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Electron Dynamics of Interatomic Coulombic Decay in Quantum Dots: Singlet Initial State

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Abstract. In this paper we investigated the interatomic Coulombic decay (ICD) of a resonance singlet state in a model potential for two few-electron semiconductor quantum dots (QDs) by means of electron dynamics. We demonstrate that ICD is the major decay process of the resonance for the singlet wave function and compare the total and partial decay widths as a function of the QD separation with that from our previous study on the corresponding triplet states [1].

1 Introduction to the Model Calculations

The elementary physical process of interatomic Coulombic decay [2] is nowadays the topic of intensive experimental and theoretical investigations. In ICD an excited electronic state localized on an atom or a molecule decays by transferring the energy to a loosely bound neighboring species and ionizing it. This decay mechanism is due to the correlated motion of electrons localized on different species. Recently, ICD has been shown to exist not only in atomic or molecular systems but also in general, non-infinite binding potentials [1,3] as schematically shown in Fig. 1.

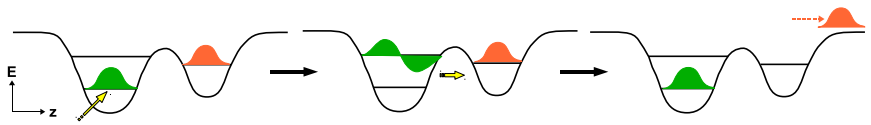

Fig. 1. Schematic view of ICD in a two-well model potential. Initially both the green (light gray) and the orange (dark gray) wave packets are bound in their wells. After excitation of the left packet to the next level, it falls back and the excess energy is used to emit the right one to the continuum.

Here, a numerical study of real-time electron dynamics using the multiconfiguration time-dependent Hartree (MCTDH) method [4] for electronic singlet wave functions was done to follow ICD. As in Ref. [1] the quasi one-dimensional model system for the QDs is a double well with separations 5 a.u. $< R \leq$ 17 a.u. in z direction. The left well contains two levels L_0 and L_1, the right well one (R_0). The two-electron resonance state that decays via ICD is termed L_1R_0. Computations have also been done for $R >$ 17 a.u., but here we used for computational efficiency a purely one-dimensional system with the effective interaction potential

$$V_{eff}^{(\omega_\perp)}(|z_1 - z_2|) = \sqrt{\frac{\pi}{2}}\frac{1}{l}e^{\zeta^2}\left(1 - erf(\zeta)\right), \tag{1}$$

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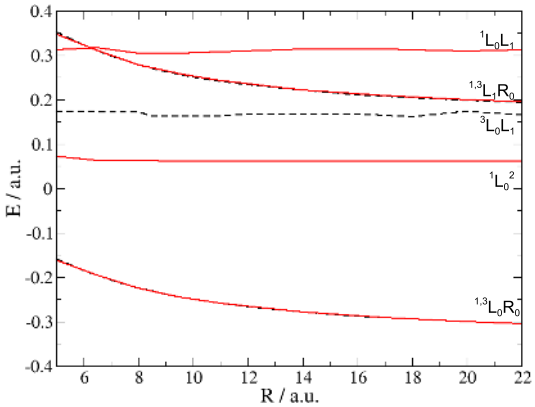

Fig. 2. Energies of the singlet (solid red) and triplet (dashed black) two-electron states of the double well as a function of R. The energies of the R_0^2 and the L_1^2 state exceed 0.4 a.u.

where $\zeta = |z_1 - z_2|/\sqrt{2}l$. The confinement scale $l = \sqrt{\hbar/m\omega_\perp}$ comes from the derivation of (1) [5] and contains $\omega_\perp$ for the transversal potential [1].

2 Results and Discussion

In Fig. 2 the energies of the localized singlet (red, solid lines) and triplet (black, dashed lines) states as functions of R are shown. For both multiplicities the ground state is L_0R_0. All other states, namely L_0L_1 and L_1R_0 as well as L_0^2, R_0^2, and L_1^2 for singlets only, are resonances. For the ICD resonance L_1R_0 at $R = 8$ a.u. the singlet-triplet splitting is $8.5 \cdot 10^{-3}$ a.u. increasing with decreasing R and vice versa. The coupling of levels is weak so that the tunneling probability among different states is negligible.

In Tab. 1 the numerical results for ICD widths and times from stabilization analysis and electron dynamics in the singlet state at $R = 8$ a.u. are given. As previously [1], total and partial widths from the more accurate electron dynamics calculations are smaller by about a factor of two. The ratio Γ_R/Γ_L shows the probability for the ICD electron to go to the right side compared to the left side.

