Comment on "New Scaling of Child-Langmuir Law in the Quantum Regime"

In their Letter on the quantum Child-Langmuir law, Ang *et al.* [1] include exchange-correlation effects within the Kohn-Sham density functional theory (DFT) [2] and explore numerically the maximum transmitted current ($J_{\max}$) in nanogaps. We show here that the calculations are in error as the exchange-correlation component of the chemical potential has been ignored while fixing the boundary conditions for the Hartree potential.

The analysis in [1] is based on solving the time-independent Schrödinger equation $-d^2\psi/dx^2 + V_{\text{eff}}\psi = E\psi$ with an effective potential energy, $V_{\text{eff}} = -eV + V_{xc} \times E_H$, where $E_H = e^2/(4\pi\epsilon_0 a_0)$ is the Hartree energy, a_0 the Bohr radius, $V_{xc} = \epsilon_{xc} - (r_s/3)d\epsilon_{xc}/dr_s$, $r_s = [3/(4\pi n)]^{1/3}$ is the Wigner-Seitz radius, n the electron number density, and e the magnitude of the electronic charge. In the above, ϵ_{xc} is the exchange-correlation energy density within the local density approximation, and V is the Hartree potential satisfying the Poisson equation $d^2V/dx^2 = en(x)/\epsilon_0 = e|\psi(x)|^2/\epsilon_0$ with boundary conditions $V(0) = 0$ and $V(D) = V_g$, where V_g is the gap voltage difference and D the gap size. These equations are solved in a nanogap with the wave function ψ and its derivative matched at the collector boundary under certain assumptions (Ref. [3] addresses one of these).

We first note the applied voltage difference, $V_g = -(\mu_C - \mu_E)/e$, where μ_C and μ_E refer, respectively, to the chemical potential at the collector and injection planes. For convenience, we consider the reference as $\mu_E = -eV(0) + V_{xc}(0) \times E_H = 0$ so that $E = 0$ refers to injection from the Fermi level. Thus, $V(0) = V_{xc}(0) \times E_H/e$. It follows that the chemical potential at the collector is $-eV(D) + V_{xc}(D) \times E_H = -eV_g$. Thus, $V(D) = V_g + V_{xc}(D) \times E_H/e$. In writing the above, we have implicitly assumed continuity of the chemical potential at the interfaces under steady-state conditions. Note that when exchange correlation is neglected altogether, the boundary conditions for V are $V(0) = 0$ and $V(D) = V_g$, respectively, as assumed in [1].

When V_{xc} is substantial, and the boundary conditions chosen are $V(D) = V_g$ and $V(0) = 0$, the results can be unphysical. As an example, consider the case $D = 1$ nm, $V_g = 0.1$ V, and $E = 0$. Using the formalism of Ref. [1], $J_{\max}$ turns out to be $J_{\max} \simeq 278 J_{\text{CL}}$, where J_{CL} is the classical Child-Langmuir current density. The corresponding effective potential energy is shown in Fig. 1. Note that V_{eff} is negative everywhere. The injection energy E has clearly no relation to the effective potential at an either end of the nanogap.

The corrected boundary conditions for V lead to $J_{\max} \simeq 3.4 J_{\text{CL}}$. The corresponding effective potential is shown in Fig. 1 (bottom). The error in determining the quantum Child-Langmuir law is therefore substantial.

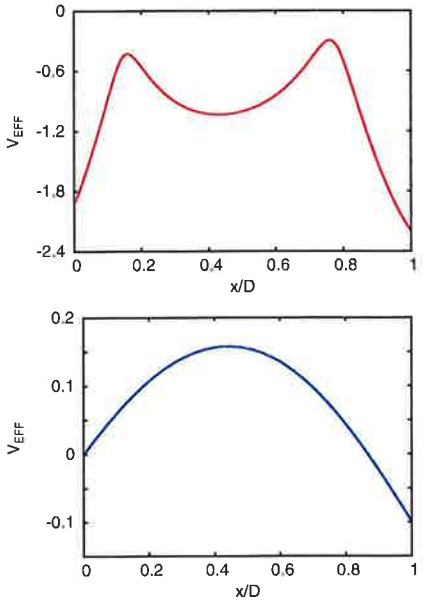

FIG. 1 (color online). The effective potential energy $V_{\text{eff}} = -eV + E_H \times V_{xc}$ for $D = 1$ nm, $V_g = 0.1$ V (top) $V(0) = 0$, $V(D) = V_g$ and $J_{\max} \simeq 278 J_{\text{CL}}$ (bottom) $V(0) = V_{xc}(0) \times E_H/e$, $V(D) = V_g + V_{xc}(D) \times E_H/e$, and $J_{\max} \simeq 3.4 J_{CL}$. The boundary conditions for ψ are the same as in Ref. [1] in both cases.

