Geometric broadening in resonant tunneling through Si quantum dots

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Abstract

The current through a resonant tunneling diode consisting of Si quantum dots embedded in a SiO$_2$ matrix is calculated and the resonance broadening effects caused by distributions of quantum dot diameter and asymmetric barrier thicknesses are simulated. It is demonstrated that a size distribution is extremely critical for the use of these structures as selective energy contacts for hot carrier solar cells, requiring precision at the order of 1 Å.

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1. Introduction

In conventional solar cells the dominant loss mechanism is relaxation of hot carriers, which a hot carrier solar cell (HCSC) [1-3] promises to avoid. Thereby a theoretical energy conversion efficiency limit of up to 85% can be approached [4]. An appropriate absorber material is required to suppress the relaxation of hot carriers in combination with selective energy contacts (SEC) to extract the charge carriers efficiently. The excited, unrelaxed charge carriers are extracted over a narrow energy range and thus their excess energy can be harvested prior to being lost to phonons. Therefore SECs have to provide thermal insulation between the absorber and the metal contacts as well as a very narrow band of allowed energies to minimize thermalization losses outside of the absorber. These conditions may be met by quantum dots (QD) embedded in a dielectric matrix, which feature discrete energy levels instead of bands. Such structures can either be used as SEC for HCSCs or as resonant tunneling diodes (RTDs) for very high speed logic [5]. A RTD requires narrow energy bands as well and therefore the suitability of a specific Si-SiO$_2$ system for SECs can be judged by its RTD performance, or more specifically by the sharpness of the resonant features.

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The band diagram of a RTD is shown in Fig. 1 for the specific system simulated. It consists of a degenerately doped $n^{++}$ Si source contact, Si QDs embedded in a SiO$_2$ double-barrier and an aluminum (Al) drain contact. The doping level of $n^{++}$ Si is adjusted to match the work function of Al (4.3 eV, [6] p. 137).

The typical current-voltage ($I$-$V$) characteristic [cf. Fig. 1(d)] is obtained if a negative voltage $V$ is applied to the source contact. If $V$ is increased the current increases until it reaches a maximum in the resonance case [$V = V_{R1}$, Fig. 1(b)]. Upon a further voltage increase less charge carriers find a state in the QD and the current decreases with increasing voltage [$V > V_{R1}$, Fig. 1(c)].

Ideally the resonance peaks in the $I$-$V$ characteristic are very sharp. Recently we reported on RTDs based on Si QDs embedded in a SiO$_2$ matrix fabricated by remote plasma enhanced chemical vapor deposition followed by high temperature annealing [7]. Features in the $I$-$V$ characteristic are clearly attributed to resonant tunneling and negative differential conductance is observed. However, the resonances are subject to substantial broadening, which, to a first approximation, is equivalent to SECs transmitting over wide energy ranges. One obvious source of broadening is temperature due to electron energies in the source contact obeying a Fermi distribution. This is minimized by measuring and simulating at 10 K. Further resonance broadening is attributed, but not limited to several non-idealities:

- Scattering: Electron energies can be changed by scattering so that tunneling into discrete QD states is allowed even if the applied voltage is off resonance, which broadens the resonance peaks.
- Size distribution (SD): The current between macroscopic contacts is averaged over many QDs with different sizes and different energy levels. Consequently the resonance condition is met by different QDs over certain voltage ranges.
Asymmetric barrier distributions (ABD): QDs may not be centered within the barrier layer, giving rise to an asymmetry between source and drain barrier thicknesses. As a consequence the voltage at which the affected QDs meet the resonance condition is changed.

In this paper we demonstrate how a QD SD and a distribution of asymmetric barriers contribute to the resonance broadening of a QD based RTD and discuss the applicability of our results to SECs. We simulate a Si-SiO$_2$ system, but our method can easily be applied to other materials by adjusting the parameters.

2. Calculation of $I-V$ curves

The $I-V$ dependence of individual QDs is calculated using the nonequilibrium Green's function method [8-12]. The details of our specific implementation and the simulation of the geometry features will be published elsewhere [13]. The QDs are modeled in 3D for accurate energy levels. Phase breaking scattering is included phenomenologically by means of a Büttiker probe [8,10,14].

The parameters used in all calculations are the following: the band structure is defined as shown in Fig. 1, the temperature is 10 K, the $n^{++}$ Si Fermi level is 0.01 eV above the conduction band edge, the QD conduction band offset $\Delta E_C$ is set to 0 and the low temperature density of states effective electron mass of Si is 1.06 $m_0$ [15]. The barriers are spherically symmetric abrupt barriers of 3.2 eV, corresponding to the offset between the conduction bands of Si and SiO$_2$ ([6] p. 790).

![Figure 2. Calculated $I-V$ curves for a 2.7 nm QD with symmetric barriers and varying amounts of scattering. Curves normalized at 0.75 V.](image)

An exemplary $I-V$ dependence of a single QD is shown in Fig. 2, calculated for a QD with a diameter of 2.7 nm and symmetric barriers. Without scattering the resonance peaks are sharp, being limited only by the intrinsic linewidth (full width at half maximum $\Delta E < 1$ meV) and the electron distribution in the source contact determined by the Fermi level ($\Delta E \approx 8$ meV). Adding Büttiker probes with a moderate scattering strength mainly increases the valley current and flattens the voltage dependence ($\Delta E \approx 8$ meV). Increasing the scattering strength by a factor of 10 reveals the peak broadening effect clearly ($\Delta E \approx 20$ meV). The smaller scattering strength is used in all further calculations to retain the distinct peak structure.

