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Computational modeling of quantum-confined impact ionization in Si nanocrystals embedded in SiO_2

C. Sevik[a,b,*], C. Bulutay[a,b]

[a]Department of Physics, Bilkent University, Ankara, 06800, Turkey
[b]UNAM - National Nanotechnology Research Center, Bilkent University, Ankara, 06800, Turkey

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Abstract

Injected carriers from the contacts to delocalized bulk states of the oxide matrix via Fowler–Nordheim tunneling can give rise to quantum-confined impact ionization (QCII) of the nanocrystal (NC) valence electrons. This process is responsible for the creation of confined excitons in NCs, which is a key luminescence mechanism. For a realistic modeling of QCII in Si NCs, a number of tools are combined: ensemble Monte Carlo (EMC) charge transport, ab initio modeling for oxide matrix, pseudopotential NC electronic states together with the closed-form analytical expression for the Coulomb matrix element of the QCII. To characterize the transport properties of the embedding amorphous SiO_2, ab initio band structure and density of states of the α-quartz phase of SiO_2 are employed. The confined states of the Si NC are obtained by solving the atomistic pseudopotential Hamiltonian. With these ingredients, realistic modeling of the QCII process involving a SiO_2 bulk state hot carrier and the NC valence electrons is provided.

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1. Introduction

Due to its indirect band gap, bulk Si is a very inefficient emitter, even at liquid He temperatures. Within the last decade, several approaches were developed towards improving the efficiency of light emission from Si-based structures. In spirit, all were based on the lifting of the lattice periodicity that introduces an uncertainty in the *k*-space and therefore altering the indirect nature of this material. Some examples are: SiGe or Si–SiO_2 superlattices [1,2] or Si nanocrystal (NC) assemblies [3]. Recently, blue electroluminescence (EL) from Si-implanted SiO_2 layers and violet EL from Ge-implanted SiO_2 layers were observed. An important process responsible for EL occurring in quantum dots and NCs is the quantum-confined impact ionization (QCII). A carrier initially at a high energy in the continuum states of the bulk structure when able to excite a valence band electron of a NC across its band gap creates an electron–hole pair (cf. Fig. 1). This process is responsible for the introduction of confined excitons in silicon NC LEDs, which is a key luminescence mechanism. In contrast to its crucial role, QCII has not been given the attention it deserves.

To model the QCII process, we start by characterizing the hot electron transport in oxides within the ensemble Monte Carlo framework. Density of states and band structure of common crystal phases of the SiO_2 used in our Monte Carlo transport calculation are obtained by using the ABINIT code [4], which is based on the density functional *ab initio* methodology. Next, we derive an analytical expression for the QCII probability in NCs that can become an instrumental result in assessing EL in the presence of other competing scattering mechanisms. The effect of QCII on bulk transport quantities is also discussed.

*Corresponding author.
E-mail addresses: sevik@fen.bilkent.edu.tr (C. Sevik), bulutay@fen.bilkent.edu.tr (C. Bulutay).

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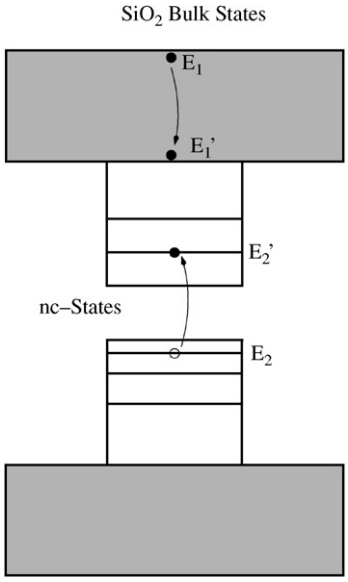

Fig. 1. Quantum-confined impact ionization in NCs.

2. Theoretical details

First-principles band structure and density of states (DOS) for SiO_2 were calculated within the density functional theory, using the pseudopotential method employing the local density approximation as implemented in the ABINIT code [4] in excellent agreement with the available results [5,6]. We demonstrate the utility and the validity of our *ab initio* DOS results by studying the high-field carrier transport in bulk SiO_2 up to fields of 10 MV/cm using the ensemble Monte Carlo technique which is currently the most reliable choice for studying hot carrier phenomena free from major simplifications [7,8]. We include the acoustic, polar and non-polar optical phonon scatterings. The corresponding scattering rates are intimately related with the band structure and the DOS of SiO_2 for which we use those of the α-quartz phase due to its strong resemblance of the amorphous SiO_2 in terms of both the short-range order and the total DOS [9]. Aiming for very high fields around 10 MV/cm, we also include the impact ionization process within the bulk SiO_2 medium; the relevant parameters were taken from the work of Arnold et al. [10].

