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Electron-LO Phonon and Electron-Photon Interactions Analogy in Semiconductor Quantum Dots

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Abstract. We discuss the relationship between the two most important types of intersublevel coupling in the polar semiconductor based quantum dots (QDs), namely Frölich electron-longitudinal optical(LO) phonon coupling and electron-light coupling. We further parametrize the Frölich coupling, giving its quantitative description in terms of QD geometric and composition parameters.

1. Introduction

Electron-longitudinal optical phonon interaction was found to be the most important intrinsic interaction in the conduction band of self-assembled QDs based on polar semiconductors. Either considered as a weak interaction modelled by Fermi golden rule [1] or as a strong interaction leading to the formation of finite-lifetime polarons [2, 3], it is responsible for the decay of excited carriers from p-like first excited state to s-like ground state [4, 5]. Therefore, understanding this interaction and its relations to other relevant quantities is of great importance for understanding the physics of conduction band electrons in QDs. In the following sections we show that this coupling can be parametrized so to relate it to optical matrix elements, giving a clear relationship between these two types of coupling of the electronic degrees of freedom, and providing a

simple intuitive picture of the dependence of Frölich coupling on QD geometric and composition parameters.

2. Initial Considerations

Consider a simple case of one spinless electron in the conduction band of a QD. At low temperatures the electron predominantly occupies s-like ground state. The strongest resonance from that state via optical excitation is with the p-like first excited state [6, 7]. In this reduced space of only two relevant electronic levels an optical LO phonon mode \mathbf{k} couples electron wavefunctions via Hamiltonian matrix element $\mathcal{H}_{ab}^{ph}(\mathbf{k}) = K \langle \psi_a | \frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{k} | \psi_b \rangle$, where $K = \sqrt{\frac{e^2 \hbar \omega_{LO}}{2V} \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_{st}} \right)}$. This matrix element will be called Frölich coupling function (FCF) for a wavevector \mathbf{k} . The most important parameter describing this interaction in both weak coupling and strong coupling regime, for such a two-level system, is the integral over the mode space \mathbf{k} of the Frölich matrix element [3, 8], $C_{ab}^2 = \int d^3\mathbf{k} |\mathcal{H}_{ab}(\mathbf{k})|^2$, which will be denoted as Frölich coupling constant.

The electron-photon interaction is described within the dipole approximation. The optical matrix element coupling these two states in the dipole approximation is $\mathcal{R}_{ab} = \langle \psi_a | \mathbf{r} | \psi_b \rangle$.

3. The Relationship

Due to confinement, only LO-phonons with small wavevectors can significantly interact with electrons. By expanding the exponential term in FCF in Taylor series around $\mathbf{k} = 0$ one obtains

$$\begin{aligned} \mathcal{H}_{ab}^{\mathbf{k}} &= K \int d^3\mathbf{r} \psi^{(a)*} \frac{\cos \mathbf{k} \cdot \mathbf{r}}{k} \psi^{(b)} + iK \int d^3\mathbf{r} \psi^{(a)*} \frac{\sin \mathbf{k} \cdot \mathbf{r}}{k} \psi^{(b)} = \\ &= \frac{K}{k} \int d^3\mathbf{r} \psi^{(a)*} \psi^{(b)} + iK \mathbf{e}_{\mathbf{k}} \cdot \int d^3\mathbf{r} \psi^{(a)*} \mathbf{r} \psi^{(b)} - \\ &- \frac{k}{2} K \int d^3\mathbf{r} \psi^{(a)*} (\mathbf{e}_{\mathbf{k}} \cdot \mathbf{r})^2 \psi^{(b)} - i \frac{k^2}{6} K \int d^3\mathbf{r} \psi^{(a)*} (\mathbf{e}_{\mathbf{k}} \cdot \mathbf{r})^3 \psi^{(b)} + \dots, \end{aligned} \quad (1)$$

where the second equation represents the Taylor series of sine and cosine functions, and $\mathbf{e}_{\mathbf{k}} = \frac{\mathbf{k}}{k}$. In the case of well defined and opposite parity for $|\psi_a\rangle$ and $|\psi_b\rangle$ we have

$$\mathcal{H}_{ab}^{\mathbf{k}} = iK \left(\mathbf{e}_{\mathbf{k}} \cdot \mathcal{R}_{ab} - \frac{k^2}{6} \int d^3\mathbf{r} \psi^{(a)*} (\mathbf{e}_{\mathbf{k}} \cdot \mathbf{r})^3 \psi^{(b)} + \dots \right). \quad (2)$$

In the limit $k \rightarrow 0$ the expression (2) becomes:

$$\mathcal{H}_{ab}^{\mathbf{k}} = iK \mathbf{e}_{\mathbf{k}} \cdot \mathcal{R}_{ab}. \quad (3)$$

Hence, the maximum of the scalar field $\mathcal{H}_{ab}^{\mathbf{k}}$ is proportional to the intensity of the dipole coupling vector, and occurs at $k \rightarrow 0$.

