Chemical Physics 482 (2017) 135–145

Contents lists available at ScienceDirect

Chemical Physics

journal homepage: www.elsevier.com/locate/chemphys

Strong field control of the interatomic Coulombic decay process in quantum dots

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ARTICLE INFO

Article history:
Available online 16 September 2016

Keywords:
Electron dynamics
Laser control
Interatomic Coulombic decay
Quantum dot
Strong field

ABSTRACT

In recent years the laser-induced interatomic Coulombic decay (ICD) process in paired quantum dots has been predicted (Bande, 2013). In this work we target the enhancement of ICD by scanning over a range of strong-field laser intensities. The GaAs quantum dots are modeled by a one-dimensional double-well potential in which simulations are done with the space-resolved multi-configuration time-dependent Hartree method including antisymmetrization to account for the fermions. As a novelty a complementary state-resolved ansatz is developed to consolidate the interpretation of transient state populations, widths obtained for the ICD and the competing direct ionization channel, and Fano peak profiles in the photoelectron spectra. The major results are that multi-photon processes are unimportant even for the strongest fields. Further, below-π to π pulses display the highest ICD efficiency while the direct ionization becomes less dominant.

1. Introduction

"ICD appears everywhere!" [1] – this slogan summarizes the nearly twenty years of success history of the ultrafast interatomic (intermolecular) Coulombic decay (ICD) process from its first theoretical prediction by Cederbaum, Zobeley, and Tarantelli [2] to its widespread theoretical and experimental observation [3,4] in various atomic and molecular systems including clusters of noble gas atoms [5–8], endohedral fullerenes [9–11], aqueous solutions [12–17], biological systems [18–23], and nanomaterials [24–26] just to name a few.

ICD can be understood as a delocalized Auger decay over two or more atomic species which is mediated by the long-range Coulomb force among electrons residing on the different sites: on one site a high-energy electron relaxes into a lower-energy state and transfers its energy to another electron on one of the neighboring sites which is then ionized. The decaying resonance excited state is typically prepared by either inner-valence ionization [3,4,27], resonant excitation [28–31], electron [4,32–36], or ion impact [4,37].

Moreover, the above slogan directs into the future. It motivates the interdisciplinary community of ICD researchers to push the frontiers towards a comprehensive fundamental understanding of ICD as well as towards novel materials. Our contributions support both directions. Firstly, we pioneered material sciences investigations of ICD by studying pairs of nano-structured semiconductors, namely quantum dots (QD) [24,25,38]. They are available from various fabrications techniques as self-assembled [39], nanowire [40], etched vertical [41], and gated two-dimensional electron gas [42] quantum dots. Such materials are attractive candidates for a device application of ICD in the field of energy conversion, e.g. as next-generation infrared photo detector or solar cell. This is because the QD pair's electronic structure, and hence the ICD performance, can be custom made through geometry control of the QDs' distance [25,38] (this holds likewise for quantum wells [26]), widths [43], and heights [43,44].

Secondly, we explicitly solve the time-dependent electronic Schrödinger equation in a space-resolved fashion [25,34] for the scenario of two correlated electrons. This renders insight into the transient electronic level occupations during ICD in addition to the decay rates which can likewise be obtained from

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http://dx.doi.org/10.1016/j.chemphys.2016.09.020
0301-0104/

non–hermitian electronic structure theory for resonances [6,27,45,46] and nuclear dynamics of the cluster explosion after ICD [47–49]. In cases less-accurate partially-correlated electron dynamics has been used in combination with hole [50] or nuclear [17] dynamics. In this paper we push the theory to another level in solving the time-dependent electronic Schrödinger equation in an alternative state-resolved ansatz to consolidate and interpret our space-resolved results.

Thirdly, we explicitly consider the radiative initiation of ICD in our description [51]. Control of ICD by field strength variation is the theme of this paper. Before we lay out the details, let us introduce the two-state-and-continuum model system of the paired QDs (Fig. 1). Each QD is represented by an electron binding potential where the two levels L_0 and L_1 of the left and the single level R_0 of the right QD are in the energetic order $E^{1e}_{L_0} < E^{1e}_{R_0} < E^{1e}_{L_1}$. The system is initially in its $|L_0R_0\rangle$ ground state with one electron in the lowest level of each QD. A laser excites the electron in the left QD such that the two-electron resonance state $|L_1R_0\rangle$ is populated. This initiates ICD where the L_1 electron relaxes to the L_0 level again while energy becomes available to ionize the right binding potential through exciting the R_0 electron into the electronic continuum with energies ε, i.e. into the state manifold $|L_0\varepsilon\rangle$ [25,51]. States with both electrons localized in the same QD were found to be irrelevant for the processes investigated [25,38]. Note as well that available paired QDs meeting this theoretical description are singly-charged e.g. by electron transport from a reservoir [39–42]. Either a triplet state, which is stable over 100 ns [40,41] or the energetically favored singlet state could be established for the full duration of ICD and likewise be calculated [25,51].

When speaking of laser control of ICD an efficient preparation of the decaying resonance state is anticipated. One control parameter is the laser focus that influences the direct ionization of the R_0 electron [51]. A few others, which will be subject to future publications, are off-resonance lasing actions, polarization effects relevant to other typical QD geometries, and pulse duration. Their combination may eventually be cast into an optimal control scheme.

