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THEORY OF THE EXCITONIC STARK SHIFT

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The exciton Stark shift is due to a coupling between the exciton and all the biexcitonic states. This coupling induces a blue shift at large detuning which goes into a red shift at small detuning, when the biexcitonic molecule is stable.

When a direct gap semiconductor is irradiated by a laser beam with wavelength in the transparency region, the exciton line blue shifts as experimentally observed two years ago by Danièle Hulin and coworkers\(^1\) in bulk GaAs and quantum wells. The shift lasts the time of the laser pulse, and so produces optical gates as short as femtoseconds.

A laser beam in the transparency region produces virtual electron hole pairs (e-h); the semiconductor-laser coupling reads, in the rotating frame,

\[
W = \lambda (U^+ + U) = \lambda \sum_k (B_k^+ + B_k^-) \tag{1}
\]

the coefficient \(\lambda^2\) being proportional to the laser intensity, \(B_k^+ = a_k^+ b_{-k}^+\) where \(a_k^+\) creates an electron in the conduction band while \(b_{-k}^+\) creates a hole in the valence band.

As the exciton line is seen in these experiments, this means that the laser intensity is in fact not very large (even if experimentalists use "intense femtoseconds laser source" to see a sizeable effect); a really intense laser pulse would have created a large amount of virtual (e-h) and destroyed the bound state. Consequently the excitonic Stark shift corresponds in fact to the exciton energy change induced by the perturbation \(W\), which can be treated to lowest order in \(\lambda\).

The exciton ground state energy is the difference between the perturbed (e-h) ground state energy \(E_{X1}'\) and the perturbed vacuum energy \(E_O'\). The perturbation \(W\) couples the vacuum to all one (e-h) states \(\ket{X_1}\), bound or unbound, with energies in the rotating frame \(E_{X1} = \omega_{X1} - \omega_p\), \(\omega_p\) being the laser frequency. \(W\) couples \(\ket{X_1}\) to all two (e-h) states \(\ket{XX_n}\) with energy \(E_{XX_n} = 2(\omega_{XX_n} - \omega_p)\) and also to the vacuum \(\ket{0}\). So that the excitonic shift \(\delta\omega_{X1}\) is to lowest order in \(\lambda\)

\[
\delta\omega_{X1} = \lambda^2 \left[ \sum_n \frac{1 < XX_n | U^+ U' X_1 > |^2}{E_{X1} - E_{XX_n}} + \frac{1 < 0 | U | X_1 > |^2}{E_{X1}} \right] - \lambda^2 \sum_i \frac{1 < X_i | U U' | 0 > |^2}{\epsilon_{X_i}'} \tag{2}
\]

where we have chosen \(E_O = 0\). Eq. (2) gives the observed excitonic Stark shift as a function of the detuning \(E_{X1}'\).
But there is a major difficulty with Eq. (2). All the terms of the last two sums, which describe the exciton-vacuum coupling, are proportional to the sample volume \( V \) (as \( |<X_1|U^+|0>|^2 = |\Sigma_k \phi_i^* (k)|^2 = V |f_j (r = 0)|^2, \phi_i (k) \) and \( f_j (r) \) being the exciton wave functions). As the exciton shift is physically independent of \( V \), these exciton-vacuum coupling terms have to be cancelled by similar terms included in the first sum. So that \( \delta \omega_{X_1} \) turns out simply to be the volume independent part of the first sum i.e the excitonic Stark shift results only from the (volume independent) exciton - "biexciton" coupling — we loosely call "biexciton" all two (e-h) states, bound and unbound.

In order to prove that the sample volume mathematically disappears from Eq. (2), one should perform the first sum of Eq. (2). But the biexcitonic states are unknown. There are however two cases when this sum can be calculated explicitly, and in which one can see indeed the disappearance of the volume.

i) If the Coulomb interaction is neglected, the exciton states are simply plane waves \( |e_k^+|0>| \) with energy \( E_k = \Omega_o + k^2/2m. \) Similarly the biexciton states are \( B_{k^+} B_{k^+}^+|0>| \) with energy \( E_k + E_{k^+}. \) Using these values into Eq. (2), as well as the expression (1) for \( U \), one finds

\[
\delta \omega_{X_1} = \sum_{k,k'} \left| \frac{|<0|B_{k'} B_{k^+}^* (\sum_k \lambda B_k^+ B_{k'}^+ |0>| - E_{k'} - E_{k^+} |0>|^2}{E_{k_1} - E_{k'} - E_{k^+} + E_{k_1}} \right| + \frac{|<0|\sum_k \lambda B_k B_k^+ |0>|^2}{E_{k_1}}
\]

\[= \sum_{k,k'} \frac{\lambda^2}{E_{k'}} + \frac{\lambda^2}{E_{k_1}} + \sum_{k,k'} \frac{\lambda^2}{E_{k'}} = \frac{2\lambda^2}{E_{k_1}} \tag{3}\]

ii) If the detuning is very large, all the denominators of Eq. (2) are essentially equal to \( E_{X_1} \), and the sum can be performed through closure relation. The last sum is simply

\[<0|UU^+|0> = \sum_{k} B_k \sum_{k'} B_{k'}^+ |0> = N \tag{4}\]

