Modeling of space charge limited current

Zhu, Yingbin

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Modeling of Space Charge Limited Current

Zhu Yingbin

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SUMMARY

Space-charge-limited (SCL) electron flow describes the maximum current density allowed for steady-state electron beam transport across a diode. It is central to the studies of high current diodes, high power microwave sources, vacuum microelectronics, and sheath physics in plasma processing, etc. It is also of interest to the contemporary studies of the nanogap and nanodiode. This thesis presents recent development and application of SCL current.

Recently, various studies were conducted in using ultrafast lasers to excite localized photofield electron emission from metallic nanotips, where a finite number of emitted electrons per pulse can be obtained. Based on a time of flight model, the one-dimensional classical Child-Langmuir (CL) law has been extended to the Coulomb blockade regime, including the effect of single-electron charging. It is found that there is a threshold of voltage equals to one-half of the single-electron charging energy for electron injection assuming zero barrier at the interface. The time-averaged single-electron injected current is equal to or higher than the 1D CL current.

The traditional Langmuir and Blodgett (LB) law for SCL current in cylindrical and spherical geometries remains numerically tabulated form. We develop a transit time model to obtain a uniform analytical form for LB law. It is found that the LB solutions may be approximated by a scaling law using the surface electric field

on the cathode of the vacuum diode. With the help of data fitting, minor empirical corrections to the scaling law is obtained and a new formula within 5% error compared with the LB law is given. We also find that the electron transport through a 100 nm gap at THz frequency. The transit time model is then used for the problem of SCL current injection into solid. The same expression for traditional theory is obtained.

Due to the development of nanotechnology, many experiments about the transport properties through nanowire and nanorod whose injection area is comparable or much smaller than the transport length were performed. In order to understand the experiment observed current enhancement, we propose a 2D model for SCL nonuniform injection. Large current density enhancement is observed due to the edge effect. The trap effect is also considered.

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CHAPTER 1

INTRODUCTION

1.1 Motivation

Charged particle dynamics deals with the motion of charged particles, which is treated as classical point charge, in static or time dependent electric and magnetic fields. Historically, the first and most prominent area of charged particle dynamics is the field of electron optics, which started from the phenomena that electrons can be focused by axially symmetric electric or magnetic field just like that of a glass lens on light. Charged particle beam is widely used in accelerator physics, where the beam current ranges from 10^{-9} A to 10^6 A and the beam energy ranges from a few electron volts (eV) to almost 1 TeV (10^{12} eV). Another main application is in the microwave tubes: klystrons, magnetron, traveling-wave tubes, etc. The research of particle beam is also stimulated by strong industrial needs, such as electron microscopes, cathode ray tubes, and television. Charged particle beams have continually expanding various applications in many different branches of research and technology. Recent active areas include beam lithography for microcircuits, thin-film technology, flat-screen cathode-ray tubes, synchrotron light sources, and free-electron lasers. In devices like

cathode ray tubes, X-ray tubes and electron microscopes, the space charge density in the beams is extremely small and its action on charged-particle motion is negligible. For charged particle beams with high currents values of order of amperes and tens of amperes, the space charge density is rather high and the potential distribution modified by the space charge should be considered, the motion of charge particles is considerably affected by the electric self-fields created by the space charge.

Recently, various studies were conducted by using ultrafast lasers to have localized photo-field electron emission from metallic nanotips [1, 2, 3, 4, 5], where a finite number of emitted electrons per pulse can be obtained depending on the laser pulse energy [5]. We are interested to study if the space charge effect of the electrons can be ignored when the electrons are emitted or injected into a gap in the limit of single electron per injection, and thus provide a single electron based model for the 1D classical Child-Langmuir (CL) law. In this case, the effects of single electron charging or Coulomb blockade must be included.

Beside the planar geometry, the other most used geometries are cylindrical and spherical diodes. About 90 years ago, the problem of space-charge-limited (SCL) current in cylindrical and spherical diode was investigated by Langmuir and Blodgett (LB) [6, 7]. Due to the difficulties in solving the nonlinear differential equations, the LB solution remains in a tabulated form till today. For practical use, it would be beneficial if we can have an analytical expression. For the application of vacuum devices such as Terahertz sources, the period of the electrons across the gap is important. Furthermore, the concept of transit time plays an important role in the SCL current. So it is useful to develop a model based on the concept of transit time. For electron field emission, the current is dependent on the field acting on the material surface. The traditional CL law is dependent explicitly on the applied voltage but not on the E-field. Thus we are interested to use the electric field on the cathode surface instead of voltage in our expression.

For SCL electron injection into solids, due to the development of current technology, the width of the contact or cathode (electron injection) becomes comparable to the length of the dielectric. The traditional SCL current model in solids only accounts for the 1D case whereby the width is much longer than the length of the SCL transport direction or diode's length. In exprement, geometrical enhancement effects have also been observed in nanowires but with a very smallle emitting area [8, 9] and a much higher enhanced value. The crossover from the 1D to 2D SCL current conduction behavior was also observed in single crystals [10]. Due to the fact that a clear theoretical model for this 2D enhancement is lacking, we developed a 2D model to include the effect of non-uniform injection and the discontinuity of dielectric constant at the vacuum-solid interface.

1.2 Objectives and ontributions

Firstly, most of the traditional SCL theory is based on a continuous fluid like model. We have extended Cthe 1D classical CL law to the Coulomb blockade (or single-electron) regime, where the applied voltage is comparable to the single-electron charging potential. The standard fourth-order Runge-Kutta method is used to solve the transport equations. Time dependent current injection induced by a single electron under a DC voltage applied to the gap is observed. It is found that there is a threshold voltage for the single-electron injected current to reach the traditional 1D CL law. The average value of the time dependent current can be much larger than the classical CL current value. Smooth transition to the traditional CL model is demonstrated at higher voltage. Our model would be useful for vacuum devices using very few electrons within one transit time. The breakdown of CL law for time dependent current injection has received a lot of interest and it is now widely investigated by many groups.

Secondly, we developed a transit time model for SCL current in a cylindrical and spherical diode. The transport current is simply approximated by $I = Q/T$, where Q is the total charge of the vacuum capacitor and T is the transit time. It is found that the current density for cylindrical and spherical geometry can be described by a universal scaling law based on the vacuum electric field on the cathode surface. Furthermore, minor empirical corrections is applied to recover the numerical LB law, which is accurate to within 5%. By using the Ramo-Shockley theorem, we improve the model to include the space charge effect without the empirical correction. It is found that the transit time frequency is in the THz range for a mesoscopic gap with $D = 100$ nm in the Coulomb Blockade regime. The model may be useful for the development of an electron gun code, where modeling of electron emission in the first numerical grid is most critical. It would also be useful for the design of THz vacuum device. The new expression of LB law is now very convenient to be used to investigate the transport properties of SCL current in cylindrical and spherical diode.

Thirdly, we apply the capacitance formulation to study the SCL current injection into solid. The same expression for the traditional Mott-Gurney law and Mark-Helfrich law can be obtained. Geometry effects are also considered. This model gives a new picture for SCL electron injection model in solid. It would be useful for researchers to understand electron transport properties under the SCL condition in solid-state devices.

Lastly, the 1D SCL current injection is generalized to a 2D non-uniform model. A numerical method in solving the nonlinear equation is used. Large current enhancement near the edge is observed due to the edge effect. This model would be helpful for the design of devices whose injection contact diameter is comparable with the electron transport length. Such non-uniform effects are not included in the existing commercial device simulator

1.3 Organization of the Thesis

In Chapter 2, we will present a brief review of the classical 1D CL and LB laws for the planar and coaxial cylindrical diodes, respectively. Both the CL and LB laws give us the maximum current that can be transmitted across a vacuum gap in planar and cylindrical geometries at steady-state condition. Recent developments and extensions of the classical 1D CL law to the quantum regime, multidimensional models, short-pulse model and relativistic regime are also introduced. Previous work on the transition from field emission to space-charge regime is also briefly discussed for completeness. Theoretical model and experimental results for SCL current injection into solid are also presented in this chapter.

Next, we will present a 1D single electron model for the SCL flow in a planar diode in Chapter 3. Numerical solution of the model is given to analyze the Coulomb blockade phenomena.

In Chapter 4, a simple capacitor model proposed by Lau is developed to become a transit time model and applied to study the SCL current in cylindrical and spherical diodes. Novel scaling is found with a universal expression for the planar, cylindrical, and spherical cases. To include the space charge effect, we develop a single electron capacitor model which gives the same numerical value as the traditional numerical tabulated LB law.

In Chapter 5, we will use the capacitor model to investigate the problem of current injection into a solid. Equations for trap free and trap filled solids will be formulated. We also include the geometrical effects in our studies.

The theory of SCL current injection into solid will be briefly reviewed in Chapter 6. Here we expand the 1D uniform model to 2D non-uniform model. A numerical method based on the finite difference method is developed to solve the nonlinear

Poisson boundary value problem. The dielectric vacuum interface effect is also considered, as well as the effect of traps in solids.

Last but not least, we will summarize and conclude our work in Chapter 7. Some suggestions for future work will also be discussed.

CHAPTER 2

LITERATURE SURVEY

2.1 Classical Regime

The classical expression for SCL current was first derived by C. D. Child in 1911 to describe the discharge from hot calcium oxide (CaO), where the current flowing through the discharge tube was limited due to the positive ion inside. [11]. In 1913, Irving Langmuir independently derived the same law for the SCL electron flow from a hot filament (thermionic cathode) in vacuum [12]. The maximum current density that can be transported through a vacuum gap with gap spacing D and applied voltage V_g under steady state conditions bear their names and is well known as the Child-Langmuir (CL) law [11, 12].

We first briefly describe the traditional solution to the SCL problem. Suppose the electrons are initially at rest at the grounded cathode and then transport through the gap of length D , with the anode biased positively at V_g . The current density J_{CL} given by the SCL flow is

$$J_{CL}(x) = \rho(x) v(x), \quad (2.1)$$

where $\rho(x)$ and $v(x)$ are the density and velocity of the electrons, respectively. According to energy conservation, $v(x)$ can be found from

$$\frac{1}{2}mv^2(x) = eV(x). \quad (2.2)$$

Due to the space charge inside the gap, the potential $V(x)$ must obey the Poisson's equation

$$\nabla^2 V(x) = \frac{-\rho(x)}{\varepsilon_0}. \quad (2.3)$$

If we eliminate $\rho(x)$ and $v(x)$ using Eqs. (2.1) and (2.2), Eq. (2.3) becomes a second-order nonlinear differential equation for the potential given by

$$\frac{d^2 V(x)}{dx^2} = \sqrt{\frac{m}{2e\varepsilon_0}} \frac{J_{CL}}{\sqrt{V(x)}}. \quad (2.4)$$

Multiplying both sides by $dV(x)/dx$, after integration, we can obtain

$$\left(\frac{dV(x)}{dx}\right)^2 = \sqrt{\frac{m}{2e\varepsilon_0}} \frac{4J_{CL}}{\varepsilon_0} \sqrt{V(x)} + \left(\frac{dV(x)}{dx}\Big|_{x=0}\right)^2. \quad (2.5)$$

If we set the electric field at the cathode to zero, we can take the square root of Eq. (2.5) and then integrate to find the potential at this SCL condition, which is

$$V(x) = \left(\frac{3}{2}\right)^{4/3} \left(\frac{J_{CL}}{\varepsilon_0}\right)^{2/3} \left(\frac{m}{2e}\right)^{1/3} x^{4/3} + C. \quad (2.6)$$

By applying the boundary condition at the cathode $V(0) = 0$ and at the anode, $V(D) = V_g$, we can solve Eq. (2.6) for the SCL current density, which gives

$$J_{CL} = \frac{4\epsilon_o}{9} \sqrt{\frac{2e}{m}} \frac{V_g^{3/2}}{D^2}. \quad (2.7)$$

The potential distribution in a vacuum diode under the SCL condition is shown in Fig. (2.1). It can be seen that, when the electron flow reaches the space charge limit, the first order derivative of the potential on the cathode surface becomes zero, no more electrons can enter the gap.

In the derivation of CL law, one has to note that the inherent assumption is a perfect vacuum inside the gap and the charged particle flow is unidirectional and

Figure 2.1: Potential profile of vacuum diode without space charge (dash line) and SCL flow (solid line).

ballistic in nature. The unipolar SCL flow has been re-investigated by using (a) the Lagrangian flow description [13, 14]; (b) the current over-injection approach (similar to the time-averaged numerical results) [15], and (c) the vacuum capacitance approach [16], which provide the same scaling of $V^{3/2}$ and D^{-2} as predicted. The corresponding SCL law for a cylindrical diode (with inner radius r_i and outer radius r_o) is the Langmuir-Blodgett (LB) law derived by Langmuir and Blodgett [17] :

$$J_{CLB} = \frac{4}{9}\epsilon_0 \sqrt{\frac{2e}{m}} \frac{V_g^{3/2}}{R_a R_c \beta^2}, \quad (2.8)$$

where β is a function of R_a/R_c , and it is tabulated numerically, R_a and R_c are the radius of anode and cathode, respectively. Similarly, the SCL current density on the cathode of a spherical diode is governed by [7]:

$$J_{SLB} = \frac{4}{9}\epsilon_0 \sqrt{\frac{2e}{m}} \frac{V_g^{3/2}}{R_c^2 \alpha^2}, \quad (2.9)$$

where α is a function of R_a/R_c and it is also tabulated numerically.

2.2 Quantum Regime

In the emerging fields of nanoelectronics, tunneling microscopy, and vacuum microelectronics, diode gaps and junctions with scales down to tens of angstroms or nm scale are being considered. With the rapid development in nanoscale fabrication, nano-size gaps between metallic electrodes with gap spacing on the order of $D < 100$ nm have been successfully fabricated [18, 19, 20]. On such a microscopic scale, quantum effects may no longer be neglected. In 1991, Lau *et. al.* developed the basic framework of a quantum CL law by solving the Schrodinger equation and Poission equation self-consistently [21]:

$$-\frac{\hbar^2}{2m} \frac{d^2\psi}{dx^2} - eV(x)\psi = E\psi, \quad (2.10)$$

$$\frac{d^2V(x)}{dx^2} = \frac{e\psi\psi^*}{\varepsilon_0}, \quad (2.11)$$

where $\hbar$ is the reduced Planck constant, ψ is the wave function of the electron, $V(x)$ is the potential distribution, and E is the energy of the injection electrons. The model assumes that the electron density in the gap is sufficiently small that single-particle wave functions do not overlap significantly. Very huge current enhancement was found due to the electron tunneling. Later, in 2003, Ang included the many electron exchange-correlation potential into the quantum model and find a new quantum scaling that the maximum current density is proportional to $V_g^{1/2}$ and D^{-4} in the deep quantum regime when the gap distance is very small or the applied gap voltage is very low, as seen in Fig. (2.2) [22].