In this study we investigate the influence of different strengths of a pulsed infrared-laser field on the ICD process in QDs. They are in the range of $3 \cdot 10^7 - 2 \cdot 10^9$ W/cm^2 [52,53] and thus lie well below the maximum intensities 10^{18} W/cm^2 typically accessible in regular laser labs for a variety of energies from ultraviolet to infrared pulses [54]. To date experiments on ICD have only been done for atomic and molecular clusters and require soft X-ray pulses of similar intensity that are only available in large-scale synchrotron facilities as e.g. BESSY II [28,55]. There the peak intensity is prescribed by the beamline used and typically allows for a sub-π pulse excitation only. Hence, to our knowledge, ICD has experimentally never been investigated with view on laser strengths. For atoms and molecules this may become possible at high-brilliance soft X-ray sources specifically prepared for intensity variation, or when laser technology advances towards generating soft X-ray pulses in regular laser labs. At that time our theoretical method will be ready to accompany such experiments. By contrast, no obvious technical obstacle may hinder a potential QD ICD lab experiment on the influence of infrared lasing strength, and indeed QD experiments have already been done on population inverting Rabi oscillations with ultraviolet to visible light in the intensity range $10^3 - 10^6$ W/cm^2 [56,57]. But there photoionization was no relevant process.

Although not ICD, the Auger decay after core-ionization of atoms has been studied in competition with photoionization as function of the strength of hard X-ray fields with $10^{14} - 10^{19}$ W/cm^2 [53,58]. In focus was the field strength dependence of the shape of the electron spectrum after irradiation with time-symmetric $n\pi$ pulses which established an n-fold multiplet structure. Such a profile can be observed when the decay is much faster than the pulse, i.e. where the spectral widths of a π pulse, or the Rabi oscillation time of any $n\pi$ pulse, is significantly larger than the decay widths [53,58]. In the inverse case [51,59–61] the photoelectron peak establishes an ideal Fano profile when no strong-field bending of continuum states occurs [60]. In our previous work we have been able to deduce the Fano rather than the multiplet regime and showed there a nearly ideal Fano profile for an $n = 1\pi$ pulse [51]. Here we will extend these findings towards higher-$n\pi$ pulses.

The course of the paper is this: In Section 2.1 we introduce the model for the QD pair followed by the two theoretical methods for space- and a state-resolved electron-dynamics calculations (Sections 2.2.1 and 2.2.2). To compare both representations we begin discussing the results with the known π-pulse-induced ICD process (Section 3.1) and then turn to stronger $n\pi$-pulses (Section 3.2.1) as well as arbitrarily strong (Section 3.2.2) and weak fields (Section 3.2.3).

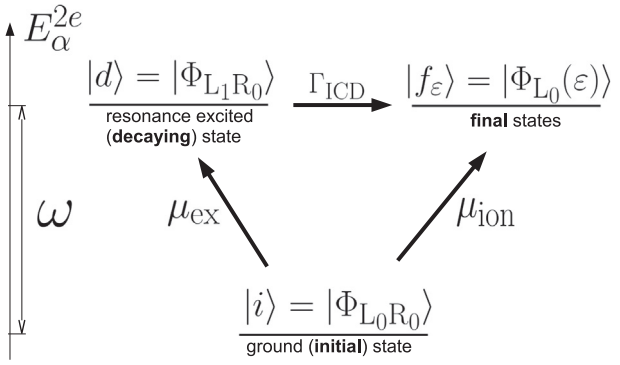

Fig. 1. Schematic representation of the relevant two-electron states in the paired QD. The $|L_0R_0\rangle$ ground state is by ω lower in energy than the equally-energetic decaying and continuum states $|L_1R_0\rangle$ and $|L_0\varepsilon\rangle$. These states can be addressed by the laser, the first through resonant excitation (ex), the second through direct ionization (ion). $|L_1R_0\rangle$ can decay into $|L_0\varepsilon\rangle$ via ICD.

2. Theory

2.1. Paired quantum dot model

The pair of QDs in which we investigate ICD is modeled by two one-dimensional negative Gaussian potentials expanding into the z-direction,

$$\hat{V}_{QD}(z) = -D_L e^{-b_L(z+\widetilde{R}/2)^2} - D_R e^{-b_R(z-\widetilde{R}/2)^2}, \quad (1)$$

where $\widetilde{R} = 8.0$ a.u. (86.68 nm) is the distance between the potential minima, $D_L = 1.0$ a.u. (10.30 meV) and $D_R = 0.8$ a.u. (8.24 meV) are the depths of the left (L) and right (R) potential wells. With $b_L = 0.25$ a.u. and $b_R = 1.0$ a.u. we determine the corresponding full width at half maximum of the two potentials via $r_{L,R} = 2\sqrt{\ln 2/b_{L,R}}$ which gives $r_L = 36.08$ nm and $r_R = 18.04$ nm. Note that our calculations were performed in atomic units. The numerical data in this paper is given in units of GaAs QDs (nm for distances, meV for energies) with a material specific effective mass and dielectric constant, $m^* = 0.063$ and $\kappa = 12.9$ [62], and the conversion equations as in previous work [43]. The parameters fulfill the requirement of keeping the spatial overlap of the Gaussian potential wells' bound one-particle states negligible, so that the two-electron wavefunctions can be approximated by an antisymmetrized Hartree product (see below).