where \( N \) is the number of \( k \) states in the sample volume. Similarly as \( |X_1> = \Sigma_k \phi_i (k) B_k^+ |0> \) the first sum is

\[<X_1|UU^+X_1> = \sum_{k,k,k,k'} \phi_i (k_1) \phi_i^* (k_2) <0|B_{k_1} B_{k_2}^+ B_{k_1}^* |0> = \sum_{k_1,k,k,k'} \phi_i (k_1) \phi_i^* (k_2) (\delta_{kk'} \delta_{k_1 k_2} - \delta_{kk'} \delta_{k_1 k_2})^2 \]

\[= \sum_{k_1} |\phi_i (k_1)|^2 \sum_k 1 + \sum_{k_1} \phi_i (k) \sum_{k_2} \phi_i^* (k_2) - 2 \sum_{k_1} |\phi_i (k_1)|^2 = N + V |f_i (o)|^2 - 2 \tag{5}\]
So that the exciton shift $\delta \omega_{X_1}$ is again $2\lambda^2/\varepsilon_{X_1}$ i.e the Stark shift of the two-level atom(2).

If one wants to show that the volume indeed disappears for all detuning, one needs to rewrite the perturbation theory using its Brillouin Wigner form. Eq. (2) is equivalent to

$$\delta \omega_{X_1} = \lambda^2 \left[ <X_1|U - \frac{1}{\varepsilon_{X_1}} H|X_1> + <X_1|U^+ X_1> + <0|U - \frac{1}{\varepsilon_{X_1}} H U^+|0> \right]$$

(6)

Writing $|X_1> = B_1^+ |0>$, one can then rewrite Eq. (6) in a compact form, introducing the commutator

$$[H, B_1^+] = E_{X_1} (B_1^+ + C_1^+)$$

(7)

One then obtains

$$\delta \omega_{X_1} = (2 + \alpha + \beta - \gamma) \lambda^2 / E_{X_1}$$

For $V_{\text{coul}} = 0$, or at large detuning, one of course recovers the previous result namely $\alpha + \beta - \gamma = 0$.

The coefficient $\alpha$ results from the fact that the two excitons forming the biexciton feel each other because they are not real bosons. Its explicit calculation requires only the knowledge of the exciton wavefunctions, as it is given by

$$\alpha = <0|U (1 - B_1 B_1^+) \frac{E_{X_1} - H}{H} U^+ |0> = \sum_i \frac{E_{X_1} - E_{X_1}}{E_{X_1}} \alpha_i$$

(8)

$$\alpha_i = 2 \sum_{k,k'} \phi_i^*(k) \phi_i(k) \phi_i(k)^2$$

At large detuning, i.e for $E_{X_1} \geq \varepsilon_{X_1}$, $\alpha$ behaves as $(\varepsilon_{X_1}/E_{X_1})^{1/2}$, where $\varepsilon_{X_1}$ is the exciton binding energy. In this limit, the main contribution to $\alpha$ comes from the high energy diffusion states. At small detuning, the state $i = 1$ dominates the sum and $\alpha$ goes to $\alpha_1 - 2 = 5$ in 3 dimensions. The Schmitt-Rink et al final(3) result corresponds only to this limit, but as seen later this limit never dominates the exciton shift value.

In the coefficients $\beta$ and $\gamma$ enters the operator $C_1^i$, which can be seen as the Coulomb interaction between the two excitons forming the biexciton. In usual problems dealing with excitons, these Coulomb interaction can be neglected but in the case of the exciton shift, this is not possible, as easily seen from a diagrammatic expansion of the shift. At large detuning, $\beta$ and $\gamma$ are negligible compared to $\alpha$, which is therefore the leading correction to the two-level atom shift $2\lambda^2/\varepsilon_{X_1}$. On the opposite limit, at small detuning, $\beta = \varepsilon_{X_1}/E_{X_1}$ and dominates $\alpha$, while $\gamma$ leads to an ever more dramatic effect in materials having a stable biexcitonic molecule. As can be easily seen directly from Eq. (2), this bound state induces a new pole $\delta \omega_{X_1} = \lambda^2 (E_{X_1} - E_{XX_1})$ associated with a red shift for the exciton line at the two photon absorption threshold ($E_{X_1} = E_{X_1} \rightarrow \omega_p + \omega_{X_1} = 2\omega_{XX_1}$). Close to this threshold, there is of course no divergence for the exciton shift, but instead the shift saturates to a value obtained from perturbation theory done in the degenerate subspace $|X_1>$ and $|XX_1>$; one finds

$$\delta \omega_{X_1} = - \lambda <XX_1|U|X_1>$$

(9)
In conclusion, we have shown that the excitonic stark shift results from the coupling between the exciton and all biexcitonic states, bound and unbound. The exciton line blue shifts at large detuning, the value of the shift being the one of a two-level atom. At small detuning, compared with the exciton binding energy, interactions between the two excitons forming the biexciton modify the value of the shift. In particular, in materials having a stable biexcitonic molecule, these interactions induce a red shift of the exciton line close to the two photon absorption threshold for formation of the molecule.

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