3. Influence of sample geometry on $I-V$ curve

To investigate the effects of geometric fluctuations an averaging process involving many QDs is used. The overall broadening is determined by these geometric features rather than the exact shape of the resonant peaks of single QDs. Although an entire ensemble is simulated, each calculation features a single QD and thus a minimum distance between the dots within the insulating matrix is assumed and interactions between QDs are neglected.
3.1. Asymmetric barriers

The influence of ABDs [Fig. 3(b)] on the $I$-$V$ characteristic is simulated at very high energy and voltage resolutions. It was found that a shift of the QD position relative to the contacts changes the voltage drop $V_{B1}$ prior to the QD state and therefore shifts the peak positions, whereas peak amplitude and shape are essentially unaffected. Hence the $I$-$V$ curve of QDs with asymmetric barriers can be simulated in a computationally efficient post processing step by calculating the data for QDs centered within the layer (i.e. with symmetric barriers) and scaling the voltage axis according to the change in $V_{B1}$. Quasi continuous variation of the barrier thicknesses is enabled, which is much more flexible than changing the number of barrier grid points. To represent a distribution of asymmetric barriers each QD is described by its center position within the layer stack. A distribution of these positions is defined, the voltages are scaled according to the position dependent values of $V_{B1}$ and the superposition of the current through all QDs is calculated.

The results shown are based on a 2.0 nm QD centered in the middle of an 8.0 nm layer. In Fig. 3(a) $I$-$V$ curves for three Gaussian ABDs with varying full width at half maximum (FWHM) are plotted. It is apparent that the asymmetry broadens the peaks and thereby strongly reduces the peak to valley current ratio (PVCR) but does not change the peak positions. The PVCR values of all configurations shown in Fig. 3 and 4 are listed in Tab. 1.

Figure 3. QDs displaced within the stack, i.e. with asymmetric barriers, illustrated in (b). Calculated $I$-$V$ curves for QD ensembles with Gaussian ABDs centered in the middle of the stack at 4.0 nm and with varying FWHM in (a) and distributions plotted in (c).
Table 1. Broadening due to different geometric features. PVCR₁ and PVCR₂ are the peak to valley current ratios for the first and second resonance peak. "ABD, FWHM" refers to the FWHM of the asymmetric barrier distributions shown in Fig. 3 and "SD, FWHM" to the FWHM of the SDs used for Fig. 4. Values for a single QD with default scattering strength (Fig. 2) are provided for reference.

<table>
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<th>PVCR₁</th>
<th>PVCR₂</th>
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3.2. Size distribution

In order to take a SD into account I-V curves of individual QDs with different diameters are calculated, weighted by a distribution function and summed up. We keep the number of lattice points constant and change the lattice resolution to yield arbitrary QD diameters. The overall layer thickness is adjusted to a constant value in a post processing step using a thickness calibration based on the assumption of direct tunneling dominated transport through the SiO₂ barriers at the low temperature of 10 K and voltages too small for Fowler-Nordheim tunneling ($qV_{\text{max}} \ll \Phi_0$). I-V curves of 80 QDs with diameters ranging from 1.5 to 3.5 nm and a total layer thickness of 8 nm are calculated and all other parameters are unchanged.

Larger QDs feature an exponentially increased current as they are surrounded by thinner barriers due to the constant layer thickness. Therefore the larger QDs in a distribution dominate the I-V curve, which causes the shift in the peak positions in Fig. 4, where the QD SD is centered at 2.0 nm and the FWHM is varied. The reduction of the peak amplitude upon an increase of the FWHM from 0.1 to 0.5 nm is caused by a smaller number of QDs per diameter due to the normalization for a constant number of QDs (see inset of Fig. 4). Furthermore strong peak broadening due to the SD is apparent. Upon an increase of the FWHM from 0.1 to 0.3 nm the PVCR is dramatically reduced from 2950 to 21 and at a FWHM of 0.5 nm the current valleys almost disappear.
4. Conclusions and consequences

This calculatory investigation defines the necessary precision in size and location QDs have to have in the Si-SiO₂ system if HCSCs with SECs are to become real operative devices. Specifically a SD with a FWHM < 0.1 nm (5%) is demanded for SECs to provide significant energy selectivity, corresponding to large PVCRs. These tantalizing requirements derived exclude nearly all known $kT$-related growth phenomena of QDs. This tyranny of SD can partially be relieved by switching to a different QD material with a smaller effective mass, which enables larger QDs to yield the same amount of confinement. However, for a successful implementation of HCSC concepts with Si based SECs alternative self organization processes have to be developed where the required size control is provided by novel physical principles.

On the other hand an ABD broadens the resonance peaks by changing the voltage drop $V_{B1}$. In a HCSC there is no voltage drop over the SEC and therefore its operation is not influenced by an ABD. Systems of QDs embedded in a dielectric can thus be more suitable for SECs than their low temperature RTD performance suggests.

Acknowledgement

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