

Some Solutions of the 3D Laplace Equation in a Layer with Oscillating Boundary Describing an Array of Nanotubes with Applications to Cold Field Emission. II. Irregular Arrays

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Abstract. As in the first part (J. Brüning, S.Yu. Dobrokhotov, D.S. Minenkov, 2011), we construct a family of special solutions of the Dirichlet problem for the Laplace equation in a domain with fast changing boundary. Using these solutions, we construct an analytic model of cold field electron emission from surfaces simulating arrays of vertically aligned nanotubes. Explicit analytic formulas lead to fast computations and also allow us to study the case of random arrays of tubes with stochastic distribution of parameters. We present these results and compare them with numerical approximations given in [1].

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1. INTRODUCTION

We propose an analytic model of certain types of cold field-emission cathodes based on arrays of carbon nanotubes, see the review [2]. We consider an array of vertically aligned nanotubes in an external field and study the dependence of its emission characteristics on its geometry. In the first part of our work [3], such a model was constructed for the case of regular arrays of identical tubes. Here we present a generalization of this model to the case of random arrays, where the tubes are not identical and have geometric characteristics and positions that are defined randomly with some dispersion, see Fig. 1. This generalization allows us to study the influence of dispersion of geometric characteristics (such as tube length and position) on the emission properties of the array.

As in [3], we are going to construct a family of exact solutions of the following Dirichlet problem for the Laplace equation,

$$\begin{aligned} \Delta u &= 0, & g(x, y) \leq z \leq d, & \quad (x, y) \in \mathcal{R}^2, \\ u|_{z=g(x,y)} &= 0, & u|_{z=d} &= U. \end{aligned} \tag{1.1}$$

Here $u(x, y, z)$ is the electric potential, d is the anode-cathode distance, and U is a bias. The lower boundary, $g(x, y)$, describes an enveloping surface of an array of vertically aligned tubes (see Fig. 1), with fast oscillation.

Having thus defined the electrical field $E = \nabla u$ near the tube apexes, we can use the Fowler–Nordheim formula for the current from a flat surface [4, 5] to calculate the emission current of a single tube or an array of tubes, by the following integral over the surface of the tube or the array [2]:

$$I = \iint AF^2 \exp \left\{ -\frac{B}{F} \right\} \Big|_{z=g(x,y)} dx dy. \tag{1.2}$$

Here $F := \partial u / \partial z$ is the z -component of the electric field on the surface $z = g(x, y)$. The constants $A := 1.54 \times 10^{-6} \times \varphi^{-1} = 0.31 \times 10^{-6} \text{ A V}^{-2}$ and $B := 6.8 \times 10^3 \times \varphi^{3/2} = 76 \times 10^3 \text{ V } \mu\text{m}^{-1}$ depend on the so-called work function φ which is determined by the tubes material and the emission conditions. Here and below, we take $\varphi = 5 \text{ eV}$.

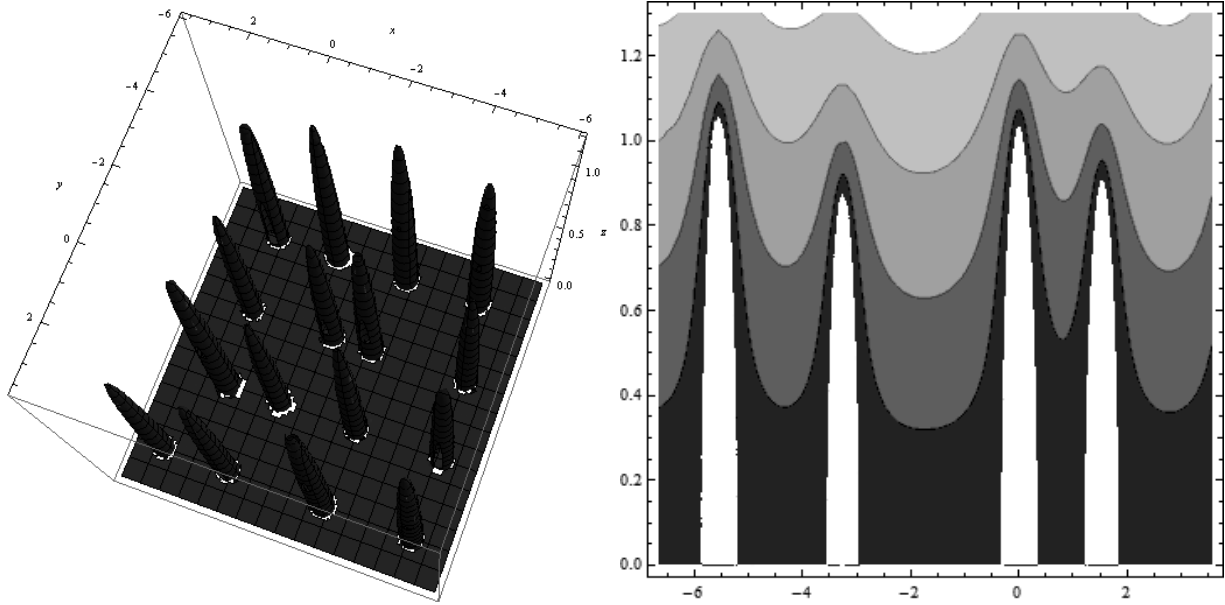


Fig. 1. Random array of vertically aligned tubes (left) and contour graph for the potential (right). Average length of the tubes is $L = 1$ and the radius of apex curvature is $R = 0.1$.

