Symmetry suppression of allowed transitions between energy eigenstates of a Hamiltonian perturbed from a high symmetry.

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Abstract

It has been observed in quantum dots with C_{3v} symmetry that certain electronic transitions allowed by the dipole approximation selection rule are unresolvable in the measured photoluminiscence (PL) spectrum. We provide a theoretical model that predicts these spectral lines to be severely weakened if the Hamiltonian is considered to be perturbed from one with a higher spatial symmetry. An expression for the suppressing factor of the spectral line intensity is given in 1st order perturbation theory.

1 Introduction

Karlsson et al [1] have studied the PL spectrum of InGaAs C_{3v}-symmetric quantum dots (QDs). Their model assumes that the QD-Hamiltonian eigenstates which interact with electromagnetic waves on the studied interval of the spectrum are given by considering the symmetry breaking of exciton complex fine structure, which splits total angular momentum J eigenstates into energy levels which transform as double group irreps. By using rudimentary group theory, the energy levels, their degeneracies, and their allowed transitions can be calculated for all excitonic complexes, as was done by this particular QD. However, it was found that the number of spectral lines resolvable in a spectral feature associated with a specific excitonic transition is smaller than the predicted number for several exciton complex

transitions, namely $X_{10} \to vac., 2X_{20} \to X_{10}, 2X_{11} \to X_{10}, 2X_{11} \to X_{01}, 2X_{21}^{+} \to X_{11}^{+}$. Since the number of allowed transitions lowers if the Hamiltonian is altered to commute with more rotations (e.g. to D_{3h} or C_{6v} point group symmetry), the effect was dubbed "symmetry elevation". However, no explanation for why symmetry elevation occurs was found.

What the previous work on symmetry elevation in QDs did not consider is the nanocrystallic lattice that constitutes the QDs inside the potential well. The InGaAs QDs are formed by a lattice which does possess a 6-fold rotation along the z-axis. Even though the full Hamiltonian still only possesses C_{3v} symmetry, since it is ultimately dominated by the lower-symmetry potential well boundary shape, it can be modelled as a high-symmetry Hamiltonian which underwent symmetry breaking with a small perturbation and enjoys certain approximate symmetries, which still turn out to have a considerate effect in 1st order expansion. To obtain a theoretical description of the effects of these approximate symmetries, we first consider 1st order perturbation theory and describe a surjective mapping of basis vectors of irreps of the larger point group onto the basis vectors of irreps of its point subgroup, which allows us to determine the mapping of high-symmetry Hamiltonian eigenstates onto the low-symmetry Hamiltonian eigenstates under symmetry breaking, and then we emply Fermi's golden rule to quantify the suppression of certain spectral lines. The manifestation of de-intensifying of specific spectral lines due to approximate symmetries will be named "symmetry suppression".

2 Theoretical model of symmetry suppression

We shall first outline a general approach to describing a non-specific minimally symmetry-broken Hamiltonian, and then argue for why the assumptions made are justified in the case of InGaAs QDs.

