Interaction effects on transport in a 1d array of silicon nanoclusters

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Abstract

The effect of Coulomb interactions on charge transport in a model of light emission from an array of silicon nanoclusters is studied. The array is sandwiched between a p-type and an n-type doped silicon crystals, and electrons and holes are driven into the array by an applied electric field. Radiative recombinations of electrons and holes take place around the center of the array producing the emission of red light. The total emission power is approximately proportional to the current injected into the system. It is found that carrier-carrier interactions play a crucial role for charge transport. Specifically, the self-interaction of charges inside a nanocluster drastically limits the current, yielding a strong non-lineal behavior between current and density of free carriers.

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I. MOTIVATION

Most microelectronics devices are made of silicon, so this material should be the natural choice for anyone designing new devices that will be integrated with microelectronic circuits. Replacement of metal by silicon optical connectors is an attractive alternative because they promise to eliminate of power dissipation problems that appear using metal connectors. However, the electronic structure of silicon does not allow it to emit light readily and efficiently. This has been partially achieved using porous forms of silicon when illuminated with an external UV light source. Today, the main goal is to produce light efficiently from the injection of charges into silicon based devices without to use of such additional light sources (see, e.g. [1–8]).

In a recent work, a prototype of a silicon nanocluster array has been introduced and studied theoretically [9]. The model consists of an assembly of silicon nanoclusters sandwiched between p- and n-type doped silicon crystals, which act as charge reservoirs of holes and electrons, respectively. The carriers are driven by an applied electric field into the silicon array where they recombine (producing red light when radiative recombinations take place). It was found that the efficiency of the light emission is quite large, between about 2% and 0.5% for fields ranging from 100 to 500 kV/cm, respectively, at the temperature $T = 300$ K. The light emission takes place near the center of the array, where nanoclusters of linear size of about 3.6 nm are located, and the total emission power is approximately proportional to the total current injected into the system.

Due to the small size of the nanoclusters, two different effects appear. One effect is quantum-confinement which allows visible light emission with rather large yield. For example, a silicon nanocluster of diameter 3.6 nm has a yield of 1/3 [6] (i.e., on average, for every three electron-hole recombinations one photon is produced) and light emission is in the visible red. A second effect is the existence of unscreened Coulomb self-interaction between charges inside a nanocluster. This interaction is not present in ordinary crystalline semiconductors because charge confinement does not take place in the bulk. Thus, we must deal with this type of interaction in the study of charge transport in nanocluster arrays.

The main purpose of the present work is to study, from a basic theoretical point of view, the effects of Coulomb interactions on charge transport in the array of nanoclusters introduced in Ref. [9]. This study is also related to the total power light emission since

current and emission power are approximately proportional to each other.

II. ARRAY GEOMETRY AND DIFFERENT INTERACTION MODELS

The geometry of the array considered here is illustrated in Fig. 1. The p and n boxes at the border represent the p- and n-type doped silicon crystals. The circles symbolize the one-dimensional chain of nanoclusters of different sizes (larger at the ends and smaller at the center of the array).

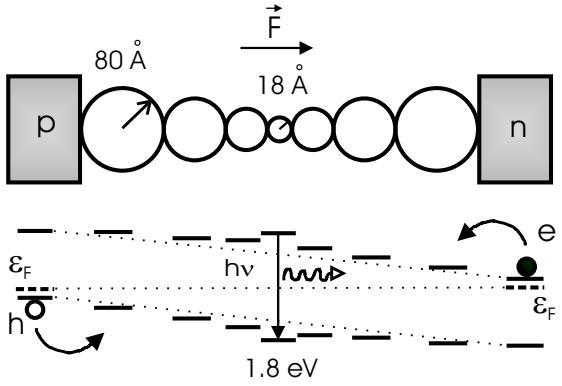

FIG. 1: Schematic illustration of the one-dimensional array of Si nanoclusters. The lower part of the figure illustrates the corresponding energy levels of the neutral system in the absence of an external field. The relation between optical gap E and radius R of each nanocluster is given by Eq. (1). The optical gaps in the boxes take their bulk value 1.17 eV. In the stationary state, most of the photons (of ≈ 1.8 eV) will be emitted from the center of the system. The Fermi energy of the array, $\varepsilon_{\rm F}$, is indicated by the short dashed line. The uppermost arrow indicates the direction of the applied electric field, $\vec{F}$, pointing in the forward bias direction to bring electrons e (small filled circle) and holes h (small open circle) from the borders to the center of the system. The dotted lines are drawn to guide the eye.