This scaling was later observed in experiments as shown in Fig. 2.3 [23]. They found the quantum scaling $J_{QCL} \propto V_g^{1/2}$ for a narrower nano gap with gap distance equals to 70 nm. While for a wider gap with distance 90 nm, the I-V character is still obey the classical scaling $J_{CL} \propto V_g^{3/2}$. The quantum model was also extended to a nanoscale cross field diode [24] and bipolar fluid [25]. Biswas and Kumar developed the model considering the self-consistent boundary conditions [26]. In

Figure 2.2: (a) The current density normalized to the 1D classical CL law as a function of λ ($\lambda = D/\lambda_0$ is gap spacing normalized to the electron de Broglie wavelength λ_0) for various ϕ_g ($\phi_g = eV_g/E_H$, E_H is the Hartree energy). (b) The values of $\mu_Q V_g^{3/2}$ as a function of gap voltage V_g (volt) for various gap spacing $D = 1, 10$, and 100 nm (solid lines: top to bottom).

Figure 2.3: $I - V$ data obtained for the 70 and 90 nm samples. This confirm the scaling $J_{QCL} \propto V_g^{1/2}$ in the quantum regime and $J_{CL} \propto V_g^{3/2}$ in the classical regime.

recent years, the field of "quantum plasmas" received a lot of interest and collective nonlinear phenomena at nanoscales were investigated [27, 28]. For the quantum diode, Shukla and Eliasson developed a nonlinear model using the steady-state quantum hydrodynamical equations [29]. They found that the quantum statistical pressure introduces a lower bound on the steady electron flow in the quantum diode, while the quantum diffraction effect allows the electron tunneling at low flow speeds.

2.3 Mutidimensional Model

The multidimensional SCL model can be divided into two categories: uniform injection model and nonuniform injection model. For uniform injection, in 1996, Luginsland *et. al.* considered uniform emission of electrons over a finite strip of width W in a planar gap of gap separation D using two commercial software: OOPIC

2.3 Mutidimensional Model

(electrostatic) and MAGIC (electromagnetic). Both of them are based on particle-in-cell (PIC) simulation. A PIC code consists two part, the field solver based on FDTD and a particle solver [30]. Firstly, the Maxwell or Poisson equation is solved without considering the particle, then the particle is introduced and their trajectories are calculated. Some weighting method is used obtained the charge density on the grid point. later, the new field is solved considering the space charge. Based on this new field, the new particle position is obtained. These process is repeated step by step and the field is filled by space charge. Steady state plasma motion is established lastly. Based on the simulation, they showed for the first time that the limited current density in two dimensions $J_{CL}(2)$ in units of the classical one-dimensional value $J_{CL}(1)$ is found to be a monotonically decreasing function of W/D given by:

$$\frac{J_{CL}(2)}{J_{CL}(1)} = 1 + \frac{0.3145}{W/D} - \frac{0.0004}{(W/D)^2}. \quad (2.12)$$

The 2D SCL current enhancement is shown in Fig. 2.4.

Figure 2.4: The SCL current density in two dimensions, in units of the one-dimensional CL value for an unmagnetized gap.

Later, Lau derived an analytical expression [31] for this 2D current enhancement which is :

$$\frac{J_{CL}(2)}{J_{CL}(1)} \approx 1 + \frac{D}{\pi W}, \quad (2.13)$$

and it is in remarkable agreement with the empirical formula given by Eq. 2.12. The 2D model was then extended to 3D emitting shapes (i.e. rectangular, elliptical and polygonal emitting areas) [32].

For nonuniform injection, Umstattd and Luginsland first investigated the characteristics of planar diode electron emission in 2D space with the emphasis on the transition between the beam and vacuum using a 2D finite element, electrostatic ray-tracing code [33]. The properties of the enhanced current density near the edges were examined. As shown in Fig. 2.5, current densities higher than the CL law was observed near the beam edges. The enhancement increased as the emission area decreased.

Figure 2.5: The SCL current density in two dimensions in units of the one-dimensional CL value for an unmagnetized gap.

Rokhlenko and Lebowitz then constructed an approximate analytic solution for the 2D SCL emission by a cathode surrounded by nonemitting conducting ledges of finite width. Similar current wings were also observed. They have later developed several works related to this topic [34, 35, 36]. A numerical algorithm was later developed by Watrous *et. al.* to solve the 2D problem in a parallel-plate geometry. Recently, this algorithm was applied for the problem of SCL emission from a circular cathode [37].

2.4 Time Dependent Current Injection and Short-Pulse SCL Flow

Today, free-electron laser facilities around the world have replaced using thermionic cathodes (operating in continuous mode) with photocathode guns (operating in pulsed mode) [38, 39, 40, 41] as photocathode guns are able to emit high currents (> 100 mA) with low emittance and low energy spread. At the photocathode operating condition, the RF field is typically on the order of 10 MV/m. When the emitted electrons are on the order of 1 nC per bunch, the space charge field induced by the emitted electrons is comparable to the RF field. So it is meaningful to investigate the space-charge-limited current of a short pulse electron beam. For a short pulse SCL electron bunch, the pulse length of the electron beam (from the photoinjector) is usually shorter than the transit time of the electrons across the gap. Thus, the classical scaling laws, such as the CL law and LB law, which were derived based on steady-state conditions (or long pulse limit) are no longer valid for short pulse SCL emission. It has also been shown that the traditional long-pulse CL law can be enhanced by a large factor at the short-pulse limit in a recent experiment, and a 1D short-pulse CL law has been

formulated and confirmed by particle-in-cell simulations [42]:

$$\frac{J_{SCL}}{J_{CL}} = 2 \frac{1 - \sqrt{1 - 3X_{CL}^2/4}}{X_{CL}^3}, \quad (2.14)$$

where J_{SCL} is the classical short-pulse current density, J_{CL} is the current density of the classical CL law, $X_{CL} = \tau/T_{CL}$ is the normalized transit time and $T_{CL} = 3D/\sqrt{2eV_g/m}$ is the gap transit time of the 1D classical CL law. Recently, a 3D simulation of molecular dynamics type was performed by Valfells *et. al* to reproduce the breakup of a single pulse electron beam with a current density beyond the CL limit, and they found that continuous injection of current into the diode gap will produce a well-defined train of electron bunches corresponding to THz frequency [43]. The computational study was extended [44], and it was found that for a fixed emitter area, the frequency of the pulse train is solely dependent on the vacuum electric field in the diode described by a simple power law. It was also found that, for a fixed value of the electric field, the frequency increases with decreasing size of the emitting spot on the cathode. For femtosecond laser excitation of electron photofield emission from a sharp tip, the electron pulse length is usually in the nanometer scale. As an example, consider femtosecond laser irradiation on a sharp emitter tip with a local dc field of $E = 0.2$ GV/m. The emitted short pulse electron beam (without space charge effects) is estimated to have a characteristic length of $\Delta x \sim \frac{e}{m} E \Delta t^2 \sim 2$ nm with a pulse length of $\Delta t = 7$ fs. The size of the electron pulse is comparable to the electron de Broglie wavelength and quantum effects of electron transport near the cathode are important. When the electron bunches are accelerated to the MV range inside the gap, relativistic effects may also become important. A consistent quantum and relativistic model of short-pulse CL law was developed by Ang and Zhang [45]. It was found that the enhancement of the short-pulse CL law over the long-pulse CL law is proportional to the inverse power of the normalized pulse length in all regimes (classical, quantum and relativistic) when the pulse length is much smaller than the gap transit time. Thus, the new scaling of the short-pulse SCL current

density is V_g , D^{-1} and τ^{-1} , as compared to the classical steady-state CL law with a scaling of $V_g^{3/2}$ and D^{-2} . A two-dimensional model of the short pulse limit has also been developed [32]. Chen investigated a electromagnetic model for two-dimensional short-pulse electron flow. They revealed that the enhancement is due to the effect of the displacement current and the self-magnetic field [46].

2.5 Transition from Field Emission to SCL Flow

Field emission (FE) process is a unique type of electron emission as it is due exclusively to the quantum-mechanical effects of tunneling of electrons into vacuum. Nowadays field emission is the principal means of generating an electron beam for many application. In order to decrease the potential barrier at the metal-vacuum interface sufficiently for the electrons to have a significant probability of tunneling from the solid into the vacuum, high electric fields in the range of $10^7 - 10^8$ V/cm are usually needed. The most conventional method to generate such high fields is by using a sharp tip with apex radius of curvature ranging from tens of angstroms to several microns. The field emission current density is usually given by the Fowler-Nordheim (FN) law:

$$J_{\text{FN}} = A' F^2 \exp \left[- B' \phi_B^{3/2} / F \right], \quad (2.15)$$

where $A' \equiv e^3 / [16\pi^2 \hbar \phi_B t^2(y)]$ and $B' \equiv \frac{4}{3e} \left(\frac{2m}{\hbar^2} \right)^{1/2} v(y)$

and $t(y)$ and $v(y)$ are functions of elliptical integrals with $y = \sqrt{e^3 F} / \phi_B$, and ϕ_B is the material work function. The numerical values of $t(y)$ and $v(y)$ are listed in the literature by Burgess, Kroemer and Houston [47]. Widely accepted approximations are $t(y) \approx (1.1)^{1/2}$ and $v(y) \approx 0.95 - y^2$. Most works in the literature for the transition from field emission or the Fowler-Nordheim (FN) regime to the space-charge or CL regime are for the 1D planar case. Experimentally the transition was first shown by

Barbour *et. al.* in 1953 for various crystal faces of clean tungsten and barium-on-tungsten cathodes [48] and the results were also reviewed by Dyke and Dolan [49]. Recently, Jensen *et. al.* have develop different models to explain the experimental data [50, 51, 52]. A universal curve (independent of cathode material) that separates the FN and CL regime for a planar diode was given by Lau *et. al.*. The inclusion of ion effects and relativistic effects of the electron for a 1D planar diode was also investigated by Lin [53] by using a self-consistent numerical method. The extension of the simple planar transition to a gated field emitter model was also discussed by Jensen *et. al.* [54]. Further comparison between the calculation results based on the geometry of the tip and experimental results have been conducted [55]. Van Veen [56] developed three different models about planar, spherical and paraboloidal tips model to compare with experiment results. Forbes has used simple dimensionless equations to exactly analyze the surface field reduction due to field-emitted vacuum space charge field in a parallel-plane geometry [57]. A 1D PIC simulation was developed by Feng and Verboncoeur to study the transition behavior, which indicated that the transition without geometrical effects is smooth and much slower than reported previously [48]. The geometric field enhancement effect is analyzed and they found that an enhancement $\beta > 10$ can accelerate the approach to the SCL at practical electric fields.

2.6 Relativistic Effects

Single gap electron extractors have operated in the MV range. These extractors are driven either by electrostatic pulsed power generators or by stacked induction linac cavities. Relativistic effects are important at $eV_g \gg m_0c^2$. In the relativistic regime, the charged particles (usually electrons) flowing across the high voltage vacuum diode typically travel close to the speed of light c . Ivey [58] and Boers *et. al.* [59] illustrated

2.6 Relativistic Effects

the relativistic CL law using a series expansion. A generalization of the relativistic CL law was also introduced by Howes [60] and Jory *et. al.* [61] with the aid of elliptic integrals, given by

$$J_{RCL} = \frac{2\varepsilon_0 m_0 c^3}{ed^2} \left[\frac{\omega (\omega^4 + 1)^{1/2}}{\omega^4 + 1} + \frac{1}{2} F(\phi, k) - E(\phi, k) \right], 0 \leq \omega \leq 1 \quad (2.16a)$$

$$= \frac{2\varepsilon_0 m_0 c^3}{ed^2} \left[\frac{\omega (\omega^4 + 1)^{1/2}}{\omega^4 + 1} - \frac{1}{2} F(\phi, k) + E(\phi, k) + K(k) - 2E(k) \right], 1 \leq \omega < \infty \quad (2.16b)$$

where $F(\phi, k)$ and $E(\phi, k)$ are the incomplete elliptic integrals of the first and second kind, respectively, while $K(k)$ and $E(k)$ are the complete elliptic integrals of the first and second kinds. The modulus $k = \sqrt{2}/2$, the argument ϕ is defined by $\sin(\phi) = 2\omega/(\omega^2 + 1)$, and $\omega = (U^2 + 2U)^{1/4}$, where $U = eV_g/m_0 c^2$. In the ultra-relativistic regime, under the approximation $2U \ll U^2$, the current density is simply given by

$$J_{URCL} = 2\varepsilon_0 \frac{V_g}{D^2} c. \quad (2.17)$$

From Fig. 2.6, it can be seen that the relativistic effect can be omitted under low voltage. While at high voltage, the relativistic current density has much lower value than that predicted by classical CL law. Nation *et. al.* derived the limited current for a relativistic foilless diode under the influence of a strong magnetic field [62, 63]. The theory of the relativistic SCL electron beam in a grounded cylinder [64] and a grounded coaxial cylinder [65] was also discussed. To understand the physics and diode properties of a rod-pinch diode, Cooperstein *et. al.* conducted both theoretical and experimental studies of a relativistic beam from SCL flow at low relativistic voltages, through the weakly-pinch regime and the magnetically limited regime [66, 67]. Biwas *et. al.* revisited the SCL theory of the planar 1D relativistic diode using the current over-injection approach [15], which is in good agreement with the results of Jory *et. al.* [61]. Lin numerically investigated the transition from field emission to relativistic SCL current [68]. A 2D/3D relativistic model (valid

Figure 2.6: The comparison of relativistic CL law and traditional CL law as a function of the gap voltage.

2.7 SCL Current Injection into a Solid

in the weakly-relativistic regime) was also developed [69]. In order to investigate the transport property of an electron pulse in vacuum diode, a theoretical model is developed for both the weakly-relativistic and ultra-relativistic regime and compared with PIC simulation.

It must be stressed that most of the theoretical treatment of relativistic SCL flow reported are restricted to the case where the self-magnetic field is ignored. The only two models that take into account of the self-magnetic field generated by the relativistic beam are the parapotential flow [63, 70] and the focused flow model [71, 72]. It has been found that the parapotential flow model is only found to be good for diode impedance prediction and is unable to take into account the electron flow near the cathode or anode. A comprehensive overview of the parapotential flow and the focused flow concept was illustrated by Miller [63].

A more complete review of the theory behind relativistic flow in the various types of diode and triode configurations are discussed extensively by Miller [63], Humphries [70], Reiser [73] and Davidson [74]. The application of a relativistic electron beam in a free electron laser (FEL) to produce different types of coherent radiation is also discussed by Miller [63, 70]. A recent study of the relativistic SCL current in a crossed-field planar diode including self-electric and magnetic fields was undertaken by Lopez *et. al.* and they have given a set of universal curves to obtain the maximum current density at various gap voltages which recover the established non-relativistic results at low accelerating voltages [75].