2.1 Symmetry breaking as basis vector surjection

The first issue we must tackle is

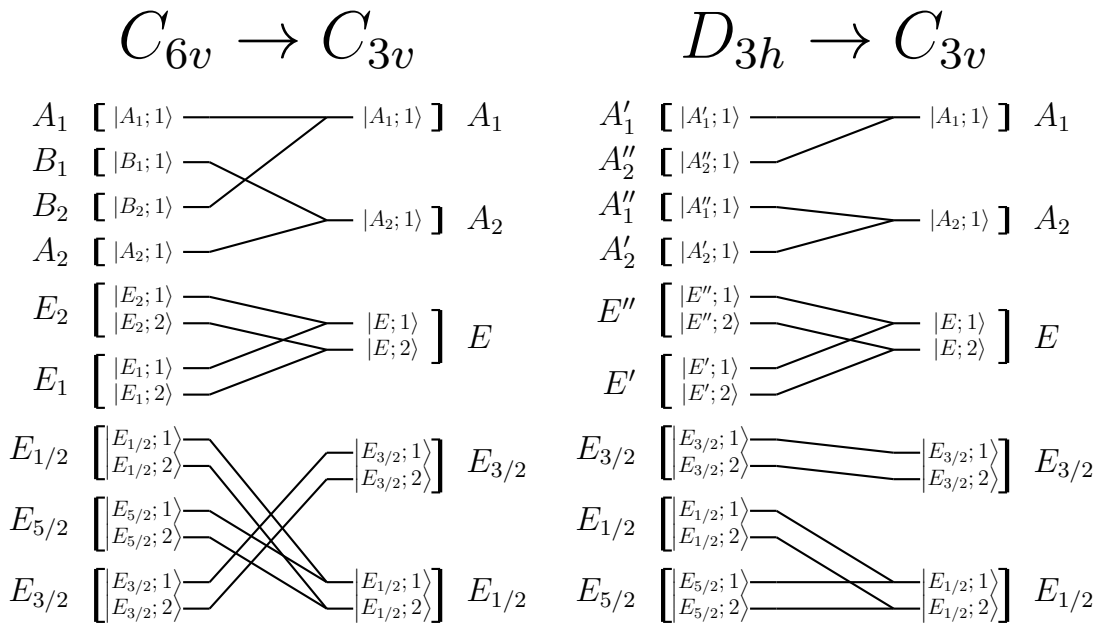

Figure 1: Surjective mapping of basis partners in irreps of C_{6v} and D_{3h} respectively onto their mutual subgroup C_{3v}. We see that both of these examples are trivial in the sense that no irrep decomposes into multiple irreps in the subgroup, which makes the mapping of partner states non-trivial. This typically happens when the high-symmetry group possesses irreps of high dimensions.

2.2 Domain alteration and perturbation theory

Suppose now we have a system described by a Hamiltonian $\hat{H}$ equipped with a group of symmetries G. Let us construct a Hamiltonian $\hat{H}_+$ which is equipped with a group of symmetries G_+ such that $G < G_+$ and $\hat{H}$ can be modelled as a perturbation of $\hat{H}_+$ like so:

$$\hat{H} = \hat{H}_+ + \lambda \hat{H}', \quad \text{where} \quad \lambda \ll 1, \langle E_i; n|\hat{H}'|E_i; n\rangle \sim E_i$$

Let us denote the eigenstates of the Hamiltonians $\hat{H}, \hat{H}_+$ as $|E_i; n\rangle$ and $\left|E^+_{i'}; n'\right\rangle$ respectively, where n, n' are the degeneracy labels. Disregarding accidental degeneracy, we know that every set of degenerate energy levels forms a basis of (i.e. transforms according to) an irrep

of the respective symmetry group:

$$\begin{pmatrix} |E_i;1\rangle \\ |E_i;2\rangle \\ \vdots \\ |E_i;d_i\rangle \end{pmatrix} \text{ forms a basis to } \Gamma^{(G)}_{r(i)}; \quad \begin{pmatrix} \left|E^+_{i'};1\right\rangle \\ \left|E^+_{i'};2\right\rangle \\ \vdots \\ \left|E^+_{i'};d^+_{i'}\right\rangle \end{pmatrix} \text{ forms a basis to } \Gamma^{(G_+)}_{r^+(i')}$$

where $d_i, d^+_{i'}$ specify the total edegeneracies of the energy levels and r, r^+ are some functions which map energy levels onto irreps. We know (TODO cite this) that under a perturbation of a set of degenerate eigenstates, the distance of the new states from the subspace $\mathcal{D}^+$ spanned by the degenerate eigenstates goes like $O(n)$. Therefore, if $|E_i;n\rangle$ arises from perturbing a degenerate subspace of energy E^+_j, there exists an unnormalized vector $\left|E^+_j;P\right\rangle \in \mathcal{D}^+_j$ for which

$$\begin{aligned} \left\langle E_i;n\middle|E^+_j;P\right\rangle &= 1 - O(\lambda) \\ \text{where} \quad \left\langle E^+_j;P\middle|E_i;n\right\rangle &= \left\langle E^+_j;P'\middle|E_i;n\right\rangle \forall \left|E^+_j;P'\right\rangle \in \mathcal{D}^+ \end{aligned}$$

