathbf{g}, 1\mathbf{g})$ are proportional to exp (ikg), where **k** is the quasi-momentum vector. Equation (8) and the analogous equation for $u_\gamma(0\mathbf{g}, 1\mathbf{g})$ are easily solved. Retaining only the two-center integrals and neglecting the exchange integrals, we obtain the energy spectrum of the exciton

$$E_\gamma \equiv E(\mathbf{k}) = \{\,|\Delta E + \widetilde{F}_1(\mathbf{k})|^2 - |\widetilde{F}_2(\mathbf{k})|^2\}^{1/2}, \quad (9)$$

where $\Delta E = L_{\mathrm{eff}}(1\mathbf{g}, 1\mathbf{g}) - L_{\mathrm{eff}}(0\mathbf{g}, 0\mathbf{g})$ is the width of the gap, while $\widetilde{F}_1(\mathbf{k})$ and $\widetilde{F}_2(\mathbf{k})$ are respectively the Fourier transforms of the functions

$$F_1(|\mathbf{g}-\mathbf{g}'|) = (n_0 - n_1)\,F(1\mathbf{g},\ 0\mathbf{g}';\ 0\mathbf{g},\ 1\mathbf{g}'),$$

$$F_2(|\mathbf{g}-\mathbf{g}'|) = (n_0 - n_1)\,F(1\mathbf{g},\ 1\mathbf{g}';\ 0\mathbf{g},\ 0\mathbf{g}'),$$

$$n_0 = \langle \overset{+}{a}_{0\mathbf{g}} a_{0\mathbf{g}} \rangle, \qquad n_1 = \langle \overset{+}{a}_{1\mathbf{g}} a_{1\mathbf{g}} \rangle.$$

The integrals $F_1(|\mathbf{g}-\mathbf{g}'|)$ and $F_2(|\mathbf{g}-\mathbf{g}'|)$ have the character of a Coulomb dipole interaction.[29] The difference between the spectrum (9) and that obtained by Heller and Marcus[29] by a variational method lies in the presence of the term $F_2(\mathbf{k})$ in (9). We note that this difference is insignificant for small k, i.e., when

$$|\tilde{F}_1(\mathbf{k})| < \Delta E, \qquad |\tilde{F}_2(\mathbf{k})| < \Delta E,$$

for in this case $E(\mathbf{k}) = \Delta E + \tilde{F}_1(\mathbf{k})$.

Mott Exciton. Let an electron, located at the site **g** in state 0, be transferred by excitation to site **h** in state 1 (for example in the S state, as in Cu_2O). In this case Eq. (5) is rewritten

$$\begin{aligned}
E_\gamma u_\gamma(1\mathbf{h},\ 0\mathbf{g}) &= \sum_{\mathbf{h}'} L_{eff}(1\mathbf{h}',\ 1\mathbf{h})\,u_\gamma(1\mathbf{h}',\ 0\mathbf{g}) \\
&- \sum_{\mathbf{g}'} L_{eff}(0\mathbf{g},\ 0\mathbf{g}')\,u_\gamma(1\mathbf{h},\ 0\mathbf{g}') \\
&+ \sum_{\mathbf{g}',\,\mathbf{h}',\,\sigma=0\mathbf{g}'',\,1\mathbf{h}''} \{[F(\sigma,\ 1\mathbf{h}';\ 1\mathbf{h},\ 0\mathbf{g}') \\
&- F(\sigma,\ 1\mathbf{h}';\ 0\mathbf{g}',\ 1\mathbf{h})]\,\langle \overset{+}{a}_\sigma a_{0\mathbf{g}} \rangle - [F(0\mathbf{g},\ 1\mathbf{h}';\ \sigma,\ 0\mathbf{g}') \\
&- F(0\mathbf{g},\ 1\mathbf{h}';\ 0\mathbf{g}',\ \sigma)]\,\langle \overset{+}{a}_{1\mathbf{h}} a_\sigma \rangle\}\,u_\gamma(1\mathbf{h}',\ 0\mathbf{g}') \\
&+ \sum_{\mathbf{g}',\,\mathbf{h}',\,\sigma=0\mathbf{g}'',\,1\mathbf{h}''} \{[F(\sigma,\ 0\mathbf{g}',\ 1\mathbf{h},\ 1\mathbf{h}') \\
&- F(\sigma,\ 0\mathbf{g}';\ 1\mathbf{h}',\ 1\mathbf{h})]\langle \overset{+}{a}_\sigma a_{0\mathbf{g}} \rangle - [F(0\mathbf{g},\ 0\mathbf{g}';\ \sigma,\ 1\mathbf{h}') \\
&- F(0\mathbf{g},\ 0\mathbf{g}';\ 1\mathbf{h}',\ \sigma)]\langle \overset{+}{a}_{1\mathbf{h}} a_\sigma \rangle\}\,u_\gamma(0\mathbf{g}',\ 1\mathbf{h}'). \qquad (10)
\end{aligned}$$

An analogous equation can be written for $u_\gamma(0\mathbf{g},\ 1\mathbf{h})$.