2.7 SCL Current Injection into a Solid

The study of SCL current in a vacuum diode has inspired the study of SCL current injection in solids [76]. For electron transport through a solid with length D under lower injection with a voltage drop V_g , the electric field within the dielectric material

is uniform everywhere with a value $E = V_g/D$. The current density is proportional to V_g and is given by the well-known Ohm's Law:

$$J_\Omega = n_0 e v = n_0 e \mu \frac{V_g}{D}, \quad (2.18)$$

where n_0 is the thermally generated electrons concentration and μ is the drift electron mobility. Once the injected charge density is larger than the thermal charge density, the current continuity and Poisson's equations have to be simultaneously solved to obtain the current-voltage characteristics of the material. In the case that the velocity increases monotonically with electric field given by $v = -\mu E$, applying the boundary condition $V(0) = 0$, $V(D) = V_g$, and $E(0) = 0$, the current density is given by the Mott-Gurney (MG) law for the SCL conduction in a trap-free solid:

$$J_{MG} = \frac{9}{8} \varepsilon_0 \varepsilon_r \mu \frac{V_g^2}{D^3}, \quad (2.19)$$

where ε_r is the relative dielectric permittivity. The theory of a single carrier MG law is based on the following assumption: (i) When the applied voltage V_g is much larger than the thermal voltage given by $k_B T/e \propto 26mV$, or the material length D is much larger than the diffusion length, the drift current dominates the transport current and the diffusion current is neglected completely; (ii) The cathode is taken to be an infinite reservoir of electrons available for injection; (iii) the contact responsible for the charge injection is an Ohmic contact, and we assume no voltage drop across the contact. The CL law for a vacuum diode is proportional to 3/2 power of gap voltage V_g and inversely proportional to the 2nd power of gap distance D ($J_{CL} \propto V_g^{3/2}/D^2$), while the MG law has a power of two to V_g and a power of -3 to D ($J_{MG} \propto V_g^2/D^3$). Their transport properties are compared in Table 2.1.

The magnitude and nature of the current flow changes markedly when a dominant trap or set of traps exist in the material. When an external bias is applied, not all of the electrons injected into the material are mobile. While some of the injected electrons occupy states in the conduction band, many of them will fill the

2.7 SCL Current Injection into a Solid

Table 2.1: Properties of SCL Current Flow.

	CL law	MG Law
electric field	$E(x) \propto x^{1/3}$	$E(x) \propto x^{1/2}$
potential	$V(x) \propto x^{4/3}$	$V(x) \propto x^{3/2}$
charge density	$n \propto x^{-2/3}$	$n \propto x^{-1/2}$
transit time	$t = 3D \sqrt{\frac{m}{2eV_g}}$	$t = \frac{4}{3} \frac{D^2}{\mu V_g}$
capacitance per unit area	$C = \frac{4}{3} \frac{\epsilon_0}{D}$	$C = \frac{3}{2} \frac{\epsilon_0 \epsilon_r}{D}$

empty trap states and, therefore, do not contribute to current flow. However, all injected electrons, whether mobile or trapped, will contribute to the voltage drop in the material. For an exponential distribution in energy level of traps, the density of traps per unit energy range E is described by:

$$h(E_t) = \frac{N_t}{k_B T_t} \exp\left(\frac{E - E_c}{k_B T_t}\right), \quad (2.20)$$

where N_t is the total trap density and T_t is a characteristic constant of the distribution, E_c is the bottom of the conduction band. Assume that the absolute temperature $T = 0$, and that the space charge $\rho(x)$ fills all the traps to a quasi Fermi level for electrons $E_f(x)$, the charge density is given by

$$\rho(x) = e \int_{-\infty}^{E_f(x)} h(E_t) dE = e N_t \exp\left(\frac{E_f(x) - E_c}{k_B T_t}\right). \quad (2.21)$$

If we now increase the temperature and assume that a Fermi distribution for the injected electrons has been established for all positions, the density of free charge in the conduction band can be given by

$$\rho_f(x) = e N_c \exp\left(\frac{E_f(x) - E_c}{k_B T_t}\right), \quad (2.22)$$

where N_c is the effective density of states in the conduction band. We can then obtain the relation between the free and the total charge density as

$$\rho_f(x) = \frac{N_c}{N_t^l} e^{1-l} [\rho(x)]^l, \quad (2.23)$$

where $l = T_c/T$. By following the same procedure to derive the CL law and the MG law, solving the Poisson equation and current continue equation, the trap limited current density becomes

$$J_{TL} = N_c \mu e^{1-l} \left[\frac{\epsilon_0 \epsilon_r l}{N_t (l+1)} \right]^l \left(\frac{2l+1}{l+1} \right)^{l+1} \frac{V_g^{(l+1)}}{D^{(2l+1)}}. \quad (2.24)$$

In Fig. 2.7, the experimental current density of four difference devices as a function

Figure 2.7: Three regimes under different voltages: Ohmic regime, trap-limited regime and the SCL regime.

of applied bias is plotted [77]. For lower bias $V_g < 1.5$ V, the current density is linear proportional to the bias and this is the Ohmic regime. For bias between 1.5 V and 2.5 V, the $J - V$ curve shows a power of 20. This is the trap-limited region with $l=19$. according to Eqs. (2.18) and (2.24), the transit voltage from the Ohmic region to the trap-limited region is given by

$$V_{\text{trap-MG}} = \frac{en_0 L^2}{\epsilon_0 \epsilon_r} \Gamma \left[\frac{9}{8} \left(\frac{l+1}{l} \right)^l \left(\frac{l+1}{2l+1} \right)^{l+1} \right]^{[1/(l-1)]}. \quad (2.25)$$

2.7 SCL Current Injection into a Solid

For bias higher than 2.5 V, the devices works in the SCL region whose value can be predict by the MG law. The cross-over voltage can be estimated using

$$V_{\Omega-trap} = \frac{en_0L^2}{\epsilon_0\epsilon_r} \left(\frac{l+1}{2l+1} \right)^{[(l+1)/l]} \frac{l+1}{l} \Gamma, \quad (2.26)$$

where $\Gamma = \frac{N_t}{n_0} \left(\frac{n_0}{N_c} \right)^{1/l}$, for the trap-limited region and the SCL region.

The value l can be obtained from the power scaling in a $\log(J_{TL})$ versus $\log(V_g)$ plot [77], that is, the slope equals to $l+1$. The value of the trap density can also be obtained using the method proposed by Kumar *et. al.* [78]. The trap limited current density can be rewritten as:

$$J_{TL} = \left(\frac{\mu N_c e V_g}{D} \right) f(l) \exp \left[-\frac{E_t}{kT} \ln \left(\frac{e N_t D^2}{2\epsilon_r \epsilon_0 V_g} \right) \right], \quad (2.27)$$

where f is given by

$$f(l) = 2^{-l} \left(\frac{2l+1}{l+1} \right)^{l+1} \left(\frac{l}{l+1} \right)^l. \quad (2.28)$$

The value of $f(l)$ is about 0.5 when $l > 2$; thus, we regard $f(l)$ as a constant (that is, 0.5). In this case, Eq. (2.27) becomes a typical Arrhenius equation, $J = J_0 \exp(-E_a/kT)$, where $E_a = E_t \ln(e N_t D^2 / 2\epsilon_r \epsilon_0 V_g)$.

Now we can see that J becomes independent of temperature when the right-hand side of Eq. (2.27) tends to zero which gives

$$V_c = \frac{e N_t D^2}{2\epsilon_r \epsilon_0}. \quad (2.29)$$

If we plot the $\log(J) - \log(V)$ characteristics at various temperatures and extrapolate the curve to higher voltages, all the extrapolated lines will converge to a critical voltage $V_c \approx 1200V$, as seen in Fig. 2.8. As the bias voltage increases, the exponentially distributed trap states within the bandgap are occupied by injected space charge and, finally, all the trap states are filled, which results in a temperature independent trap-free SCL current. Therefore, at voltage V_c , all the measured J values are expected to be independent of the temperature. Using Eq. (2.29) we can obtain the value of trap density N_t .

Figure 2.8: Linear extrapolation of the $\log(J) - \log(V)$ characteristics to obtain the cross-over voltage V_c .

Recently, there are renewed interests in SCL current transport in many novel devices involving either inorganic or organic materials, such as graphene oxide sheets [79], light emitting diode [80], organic device [81], polymer transistor [82], nanowire [8, 83, 84], magnetoresistance [85], photocurrent [86], and nanocrystallites embedded silicon Schottky junction [87]. The effects on traps have also been extensively studied [88, 89, 90, 91, 92, 93] as the transport of SCL current as a function of voltage is very useful to characterize the properties of the traps and to also indirectly measure the mobility of charge carrier. Other studies include temperature dependence [94, 93, 95], single electron model [96], and thermoelectric efficiency [97].

In summary, the transport properties of SCL current in vacuum diode and solid received a lot of interest in recent years. Many works about theoretical model, numerical simulation and experiment are developed not only in the field of plasma, but also in semiconductor.

CHAPTER 3

CHILD-LANGMUIR LAW IN THE COULOMB BLOCKADE REGIME

3.1 Introduction

In recent years, several groups succeed in using ultrafast femtosecond laser to obtain photo-field electron emission from tungsten or gold nanotips [1, 2, 3, 4, 5]. By controlling the laser pulse intensity, finite number of emitted electrons per pulse can be obtained[5]. Attosecond scale control of electrons emitted from tip is even reached. Space charge effect is showed to be important in this femtosecond pulse induce electron emission. While the continue model of space charge effect is clear as discussed in Chapter. 2, the Coulomb interaction between few electron has not been investigated. In the research field known as surface plasmonic, there is a great interest about the quantum plasmonic effect. In a recent work about the so called charge transfer plasmonic between two nano particles, the space charge effect becomes very important and induce charge transfer plasmonic [98]. It can be calculated that with one transit time, there is a finite number of electron inside the nano gap. As

introduced in Chapter. 2, the classical CL law has been extended to 2D model, quantum model, short pulsed model and etc. It would be interesting to develop a single electron based model to investigate the effect of Coulomb interacting between few electrons in the Coulomb blockade regime.

In this chapter, we are interested to study if the space charge effect of the electrons can be ignored when the electrons are emitted or injected into a gap in the limit of single electron per injection, and thus provide a single electron based model for the 1D classical CL law. In this case, the effects of single electron charging or Coulomb blockade must be included.

Our approach is based on a time of flight (TOF) model [99] formulated earlier to study SCL ion flow in a solid [100]. Such TOF models have also been used extensively to directly measure the mobility of charge carriers in solids, such as Refs. [101, 102].

In this chapter, the model will be discussed in Sec. 3.2, and the subsequent numerical results will be presented in Sec. 3.3. Finally, some conclusion remarks will be given in Sec. 3.4.

3.2 Single Electron Model

3.2.1 Model Schematic and Mathematical Formulation

In the model, we consider a capacitor with two large parallel plates separated by distance D . A constant voltage V_g is applied to the plate at $x = 0$ and the voltage of another plate at $x = D$ is set to be zero. The charge of the electron is $q = e$. At time $t = 0$ there are N electrons located at the cathode $x = 0$, get ready to be injected into the gap travelling towards the anode at $x = D$, and the order of the injection electron is labeled by $n = 1, 2, \dots, N$. Here, we have assumed an unlimited supply of electrons from the cathode, so N is an input parameter, which is independent of the

3.2 Single Electron Model

gap voltage and material. At low voltage close to a threshold voltage (see below), we can even reach the CL law even with small $N(<10)$. When the electric field acting on the electron is non-zero, the electron will start to be injected into the gap. The launch time of each n -th electron is determined by the consistent electrical field acting on the electron controlled by Eq. (3.5) below.

The initial charge on the plate is $Q = CV_g$, where $C = \epsilon_o A/D$ is the capacitance and A is the area of the plate. Consider the case only one electron inside the gap. When an electron of charge $q = e$ is injected from the cathode ($x = 0$) to a location $x(t) \leq D$ inside the gap, the accelerating electric field at the front of the electron is [99]

$$E_f(t) = \frac{V_g}{D} + \frac{e}{\epsilon_o A} \frac{x(t)}{D}. \quad (3.1)$$

At the back of the electron, the field is given by

$$E_b(t) = \frac{V_g}{D} + \frac{e}{\epsilon_o A} \left(\frac{x(t)}{D} - 1 \right). \quad (3.2)$$

The field acting on the electron is given by the average value of the field at the front and the back:

$$E(t) = \frac{1}{2} (E_f(t) + E_b(t)) = \frac{V_g}{D} + \frac{e}{\epsilon_o A} \left(\frac{x(t)}{D} - \frac{1}{2} \right). \quad (3.3)$$

Due to the time-varying of the electric fields at $x(t)$, there will be a redistribution of charge on the electrode. For anode, the charge will increase by an amount $\Delta q = e \times x(t)/D$ [see Eq. (3.1)]. Including all the injected electrons ($n = 1, 2, \dots, N$), the charge collected at the anode as a function of time is [100]

$$q(t) = CV_g + \frac{e}{D} \sum_{n=1}^N x_n(t), \quad (3.4)$$

where $x_n(t)$ is the location of the n -th electron at time t .

To include space charge effects, we solve Gauss's law for every electron and obtain the electric field acting on the n -th electron, which gives

$$E_n(t) = \sum_{m=1}^{n-1} E_{mb} + \sum_{m=n+1}^N E_{mf} + E_{self} = \frac{1}{CD} [q(t) - e(n - 1/2)], \quad (3.5)$$

where E_{mb} and E_{mf} are respectively the back and front field of m -th electron, E_{self} is the field due to the n -th electron itself given by Eq. (3.3). The dynamic of the electron is then determined by the equation of motion given by

$$\frac{d^2 x_n}{dt^2} = \frac{e}{m} E_n(t). \quad (3.6)$$

For the first electron ($n = 1$) at time $t = 0$, Eq. (3.5) becomes $E_1(0) = V_g/D - e/(2A\epsilon_0)$, where the first term is the external applied field, and the second term is the electric field attributed to the Gauss law. This expression can be also seen from Eq. (3.3). It can be seen that the electric field $E_1(0)$ will be zero unless we have $q(0) > e/2$. By using $q(0) = e/2$ and $q(0) = CV_g$ [from Eq. (3.4)], the threshold voltage for the injection of electrons is

$$V_{th} = \frac{e}{2C}, \quad (3.7)$$

which equals one-half value of the single electron charge potential $V_c = e/C$. The collected current is obtained by

$$I(t) = \frac{dq}{dt} = \frac{e}{D} \sum_{n=1}^N \dot{x}_n(t). \quad (3.8)$$

This expression is just the Ramo-Schockley theorem about single electron induce current in 1D planar gap [103, 104]. It is important to note that according to the traditional CL law, there is no such threshold voltage in the formulation. For any given voltage ($V_g > 0$), there is always a CL current assuming there is an unlimited electron supply from the cathode. This threshold voltage is due to the effect of single electron charging (included in our model), which was also not found in a recent capacitance model of the CL law [16]. We will show later (in Figs. 3.2 and 3.3) that it is possible to have only a single electron inside the gap (at an instant of time) with an time-average current close to (or greater than) the CL law in the range of $1 < V_g/V_{th} < 2$.