We can construct this state by projecting the perturbed eigenstate onto the original degenerate subspace:

$$\begin{aligned} \hat{P}^+_j &= \sum_{m=1}^{d^+_j} \left|E^+_j;m\right\rangle\!\left\langle E^+_j;m\right| \\ \left|E^+_j;P\right\rangle &= \hat{P}^+_j \left|E_i;n\right\rangle = \sum_{m=1}^{d^+_j} \left|E^+_j;m\right\rangle \left\langle E^+_j;m\middle|E_i;n\right\rangle \end{aligned}$$

The projection can be calculated directly for any choice of basis of $\mathcal{D}^+$, but we can simplify the calculation. Because $\left\langle E_i;n\middle|E^+_j;P\right\rangle \approx 1$, we expect both of these states to transform according to $\Gamma^{(G)}_{r(i)}$ under rotations in G. In general, the perturbation might reduce the degeneracy of $\mathcal{D}^+$ and render $d_i < d^+_j$ (even though it is also possible to have $d_i = d^+_j$, which is a special case). Then we can identify the subset of basis vectors of $\mathcal{D}^+$ which transform according to $\Gamma^{(G)}_{r(i)}$ by reducing $\Gamma^{(G_+)}_{r^+(j)}$ into G and considering the basis vector surjection, as described above. Then we can approximate $\left\langle E^+_j;m\middle|E_i;n\right\rangle \approx 0$ for m which are not mapped onto $\Gamma^{(G)}_{r(i)}$.

Now we can express a general eigenstate of $\hat{H}$ as a superposition of an eigenstate of $\hat{H}^+$ with a norm that goes like $1 - O(\lambda)$ and a remainder. We normalize the components and introduce superposition coefficients:

$$|E_i; n\rangle = c_{ES} \left| E_j^+; ES \right\rangle + c_R \, |R\rangle$$

where the *elevated symmetry component* is defined as

$$\begin{aligned}
\left| E_j^+; ES \right\rangle &= \left| E_j^+; P \right\rangle \left| \left\langle E_j^+; P \middle| E_j^+; P \right\rangle \right|^{-1/2} \\
&= \left| E_j^+; P \right\rangle \left(\sum_{m=1}^{d_j^+} \left| \left\langle E_j^+; m \middle| E_i; n \right\rangle \right|^2 \right)^{-1/2} \\
c_{ES} &= \left\langle E_j^+; ES \middle| E_i; n \right\rangle \\
&= \sqrt{\sum_{m=1}^{d_j^+} \left| \left\langle E_j^+; m \middle| E_i; n \right\rangle \right|^2} \\
&\sim 1 - O\left(\lambda^2\right)
\end{aligned}$$

and the *residual component* then becomes

$$|R\rangle = c_R^{-1} \left(|E_i; n\rangle - c_{ES} \left| E_j^+; ES \right\rangle \right)$$

By demanding norm 1, we obtain:

$$\begin{aligned}
1 &= c_R^{-2} \left(\langle E_i; n | E_i; n \rangle + c_{ES}^2 \left\langle E_j^+; ES \middle| E_j^+; ES \right\rangle - 2 c_{ES} \left\langle E_i; n \middle| E_j^+; ES \right\rangle \right) \\
c_R &= \sqrt{1 - c_{ES}^2} \\
&\sim O(\lambda) \\
|R\rangle &= \left(1 - c_{ES}^2\right)^{-1/2} \left(|E_i; n\rangle - c_{ES} \left| E_j^+; ES \right\rangle \right)
\end{aligned}$$

A note about the ambiguity of construction of $\hat{H}_+$. We see that the construction of $\hat{H}_+$ is not unique, and indeed there may be multiple options for the general shape of $\hat{H}_+$. It is important to note that we need not consider all options of $\hat{H}_+$ as if their effects were cumulative– the formulation of $\hat{H}$ as a perturbation of $\hat{H}_+$ should retrieve the same physics regardless of $\hat{H}_+$ should an infinite-order (no approximation) perturbation theory be used. However, the error associated with a 1st

order perturbation analysis is inversely proportional to $\left\langle E_i; n \middle| E_j^+; P \right\rangle$, and thus a choice of $\hat{H}_+$ which maximises the norm of the elevated symmetry component will be the most precise one. Then, if a certain symmetry elevation suppresses a certain transition, then the symmetry suppression factor for a particular choice of $\hat{H}_+$ is in general a lower bound on the true symmetry suppression factor, as there might exist a different choice of $\hat{H}_+$ with a higher norm of the elevated symmetry component.