In intrinsic silicon nanoclusters the optical gap E between electron and hole energy levels depends on its size approximately as,

$$R(E) = \frac{13\,({\rm eV})^{0.72}}{(E - E_{\rm Bulk})^{0.72}}\,{\rm Å}. \qquad (1)$$

Electron-hole recombinations are assumed to take place when both the electron and the hole are present inside the same nanocluster. The recombination can be radiative (producing a

photon) or non-radiative (producing a phonon). For E in the range 1.2 eV $< E <$ 2 eV, and at a temperature $T = 300$ K, the radiative recombination time is estimated to be [12]

$$\tau_{\rm rad}(E) = \frac{7.7 \times 10^{-4}\,({\rm eV})^{1.38}}{(E - E_{\rm rad})^{1.38}}\,{\rm s}, \tag{2}$$

where $E_{\rm rad} = 1.137$ eV. From [6] the non-radiative recombination time, $\tau_{\rm nrad}(E)$ can be calculated (for more details see [9]) as

$$\tau_{\rm nrad}(E) = \frac{Y(R)}{1 - Y(R)}\tau_{\rm rad}(E), \tag{3}$$

where the photoluminescence yield, $Y(E) \equiv \tau_{\rm nrad}/(\tau_{\rm rad} + \tau_{\rm nrad})$, behaves as $Y(E) = 4.4\exp(-2R/14\text{Å})$, for $R \geq 18$ Å.

From Eqs. (1)-(3) and for a given value of the gap E, one obtains the radius R of the nanocluster and its radiative and non-radiative recombination times, $\tau_{\rm rad}$ and $\tau_{\rm nrad}$.

In the models, hopping of carriers is studied by Monte Carlo simulations. Hopping processes occur between nearest-neighbor NC, or between a box and its neighboring NC. The corresponding hopping time is taken as $\tau_{\rm hop} = 5 \times 10^{-3}$ s. If a particle (electron or hole) is selected, say at site i, it can either hop to one of the two neighboring sites or remain at i. A hop occurs with a probability

$$\begin{aligned} &p_{i\to j} = p_{\rm hop}\,\exp(-\beta\Delta E_{\rm tot}), \quad \text{for} \quad \Delta E_{\rm tot} > 0, \quad \text{and} \\ &p_{i\to j} = p_{\rm hop}, \quad \text{otherwise}, \end{aligned} \tag{4}$$

where $p_{\rm hop} = \tau_0/\tau_{\rm hop}$, τ_0 is the time unit, $\Delta E_{\rm tot} = E_{\rm tot}^{\rm (after)} - E_{\rm tot}^{\rm (before)}$ the variation of the total energy of the system for the attempted jump from site i to j, and $\beta = 1/K_{\rm B}T$.

The $E_{\rm tot}$ is given by

$$\begin{aligned} E_{\rm tot}(\{n_{\rm e}, n_{\rm h}\}) = &\sum_{i=1}^{N}\left[\left(\frac{E_i}{2} + \frac{E_{\rm Bulk}}{2L_{\rm NC}}\left(L_{\rm NC} - 2x_i\right)\right)n_{\rm e}(i) + \left(\frac{E_i}{2} + \frac{E_{\rm Bulk}}{2L_{\rm NC}}\left(2x_i - L_{\rm NC}\right)\right)n_{\rm h}(i)\right] \\ &+E_{\rm Bulk}(n_{\rm e}(0) + n_{\rm h}(N+1)) + \sum_{i=1}^{N}\sum_{j=i}^{N}V_{i,j} - \sum_{i=1}^{N}Fx_iq_i - eU(n_{\rm h}(N+1) - n_{\rm e}(N+1)) \,, \end{aligned} \tag{5}$$

where the first two terms account for the energy of the particles occupying site i with respect to the Fermi level, the third term represents the total Coulomb interaction, and the last two terms the potential energy of carriers due to the external field.