3.2.2 Normalized Equations

In order to perform convenient numerical calculations, we introduce the following normalized parameters: $\bar{x} = x/D$, $\bar{V}_g = V_g/V_{th}$, $\bar{E}_n = E_n/E_c$, $\bar{q} = q/e$, $\bar{t} = t/t_c$, and $\bar{I} = I/I_c$. The normalized constant $V_{th} = 0.5 \times V_c$ is the threshold voltage [see Eq. 3.7], $V_c = e/C$ is the single electron charging potential, $E_c = V_c/D$, $t_c^{-1} = e/\sqrt{m\epsilon_o AD}$ is the plasma frequency for one electron per unit volume of $A \times D$, and $I_c = e/t_c$. Using the normalized parameters, the governing equations become

$$\bar{q}(\bar{t}) = \bar{V}_g/2 + \sum_{n=1}^N \bar{x}_n(\bar{t}), \quad (3.9)$$

$$\bar{E}_n(\bar{t}) = \bar{q}(\bar{t}) - (n - 1/2), \quad (3.10)$$

$$\frac{d^2 \bar{x}_n}{d\bar{t}^2} = \bar{E}_n(\bar{t}), \quad (3.11)$$

$$\bar{I}(\bar{t}) = \sum_{n=1}^N \dot{\bar{x}}_n(\bar{t}). \quad (3.12)$$

Using the normalized equations, the time dependent particle position $\bar{x}_n$ and $\bar{I}$ are solved by the standard fourth-order Runge-Kutta method with the initial conditions that every electron is at rest at the cathode surface, which can be described as $\bar{x}_n(0) = 0$ and $\dot{\bar{x}}_n(0) = 0$.

To compare with the steady-state 1D CL law, our results will be presented in terms of the 1D CL current and its transit time: $I_{CL} = A \times J_{CL}$, and $T_{CL} = 3D/\sqrt{2eV_g/m}$. For a gap of $D = 100$ nm with an area $A = D^2$, the threshold voltage is about $V_{th} = 0.09$ V. At $V_g/V_{th} = 1.02$, we have $I_{CL} = 62.91$ nA and $T_{CL} = 1.68$ ps. While we have assumed $D = 100$ nm and $A = D^2$ to illustrate some practical values, the normalized results (I/I_{CL} at different V_g/V_{th}) shown below are independent of the exact values of D .

3.2.3 Numerical Results

In Fig. 3.1, we plot the particle position normalized to the gap spacing $\bar{x} = x_n(t)/D$ for the first five electrons ($n = 1$ to 5, from left to right) across the gap of distance $D = 100$ nm as a function of normalized time t/T_{CL} at $V_g/V_{th} =$ (a) 1.02 and (b) 3. As mentioned before, when the electrical field acting on the n -th electron is larger than zero, the electron will enter the gap and then transport; before that time, the electron is punched on the cathode. At low voltage slightly greater than the threshold voltage, for example $V_g/V_{th} = 1.02$ shown in Fig. 3.1a, there is only one electron inside the gap during its transit time between $0 < t/T_{CL} < 1.7$ for the first electron, $1.7 < t/T_{CL} < 3.5$ for the second electron and so on. The $(n+1)$ -th electron will only enter the gap when the preceeding n -th electron is about to reach the anode. At a higher $V_g/V_{th} = 3$, the subsequent electron can enter the gap earlier, and there will be two electrons inside the gap with a transit time of about $t/T_{CL} = 0.75$, which is shown in Fig. 3.1b. Keep on increasing the applied voltage, there can be much more electrons inside the gap within one transit time.

The time evolution of the injected single electron based current $I(t)$ in terms of the 1D CL current is shown in Fig. 3.2 for various voltages $V_g/V_{th} =$ (a) 1.02, (b) 2, and (c) 500. To study the time to reach the CL current, we define the average value of the time dependent current as

$$I_{ave}(t) = \frac{1}{t} \int_0^t I(\tau) d\tau. \quad (3.13)$$

The calculated value is also plotted using dashed lines in Fig. 3.2. From Fig. 3.2a, it is clear that the current $I(t)$ will increase when the first electron is accelerated across the gap and its velocity keep on increasing. At the moment the electron reach the anode, the current drops to zero. Almost at the same time, the second electron is injected into the gap and induce a time increasing current again. Note there is

Figure 3.1: The time-dependence of the normalized position of the first 5 injected electrons: x_n/D for $n = 1, 2, 3, 4$ and 5 (left to right) at $V_g/V_{th} =$ (a) 1.02 and (b) 3.

only 1 electron at this low voltage case of $V_g/V_{th} = 1.02$, and we see that the time-average current is approaching the CL current (dashed line) after a transit time of $t/T_{CL} = 1.7$.

Similar behavior is found from Fig. 3.2b at higher voltage $V_g/V_{th} = 2$ but with a shorter transit time of about $t/T_{CL} = 1$. When the first electron reaches the anode, the current doesn't drop to zero because the second electron has already gone into the gap and induces a current. By increasing the voltage to values much larger than V_{th} [$V_g/V_{th} = 500$] as shown in Fig. 3.2c, the time varying current $I(t)$ will reach a steady state value equal to the 1D CL law. At this very high voltage limit, it is very similar to the traditional CL model. The transient behavior at $t/T_{CL} < 0.6$ shows that the current is increasing until the first electron to be collected at the anode, and it will reach a steady state value ($I(t)/I_{CL} = 1$) after a few oscillations.

Another way to compare with the 1D CL law is by taking the average current value of N electron injections, given by

$$\langle I(t) \rangle = \lim_{N \rightarrow \infty} \frac{Ne}{t_N}, \quad (3.14)$$

where t_N is the cumulative transit time of all N injected electrons. The numerator is the summation of the total transport charge. The normalized results ($\langle I \rangle / I_{CL}$) are shown in Fig. 3.3 in the range $1 < V_g/V_{th} < 4$ by using charge numbers $N = 500$ (circles) and $N = 50$ (circles).

As mentioned earlier, the value of N is not important as long as N is sufficiently large to reach a numerically stable result, where N can be as small as $N < 10$ for small gap voltage close to the threshold voltage (see Fig. 3.2a also). Note that, even there is only 1 electron passing the gap in the range of $1 < V_g/V_{th} < 2$ (or $e/2C < V_g < e/C$), it is possible to reach the 1D CL law. In this single-electron limit, the space charge field is induced through the modification of charges on the electrodes during the transit of the electron. From the figure, it is interesting to see

Figure 3.2: The time-dependence of the normalized injected current $I(t)/I_{CL}$ at $V_g/V_{th} =$ (a) 1.02, (b) 2, and (c) 500. The dashed lines are the time-average current of $I(t)$.

Figure 3.3: The normalized calculations of $\langle I \rangle / I_{CL}$ (in symbols) as a function of $1 < V_g/V_{th} < 4$ with $N = 500$ (circles) and 50 (diamonds).

that we may have a current higher than the 1D CL current ($\langle I \rangle / I_{CL} > 1$) in the range of about $V_g/V_{th} = 1.06$ to 2. For example, the maximum ($\langle I \rangle / I_{CL} \approx 1.13$) occurs at around $V_g/V_{th} = 1.4$. We confirm that this maximal current is not due to numerical error, and the results are consistent when using different electron number $N = 10$ to 500 at $V_g/V_{th} = 1.4$ case. This increment is probably due to the nature of time-evolution of the current, which may have an higher upper bound of time-average CL law as suggested in a recent paper [105].

3.3 Discussion

To observe the proposed 1D classical CL law in the Coulomb blockade (or single electron) regime, it is assumed that there is an unlimited amount of electrons to be injected into the gap without having any potential barrier at the cathode interface. Assuming a planar gap with an emission size ($\sqrt{A}$) and gap spacing (D) in sub-micrometer scale, the magnitude of the threshold voltage (V_{th}) or the single electron charging potential is less than 1 V. At this low voltage regime, the applied electric field V_{th}/D is less than 10^6 V/m, and it is not sufficient for the emission of electrons at room temperature, due to the existence of the work function (or potential barrier) of the material. Some possible methods to increase electron emission at low voltage regime include using lasers to have a finite number of electron photo-field emission [5], or using sharp field emitters to enhance the tip's local field.

3.4 Chapter Summary

In summary, we have extended the 1D classical CL law to the Coulomb blockade (or single-electron) regime, where the applied voltage is comparable to the single electron

charging potential. It is found that there is a threshold voltage in order for the single-electron injected current to reach the 1D CL law. The single electron limit of the 1D classical CL law is demonstrated by having only one electron being transported across a gap when the applied voltage is slightly greater than the threshold value. A smooth transition to the traditional CL model is demonstrated.

CHAPTER 4

NOVEL SCALING LAWS FOR THE LANGMUIR-BLODGETT SOLUTIONS IN CYLINDRICAL AND SPHERICAL DIODES

4.1 Introduction

For 1D planar diode, the maximum current is given by the CL law [see Eq. (2.7)]. While for cylindrical and spherical diode, the space-charge-limited problem can not be solved analytically. The LB solutions given by Eq. (2.8) and Eq. (2.9) are in numerical form, and it is highly desirable to derive an accurate approximation with an adequate physical picture describing their underlying scalings. With the emergence of nano-particles and nano-tubes, scaling laws in these geometries would be of interest.

The CL current may be simply approximated by $I = Q/T$, where $Q = CV_g$ is the total bound surface charge on the cathode, C is the diode capacitance and T is the transit time of an electron to cross the gap subjected only to the vacuum field [106]. This capacitance model, or transit time model, yields a current density also given by Eq. (2.7) for the planar gap, except the numerical factor $4/9$ is replaced by

$1/2$, thus committing an error of only 12.5 percent [106]. That is, we need to multiply the current density obtained from the transit time model by the numerical factor $8/9$ to obtain the correct CL law in a planar gap. Because of its simplicity and accuracy, this capacitance or transit time model is of great utility to study a short pulse diode [42], and more recently the quantum regime [52].

To our knowledge, there is no simple analytical approximation for the LB solution (Eq. (2.8)), except for a recent model [107] that approximately solved the Poisson equation for a cylindrical diode. In this chapter, we apply the transit time model to the space-charge-limited problem in cylindrical and spherical geometries, novel scaling is obtained in Sec. 4.2. Later, the single electron model in Chapter. 3 for planar diode is extended to cylindrical and spherical diode in Sec. 4.3. Finally, we provide concluding remarks in Sec. 4.4.

4.2 Transit Time Model

4.2.1 Novel Scaling

Let us consider a cylindrical diode with an outer cathode located at $r = R_c$ and inner anode at $r = R_a (< R_c)$ as shown in Fig. 4.1a. The diode is applied with a voltage V_g across the gap with a spacing $D = R_c - R_a$ and a grounded cathode. For a cylindrical capacitor, the bound surface charge Q_b per unit length is

$$Q_b = CV_g, \quad (4.1)$$

where $C = 2\pi\epsilon_0/\ln(R_c/R_a)$ is the capacitance (per unit length). According to Gaussian law, the radial electric field inside the capacitor is

$$E_r(r) = \frac{Q_b}{\epsilon_0 2\pi r}, \quad (4.2)$$

Figure 4.1: Schematic for the case $R_a < R_c$ for (a) coaxial cylindrical diode and (b) concentric spherical diode.

and the electrical potential inside the gap is calculated by

$$V(r) = - \int_{R_c}^r E_r(r) dr = \frac{V_g}{\ln(R_c/R_a)} \ln(R_c/r). \quad (4.3)$$

Assuming all the electrons are liberated from the cathode at zero initial velocity, and they will reach the anode after a transit time T given by

$$T = \int_{R_c}^{R_a} \frac{dr}{-v} = - \int_{R_c}^{R_a} \frac{dr}{\sqrt{\frac{2eV(r)}{m}}}. \quad (4.4)$$

Here the velocity v is moving in the negative radial direction as the cathode is located outside the anode. By substituting Eqs. (4.3) into (4.4), we have

$$T = - \sqrt{\frac{m}{2e}} \sqrt{\frac{\ln(R_c/R_a)}{V_g}} \int_{R_c}^{R_a} \frac{dr}{\sqrt{\ln(R_c/r)}}, \quad (4.5)$$

and it can also be written as (by setting $x = R_c - r =$ distance measured from the cathode)

$$T = \sqrt{\frac{m}{2e}} \sqrt{\frac{\ln(R_c/R_a)}{V_g}} \int_0^D \frac{dx}{\sqrt{-\ln\left(1 - \frac{x}{R_c}\right)}}, \quad (4.6)$$

which can be solved numerically. However, we expect that space charge field of the emitted electrons plays more dominant roles near to the cathode region where $r \approx R_c$ or $x/R_c \ll 1$. Under this limit $x/R_c \ll 1$, we have $-\ln(1 - x/R_c) \approx x/R_c$, and the electron transit time or Eq. (4.6) becomes

$$T \approx \sqrt{\frac{m}{2e}} \sqrt{\frac{\ln(R_c/R_a)}{V_g}} 2\sqrt{R_c} D \quad (4.7)$$

Based on Eqs.(4.1) and (4.7), the electron current density [A/m²] emitted from the cathode is given by

$$J_c = \frac{Q_b}{T 2\pi R_c} = \frac{1}{2} \varepsilon_0 \sqrt{\frac{2e}{m}} \frac{V_g^{3/2}}{D^{1/2} R_c^{3/2} [\ln(R_c/R_a)]^{3/2}}. \quad (4.8)$$

Note this equation recovers the same scaling reported by Chen [107] [see Eq. (16)].

The difference is only the numerical constant in front of the constant, which is 1/2

4.2 Transit Time Model

as compared of 4/9. This minor discrepancy was also reported in the capacitor formulation of 1D planar case or CL law [106].

Thus by introducing a correction factor K into Eq.(4.8), and it becomes

$$J_c = K \frac{1}{2} \varepsilon_0 \sqrt{\frac{2e}{m}} \frac{V_g^{3/2}}{D^{1/2} R_c^{3/2} [\ln(R_c/R_a)]^{3/2}} = K \varepsilon_0 \sqrt{\frac{e}{2m}} \frac{E_c^{3/2}}{\sqrt{D}}. \quad (4.9)$$

where $K = 8/9$, and E_c is the surface electric field at the cathode given by

$$E_c = \frac{V_g}{R_c \ln\left(\frac{R_c}{R_a}\right)}, \quad (4.10)$$

which can also clearly be determined by using Eq.(4.2) at $r = R_c$ and Eq.(4.1).

By reversing the locations of the cathode and anode, so that the former becomes the inner electrode of the cylindrical diode, we may repeat the same method to obtain the analytical formula for the case where $R_c < R_a$, given by

$$J_c = K \frac{1}{2} \varepsilon_0 \sqrt{\frac{2e}{m}} \frac{V_g^{3/2}}{D^{1/2} R_c^{3/2} [\ln(R_a/R_c)]^{3/2}} = K \varepsilon_0 \sqrt{\frac{e}{2m}} \frac{E_c^{3/2}}{\sqrt{D}}. \quad (4.11)$$

Finally, using the current continuity condition, we obtain the current density at any position of r inside the cylindrical diode: $J_c(r) = J_c \times R_c/r$, where J_c is either from Eq. (4.9) or (4.11).

