Theory of quasiballistic transport through nanocrystalline silicon dots

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A model to describe the underlying physics of high-energy electron emission from a porous silicon diode is presented. The model is based on an atomistic tight-binding method combined with semiclassical Monte Carlo simulation. It well reproduces essential features of experimental findings. An initial acceleration region is shown to play a crucial role in generating quasiballistic electron emission. © *2011 American Institute of Physics*. [doi:10.1063/1.3553501]

Nanocrystalline silicon (nc-Si), crystalline silicon with a size below several nanometers, shows characteristic of electronic and photonic effects originated in strong quantum confinement of electrons. These effects have been applied to numerous device applications.[1,2] One of the important applications is a surface-emitting diode,[3] whose active region consists of porous Si (PS) comprising treelike network of nc-Si dots. When high voltage is applied to a PS diode, electrons are quasiballistically emitted from the diode surface. In spite of the fact that high-energy electron emission from a PS diode is clearly observed, the detailed mechanism has not been fully understood and explained. One of the difficulties associated with theoretical analysis of the high-energy electron emission is due to coexistence of the strong quantum confinement in nc-Si dots and semiclassical hot-electron transport under high bias condition. In this letter, we present a model[4] to capture the essential experimental features of the quasiballistic electron emission. Our model is based on an atomistic tight-binding method combined with semiclassical Monte Carlo simulation, which enables us to treat the quantum confinement and the high-energy transport in a unified manner.

We consider a PS diode whose schematic diagram is given in Fig. 1(a). The active region of ~ 1 μm in length consists of nc-Si dots connected through thin SiO_2 layers. The typical size of a nc-Si dot is ~ 4 nm in diameter, and the oxide thickness is ~ 1 nm. When a positive bias voltage V_{ps} is applied to the thin Au electrode with respect to the n-type Si substrate, electrons are emitted from the surface. As schematically shown in Fig. 1(b), major voltage drop occurs across the PS region due to its high resistivity. The energy distribution of the emitted electrons has been measured in various device structures with different fabrication processes. From the measured distribution and by considering the Au work function, the energy loss of the emitted electrons, E_{loss}, has been obtained, which indicates that electrons quasiballistically travel through the active region.

The experimental features of the energy loss, E_{loss}, described in Refs. 1 and 5 can be summarized as follows: (1) E_{loss} (≈ 10 eV) is nearly independent of V_{ps} for 10 V $\lesssim V_{ps} \lesssim 30$ V; (2) E_{loss} can vary from sample to sample but is a virtually constant parameter of each individual device; and (3) E_{loss} becomes smaller when temperature is lowered from room temperature to low temperature ($T=100$ K).

Our model for the quasiballistic transport through a PS diode consists of three parts: (1) the atomistic tight-binding calculation of electronic states and phonon scattering rates in a single nc-Si dot; (2) the Kronig–Penny model for tunneling between neighboring nc-Si dots; and (3) the Monte Carlo simulation of electron dynamics inside a diode.

Since electrons are strongly confined in a nc-Si dot, the electronic states should be calculated by taking into account the full-band structure of Si. We use an empirical $sp^3s^*d^5$ tight-binding method[6] to obtain the energy levels E_i ($i=1,2,\ldots$) in a nc-Si dot. We assume that the dot has a spherical shape. Figure 2(a) shows E_i of a nc-Si dot with a diam-

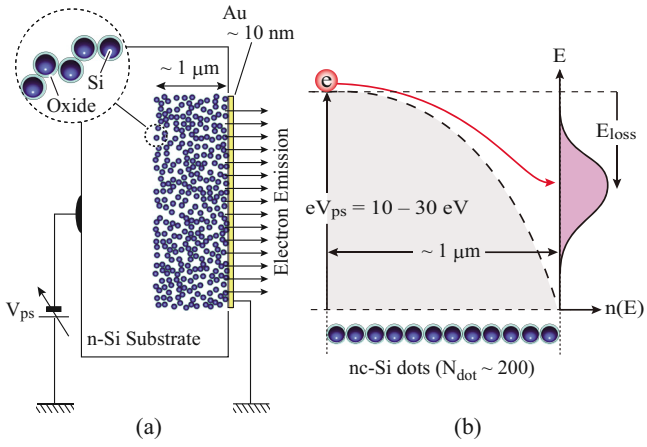

FIG. 1. (Color online) (a) Schematic illustration of the porous silicon surface-emitting diode. Electrons are emitted from the surface under positive bias V_{ps}. (b) Conduction band profile (dashed line) and electron distribution at the diode surface, $n(E)$. The energy loss, E_{loss}, is the energy difference between the substrate Fermi level and the peak energy of the electron distribution $n(E)$.