Our modeling for QCII is an extension of the approach by Kehrer et al. [11] who have dealt with the high-field impurity breakdown in n-GaAs. We assume the impacting carrier to be an electron; however, all the formulation can be reiterated by starting with an impacting high-energy hole in SiO_2. Above the mobility edge that is well satisfied for an energetic electron in SiO_2 the bulk SiO_2 wave function will be of the Bloch form

$$\psi_b = \frac{1}{\sqrt{V}} u_k(\mathbf{r}) e^{i\mathbf{k}\cdot\mathbf{r}}, \tag{1}$$

whereas for the NC wave function we use a simple hydrogenic form [11],

$$\psi_n = \frac{\alpha_v^{3/2}}{\sqrt{\pi}} u_v(\mathbf{r}) e^{-\alpha|\mathbf{r}|}, \tag{2}$$

Some remarks will be in order, regarding the choice of these wave functions. Even though the embedding medium is usually an *amorphous* oxide, for high-field transport purposes well above the mobility edge, one can safely use crystalline states (i.e., Bloch functions) [9,10]. On the other hand, the use of hydrogenic wave function, which is well suited for the impurity problem was preferred solely due to its analytical convenience. The latter can be relaxed in case a closed-form expression is not aimed for.

Furthermore, we are neglecting the exchange interaction between the impacting electron and the valence NC electron due to huge energy difference between them [12]. The scattering matrix element that is due to the Coulomb interaction between the two electrons is given by

$$M = \int d^3\mathbf{r}_1 \int d^3\mathbf{r}_2 \frac{\alpha_c^{3/2}}{\sqrt{\pi}} u_c^*(\mathbf{r}_1) e^{-\alpha_c|\mathbf{r}_1|} \frac{1}{\sqrt{V}} u_{k'}^*(\mathbf{r}_2) e^{i\mathbf{k}'\cdot\mathbf{r}_2} \times \frac{e^2}{4\pi\varepsilon\varepsilon_0} \frac{e^{-\lambda|\mathbf{r}_1-\mathbf{r}_2|}}{|\mathbf{r}_1-\mathbf{r}_2|} \frac{1}{\sqrt{V}} u_k(\mathbf{r}_2) e^{i\mathbf{k}\cdot\mathbf{r}_2} \frac{\alpha_v^{3/2}}{\sqrt{\pi}} u_v(\mathbf{r}_1) e^{-\alpha_v|\mathbf{r}_1|},$$

yielding

$$|M|^2 = \left[\frac{64e^4\alpha_c^3\alpha_v^3\alpha^2}{(\varepsilon\varepsilon_0 V)^2}\right] |F_{cv}|^2 |F_{k'k}|^2 \times \frac{1}{[|\mathbf{k}-\mathbf{k}'|^2+\lambda^2]^2} \frac{1}{[|\mathbf{k}-\mathbf{k}'|^2+\alpha^2]^4}, \tag{3}$$

where

$$F_{cv} = \int_{\text{cell}} u_c^*(\mathbf{r}_1) u_v(\mathbf{r}_1) d^3\mathbf{r}_1,$$

$$F_{k'k} = \int_{\text{cell}} u_{k'}^*(\mathbf{r}_2) u_k(\mathbf{r}_2) d^3\mathbf{r}_2,$$

and $\alpha = \alpha_c + \alpha_v$. By using Fermi's golden rule, we can write

$$P(k) = \sum_{\text{NC}} \sum_{k'} \frac{2\pi}{\hbar} |M|^2 \delta\left[\frac{\hbar^2k^2}{2m_k} - E_v - E_c - E_g - \frac{\hbar^2k'^2}{2m_{k'}}\right] f_{\text{NC}}, \tag{4}$$

where E_g is that bandgap of the NC which is absorbed into the value of E_c. Here E_v is taken as positive hole energy. Taking $A = ((m_{k'}k^2)/m_k) - ((2m_{k'}E_v)/\hbar^2) - ((2m_{k'}E_c)/\hbar^2) - ((2m_{k'}E_g)/\hbar^2)$ and assuming

$$\sum_{\text{NC}} f_{\text{NC}} = N_{\text{NC}} = n_{\text{NC}} V, \tag{5}$$

where n_{NC} is the density per unit volume and in terms of the NC filling ration n_{NC} is

$$n_{\text{NC}} = \frac{f}{V_{\text{NC}}}, \tag{6}$$