We can use the same approach to derive the SCL current for a spherical diode with the cathode and anode located, respectively at $r = R_c$ and $r = R_a (< R_c)$ as shown in Fig. 4.1b. The capacitance of the diode is given by

$$C = \frac{\varepsilon_0 4\pi R_a R_c}{(R_c - R_a)}, \quad (4.12)$$

and the electrical potential is

$$V(r) = \frac{V_g R_a R_c}{(R_c - R_a)} \left(\frac{R_c - r}{R_c r} \right). \quad (4.13)$$

For an electron emitted from the cathode, the transit time across the gap is

$$T = -\sqrt{\frac{m}{2e}} \sqrt{\frac{(R_c - R_a)}{V_g R_a R_c}} \int_{R_c}^{R_a} \left(\frac{R_c r}{R_c - r} \right)^{1/2} dr \quad (4.14)$$

$$= \sqrt{\frac{m}{2e}} \sqrt{\frac{(R_c - R_a)}{V_g R_a R_c}} \int_0^D \left[\frac{R_c (R_c - x)}{x} \right]^{1/2} dx. \quad (4.15)$$

Under same limit $x \ll R_c$, the transit time becomes

$$T \approx \sqrt{\frac{m}{2e}} \sqrt{\frac{(R_c - R_a)}{V_g R_a R_c}} 2R_c D^{1/2}, \quad (4.16)$$

which can be used to obtain the SCL current density:

$$J_c = \frac{1}{2} \varepsilon_0 \sqrt{\frac{2e}{m}} \left(\frac{R_a}{R_c} \right)^{3/2} \frac{V_g^{3/2}}{D^2}. \quad (4.17)$$

Similarly Eq.(4.17) can be rewritten as

$$J_c = K \frac{1}{2} \varepsilon_0 \sqrt{\frac{2e}{m}} \left(\frac{R_a}{R_c} \right)^{3/2} \frac{V_g^{3/2}}{D^2} = K \varepsilon_0 \sqrt{\frac{e}{2m}} \frac{E_c^{3/2}}{\sqrt{D}}, \quad (4.18)$$

where $K = 8/9$ is the same correction factor, and E_{sc} is the surface electric field at the cathode given by

$$E_{sc} = \frac{R_a}{R_c} \frac{V}{D}. \quad (4.19)$$

For the case $R_c < R_a$, the current density on the cathode is simply

$$J_c = K \frac{1}{2} \varepsilon_0 \sqrt{\frac{2e}{m}} \left(\frac{R_c}{R_a} \right)^{3/2} \frac{V_g^{3/2}}{D^2} = K \varepsilon_0 \sqrt{\frac{e}{2m}} \frac{E_c^{3/2}}{\sqrt{D}}. \quad (4.20)$$

Inside the spherical diode, the SCL current density at any position r is $J_c(r) = J_c \times R_c^2/r^2$.

The poor agreement between the transit time model and the exact LB solutions for the cylindrical and spherical diodes, which was also noted independently by Carter [108], prompted us to look for a more accurate approximation to the LB solutions. We find that, Eqs. (4.9), (4.11), (4.18) and (4.20) has the same form. So, over a fairly large range of R_c/R_a , the LB solutions may be approximated by

$$J_{app} = \frac{4}{9} \varepsilon_0 \sqrt{\frac{2e}{m}} \frac{E_c^{3/2}}{\sqrt{D}}, \quad (4.21)$$

for both cylindrical and spherical diode. Here E_c is the cathode surface electric field of a vacuum diode and D is the gap spacing, irrespective of whether the cathode is inside or outside the anode. Note that the very different new scaling, Eq. (4.21),

Figure 4.2: Comparison between the LB law (solid line) and its various approximations for a cylindrical diode over a wide range of R_c/R_a . The inner radius either R_c or R_a is 1 cm. The applied voltage is $V_g = 1$ V. Symbols represent the approximate expressions: crosses for transit-time model (TTM) using T given by Eq. (4.4); squares for the new scaling Eq. (4.21); circles for Eq. (4.22), the new expression of LB law; and triangles for the single electron capacitor model (SECM). Also plotted is the error in the approximate formulas compared with the LB law.

Figure 4.3: Same as Fig. (4.2), except for a spherical diode.

4.2 Transit Time Model

from Eq. (2.7). Note also that Eq. (4.21) becomes the exact CL solution, Eq. (2.7), in a planar cathode, for which $E_c = V_g/D$.

The approximate solution, Eq. (4.21), gives a novel scaling (NS) and is also presented in Figs. (4.2) and (4.3). Its percentage error in comparison with the exact LB model, is shown by the dash curves in Figs. (4.2) and (4.3). These figures show that Eq. (4.21) is accurate to within 30 percent, for $0.002 < R_a/R_c < 20$, whether the diode is cylindrical or spherical. It is far more accurate than the transit time model, and over a much wider range of R_a/R_c .

Fig. (4.2) and Fig. (4.3) show the LB solution for the cylindrical (Eq. (2.8)) and spherical (Eq. (2.9)) diode respectively. In these figures, we set $V_g = 1$ V, and the inner radius of the diode at 1 cm, whether it be the anode or cathode radius. It is clear from Eqs. (2.8) and (2.9) that, once the gap voltage and the inner radius are fixed, J_{LB} depends only on the radius ratio R_a/R_c ; and J_{LB} at other values of gap voltage and inner radius may be obtained accordingly. The transit-time model (TTM) with Eq. (4.4), is presented in Figs. (4.2) and (4.3) to compare with the exact LB solutions. The percentage error in the transit-time model is shown by the dash-dot curves in Figs. (4.2) and (4.3). Over much of the ranges of R_a/R_c that were considered by LB, these percentage errors greatly exceed the 12.5 percent error that is incurred in the transit-time model for the planar diode [106].

4.2.2 New Expression of LB law

Here, we present an empirical correction to Eq. (4.21) that fits the exact LB solution over the large ranges of R_a/R_c considered by LB [109, 7],

$$J_{app} = \frac{4}{9} \epsilon_0 \sqrt{\frac{2e}{m}} \frac{E_c^{3/2}}{\sqrt{D}} [1 + F(R_c/R_a)], \quad (4.22)$$

Here, the correction factor F within the range $3 \times 10^5 < R_c/R_a < 500$ is given, for a cylindrical diode, by

$$F\left(\frac{R_c}{R_a}\right) = \exp\left[\frac{(s - s_0)(7s + 23)(8s - 417)}{143742}\right] - 1, \quad (4.23)$$

and, for a spherical diode, by

$$F\left(\frac{R_c}{R_a}\right) = \exp\left[\frac{(s - s_0)(9s - 37)(4s + 143)}{42092}\right] - 1, \quad (4.24)$$

where $s = \ln[\ln(1 + R_c/R_a)]$ and $s_0 = \ln[\ln(2)]$. Note that $F = 0$ when $R_c/R_a = 1$, as expected.

The improved approximate solution, Eq. (4.22), is also presented in Figs. 4.2 and 4.3. Its percentage errors in comparison with the exact LB model, Eq. (4.2), are shown by the dotted curves in Figs. 4.2 and 4.3. These figures show that Eq. (4.8) is accurate to within 2.5 percent for $0.002 < R_a/R_c < 30000$, whether the diode is cylindrical or spherical. Note from Figs. 4.2 and 4.3 that, over this range of R_a/R_c , the LB solution varies over many orders of magnitude.

The value of the correction factor F in Eq. (4.22) is small compared with unity when the cathode is outside; in fact, it is always less than 0.2 for $1 < R_c/R_a < 500$ according to Figs. 4.2 and 4.3. That is, Eq. (4.21) provides the dominant scaling if the cathode is closer to the planar geometry relative to the anode surface. On the other hand, if the cathode is inside and the anode is outside at a large distance ($R_a/R_c \gg 1$, as in Figs. 4.2 and in particular Fig. 4.3), the cathode surface is poorly approximated by a planar surface and the correction factor F is of order unity or larger. Thus, the dominant scaling, Eq. (4.21), may be deduced by concentrating at the close proximity of the cathode surface, where the local curvature effects of the cathode can be neglected, and the anode would play a secondary role.

4.3 Single Electron Capacitor Model

The use of the vacuum field ignores the space charge effect that is empirically modeled by the correction factor F in Eq. (4.8). To correctly include the space charge effect in the transit time model, we inject a single electron (in the form of an infinitesimally thin cylindrical or spherical shell) into the diode, one at a time [110]. At time $t = 0$, there are N electrons located on the cathode, ready to be injected into the gap, and the order of injection is labeled by $n = 1, 2, \dots, N$. The initial condition of the electron can be described as: $r_n(0) = R_c$, $\frac{dr_n(0)}{dt} = 0$. An electron (shell) is injected into the gap only if the total electric field acting on it would initiate an acceleration toward the anode. For the first electron to be injected, the imposed gap voltage must exceed a threshold voltage, $V_{th} = e/2C$, where C is the capacitance in the cylindrical or spherical diode. (This threshold voltage leads to Coulomb blockade in the single-electron regime.) The radial position of the n -th electron shell, denoted as $r_n(t)$, is computed from the total force acting on it. For a cylindrical diode with $R_c < R_a$, the induced charge on the capacitor $q(t)$ is given in terms of $r_n(t)$ by

$$q(t) = CV_g + \frac{e}{\ln\left(\frac{R_c}{R_a}\right)} \sum_{n=1}^N \ln\left(\frac{R_c}{r_n(t)}\right). \quad (4.25)$$

The Ramo-Shockley theorem [103, 104] yields the instantaneous current through the diode, $I(t) = dq(t)/dt$, which is given by

$$I(t) = \frac{e}{\ln\left(\frac{R_a}{R_c}\right)} \sum_{n=1}^N \frac{1}{r_n(t)} \frac{dr_n(t)}{dt}, \quad (4.26)$$

On application of Gauss's law, the field acting on the n -th electron is

$$E_n(t) = \frac{q - e(n - 1/2)}{2\pi r_n(t) L \varepsilon_0}. \quad (4.27)$$

The dynamic motion of electron is governed by

$$\frac{d^2 r_n(t)}{dt^2} = \frac{e}{m} E_n(t). \quad (4.28)$$

The time-average current for the N injected electrons is equal to $\langle I \rangle = eN/t_N$, where t_N is the total time measured from the first electron $n = 1$ when it is injected from the cathode and the time when the N -th electron arrives at the anode.

For a cylindrical diode with $R_c > R_a$, the induce charge is given by:

$$q(t) = CV_g + \frac{e}{\ln\left(\frac{R_c}{R_a}\right)} \sum_{n=1}^N \ln\left(\frac{R_c}{r_n(t)}\right), \quad (4.29)$$

The, electric field acting on the electron is the same as that for the case $R_c < R_a$ and the dynamic equation is

$$\frac{d^2 r_n(t)}{dt^2} = -\frac{e}{m} E_n(t). \quad (4.30)$$

For Spherical diode, the charge can be described as:

$$q(t) = CV_g + e \frac{R_a R_c}{R_a - R_c} \sum_{n=1}^N \frac{r_n(t) - R_c}{r_n(t) R_c}, \quad (4.31)$$

for the case $R_c < R_a$ and

$$q(t) = CV_g + e \frac{R_a R_c}{R_c - R_a} \sum_{n=1}^N \frac{R_c - r_n(t)}{r_n(t) R_c}, \quad (4.32)$$

for the case $R_c > R_a$. The field acting on the electrons is

$$E_n(t) = \frac{q - e(n - 1/2)}{\varepsilon_0 4\pi r_n^2(t)}. \quad (4.33)$$

The time-average current density on the cathode is presented in Figs. (4.2) and (4.3), designated as “single electron capacitor model(SECM)”. Its percentage error in comparison with the exact LB model, is shown by the dash-dot-dot curves in Figs. (4.2) and (4.3). These figures show that the SECM is accurate to within 10 percent for $0.002 < R_a/R_c < 30000$, an acceptable result in these particle-in-cell like simulations.

We shall next show that the LB law also applies to the Coulomb blockade regime, at low numbers of N . In Fig. (4.4), we show the normalized $\langle I \rangle$ relative to the LB value (solid line) for $R_c = 100$ nm, $D = R_a - R_c = 100$ nm and $N = 50$ with $E_c/E_{th} = 1.02$ to 3.5 . The higher than LB value shown in the range of $1 < E_c/E_{th} < 2$,

Figure 4.4: Normalized time-average current from the single electron capacitor model as a function of normalized electric field at cathode E_c/E_{th} for a cylindrical diode of $R_c = 100$ nm, and gap spacing $D = R_a - R_c = 100$ nm. The insets show the time-dependent current $I(t)$ [relative to the LB solution] for $E_c/E_{th} = 1.02$ at $N = 10$ and $E_c/E_{th} = 3$ at $N = 20$.

due to the Coulomb blockade effect [110], and it has been confirmed by Griswold et al. [111] in their particle-in-cell simulation.

The insets in Fig. (4.4) show the temporal evolution for the two cases, $E_c/E_{th} = 1.02$ at $N = 10$, and $E_c/E_{th} = 3$ at $N = 20$. Note that for $E_c/E_{th} = 1.02$, there is only one electron transported through the gap within one transit time T . Subsequent electrons will only be injected into the gap while the previous electron nearly arrives at the anode. For $E_c/E_{th} = 3$, there is more than one electron (between 0.5 and 1.5) per transit time for which $I(t)$ is nonzero.

In Fig. (4.5), we plot the corresponding the calculated transit time T_s normalized with the vacuum-gap transit time T as a function of $E_c/E_{th} = 1.02$ to 4 at different gap spacing $D = R_a - R_c = 50, 100$ and 200 nm at fixed $R_c = 100$ nm. From the figure, we can see that the space charge effect will in general increase the vacuum transit time by 40% or more. In the range of $E_c/E_{th} > 3$ that $\langle I \rangle$ equals to the LB solution as shown in Fig. (4.4)a, the values of T_s/T are about 1.45 to 1.5. As compared to a gap of $D = 1$ cm used earlier in Figs. (4.2) and Fig. (4.3), we have chosen a gap of $D = 50$ to 200 nm in the presentation of Fig. (4.5) as microdiode can have space charge modulation at THz frequency that may be useful for the development of vacuum devices operating at THz frequency. In Fig. (4.5), the inverse of the transit time $f = 1/T_s$ corresponds to a frequency range of 0.1 to 1.5 THz for $D = 100$ nm from $E_c/E_{th} = 1.02$ to 4.

4.4 Summary

In summary, we generalize the simple model using capacitance for 1D planar diode to a transit time model and apply it to the SCL current in cylindrical diode and spherical diode. we present semi-empirical formulas which provide an excellent approximation to the LB solutions for cylindrical and spherical diodes, essentially for the entire

Figure 4.5: The corresponding transit time T_s (with space charge effects) normalized to the vacuum transit time T as a function of E_c/E_{th} at different $D = R_a - R_c = 50, 100$ and 200 nm for fixed $R_c = 100$ nm.

range of R_a/R_c that is of practical significance. This simple dependence given in Eq. (4.21) was qualitatively derived by focusing on the approximate transit time of an electron sheet, which is spent mostly in the vicinity near the cathode surface. Different from the well-known Child-Langmuir's $V_g^{3/2}/D^2$ scaling given by Eq. (2.7), the dominant dependence is given by Eq. (4.21) depends on the vacuum electric field on the cathode surface. This dominance of the vacuum field in the immediate vicinity of the cathode surface may have some implication in the contemporary development of electron gun codes, where modeling of electron emission in the first numerical grid proves most critical [112]. Corrections to the transit time including the space charge effects are considered by developing a single electron capacitor model, down to the Coulomb blockade regime where electrons numbered in the single digits are present in the diode.