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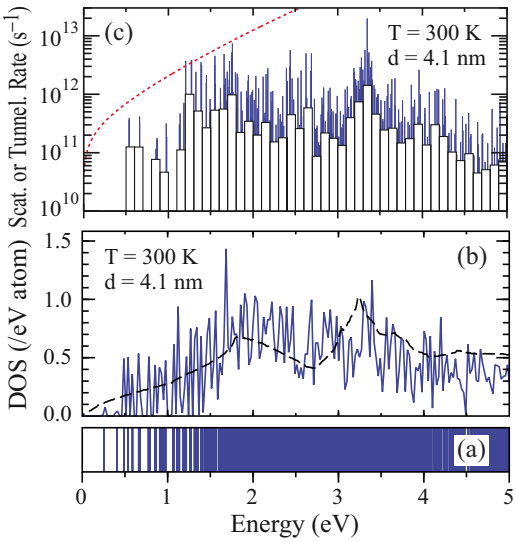

FIG. 2. (Color online) (a) Energy levels of a nc-Si dot with a diameter of d=4.1 nm. (b) Density of states of the nc-Si dot (solid line) and bulk Si (dashed line). (c) Optical-phonon emission rate (vertical lines). The box shows an average over a 100 meV interval. Tunneling escape rate between neighboring nc dots is also plotted for comparison (dashed line).

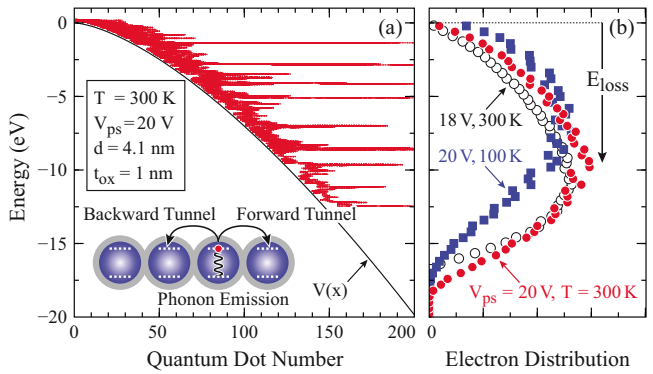

FIG. 3. (Color online) (a) An example of electron trajectories (dots), together with the potential profile (solid line). The inset shows the schematic illustration of the Monte Carlo simulation. (b) Electron distribution at the diode surface for a nc-Si dot array with d=4.1 nm and t_{ox}=1 nm.

eter of d=4.1 nm. The Si/SiO_2 interface is treated with the H termination model.[7] The quantum confinement makes the level separation larger for lower energy region $E \lesssim 1.2$ eV. However, the level separation becomes quite small ($\Delta E < 10$ meV) for higher energy region. As will be shown later, this emergence of the two regions of the low-energy discrete and the high-energy quasicontinuous regions plays an essential role in generating the electron emission from a PS diode.

The density of states per Si atom is calculated from E_i as $g(E)=(2/N)\Sigma_i \delta(E-E_i)$. Here, N is the number of Si atoms in a nc-Si dot. This $g(E)$ diverges at $E=E_i$. In actual systems, scatterings will remove the divergences in $g(E)$. For nc-Si dots, acoustic-phonon scattering is considered to be a dominant process causing the collisional broadening, and the level broadening, Γ, is given by $\Gamma=D_{ac}(kT/\rho s_l^2 \Omega_D)^{1/2}$. Here, D_{ac} is the acoustic deformation potential, kT is the thermal energy, ρ is the mass density, s_l is the sound velocity, and Ω_D is the volume of a nc-Si dot. Γ is introduced by replacing the delta function in $g(E)$ with $\exp[-(E/\Gamma)^2]/(\sqrt{\pi}\Gamma)$. $g(E)$ at T =300 K is shown in Fig. 2(b) together with the density of states of bulk Si for comparison. The strong quantum confinement makes $g(E)$ discrete for the lower energy region. On the other hand, $g(E)$ for higher energy shows a quasicontinuous nature and resembles bulk Si density of states, suggesting the fact that upper states are less affected by the quantum confinement.

The optical-phonon emission rate, W_i, for an electron in the ith energy level is calculated within Fermi's golden rule and is given by $W_i=(\pi D_{op}^2/2M\omega_0)g(E_i-\hbar\omega_0)$, where D_{op} is the optical deformation potential, ω_0 is the optical-phonon frequency, and M is the mass of a Si atom. Since D_{op} for high-energy electrons can be different from that for electrons near the conduction band edge, we estimate D_{op} from an energy shift δE_i induced by lattice displacement δa with $D_{op}=\delta E_i/\delta a$ for each energy level i. Figure 2(c) shows W_i for a nc-Si dot with d=4.1 nm at T=300 K. Optical-phonon emission is suppressed for the lower energy region because of the discrete nature of the energy levels.[8] Note that the energy levels with $W_i < 10^{10}$ s^{-1} do not appear in Fig. 2(c).