Retaining only the two-center integrals, we rewrite (10) in the form

$$\begin{aligned}
E_\gamma u_\gamma(1\mathbf{h},\ 0\mathbf{g}) &= \sum_{\mathbf{h}'} L_{eff}(1\mathbf{h}',\ 1\mathbf{h})\,u\ (1\mathbf{h}',\ 0\mathbf{g}) \\
&- \sum_{\mathbf{g}'} L_{eff}(0\mathbf{g},\ 0\mathbf{g}')\,u_\gamma(1\mathbf{h},\ 0\mathbf{g}') + [F(0\mathbf{g},\ 1\mathbf{h};\ 1\mathbf{h},\ 0\mathbf{g}) \\
&- F(0\mathbf{g},\ 1\mathbf{h};\ 0\mathbf{g},\ 1\mathbf{h})]\,(n_0 - n_1)\,u_\gamma(1\mathbf{h},\ 0\mathbf{g}), \qquad (11)
\end{aligned}$$

where

$$n_0 = \langle \overset{+}{a}_{0\mathbf{g}} a_{0\mathbf{g}} \rangle, \qquad n_1 = \langle \overset{+}{a}_{1\mathbf{h}} a_{1\mathbf{h}} \rangle.$$

In such an approximation the equations for the functions $u_\gamma(1\mathbf{h},\ 0\mathbf{g})$ and $u_\gamma(0\mathbf{g},\ 1\mathbf{h})$ are not coupled to each other. For excitons of large radii the difference equation (11) can be replaced by a differential equation. This can be done in the following way. We first separate the motion of the center of gravity, putting

$$u_\gamma(1\mathbf{h},\ 0\mathbf{g}) = \exp\{i\mathbf{k}(\alpha\mathbf{g} + \beta\mathbf{h}\}\,\overline{u_\gamma}(\mathbf{g}-\mathbf{h}),$$

where **k** is the exciton momentum. In the momentum representation we obtain for the function $\bar{u}_\gamma$ the equation

$$\begin{aligned}
E_\gamma \overline{u_\gamma}(\mathbf{p}) &= (\varepsilon^{(1)}(\mathbf{p} - \beta\mathbf{k}) - \varepsilon^{(0)}(\mathbf{p} + \alpha\mathbf{k}))\,\overline{u_\gamma}(\mathbf{p}) \\
&- \sum_{\mathbf{p}'} \bar{V}(\mathbf{p} - \mathbf{p}')\,\overline{u_\gamma}(\mathbf{p}'), \qquad (12)
\end{aligned}$$

and the dispersion laws for an electron have the following form in the conduction and valence band respectively.

$$\varepsilon^{(1)}(\mathbf{p}) = \sum_{\mathbf{h}-\mathbf{h}'} L_{eff}(1\mathbf{h},\ 1\mathbf{h}')\,e^{-i\mathbf{p}(\mathbf{h}-\mathbf{h}')},$$

$$\varepsilon^{(0)}(\mathbf{p}) = \sum_{\mathbf{g}-\mathbf{g}'} L_{eff}(0\mathbf{g},\ 0\mathbf{g}')\,e^{-i\mathbf{p}(\mathbf{g}-\mathbf{g}')}.$$

In (12) we denote by $\bar{V}(\mathbf{p})$ the Fourier transform of the function

$$\begin{aligned}
V(\mathbf{g}-\mathbf{h}) &= (n_0 - n_1)\,[F(0\mathbf{g},\ 1\mathbf{h};\ 0\mathbf{g},\ 1\mathbf{h}) \\
&- F(0\mathbf{g},\ 1\mathbf{h};\ 1\mathbf{h},\ 0\mathbf{g})].
\end{aligned}$$

In the effective-mass approximation

$$\varepsilon^{(1)}(\mathbf{p}) = \Delta E + \mathbf{p}^2/2\mu_1, \qquad \varepsilon^{(0)}(\mathbf{p}) = -\mathbf{p}^2/2\mu_0,$$

where ΔE is the energy gap and μ_0, μ_1 are the effective masses.

Putting $\alpha = \mu_0/(\mu_0 + \mu_1)$ and $\beta = \mu_1/(\mu_0 + \mu_1)$, we write down Eq. (12) in the coordinate representation in the form

$$E_\gamma \overline{u_\gamma}(\mathbf{r}) = \left[\Delta E + \frac{\mathbf{k}^2}{2(\mu_0+\mu_1)} - \frac{\hbar^2}{2\mu}\Delta_{\mathbf{r}}\right]\overline{u_\gamma}(\mathbf{r}) - V(\mathbf{r})\,\overline{u_\gamma}(\mathbf{r}),$$

where μ is the reduced mass.

We thus arrive at an equation that defines the discrete structure of the exciton spectrum. The approximation adopted in going from Eq. (10) to Eq. (11), which consists in neglecting configurational integrals above the second order, means that the resonance mechanism of transferring the exciton excitation is disregarded. An account of these integrals alters the dependence of E_γ on **k**.[30] We shall remark only that a correct account of the resonance transfer of exciton excitation must be accomplished by using Eq. (10).

We note now that the exciton spectrum depends on the temperature factor $n_0 - n_1$, indicating a decrease of the degree of population of the ground level with increasing temperature. In the case of the Mott exciton the presence of this factor leads to a decrease of the effective Coulomb interaction between the electron and the hole. Furthermore, from a comparison of the results obtained here with those which Heller and Marcus[29] obtained by a variational method, it follows that for small **k** the use of the variational method is justified.

The approximation (5) does not permit the finding of the attenuation for the exciton level. To find that attenuation it would be necessary to consider more accurately the three-particle Green's function, which is a cumbersome matter. This is all the more so, because there is no attenuation at small **k**, for, as perturbation theory shows, the attenuation is due to the transition of the exciton to two electron-hole pairs, which is impossible when $E_{\gamma}(\mathbf{k}) < 2\Delta E$. For small **k** the attenuation is largely determined by the interaction of the excitons with the lattice vibrations and with impurity centers.

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