The interesting connections between nano-particles, nano-cavities, Coulomb blockade, and the THz regime, remain to be explored. This model might be extended to the quantum regime [21, 22, 113], whose quantum CL law was recently used to study the transfer of plasmon charges between two nearly touching metallic nano-particles [98]. The proposed scaling might be useful to other space charge dominated systems, such as organic semiconductors [114], metal-molecule-metal junctions [115], optical field emission at high field [5], nanowires [116] and compact high power THz radiation sources [117].

CHAPTER 5

ANALYTICAL RE-DERIVATION OF SPACE CHARGE LIMITED CURRENT IN SOLIDS USING THE CAPACITOR MODEL

5.1 Introduction

For a 1D trap-free solid, the corresponding SCL current density is known as the Mott-Gurney (MG) law[76] given by Eq. (2.19) in Chapter. 2. If the solid has exponentially distributed traps (in energy), it is known as the Mark-Helfrich (MH) law [76, 118], or the trap-limited SCL current density given by Eq. (2.24). There are renewed interests in SCL conduction found in many novel devices, such as graphene oxide sheets [79], light emitting diodes [80], GaN nanorods [8], organic devices [81, 119, 120, 121], polymer transistor [82], nanowire [83], magnetoresistance [85], photocurrent [86], and nanocrystallites embedded silicon Schottky junction [87]. New models of the MG law have also been constructed to study the effect of finite emission area [122] and the presence of free space between a solid and electrode [123].

The traditional approaches in the formulation of the SCL current density in both free space and solids include solving the Poisson equation, continuity equation, and equation of motion with the related boundary conditions at the cathode ($z = 0$) and anode ($z = D$): $V(z = 0) = 0$, and $V(z = D) = V_g$, and $dV/dz[z = 0] = 0$ (electric field is zero at the cathode) .

It was shown in Chapter. 4 that the formulation of the CL law for planar diode and LB law for cylindrical and spherical diode can be reproduced by using a simple capacitor model at steady-state conditions [16] and in the single-electron limit [110]. In this chapter, we are interested to show that the SCL current transport in solids, namely the 1D analytical MH and MH law, can be reproduced by using a capacitor model for both the planar and cylindrical diode.

5.2 Trap-free Solid Case

First, consider a trap-free solid diode as a parallel plate capacitor with a total bound charge of Q_b given by

$$Q_b = CV_g, \quad (5.1)$$

where $C = \varepsilon_0 \varepsilon_r A/D$ is the capacitance, A is the area of the plate, and D is the separation between the plates of the capacitor. Without space charge effects, the electric field inside the solid between the plates is $E = V_g/D$. When the electron transports inside the solid, its velocity is determined by mobility of the solid and the electric field given by $v = \mu \times E = \mu V_g/D$. We assume all the negative bound charges (electrons) are injected from the cathode at zero-initial velocity with a transit time (across the trap-free solid) given by

$$\tau = \frac{D}{v} = \frac{D^2}{\mu V_g}. \quad (5.2)$$

5.2 Trap-free Solid Case

From Eqs. (5.1) and (5.2), the electron current density J is simply given by

$$J = \frac{Q_b}{\tau A} = \mu \varepsilon_0 \varepsilon_r \frac{V_g^2}{D^3}, \quad (5.3)$$

which has the same expression as the 1D MG law [see Eq. (2.19)] except for the numerical constant $9/8$. Note that this numerical constant can be obtained by using the continuity equation and the Poisson equation (see below).

From the 1D steady-state continuity equation, $dJ/dz = 0$, one can find that J is independent of z , and Eq. (5.3) implies that the electrical potential distribution has a scaling

$$\phi(z) = \frac{V_g}{D^{3/2}} z^{3/2}. \quad (5.4)$$

The electric field is given by:

$$E(z) = -\phi'(z) = -\frac{3}{2} \frac{V_g}{D^{3/2}} z^{1/2}. \quad (5.5)$$

By inserting Eq. (5.4) into the Poisson equation, we obtain the electron charge density ρ , which is

$$\rho(z) = \varepsilon_0 \varepsilon_r \phi''(z) = \varepsilon_0 \varepsilon_r \frac{3}{4} \frac{V_g}{D^{3/2}} z^{-1/2}. \quad (5.6)$$

Finally, the electron current density can be determined by evaluating the velocity and the electron charge density collected at the the anode ($z = D$), which are $v(D) = \frac{3}{2} \mu V_g / D$, and $\rho(D) = \frac{3}{4} \varepsilon_0 \varepsilon_r V_g / D^2$, respectively. In doing so, we recover the 1D SCL current density for a trap-free solid or the 1D MG law given by

$$J = \rho(D) v(D) = \frac{9}{8} \mu \varepsilon_0 \varepsilon_r \frac{V_g^2}{D^3}. \quad (5.7)$$

Note that in this method of formulation, there is no need to solve the Poisson equation with the boundary conditions (as done in the traditional approach), except for a simple differentiation as shown above in Eq. (5.6).

The 1D MG law for a planar diode has also been extended to a cylindrical model [124, 125, 126], where the analytical formula has been obtained at the limit case of

very small cathode radius as compared to the anode radius ($r_c \ll r_a$). Here, we will use the same capacitor model to reproduce the analytical formulas for the trap-free solids. For a cylindrical diode, the capacitance (per unit length) is

$$C = \frac{2\pi\epsilon_r\epsilon_0}{\ln(r_a/r_c)}. \quad (5.8)$$

The mean velocity for an electron to transit across the cylindrical gap is

$$\langle v \rangle = \frac{\int_{r_c}^{r_a} v(r) dr}{r_a - r_c} = \mu \frac{Q}{2\pi\epsilon_r\epsilon_0} \frac{\ln(r_a/r_c)}{r_a - r_c}, \quad (5.9)$$

with a transit time of $\langle \tau \rangle = (r_a - r_c)/\langle v \rangle$. The total current line density I [A/m] = $Q/\langle \tau \rangle$ is

$$I = \frac{Q}{r_a - r_c} \langle v \rangle = 2\pi\epsilon_r\epsilon_0\mu \frac{V_g^2}{\ln(r_a/r_c) (r_a - r_c)^2}. \quad (5.10)$$

In the limit of $r_a \gg r_c$, Eq. (5.10) can be approximated as

$$I \approx 4\pi\epsilon\mu \frac{V_g^2}{-\left(\frac{r_c}{r_a} - 1\right) (r_a - r_c)^2}. \quad (5.11)$$

Eq. (5.11) can be further approximated as:

$$I = 4\pi\epsilon\mu \frac{V_g^2}{r_a^2 \left(1 - \frac{r_c}{r_a}\right) \left(1 - \frac{r_c}{r_a}\right)^2} = 4\pi\epsilon\mu \frac{V_g^2}{r_a^2 \left(1 - \frac{r_c}{r_a}\right)^3} \approx 4\pi\epsilon\mu \frac{V_g^2}{r_a^2}. \quad (5.12)$$

The current expression implies that the electrical potential is of the form of

$$\phi(r) = \frac{V_g}{r_a} r. \quad (5.13)$$

Finally, by evaluating the electron's velocity and density at $r = r_a \gg r_c$, as shown below

$$v(r_a) = \mu \left| \frac{d\phi(r)}{dr} \right|_{r=r_a} = \mu \frac{V_g}{r_a}, \quad (5.14)$$

$$\rho(r_a) = \epsilon_0\epsilon_r \frac{1}{r} \frac{d}{dr} \left[r \frac{d\phi(r)}{dr} \right]_{r=r_a} = \epsilon_0\epsilon_r \frac{V_g}{r_a^2}, \quad (5.15)$$

we recover the space-charge-limited current line density for a cylindrical trap-free diode [124]:

$$I = 2\pi r_a \rho(r_a) v(r_a) = 2\pi\epsilon_0\epsilon_r\mu \frac{V_g^2}{r_a^2}. \quad (5.16)$$

5.3 Trap-filled Solid Case

Similarly, the same approach can be used to recover the 1D MH law for a trap-filled solid. Suppose a material has an exponential trap distribution given by

$$N(E) = N_0 \exp[(E - E_c)/kT_t], \quad (5.17)$$

where $N_0 = N_t/kT_t$, N_t is the total trapped electron density, T_t is the temperature of the trap distribution, E_c is the energy at the bottom of the conduction band, and k is the Boltzmann constant. For a constant applied electric field $E = V_g/D$, we assume that the quasi Fermi level across the sample $E_f(z)$ is also constant defined as E_f . Under this condition, the density of the trapped electrons is

$$n_t = \rho_t/e = N_t \exp[(E_f - E_c)/kT_t]. \quad (5.18)$$

Similarly, the density of the free electrons at the valence band is

$$n_f = \rho_f/e = N_c \exp[(E_f - E_c)/kT], \quad (5.19)$$

where N_c is the effective density of states at the valence band. By eliminating E_f from Eqs. (5.18) and (5.19), the relationship between n_f and n_t can be obtained as

$$n_f = N_c \left(\frac{n_t}{N_t} \right)^l. \quad (5.20)$$

Consider that the injected electrons are from the bound charge on the capacitor $Q_b = CV_g$, which will fill the traps giving an expression of n_t :

$$n_t = \frac{CV_g}{eDA} = \frac{\varepsilon_0 \varepsilon_r V_g}{eD^2}. \quad (5.21)$$

By combining Eqs. (5.20) and (5.21), the charge density of the free electrons is

$$\rho_f = eN_c \left(\frac{\varepsilon_0 \varepsilon_r V_g}{eN_t D^2} \right)^l. \quad (5.22)$$

By knowing the velocity of the electrons $v = \mu V_g/D$, the electron current density $J = \rho_f \times v$ becomes

$$J = e\mu N_c \left(\frac{\varepsilon_0 \varepsilon_r}{eN_t} \right)^l \frac{V_g^{l+1}}{D^{2l+1}}. \quad (5.23)$$

From the continuity equation (J is a constant), Eq. (5.23) yields a scaling of

$$J \propto \frac{\phi^{l+1}(z)}{z^{2l+1}} = \frac{V_g^{l+1}}{D^{2l+1}}, \quad (5.24)$$

which implies that the electrical potential distribution is in the form of

$$\phi(z) = \frac{V_g}{D^{(2l+1)/(l+1)}} z^{(2l+1)/(l+1)}. \quad (5.25)$$

So that the electrical field can be derived as

$$E(z) = -\phi'(z) = -\frac{2l+1}{l+1} \frac{V_g}{D^{(2l+1)/(l+1)}} z^{l/(l+1)}. \quad (5.26)$$

From Eq. (5.25) and the Poisson's equation, the trapped charge density can be obtained as

$$\rho_t(z) = \varepsilon_0 \varepsilon_r \phi''(z) = \varepsilon_0 \varepsilon_r \frac{l(2l+1)}{(l+1)^2} \frac{V_g}{D^{(2l+1)/(l+1)}} z^{-1/(l+1)}. \quad (5.27)$$

From Eq. (5.20) and Eq. (5.27), we get derive the free electron density as

$$\rho_f(z) = eN_c \left[\frac{\varepsilon_0 \varepsilon_r (2l+1) l}{eN_t (l+1)^2} \frac{V_g}{D^{(2l+1)/(l+1)}} z^{-1/(l+1)} \right]^l. \quad (5.28)$$

Finally, by knowing that the drift velocity at the anode ($z = D$) is $v(D) = \mu |E(D)| = \mu \frac{2l+1}{l+1} \frac{V_g}{D}$, and by determining the free charge density [Eq.(5.28) with $z = D$] as $\rho_f(D) = eN_c \left[\frac{\varepsilon_0 \varepsilon_r (2l+1) l}{eN_t (l+1)^2} \frac{V_g}{D^2} \right]^l$, we recover the space-charge-limited current for a trap-filled solid or [the 1D MH law as shown in Eq.(2.24)] as $J_{MH} = \rho_f(D) v(D)$:

$$J_{MH} = N_c \mu e^{(1-l)} \left[\frac{\varepsilon_0 \varepsilon_r l}{N_t (l+1)} \right]^l \left(\frac{2l+1}{l+1} \right)^{(l+1)} \frac{V_g^{(l+1)}}{D^{(2l+1)}}. \quad (5.29)$$

To develop the cylindrical model developed for a trap-filled solid, we first suppose the injected electrons will fill the traps in a capacitor model, given by

$$Q_t = \pi (r_a^2 - r_c^2) \rho_t = CV_g. \quad (5.30)$$

From Eq. (5.20), the line charge density of the free electrons is

$$Q_f = \pi (r_a^2 - r_c^2) N_c e \left[\frac{CV_g}{\pi (r_a^2 - r_c^2) eN_t} \right]^l. \quad (5.31)$$

5.3 Trap-filled Solid Case

From Eq. (5.9), we may solve for the mean velocity $\langle v \rangle$ with $Q = Q_t$ [from Eq.(5.30)], and derive the current line density as

$$I = \frac{Q_f}{r_a - r_c} \langle v \rangle = \frac{(2\pi\epsilon_0\epsilon_r)^l \mu N_c e \pi}{(N_t e \pi)^l} \frac{V_g^{l+1}}{(r_a - r_c)^2 \ln\left(\frac{r_a}{r_c}\right)^l (r_a^2 - r_c^2)^{l-1}}. \quad (5.32)$$

In the limit of $r_a \gg r_c$, Eq. (5.32) becomes

$$I \approx \frac{(2\pi\epsilon_0\epsilon_r)^l \mu N_c e \pi}{(N_t e \pi)^l} \frac{V_g^{l+1}}{r_a^{2l}}. \quad (5.33)$$

Eq. (5.34) implies the current scaling:

$$I \propto \frac{V_g^{l+1}}{r_a^{2l}} = \frac{\phi(r)^{l+1}}{r^{2l}}, \quad (5.34)$$

which implies that the potential distribution is of the form of

$$\phi(r) = \frac{V_g}{r_a^{2l/(l+1)}} r^{2l/(l+1)}. \quad (5.35)$$

By using the electron mobility equation, the Poisson equation and Eq. (5.20), we can obtain the electron's velocity, the trap charge density, and free electron density (all at $r = r_a \gg r_c$), respectively, as

$$v(r_a) = \mu \frac{V_g}{r_a} \frac{2l}{l+1}, \quad (5.36)$$

$$\rho_t(r_a) = \epsilon_0 \epsilon_r \left(\frac{2l}{l+1} \right)^2 \frac{V_g}{r_a^2}, \quad (5.37)$$

$$\rho_f(r_a) = e N_c \left[\frac{\epsilon_0 \epsilon_r}{e N_t} \left(\frac{2l}{l+1} \right)^2 \frac{V_g}{r_a^2} \right]^l. \quad (5.38)$$

Finally, using Eqs. (5.36) and (5.38), we recover the space-charge-limited current line density for a cylindrical trap-filled diode [124]:

$$I = 2\pi e N_c \mu \left[\frac{\epsilon_0 \epsilon_r}{e N_t} \right]^l \left(\frac{2l}{l+1} \right)^{2l+1} \frac{V_g^{l+1}}{r_a^{2l}}. \quad (5.39)$$

5.4 Summary

In summary, we have confirmed that the capacitor approach [16] that was successful in the re-derivation of the space-charge-limited (SCL) current in free space or the 1D Child-Langmuir (CL) law, is also applicable for the SCL conduction in both trap-free and trap-filled solids, which are known as the 1D Mott-Gurney (MG) law and 1D Mark-Helfrich (MH) law, respectively. It will be of interest for further investigations to apply the capacitor model to other SCL models such as bipolar SCL (electron and hole) conduction in organic electronics, cylindrical CL law, 2D models, and also to different physical regimes including relativistic and quantum effects, just to name a few. It is noted that this effort might provide insight for the material-by-design community by clearly showing what physical effects are important in the engineering of densities of states in novel materials and geometries.