FCF is not well defined at $k = 0$ where the vector \mathbf{k} does not have a defined direction, and therefore the expression $\mathbf{e}_{\mathbf{k}} \cdot \mathcal{R}_{ab}$ is not defined. Strongly anisotropic behaviour of the distribution $\mathcal{H}_{ab}^{\mathbf{k}}$ arises from the factor $\mathbf{e}_{\mathbf{k}} \cdot \mathcal{R}_{ab}$. For \mathbf{k} pointing in the direction of the dipole coupling vector, the FCF exhibits the weakest negative slope. This slope is increasing with increasing angle between \mathbf{k} and the FCF, and decays rapidly to zero when \mathbf{k} becomes almost perpendicular to the dipole coupling vector, and in the limit where \mathbf{k} is exactly perpendicular the FCF equals zero. All these features prove that the FCF has a p-orbital like shape. This was indeed expected, since the FCF is essentially the Fourier transform of an odd function.

Therefore, the Frölich coupling function has been parameterized by a bell-shaped distribution function, with the maximal value at $k \rightarrow 0$ proportional to the intensity of the dipole coupling

vector and multiplied by the $\frac{k_i}{k}$, where i denotes the direction of the dipole coupling vector. This model gives:

$$|\mathcal{H}_{ab}^k| = |\mathcal{R}_{ab}| K \frac{k_z}{\sqrt{k_x^2 + k_y^2 + k_z^2}} \mathcal{I}(\mathbf{k}), \quad (4)$$

where $\mathcal{I}(\mathbf{k})$ is the anisotropic distribution function with maximum $\mathcal{I}(0) = 1$.

The general shape of the distribution function $\mathcal{I}(\mathbf{k})$ reduced to two dimensions with HWHMs σ_x and σ_z is shown in Fig. 1. The maximal value is always $\mathcal{I}(\mathbf{k} = 0) = 1$. The exact lineshape

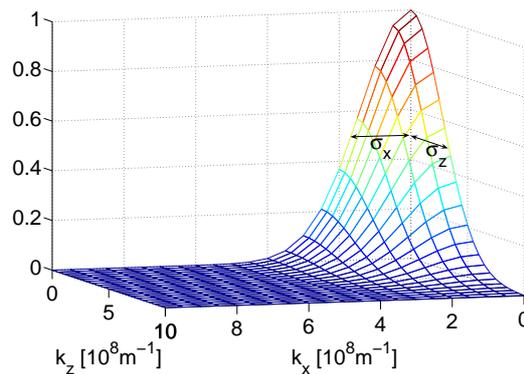


Figure 1. The distribution function for a parabolic QD. The figure is reduced to two dimensions (z and x) without losing generality. Linewidths σ_x and σ_z are inversely proportional to dot sizes in the corresponding directions.

depends on the particular QD and its geometric and composition parameters. A more extended dot in a particular direction has a narrower distribution function in that direction, i.e. σ_x and σ_z are decreasing functions of the dot sizes in x and z directions. This is clear from the fact that FCF is a Fourier transform of the product of the s-like and p-like wavefunction divided by k . It is well known that the transform of an expanded function shrinks, and vice versa. For the parabolic potential QD the distribution function is a Gaussian with HWHMs inversely proportional to the extensions of the parabolic confinement. By changing the QD material composition, i.e. increasing the dot depth, one increases the confinement of the bound wavefunctions. Therefore, for the same reason as above, the HWHM is an increasing function of the dot depth.

The FCF has been parametrized via two factors, namely the dipole coupling vector \mathcal{R}_{ab} , which is a well known spectroscopic quantity, and the distribution function with the property $\mathcal{I}(0) = 1$. Using this approximation Frölich coupling constant becomes:

$$C_{ab}^2 = \frac{e^2 \hbar \omega_{LO}}{16\pi^3} \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_{st}} \right) |\mathcal{R}_{ab}|^2 \int d^3 \mathbf{k} \frac{k_z^2}{k_x^2 + k_y^2 + k_z^2} \mathcal{I}^2(\mathbf{k}) \quad (5)$$

In calculating the Frölich coupling constant, the exact lineshape of the distribution function loses its significance due to the integration. The HWHM of the distribution function and the dipole coupling vector pre-factor remain as the main factors affecting the value of Frölich coupling constant. The integral in the last equation is affected directly by variations of QD geometric and composition parameters. Being the Fourier-transform-like image of the product of s-like and p-like wavefunctions, it shrinks and expands by expansion and reduction of the QD size. The dipole coupling vector can, at the same time, exhibit different behaviour, preventing the entire FCF to change significantly. However, this will still influence the QD optical properties. The larger the magnitude of the dipole coupling vector, the stronger is the s-p-like optical resonance.

4. Conclusion

In summary, we have parameterized the FCF via the dipole coupling vector (optical matrix element) and the distribution function whose widths were related to geometric and composition properties of the QD. There are a number of advantages of such parameterization. By using the expansion formula for $\mathcal{H}_{ab}(\mathbf{k})$ one can significantly reduce the numerical effort in calculation of all coupling constants relevant to electron-phonon interaction. The presented model can be used as the basis for estimating the ratio between radiative and non-radiative lifetimes as a function of the dot geometric and composition parameters, which is very important in future designs of QD based intersublevel emitters. By measuring the absorption one can extract the values of the dipole coupling vector and the level separation, which can be further used in combination with the presented model to get an insight into the electron-LO phonon interaction.

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