In regimes where V_{xc} is negligible (large V_g or D), $V(D) = V_g$ and $V(0) = 0$ are approximately the correct boundary conditions within the formalism of Ref. [1] as we have verified for $D = 50$ nm, $V_g = 50$ V, and $E = 0$.

We note in conclusion that models with additional inputs such as the emission mechanism or the image potential, may also require an appropriate interpretation of the chemical potential for the determination of the boundary conditions for the Hartree potential.

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Ang Replies: In the preceding Comment [1], the author raises concerns regarding to the boundary conditions assumed in our previous published model regarding the scaling laws of Child-Langmuir (CL) in the quantum regime [2,3].

In their recent publication [4], they also claimed that the reported quantum scaling law for the CL law, $J_{QCL} \propto V_g^{1/2}/D^4$, only holds under the assumption that the potential is constant beyond the gap [or at the anode], where D and V_g is the gap spacing and voltage across the gap.

While we acknowledge that the improved boundary conditions pointed out by the authors may affect the magnitude of the calculated quantum CL law; however more realistic and consistent boundary conditions related to the emission mechanisms at the cathode [5,6], and also many other issues such as image charge barrier due to exchange correlation potential, influence of atomic surface on work function, and scattering events at the surface.

In the two experiments conducted by others [7,8], the voltage $V_g^{1/2}$ scaling has been confirmed in nanogaps of 50 and 70 nm, which is sensitive to the cathode conditions. Thus, the dependence of the quantum CL law is more related to the physical boundary conditions related to the emission process, which will be explained below.

In the classical CL law [9], the emission mechanism from the cathode is due to thermionic emission and the classical CL law is reached when the amount of emitted current is sufficiently high to suppress the electric field at the cathode to zero. At this condition, the classical scaling is $J_{CL} \propto V_g^{3/2}/D^2$.

When the gap spacing D is decreased to nanometer scale for which the electron de Broglie wavelength (λ_e) is comparable or smaller than D (or $\lambda = D/\lambda_e < 1$), it is expected that we can have a higher space charge limited current than the classical CL law due to the tunneling of electrons through the space charge field.

For a nanogap, the emission mechanism from the cathode is due to field emission (also a tunneling process), and the emitted current at the cathode is very sensitive to the electric field or potential profile near to the cathode, which provides a more realistic boundary condition at the cathode.

As mentioned in our earlier works in 2003 and 2004 [2,3], we have ignored these emission condition for simplicity, and we have later improved in our subsequent papers [5,6]. Here, the image charge potential (based on the Thomas-Fermi approximation) on both cathode and anode have been included as shown in Fig. 1(a) in the paper [6]. The effects of average emission energy and cathode's temperature are also included as shown in Fig. 3 in the paper [5].

As compared to our previous model [2], the improved calculation [5,6] shows that the enhancement over the classical CL law is around $J_{QCL}/J_{CL} = 2$ to 5 for a 1 nm gap of both metallic (Tungsten or Barium or Scandate) electrodes, which may resolve the high enhancement factor raised in the comment.

Note that these boundary conditions are more realistic as compared to the improved models suggested by the author, who claims an enhancement of $J_{QCL}/J_{CL} = 3.4$ in their comment.

While the enhancement over the classical CL law is properly over estimated in our earlier models [2], the proposed quantum scaling $J_{QCL} \propto V_g^{1/2}/D^4$ may remains valid as it can be derived from a simple dimensional analysis [3], which is *independent* on any boundary conditions.

In summary, the improved boundary condition suggested by the author is a step better than our previous model, which was the first step in the extension of the classical CL law to quantum regime. However, that it is not the final word on the space charge limited flow in a nanogap as many more related physical issues at the emitting surface will require new revision for a better understanding. The proposed boundary condition in this comment remains too simplified to capture the real physical picture.

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