CHAPTER 6

NON-UNIFORM SPACE-CHARGE-LIMITED CURRENT INJECTION INTO SOLID

6.1 Introduction

In 2007, the 1D MG law (Eq. 2.19) was extended to a 2D uniform model [24] for an infinitely long emitting strip with width W , which revealed a geometrical enhancement of

$$\frac{J_{MG}(2D)}{J_{MG}(1D)} = 1 + \frac{4/3}{\pi(W/D)} \quad (6.1)$$

for $W/D \geq 1$, and it agrees well with a device simulator (with error less than 5%). It was also found that the enhancement factor decreases at larger $l > 2$ as compared to the trap-free case of $l = 1$ [122]. Such geometrical enhancement effects had also been observed in nanowires, but with a much smaller emitting area [8, 9] and much higher enhanced value. The crossover from 1D to 2D SCL current conduction behavior was also observed in single crystals [10].

However, the 2D models [8, 122] have assumed uniform SCL current injection into the solid, which may be not valid due to edge effects, especially when the typical

size of the injecting area is much smaller than the diode spacing. Based on the definition of the SCL condition, in order to drive the electric field at the edge to zero, higher current is required to be emitted near the edge. This can be seen in a later discussion based on a mathematical point of view.

Thus, there is a need to construct a non-uniform 2D model for SCL current injection into a solid as the uniform injection model may be not valid in many experiments using nanoscale materials. For simplicity, we will focus on a 2D non-uniform model having a finite emitting width W in only 1 direction (see Fig. 6.1 below). The approach presented in this chapter can be extended to different geometries such as nanowires and nanorods, which will serve for future works.

Before proceeding forward, it is worth to emphasize that 2D uniform [127, 31, 32] and non-uniform [33, 128, 129] models of SCL current conduction were first studied in a vacuum diode known as the 1D Child-Langmuir (CL) law (see Eq. (2.7)) [11, 12]. In the vacuum diode, similar geometrical enhancement effects have been calculated and observed.

In this chapter, a 2D model for space-charge-limited injection will be given in Sec. 6.2. The trap effect on current enhancement will be discussed in Sec. 6.3. Lastly, a short summary will be presented in Sec. 6.4.

6.2 2D Model for a Trap-free Solid

6.2.1 Theoretical Model

As shown in Fig. 6.1, we first consider a trap-free solid of length D and width W , which is sandwiched between two metallic electrodes with a bias voltage of V_g . The solid is assumed to be infinite in the z -direction. The size of both electrodes is L , which is assumed to be much larger than W . The electrons are injected from the lower

Figure 6.1: Schematic diagram of SCL electron injection into a solid inside a planar diode with spacing D . The solid has a finite width of W and is infinitely long in the z direction.

electrode ($y = 0$ and $-W/2 < x < W/2$) into the solid. The interface is assumed to be an Ohmic contact. The effect of the Schottky barrier on SCL current injection [130] is ignored.

From the Poisson equation, we obtain the following normalized differential equations:

$$\nabla^2 \phi(x, y) = 0, \text{ for } |x| > W/2, \quad (6.2a)$$

$$\nabla \bullet [\varepsilon(y) \nabla \phi(x, y)] = \frac{J}{\mu \frac{\partial \phi(x, y)}{\partial y}}, \text{ for } |x| \leq W/2, \quad (6.2b)$$

where ε is the dielectric constant. Eq. (6.2) can be solved numerically with the following boundary conditions:

$$\phi(x, y) = 0, \text{ for } y = 0, |x| < L/2, \quad (6.3a)$$

$$\frac{\partial \phi(x, y)}{\partial y} = 0, \text{ for } y = 0, |x| < W/2, \quad (6.3b)$$

$$\phi(x, y) = V_g, \text{ for } y = D, |x| < L/2, \quad (6.3c)$$

$$\phi(x, y) = V_g \left(\frac{y}{D} \right), \text{ for } |x| = L/2, \quad (6.3d)$$

In our model, the non-uniform injection of current density is characterized by Eq. (6.2b). Its dependence on x is required to be determined numerically subjected to the boundary conditions. Here, SCL current injection is defined by having the electric field equal to zero at all the injection points at the lower electrode, as shown in Eq. (6.3b). Note it is not trivial to solve Eqs. (6.2) and (6.3) due to this unknown parameter. From the analysis, we find that only two of the boundary conditions given in Eqs. (6.3a), (6.3b) and (6.3c) can be satisfied if a constant current density has been assumed in solving Eq. 6.2, which explains mathematically why a non-uniform model is required. Thus, one can imagine that the current density would have a non-uniform profile, like having higher value near the edge in order to satisfy the boundary conditions.

6.2.2 Numerical Method and Results

In this chapter, we use the numerical method developed in the calculation of the 2D non-uniform CL law for a vacuum diode [128]. However, there are two major differences in our model compared to Ref. [128]. The first difference is that the transport mechanism is different from a vacuum diode for which a new profile of the surface electric field is required to solve the equations. The second one is due to the difference of the dielectric constant (ε) at the solid-vacuum interface [indicated in the box of Fig. 6.1], which implies that ε is not a constant in Eq. (6.2b).

In our calculation, we first assume a specified J in an inner iteration step, and solve Eqs. (6.2) and (6.3) numerically for $\phi(x, y)$. We discretize the computational domain with a square grid. The Laplacian in finite difference form on a typical five-point stencil is given by

$$\nabla^2 \phi = \frac{\phi_{i+1,j} - 2\phi_{i,j} + \phi_{i-1,j}}{h^2} + \frac{\phi_{i,j+1} - 2\phi_{i,j} + \phi_{i,j-1}}{h^2}, \quad (6.4)$$

and the first order differential is approximated using

$$\frac{\partial \phi(x, y)}{\partial y} = \frac{\phi_{i,j+1} - \phi_{i,j-1}}{2h}, \quad (6.5)$$

where h is the grid size. Eq. (6.2) is then rewritten as

$$\begin{aligned} \phi_{i,j}^{n+1} = & \phi_{i,j}^n + \frac{\theta}{4\varepsilon_{j-1} + \varepsilon_{j+1}} \left[\frac{\varepsilon_{j-1} + \varepsilon_{j+1}}{2} \phi_{i-1,j}^n + \frac{\varepsilon_{j-1} + \varepsilon_{j+1}}{2} \phi_{i+1,j}^{n+1} + \varepsilon_{j-1} \phi_{i,j-1}^n \right. \\ & \left. + \varepsilon_{j+1} \phi_{i,j+1}^{n+1} - 4 \frac{\varepsilon_{j-1} + \varepsilon_{j+1}}{2} \phi_{i,j}^n - \delta_j \frac{h^2 J}{\mu} \left(\frac{\phi_{i,j+1}^n - \phi_{i,j-1}^{n+1}}{2h} \right)^{-1} \right], \end{aligned} \quad (6.6)$$

where $n+1$ and n are the $(n+1)$ -th and n -th iterative steps, respectively. δ_j is 0, 1 and 0.5, respectively, for the grid point in vacuum, dielectric and vacuum dielectric interface. Here, θ (a value between 1 and 2), is a relaxation parameter used to accelerate the iteration. We use

$$\theta = \frac{2}{1 + \sqrt{1 - \left[\frac{\cos(\pi/m) + \cos(\pi/n)}{2} \right]^2}}, \quad (6.7)$$

where m and n is the element numbers in the x and y direction.

From the numerically solved potential distribution $\phi(x, y)$, one can calculate the surface electric field at the injecting electrode. In the SCL regime, the electric field varies rapidly near the electrode, which makes the calculation of the surface electric field difficult. This problem can be solved by applying the following extrapolating function,

$$f(y) = A + By^{1/2} + Cy^{2/2}, \quad (6.8)$$

where A is defined as the normal component of the electric field. By using Eq. (6.8) and the calculated field, at the 3 nearest half-grid points (to the injecting electrode), we can numerically solve the following equations for A , B and C , and thus obtain the surface electric field through the value of A :

$$E_y(y = h/2) = A + B(h/2)^{1/2} + C(h/2)^{2/2}, \quad (6.9a)$$

$$E_y(y = 3h/2) = A + B(3h/2)^{1/2} + C(3h/2)^{2/2}, \quad (6.9b)$$

$$E_y(y = 5h/2) = A + B(5h/2)^{1/2} + C(5h/2)^{2/2}, \quad (6.9c)$$

where $E_y(y = h/2) = (\phi_{i,2} - \phi_{i,1})/h$, $E_y(y = 3h/2) = (\phi_{i,3} - \phi_{i,2})/h$, $E_y(y = 5h/2) = (\phi_{i,4} - \phi_{i,3})/h$. Note Eq. (6.8) is different to the one used in the CL model [128], as the electric field for SCL current in solids has a different scaling: $E_y(y) \propto y^{1/2}$.

In order to push the electric field to zero, we have another outer iteration process to update the spatial profile of $J(x)$ self-consistently. Firstly, we start with two uniform current densities which are a fraction of the current density described by the 1D MG law. For example, we normally use $J_1 = 0.2$ and $J_2 = 0.4$. We then solve the restricted nonlinear Poisson equation by considering only the Dirichlet boundary condition to obtain $\phi(x, y)$ for a given current density $J(x)$ (in the inner loop). The corresponding electrical fields on the surface are then numerically calculated as E_1 and E_2 , respectively. By using a secant method, we obtain an intermediate current

6.2 2D Model for a Trap-free Solid

Figure 6.2: The numerical evolution (indicated by the arrows) of (a) the normalized electric field at the injecting surface and (b) the normalized current density at $W/D = 1$. Here we have fixed $L/W = 4$.

density J_* given by

$$J_* = -\frac{(E_1 + E_2)}{2} \left(\frac{J_2 - J_1}{E_2 - E_1} \right) + \frac{(J_1 + J_2)}{2}, \quad (6.10)$$

and the new current density is updated by

$$J_{new} = 0.1J_* + 0.9J_{old}. \quad (6.11)$$

This numerical process will continue until convergence in the calculated electric field is better than 0.01%.

In Fig. 6.2, we show the numerical evolution of the normalized electric field, and its corresponding current density at $W/D = 1$. From the figure, we clearly

see a wing-like profile, which shows a larger current enhancement of J near the edge, qualitatively identical to the 2D non-uniform CL law [128]. We repeat the calculation at $W/D = 0.5$ and 1, the results of which are plotted in Fig. 6.3(a). For each W/D case we calculate the effects due to the dielectric constant ε_r at the interface: $\varepsilon_r = 3$ (dashed lines) for PPV film [131], $\varepsilon_r = 9$ (dashed-dotted lines) for GaN [8] and without this interface effect (solid lines). It is clear the enhancement near the edge reduces at large ε_r due to the reduction of the effective electric field at the interface given by $(V_g/D)/\varepsilon_r$.

By using $J(x)$, the total SCL current is calculated by integrating $J(x)$ over the width of the injecting surface. The results normalized to the 1D MG law [see Eq. (6.1)], I/I_{MG} are plotted in Fig. 6.3(b) in the range of $0.05 \leq W/D \leq 2.5$. The results may be fitted to an empirical scaling of

$$I/I_{MG} = \alpha_1 + \frac{\alpha_2}{(W/D)} - \frac{\alpha_3}{(W/D)^2}. \quad (6.12)$$

Here, the three constants $(\alpha_1, \alpha_2, \alpha_3)$ are, respectively, (1.0384, 0.3647, -0.0025), (1.016, 0.1651, -0.0013) and (1.0102, 0.0524, 0.0001) for no interface effect (circle), $\varepsilon_r = 3$ (square), and $\varepsilon_r = 9$ (diamond), respectively.

6.3 The Trap Effect

To include the effects of traps in a solid, we assume that the energy state of the traps in a solid is described by an exponential function $N(E) = (N_t/kT_c) \exp[(E - E_c)/kT_c]$. Based on Eq. (6.2), Eq. (6.5b) becomes

$$\nabla \bullet [\varepsilon(y) \nabla \phi(x, y)] = N_t e^{(l-1)/l} \left(\frac{J(x)}{N_0 \mu} \right)^{1/l} \left[\frac{\partial \phi(x, y)}{\partial y} \right]^{-1/l}, \text{ for } |x| \leq W/2, \quad (6.13)$$

Note, due to the different scaling for electric field at different l , Eq. (6.8) also becomes

$$f(y) = A + By^{l/(l+1)} + Cy^{2l/(l+1)}. \quad (6.14)$$

Figure 6.3: (a) The non-uniform profile of the normalized SCL current density for a trap-free solid at $W/D = 0.5$ and 1 for PPV films ($\epsilon_r = 3$) and GaN ($\epsilon_r = 9$). (b) The geometrical enhancement of the 2D non-uniform MG law over the 1D MG law as a function of W/D .

The electric field on the injection cathod surface can be obtained by solving the following equations:

$$E_y(y = h/2) = A + B(h/2)^{l/(l+1)} + C(h/2)^{2l/(l+1)}, \quad (6.15a)$$

$$E_y(y = 3h/2) = A + B(3h/2)^{l/(l+1)} + C(3h/2)^{2l/(l+1)}, \quad (6.15b)$$

$$E_y(y = 5h/2) = A + B(5h/2)^{l/(l+1)} + C(5h/2)^{2l/(l+1)}. \quad (6.15c)$$

Using the same numerical approach described above, we then calculate the 2D non-uniform trap-limited current density as a function W/D for different l and ε_r .

In Fig. 6.4(a), take Si as an example ($\varepsilon_r = 11.8$), we show the profile of $J(x) = J(x)/J_{TL}$ at $l = 2, 3, 4$ for various $W/D = 0.5$ and 1. One can see that the current density also increases at the edge, and the enhancement increase with larger l . Compared to the $l = 1$ case (like the trap-free case as shown in Fig. 3a), enhancement for $l = 2$ and $l = 3$ is more significant. Similarly the 2D total trap-limited current in terms of Eq. (6.2) is shown in Fig. 6.4(b). Using Eq. (6.11), we also obtain the fitting constants, which are (1.0817, 0.0335, 0.0076), (1.0537, 0.1319, 0.0082), (1.0218, 0.2478, 0.0137) for $l = 2, 3$ and 4 respectively. It is interesting to note that the geometrical enhancement is more important for trap-limited current for the non-uniform model (as presented here) as compare the 2D uniform model presented earlier [130]. For the uniform model, it was found that the geometrical enhancement decreases as l increases from $l = 1$ to $l = 2$ and 3 (see Fig. 3 in Ref. [130]).

From recent experimental results [8], it was suggested that the threshold voltage to reach the SCL limited current will be lower for nanowires. Thus, the non-uniform model presented in this thesis should be useful to characterize the high current SCL transport in nanowires at the regime when SCL conditions are reached at all surfaces.

