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A Stochastic Theory of Exciton Transfer

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Abstract

"Exciton" is one of the elementary excitations in solids. Especially the Frenkel exciton is considered to be an energy transfer from one atom to another: When one electron is excited at a certain lattice site in crystals, its excited energy can be transfered to the neighbouring atoms via interactions of atoms. The process of energy excitation can be dealt with a particle picture introducing the method of second quantization.

The concept of "exciton" was first proposed by Frenkel in 1931: He didn't consider the transfer of the energy excitation as a particle, rather he thought it to be a localized "excitation wave" in crystal.

In 1937 Wannier introduced another kind of exciton which is localized in crystal; an electron-hole pair similar to a hydrogen atom.

Since 1972 Haken and others have discussed several effects on the exciton motion due to lattice vibration. This is done with the use of a stochastic model:

$$H(t) = \sum_{mn} J_{mn} b_m^+ b_n + \sum_{mn} \Delta_{mn}(t) b_m^+ b_n, \tag{1}$$

where $\Delta_{mn}(t)$ is an overlap integral between wave functions of lattice sites m and n; the stochastic behaviour of $\Delta_{mn}(t)$ is due to the lattice vibrations and so on. Haken assumed that $\Delta_{mn}(t)$ is the white Gaussian process, and discussed several physical consequences derived thereby.

Because of the strong assumption imposed by the "whiteness" condition, several authors have made attempts to relax the condition: The coherent potential approximation is extended to dynamical systems. Namely, N. Ohata used the two-state-jump Markoff process whereas H. Sumi made calculations with the Gaussian Markoff process. They used the method of Green's function and discussed optical absorption. Y. Inaba calculated diffusion coefficient for the two-state-jump Markoff process from the equation of motion for the density matrix. In these treatments, they have only taken into account the diagonal randomness in $\Delta_{mn}(t)$.

In the first part of this thesis, we review the Frenkel's investigations on the exciton, and the various stochastic treatments on the exciton dynamics.

In the second part, we formulate the theory of optical absorption for excitons by means of the damping theoretical expansion formula. The method based on the damping theory is very simple and systematic, and moreover the method enables us to expect further developments of the exciton theory.

In our formulation we start from the Hamiltonian:

$$\begin{aligned} H(t) &= \sum_{<mn>} J(|\vec{R}_m - \vec{R}_n|) b_m^+ b_n + \sum_{mn} \Delta_{mn}(t) b_m^+ b_n \\ &= \sum_k \omega_k a_k^+ a_k + \sum_{mn} \Delta_{mn}(t) b_m^+ b_n, \end{aligned} \tag{2}$$

where a_k^+ and a_k represent the creation and annihilation operators of an exciton in the Bloch state and they are given by the Fourier transform of b_m^+ and b_m which are defined in the Wannier state. We treat the problem for the two different models of $\Delta_{mn}(t)$; one for the two-state-jump Markoff process and the other for the Gaussian Markoff process.

With the use of the damping theoretical formulae, we can find equations for averaged density matrix immediately. From the equations, we obtain moment equations for a_k from which we can determine response functions, power spectrum and so on, systematically.

Thus we can discuss various effects due to random perturbations on the exciton motion from the view point of "coherence v.s. incoherence".

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