2.3 Evaluation of transition rates

Consider now two eigenstates of $\hat{H}$ at different energy levels, $|E_i; n_i\rangle, |E_f; n_f\rangle$. If we perturb the system with an interaction Hamiltonian $\hat{H}'$, the rate of transition between these two states is given by Fermi's golden rule

$$\Gamma_{|E_i;n_i\rangle \to |E_f;n_f\rangle} = \frac{2\pi}{\hbar} \left| \langle E_f; n_f | \hat{H}' | E_i; n_i \rangle \right|^2$$

Since the energy levels may be degenerate, the full transition rate between the two sets of degenerate states becomes

$$\Gamma_{E_i \to E_f} = \sum_{n_i=1}^{d_i} \sum_{n_f=1}^{d_f} \Gamma_{|E_i;n_i\rangle \to |E_f;n_f\rangle}$$

Let the energy levels E_i, E_f be chosen such that the direct product $\Gamma^{(G)}_{r(i)} \otimes \Gamma^{(G)}_{\hat{H}'} \otimes \Gamma^{(G)}_{r(f)}$ contains the identity irrep of G, and hence the matrix elements do not vanish due to the selection rule. Let us now decompose the two eigenstates into elevated symmetry and residual components:

$$\begin{aligned} |E_i; n_i\rangle &= c^i_{ES} \left| E^+_{i'}; ES \right\rangle + c^i_R |R_i\rangle \\ |E_f; n_f\rangle &= c^f_{ES} \left| E^+_{f'}; ES \right\rangle + c^f_R |R_f\rangle \end{aligned}$$

where $E^+_{i'}, E^+_{f'}$ denote the energy levels of the high-symmetry Hamiltonian $\hat{H}^+$ which get perturbed into E_i, E_f respectively.

Let us now calculate the matrix element under the interaction Hamiltonian:

$$\begin{aligned} &\langle E_f; n_f | \hat{H}' | E_i; n_i \rangle = \\ &\left(c^f_{ES}\right)^* c^i_{ES} \left\langle E^+_{f'}; ES \middle| \hat{H}' \middle| E^+_{i'}; ES \right\rangle + \left(c^f_{ES}\right)^* c^i_R \left\langle E^+_{f'}; ES \middle| \hat{H}' \middle| R_i \right\rangle + \\ &\left(c^f_R\right)^* c^i_{ES} \langle R_f | \hat{H}' | E^+_{i'}; ES \rangle + \left(c^f_R\right)^* c^i_R \langle R_f | \hat{H}' | R_i \rangle \end{aligned}$$

We know that $\left(c_R^f\right)^* c_R^i$ goes like $O\left(\lambda^2\right)$, and hence we will disregard the corresponding term. There are now two possibilities for what may occur:

1. *The elevated symmetry matrix element does not vanish.* If $\Gamma^{(G_+)}_{r^+(i')} \otimes \Gamma^{(G_+)}_{\hat{H}'} \otimes \Gamma^{(G_+)}_{r^+(f')}$ contains the identity irrep of G_+, the leading term matrix element does not vanish, and forms the main contribution to the total matrix element. The leading coefficient goes like $1 - O(\lambda)$.

2. *The elevated symmetry matrix element vanishes.* If $\Gamma^{(G_+)}_{r^+(i')} \otimes \Gamma^{(G_+)}_{\hat{H}'} \otimes \Gamma^{(G_+)}_{r^+(f')}$ does not contain the identity irrep of G_+, the leading term matrix element vanishes due to the selection rule (since it must transform as a scalar). The only non-zero terms are now the cross-terms, which both go like $O(\lambda)$.

References

[1] Karlsson, K. F. *et al* (2015), Spectral signatures of high-symmetry quantum dots and effects of symmetry breaking. *New Journal of Physics*, **17** 103017

[2] Altmann, S. L., Herzig, P. (1994), *Point-Group Theory Tables.* Oxford: Clarendon Press