The left panel of Fig. 3 displays the decay widths Γ from electron dynamics for singlet and triplet states. For short R, i.e. overlapping single-electron eigenfunctions, correlation energy is higher in singlet states and they decay faster. This can be understood by the relatively lower spatial separation of opposite-spin singlet electrons compared to same-spin triplet electrons that leads to stronger electron repulsion in singlets. With increasing R this behavior becomes less pronounced. In the $R \to \infty$ asymptotic region $\Gamma \propto 2\pi|\langle L_0 k|r_{12}^{-1}|L_1R_0\rangle|^2\delta(E_k - E_{L_1L_0} + E_{L_0})$ applies [6]. To calculate the direct matrix element including the continuum states k the usual expansion of the Coulomb potential between separated charges is used:

$$r_{12}^{-1} \to \frac{1}{R} - \frac{1}{R^3}\sum_{m=-1}^{1}(2-m^2)\mathbf{r}_1 Y_1^m(\Omega_1)\mathbf{r}_2 Y_1^m(\Omega_2)(4\pi/3) + \ldots. \tag{2}$$

Table 1. Total and partial ICD widths Γ, Γ_R, and Γ_L, ratio Γ_R/Γ_L, and decay time τ for singlets at $R = 8$ a.u.

Method	Γ/meV	τ/fs	Γ_R/meV	Γ_L/meV	Γ_R/Γ_L
Stabilization	41(27)	16(10)	33(2)	8(5)	4(2)
Dynamics	17.2346	38.19	15.0746	2.1600	6.98

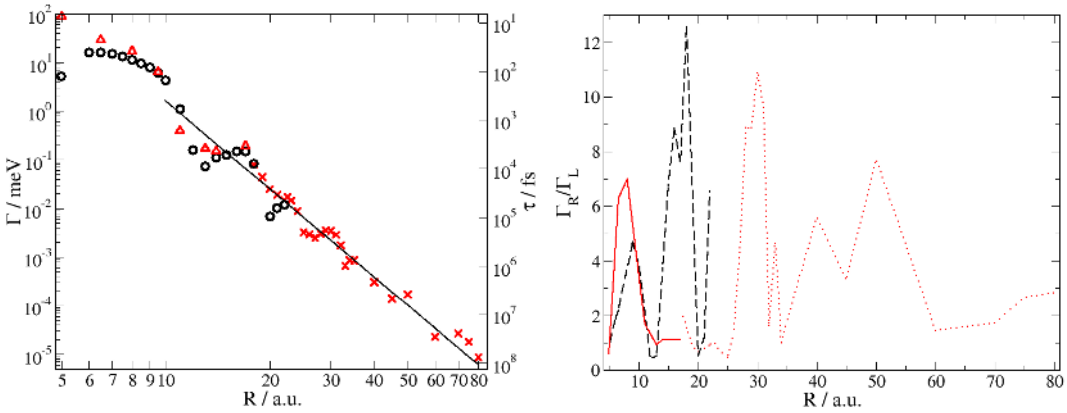

Fig. 3. Left panel: Double logarithmic representation of the total ICD widths of the L_1R_0 state for triplet (black circles) and singlet (red triangles) states as a function of R for $R \leq 17$ a.u.; for $R > 17$ a.u. results for singlet states in the one-dimensional model (red crosses) are shown. The line represents $\Gamma \propto R^{-6}$ (see text). Right panel: Ratio Γ_R/Γ_L as a function of R for triplet (dashed black) and singlet states (red lines: solid for quasi-1d, dotted for1d).

With this the relation $\Gamma \propto R^{-6}$ is obtained and displayed as a line in Fig. 3 (left). Calculated Γ follow this behavior, although overlaid by oscillations. These go back to multiple scattering and recapture effects of the continuum electron due to its reflection at the potential barrier caused by the other electron bound to the L_0 level [1]. Only for small R the kinetic energy of the continuum electron exceeds this potential barrier.

In the right panel of Fig. 3 the ratios Γ_R/Γ_L are compared. Their oscillations follow those of Γ indicating that small Γ originate in small Γ_R. For small R the ratio is always larger for singlet than for triplet states, because the emitted ICD electron is repelled more strongly by the L_0 electron.

3 Conclusion

MCTDH electron dynamics calculations of the ICD process in a quasi one-dimensional QD model potential with singlet initial states were presented. We found that at short distances between the QDs, ICD is faster in singlets than in triplets studied before and its rates behave as R^{-6} at long distances. For singlet and triplet states ICD is the dominant decay process.

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