Dwell time, τ_d, in a nc-Si dot is calculated by using a one-dimensional Kronig–Penny model.[9] It provides a miniband width Δ, which is then converted to τ_d as $\tau_d=4\hbar/\Delta$. Calculated tunneling escape rate, τ_d^{-1}, is plotted in Fig. 2(c). For the lower energy region, τ_d is rather long ($\tau_d \gtrsim 10^{-12}$ s) because of the small electron energy compared to the SiO_2 barrier height (U=3 eV). The suppression of the optical-phonon scattering, however, makes W_i much smaller than τ_d^{-1} for that energy region. We thus expect high probability for an electron in the lower energy region of a nc-Si dot to tunnel out the dot before relaxing to lower energy levels. This initial acceleration plays a crucial role as will be shown later. For higher energy, τ_d^{-1} becomes much larger than W_i and approaches the ballistic limit of $\tau_d^{-1}=v_{free}/L$, where v_{free} is the free electron velocity and L is the periodicity of a nc-Si dot array.

The Monte Carlo calculation has been done to simulate electron dynamics in a PS diode. An electron in a nc-Si dot has a probability to emit an optical phonon and a probability to escape from the dot via tunneling. Using W_i and τ_d^{-1} obtained above, the electron motion can be simulated. An example of the electron trajectories is plotted in Fig. 3(a), with the potential profile, $V(x)$, used for the simulation. $V(x)$ is assumed to be of the form $\propto x^{3/2}$ as in the space-charge-limited operation.[10] We considered the case that 200 nc-Si dots are in an array. We find clear quasiballistic nature in the electron distribution $n(E)$ at the diode surface [Fig. 3(b)]. Note that the distribution does not include effects of the Au work function.

Energy loss, E_{loss}, is calculated from the half-height point of the cumulative distribution. Figure 4(a) shows the applied voltage V_{ps} dependence of E_{loss}. We find that E_{loss} is almost independent of V_{ps}. This is consistent with the experimental findings. The energy loss can be attributed to energy dissipation to optical phonons when electrons are in the initial acceleration region. This leads to a constant energy loss of being independent of V_{ps}. E_{loss} shows weak dependence on the dot diameter, d, and becomes minima at around d =4 nm [Fig. 4(b)]. For smaller d, τ_d becomes shorter because of larger subband energy. This results in the increase of E_{loss} when d becomes small. On the other hand, for larger d, the subband separations become smaller leading to stronger phonon emission rate. This results in the increase of E_{loss} when d becomes large. Note that in an actual device structure

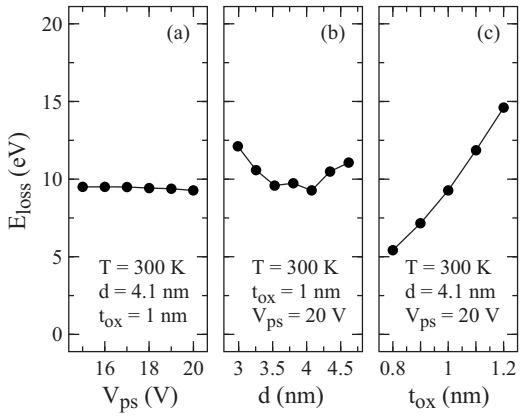

FIG. 4. (a) Energy loss, E_{loss}, as a function of applied voltage, V_{ps}, for a nc-Si dot array with d=4.1 nm and t_{ox}=1 nm at T=300 K. (b) Dot diameter dependence. (c) Oxide thickness dependence.

dot-size fluctuations are always present. They will, however, not significantly affect the present results because electrons sequentially tunnel between neighboring nc-Si dots in the present model. For analyzing experimental results, one may have to regard the parameter d of this letter as a kind of an average value of actual dot sizes in the initial acceleration region. The oxide thickness, t_{ox}, has a strong impact on E_{loss} as shown in Fig. 4(c). When t_{ox} becomes thicker, electron dwell time increases, resulting in more phonon emission and larger E_{loss}. These dependences of E_{loss} on t_{ox} and d may cause the observed E_{loss} differences among different samples.

Temperature dependence of emitted electron distribution can also be explained in our model. The level broadening Γ due to acoustic-phonon scattering becomes smaller at lower temperature. For example, Γ=7 meV at 300 K and Γ =4 meV at 100 K for d=4 nm. This small change in Γ affects $g(E)$, especially for the lower energy region, because of the discrete nature of the energy levels. This, in turn, modifies and reduces W_i of the low energy region, resulting in enhancement of the initial acceleration and reduction of E_{loss} at lower temperature [see Fig. 3(b)].

In summary, we have performed the Monte Carlo simulation of electron motion in a PS diode to clarify the underlying physics of the high-energy electron emission. The results clearly show that electrons are quasiballistically emitted from a diode. We find that the initial electron acceleration, which is enhanced in a nc-Si dot due to the discrete nature of the lower energy levels, plays an essential role in generating high-energy electron emission.

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