Figure 6.4: (a) The non-uniform profile of the normalized SCL current density for a trap-filled solid like Si ($\epsilon_r = 11.8$) with different $l = 2, 3, 4$ at $W/D = 0.5$ and 1. (b) The geometrical enhancement of the 2D non-uniform trap-limited current over the 1D model as a function of W/D .

6.4 Summary

In summary, we have presented a 2D model of non-uniform SCL current injection into a solid (of width W) with and without traps. Empirical scaling has been provided for the geometrical enhancement factor as a function of W/D for different dielectric constants and trap distributions. For fixed W/D , it is found that the enhancement (over the 1D model) is higher for a solid with smaller dielectric constant and with larger energy spread in the trap distribution. Compared to the earlier uniform model, we observed higher local current density near the edges. This model will be useful for the characterization of material's properties, such as mobility and trap distribution. The non-uniformity of the SCL current injection presented here will be relevant to any injecting surface, especially for small areas such as nano-structures or small sized organic materials.

7.1 Conclusions

This thesis focused on the modeling of electron transport properties in vacuum diode and injection behavior in solids. We extend the traditional theories to new regimes, new scallings and investigate novel phenomena.

In Chapter 3, the one-dimensional (1D) classical Child-Langmuir (CL) law was extended to the Coulomb blockade (single to few electrons) regime, including the effect of single electron charging. It was found that there is a threshold of voltage (V_{th}) equals to one-half of the single electron charging energy for electron injection assuming zero barrier at the interface. For voltages in the range of $1 < V/V_{th} < 2$, there is only 1 electron inside the gap, and the time-average of the single electron injection may be equal to or greater than the 1D CL current.

In Chapter 4, it was found that the Langmuir-Blodgett (LB) solutions for the space-charge-limited current density, for both cylindrical and spherical diodes, may

be approximated over a wide range of parameters, where E_c is the surface electric field on the cathode of the vacuum diode and D is the anode-cathode spacing. This dependence is valid whether R_a/R_c is greater than or less than unity, where R_a and R_c is the anode and cathode radius, respectively. Minor empirical corrections to the above scaling yield fitting formulas that are accurate to within 2.5 percent for $0.002 < R_a/R_c < 30000$. An explanation of this scaling was given. An accurate transit time model yields the LB solutions even in the Coulomb blockade regime for a nanogap, where the number of electrons may be in the single digit, and the transit time frequency is in the THz range.

In Chapter 5, we have used a capacitor model to reproduce the known analytical formulas of space-charge-limited current transport inside both trap-free and trap-filled solids in planar geometry. It is found that the approach is simple as compared to the traditional method, as the latter involves a solving second order differential equation. Exact analytical results can also be obtained for cylindrical diode with an outer radius much larger than the inner radius.

In Chapter 6, we have constructed a two-dimensional (2D) non-uniform model to study the space-charge-limited current injection into a trap-filled solid. A non-uniform current density profile due to edge effects is observed. For a diode of length D with an infinitely long emitting strip, the enhancement over the 1D model depends on W/D , and also on material properties, such as the dielectric constant and the trap distribution. The findings here are different from the earlier uniform 2D model, and the difference can be significant for small W/D . The model can be readily extended to other geometrical settings.

7.2 Future Recommendations

Some possible extensions of the topics of this thesis are given as follows:

7.2 Future Recommendations

- In the single electron model, the image charge and the surface potential is completely ignored and the model is only 1D. In order to realistically model single electrons emission from sharp tips, three-dimensional effects should be taken into account. Therefore a multidimensional model including the realistic potential should be developed.
- The electron emission process is not considered for the single electron model. It would be interesting to investigate the transient behavior from ultrafast laser induced electron emission to SCL in the Coulomb blockade regime.
- The average current for space-charge-limited current in the Coulomb blockade regime can be much larger than the CL law value. Is it due to the time dependent current injection? This mechanism is still not very clear and requires further study.
- The novel scaling using the electrical field on the cathode surface now works for planar, cylindrical and spherical diodes. It would be of great interest to extend this scaling to much more complicate geometries such as a sharp tip.

AUTHOR'S PUBLICATIONS

Journal Papers

1. Y. B. Zhu, P. Zhang, A. Valfells, L. K. Ang and Y.Y. Lau. “Novel Scaling Laws for the Langmuir-Blodgett Solutions in Cylindrical and Spherical Diodes”, *Phys. Rev. Lett.*, **110**:265007 (2013).
2. Y. B. Zhu and L. K. Ang. “ChildLangmuir law in the Coulomb blockade regime”, *Appl. Phys. Lett.*, **98**:051502 (2011).
3. Y. B. Zhu and L. K. Ang. “Analytical re-derivation of space charge limited current in solids using capacitor model ”, *J. Appl. Phys.*, **110**:094514 (2011).
4. Y. B. Zhu and L. K. Ang. “Non-uniform space charge limited current injection into solid”, *Submitted*.

Conference Papers

1. Y. B. Zhu P. Zhang, A. Valfells, L. K. Ang and Y. Y. Lau “Transit Time Model for Space Charge Limited Current”, *IEEE Pulsed Power and Plasma Science Conference, San Francisco, USA*, 3B-7 (2013)
2. Y. B. Zhu and L. K. Ang. “Nonuniform 2d Mott-Gurney Law”, *IEEE Pulsed Power and Plasma Science Conference, San Francisco, USA*, P4-42 (2013)
3. Y. B. Zhu and L. K. Ang. “Nonequilibrium Effect for Shot Noise in Ultrafast Laser-Induced Electron Emission”, *IEEE Pulsed Power and Plasma Science Conference, San Francisco, USA*, P4-48 (2013)
4. Y. B. Zhu P. Zhang, A. Valfells, L. K. Ang and Y. Y. Lau “Simple scaling laws for space charge limited current flow in cylindrical and spherical diode”, *14th International Vacuum Electronics Conference, Paris, France*, (2013)
5. Y. B. Zhu and L. K. Ang. “Single-Electron Based Model of the Child-Langmuir Law”, *38th IEEE International Conference on Plasma Science, Chicago, USA*, IO1A-5 (2011)
6. Y. B. Zhu and L. K. Ang. “Space Charge Limited Current in a Gap Combined of Several Kinds of Medium”, *38th IEEE International Conference on Plasma Science, Chicago, USA*, IP1D-22 (2011)

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APPENDIX A

FINITE DIFFERENCE TREATMENT OF INTERFACE

The Gauss law for the electric field states that the electric flux leaving a Gaussian surface is equal to the free charge enclosed by the surface. That is

$$-\oint \varepsilon \nabla \varphi \bullet d\mathbf{c} = -q, \quad (\text{A.1})$$

where n is the unit outward normal to the contour. As shown in Fig. A.1, the flux leaving the right side of the contour is

$$\psi_R = \varepsilon_1 \frac{\phi_{i,j+1} - \phi_{i,j}}{h} \frac{h}{2}, \quad (\text{A.2})$$

where ε_1 is the dielectric constant of the material. Similarly, the flux leaving the left side is

$$\psi_L = \varepsilon_0 \frac{\phi_{i,j-1} - \phi_{i,j}}{h} \frac{h}{2}, \quad (\text{A.3})$$

ε_0 is vacuum dielectric constant. The flux leaving the top side is

$$\psi_T = \varepsilon_0 \frac{\phi_{i+1,j} - \phi_{i,j}}{h} \frac{h}{2} + \varepsilon_1 \frac{\phi_{i+1,j} - \phi_{i,j}}{h} \frac{h}{2}, \quad (\text{A.4})$$

and the flux leaving the bottom side is

$$\psi_B = \varepsilon_0 \frac{\phi_{i-1,j} - \phi_{i,j}}{h} \frac{h}{2} + \varepsilon_1 \frac{\phi_{i-1,j} - \phi_{i,j}}{h} \frac{h}{2}, \quad (\text{A.5})$$

Figure A.1: Nodes at the interface of vacuum and dielectrics.

The source term is given by

$$-q = \frac{J}{\mu E} h \frac{h}{2}, \quad (\text{A.6})$$

where $J/\mu E$ is the charge density, $h \cdot h/2$ is the half cell that contains charge. The finite difference version is then given by

$$\begin{aligned} & \varepsilon_1 \frac{\phi_{i,j+1} - \phi_{i,j}}{h} \frac{h}{2} + \varepsilon_0 \frac{\phi_{i,j-1} - \phi_{i,j}}{h} \frac{h}{2} + \varepsilon_0 \frac{\phi_{i+1,j} - \phi_{i,j}}{h} \frac{h}{2} + \varepsilon_1 \frac{\phi_{i+1,j} - \phi_{i,j}}{h} \frac{h}{2} \\ & + \varepsilon_0 \frac{\phi_{i-1,j} - \phi_{i,j}}{h} \frac{h}{2} + \varepsilon_1 \frac{\phi_{i-1,j} - \phi_{i,j}}{h} \frac{h}{2} = \frac{J}{\mu E} h \frac{h}{2}, \end{aligned} \quad (\text{A.7})$$

Rearranging the term gives

$$\frac{\varepsilon_1 + \varepsilon_0}{2} \phi_{i,j} = \frac{1}{4} \left(\varepsilon_0 \phi_{i,j-1} + \varepsilon_1 \phi_{i,j+1} + \frac{\varepsilon_0 + \varepsilon_1}{2} \phi_{i-1,j} + \frac{\varepsilon_0 + \varepsilon_1}{2} \phi_{i+1,j} \right) - \frac{J}{\mu E} \frac{h^2}{2}, \quad (\text{A.8})$$

APPENDIX B

CAPACITOR MODEL FOR RELATIVISTIC CL LAW IN 1D PLANAR DIODE

B.1 SCL Current in the Ultrarelativistic Regime

Consider a gap D with potential V_g , use a normalized potential U defined as

$$U(z) = \frac{e\phi(z)}{m_0c^2}, \quad (\text{B.1})$$

where ϕ is the potential at every point z , it has the form as $\phi(z) = V_g/D * z$.

According to energy conversation $e\phi = mc^2 - m_0c^2$, we have

$$\frac{v}{c} = \frac{[U(U+2)]^{1/2}}{(1+U)}, \quad (\text{B.2})$$

suppose $U^2 \gg 2U$, use Taylor expansion, we have

$$\frac{v}{c} = \frac{U[(1+2U/U^2)]^{1/2}}{(1+U)} = \frac{U(1+1/U)}{(1+U)} = 1, \quad (\text{B.3})$$

The current density is then given by

$$J_c = \frac{Q}{\tau} = \frac{\varepsilon_0 V_g}{D} \frac{c}{D}, \quad (\text{B.4})$$

Due to the current continue, we can find that

$$J_c \propto \frac{\phi(z)}{z^2} = \frac{V_g}{D^2}, \quad (\text{B.5})$$

We can then obtain the potential distribution as

$$\phi(z) = \frac{V_g}{D^2} z^2, \quad (\text{B.6})$$

Using Poission's equation, we can find

$$\rho(z) = \varepsilon_0 \phi''(z) = 2\varepsilon_0 \frac{V_g}{D^2}, \quad (\text{B.7})$$

The current density is lastly given by:

$$J = 2\varepsilon_0 \frac{V_g}{D^2} c, \quad (\text{B.8})$$

It is just the ultra-relativistic CL law for very very large velocity. It is the result Eq.(14) in Jory's paper [61]. (JAP, 40, 3924(1969).)

B.2 SCL Current in the Whole Relativistic Regime

We still use the ormalized potential $U(z) = e\phi/m_0c^2$. Define a parameter $\gamma = 1 + U$,

We can obtain the velocity of electron as:

$$\frac{v}{c} = \frac{(\gamma^2 - 1)^{1/2}}{\gamma}, \quad (\text{B.9})$$

The average velocity of the electron transport through the gap is given by:

$$\langle v \rangle = \frac{\int_0^D v dz}{\int_0^D dz} = \frac{c}{D} \int_0^D \frac{(\gamma^2 - 1)^{1/2}}{\gamma} dz \quad (\text{B.10})$$

$$= \frac{m_0 c^3}{e V_g} \int_1^{\gamma_g} \frac{(\gamma^2 - 1)^{1/2}}{\gamma} d\gamma \quad (\gamma = 1 + \frac{e}{m_0 c^2} \frac{V_g}{D} z, d\gamma = \frac{e V_g}{m_0 c^2 D} dz) \quad (\text{B.11})$$

$$= \frac{m_0 c^3}{e V_g} \left[\sqrt{\gamma^2 - 1} - \arccos \left(\frac{1}{\gamma} \right) \right]_1^{\gamma_g} \quad (\text{B.12})$$

$$= \frac{m_0 c^3}{e V_g} \left[\sqrt{\gamma_g^2 - 1} - \arccos \left(\frac{1}{\gamma_g} \right) \right] \quad (\text{B.13})$$

B.2 SCL Current in the Whole Relativistic Regime

where γ_g is related to the applied gap voltage given by $\gamma_g = 1 + eV_g/m_0c^2$. According to the definition of current density, we can find

$$J_c = \frac{Q}{\tau} = \frac{\varepsilon_0 V_g}{D} \frac{\langle v \rangle}{D} = \frac{\varepsilon_0 m_0 c^3}{e} \frac{\left[\sqrt{\gamma_g^2 - 1} - \arccos\left(\frac{1}{\gamma_g}\right) \right]}{D^2} \quad (\text{B.14})$$

$$= \frac{\varepsilon_0 m_0 c^3}{e} \frac{\left[\sqrt{\gamma^2 - 1} - \arccos\left(\frac{1}{\gamma}\right) \right]}{z^2}, \quad (\text{B.15})$$

Due to the current continue equation, We can obtain

$$\left[\sqrt{\gamma^2 - 1} - \arccos\left(\frac{1}{\gamma}\right) \right] = \frac{\left[\sqrt{\gamma_g^2 - 1} - \arccos\left(\frac{1}{\gamma_g}\right) \right]}{D^2} z^2, \quad (\text{B.16})$$

Take the first order derivation, we can find

$$\frac{\sqrt{\gamma^2 - 1}}{\gamma} \gamma' = 2 \frac{\left[\sqrt{\gamma_g^2 - 1} - \arccos\left(\frac{1}{\gamma_g}\right) \right]}{D^2} z, \quad (\text{B.17})$$

Keep on performing the second order derivation, we obtain:

$$\frac{\sqrt{\gamma^2 - 1}}{\gamma} \gamma'' + \left(\frac{1}{\gamma^2 \sqrt{\gamma^2 - 1}} \right) \gamma'^2 = 2 \frac{\left[\sqrt{\gamma_g^2 - 1} - \arccos\left(\frac{1}{\gamma_g}\right) \right]}{D^2}, \quad (\text{B.18})$$

Lastly, the current density is given by

$$J = \rho v = \varepsilon_0 \phi'' v = \varepsilon_0 \frac{m_0 c^2}{e} \gamma_g'' v_g. \quad (\text{B.19})$$

The comparison of the current density given by the capacitor model is shown in Fig. B.1. We can see that it works very well in the weak-relativistic and the ultra-relativistic regime, but in the medium regime, the capacitor model is not very good. So the capacitor model should be still improved. We present the current results obtain first.

Figure B.1: Comparison of the capacitor model and Jory's relativistic CL law.