The effective Coulomb interaction between the net charges q_i at x_i and q_j at x_j is given by

$$V_{i,j} = \frac{q_iq_j}{\varepsilon_{\rm eff}|x_i - x_j|} \;, \qquad \text{for} \quad i \neq j \;, \tag{6}$$

where $q_i = e(n_{\rm h}(i) - n_{\rm e}(i))$, and $n_{\rm h}(i)$, $n_{\rm e}(i)$ are the occupation numbers of h and e at the ith NC ($1 \leq i \leq N$), and $e > 0$ is the unit electric charge. To be noted is that the occupation numbers $n_{\rm h}(0)$ and $n_{\rm e}(N+1)$ (i.e. the sources of electric charges in the boxes) are kept constant all over the simulations. Here we use $N = 400$.

Inside a single NC, the self-interaction of charges is estimated as

$$V_{i,i} = \frac{q_i^2}{\varepsilon_{\rm eff} R_i}. \tag{7}$$

We take $\varepsilon_{\rm eff} = 2$ which is close to the value of the dielectric constant in porous Si [14].

In order to study the effects of the Coulomb interaction, the following three different models have been studied using Monte Carlo simulations:

Model A: In this model we use the complete Hamiltonian given by Eq. (5) where the terms $V_{i,j}$ are present for all i and j. (i.e., the full Coulomb interactions are present).

Model B: In this model we use the Hamiltonian (5) with all $V_{i,j} = 0$ (i.e., without Coulomb interaction).

Model C: In this model we use the Hamiltonian (5) with $V_{i,j} = 0$ for all i different from j ($i \neq j$) (i.e., only Coulomb self-interactions are present).

III. RESULTS AND CONCLUSIONS

For illustrative purposes, we consider here the case in which $n_{\rm h}(0) = n_{\rm e}(N+1) = 8$. For models A, B and C the results of the current density J as a function of the electric field F are shown in Fig. 2. In the limit $F \to \infty$ all the $n_{\rm h}(0)$ and $n_{\rm e}(N+1)$ in the p and n-boxes, respectively, jumps to the array in a time $\tau_{\rm hop}$. Then the maximum value $J_{\rm max} = e n_{\rm h}(0)/\left(d_0^2 \tau_{\rm hop}\right) = 6.4 \times 10^{-5}$ A/cm^2 is reached (d_0 is the size associated to the p- and n-type semiconductors). From Fig. 2 one can see that this value is obtained for the three models and large enough values of F.

Large difference between the values of J for the cases with (model A and C) and without (model B) Coulomb interaction are obtained. Specially in the region of $F < 800$ kV/cm which corresponds to experimentally realistic values of fields. For example, for $F = 100$ kV/cm one obtains $J = 0.92 \times 10^{-6}$ A/cm^2, and $J = 49.4 \times 10^{-6}$ A/cm^2 for models A and B, respectively (i.e., the ratio between them is greater than one order of magnitude). And for $F = 500$ kV/cm one obtains $J = 5.7 \times 10^{-6}$ A/cm^2, and $J = 63.8 \times 10^{-6}$ A/cm^2

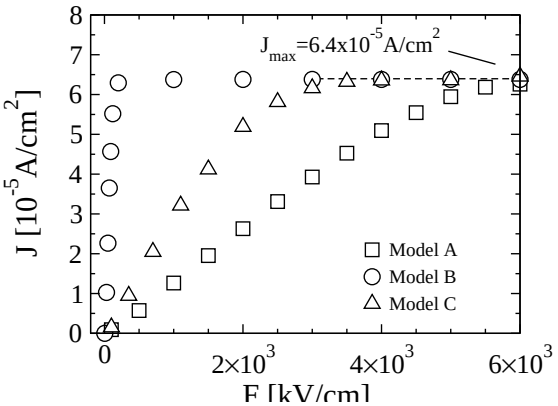

FIG. 2: Current density J versus applied electric field F. The maximum value $J_{\max}$ is obtained when the $n_{\rm e}(N+1) = 8$ electrons and the $n_{\rm h}(0) = 8$ holes hop in the time $\tau_{\rm hop}$ from n- and p-boxes to the array, respectively.

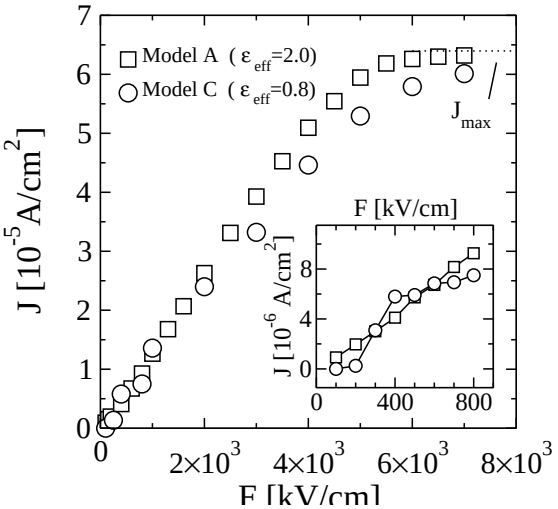

FIG. 3: Current density J versus applied electric field F for models A and C. The data were obtained using $\varepsilon_{\rm eff} = 2.0$ for model A and $\varepsilon_{\rm eff} = 0.8$ for model C (see Eqs. (6), (7), and (5)). The inset shown the same results but for F ranging from 100 to 800 kV/cm.

for models A and B, respectively. Then, the carrier-carrier interactions must be taken into account in realistic simulations of charge transport in nanoclaster arrays. Specifically, these interactions can be considered in two terms, the long-range Coulomb interaction (see Eq. (6)

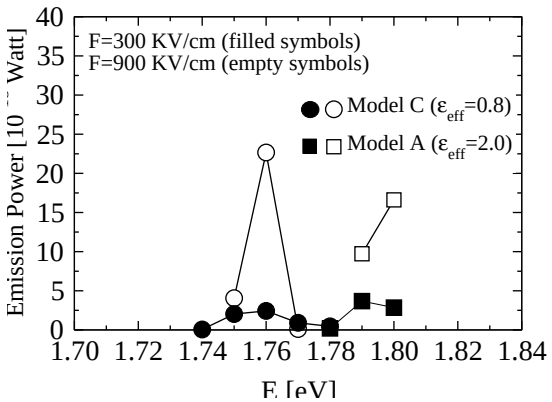

FIG. 4: Light emission spectrum of the NC array versus energy E for model A (squares) and model C with $\varepsilon_{\rm eff} = 0.8$ (circle). These results were obtained for two different values of F: $F = 300$ kV/cm (filled symbols), and $F = 900$ kV/cm (empty symbols). The total emission in each case is found to be: $P_{\rm ph} = 6.73 \times 10^{-18}$ W (filled squares), $P_{\rm ph} = 5.88 \times 10^{-18}$ W (filled circles), $P_{\rm ph} = 2.68 \times 10^{-17}$ W (empty squares) and $P_{\rm ph} = 2.63 \times 10^{-17}$ W (empty circles). Although the total emission for the same value of F are similar the spectrum is shifted to the red when the terms $V_{i,j} = 0$ for $i \neq j$.

and the Coulomb self-interaction (see Eq. (7)). The second term is much important than the first one, because:

(a) If the Coulomb self-interaction is considered, the effects of $V_{i,j}$ for $i \neq j$, are of minor importance. The results obtained from model C are not too much different from those obtained from model A. Moreover, one can obtain similar results neglecting the long-range interaction term, and using a modified value of $\varepsilon_{\rm eff}$ in Eq. (7) (see Figs. 3 and 4).

(b) If the self-interaction is neglected and also the long-range interactions are not taken into account, one has the model B. And the results obtained from model B and A are quite different (see Fig. 2). The Coulomb self-interaction limits the injection of carriers into the nanocluster array from the p- and n-boxes.

One concludes that the Coulomb self-interaction plays a crucial role in charge transports in nanocluster arrays.

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