The aim of the paper is to calculate the emission current (1.2) in the case of random arrays and to study its behavior under dispersion of the geometric characteristics.

2. THE ANALYTIC MODEL

2.1. Solution of the Laplace Equation

From the field emission point of view, one is interested in the situation when the tube length L is much larger than the diameter $D = 2R$, i.e., when the “aspect ratio” L/R is high. In this case, the solution of problem (1.1) cannot be extended to the layer $0 < z < d$ without introducing singularities; they correspond to charges appearing in the tubes as an effect of the applied voltage, see [6]. This leads to the idea of introducing a family of solutions of the Poisson equation in our model, thus incorporating the effect of charges situated outside the considered area, $g(x, y) \leq z \leq d$.

Let us consider the potential V of a vertically oriented charged segment, $z \in [-L, L]$, with “charge density” $\rho(z + \nu z^3/L^2)\delta(x)\delta(y)$, where $\rho > 0$, $\nu > -1$ are dimensionless parameters. Then we have

$$V(x, y, z) = \frac{U}{d} \rho X(x, y, z), \quad X(x, y, z) := \int_{-L}^L \frac{(\zeta + \nu \zeta^3/L^2) d\zeta}{\sqrt{x^2 + y^2 + (z - \zeta)^2}}.$$

(The integral here can be expressed in terms of elementary functions, see the appendix.) Adding the potential $-V(x, y, 2d - z)$ of the “reflected charges” (with respect to $z = d$), we obtain the following solution of the Dirichlet problem (1.1):

$$u(x, y, z) = \frac{U}{d} (z - \rho X(x, y, z) + \rho X(x, y, 2d - z)),$$

for a given surface $z = g(x, y)$ describing a single tube.

Next, to approximate an array of N tubes, we consider a family of solutions to (1.1).

Proposition 1. Consider constants $L_n > 0$, $\rho_n > 0$, $\nu_n > -1$, $n = 1, 2, \dots, N$. The potential

$u(x, y, z)$ defined by the formulas

$$u(x, y, z) = \frac{U}{d} \left(z - \sum_{n=1}^N \rho_n X_n(x, y, z) + \sum_{n=1}^N \rho_n X_n(x, y, 2d - z) \right), \tag{2.1}$$

$$X_n(x, y, z) = \int_{-L_n}^{L_n} \frac{(\zeta + \nu_n \zeta^3 / L_n^2) d\zeta}{\sqrt{(x - x_n)^2 + (y - y_n)^2 + (z - \zeta)^2}},$$

solves (1.1) for the surface $z = g(x, y)$ and describes an enveloping surface of the given array of N tubes.

The function

$$V_n(x, y, z) = \frac{U}{d} \rho_n X_n(x, y, z)$$

is the potential at (x, y, z) generated by the n th charged segment $x = x_n, y = y_n, z \in (-L_n, L_n)$. This was considered in [3] for the case of a regular array of identical tubes, with $L_n = L, \rho_n = \rho, \nu_n = \nu$, placed at the square lattice points $x_{n_{ij}} = iS, y_{n_{ij}} = jS$, where S is the minimal distance between tubes.

Below, we assume that the tube lengths are much smaller than the anode-cathode distance $L/d \ll 1$ (or that the enveloping surface is placed in a uniform external field $F_0 = U/d$). Hence, in what follows, we can drop the reflected potential since, asymptotically,

$$-V(x, y, 2d - z) = O\left(\frac{L^3}{d(2d - z)^2}\right)$$

for $z \sim L$.

Following [3], it is easy to show that the enveloping surface $z = g(x, y)$ from Proposition 1 effectively describes an array of tubes with lengths L_n and positions (x_n, y_n) which are, individually, almost elliptic near the base and almost parabolic near the apex.

Proposition 2. Consider an array of tubes with length L_n and constants ρ_n, ν_n at each position (x_n, y_n) . Then, near the apex of any given tube with data L, ρ, ν , the position $(0, 0)$ (wlog) and the index n_0 , the surface $z = g(x, y)$ is asymptotic to a paraboloid,

$$z = -\frac{1}{2}\kappa(x^2 + y^2) + L + \frac{1}{2\kappa},$$

$$\kappa \approx \frac{1}{4L} \exp \left\{ \frac{1 + 2\rho(1 + \frac{4}{3}\nu)}{\rho(1 + \nu)} - \frac{1}{\rho(1 + \nu)} \sum_{n \neq n_0} \rho_n X_n(0, 0, L + \frac{1}{2\kappa}) \right\} > 0. \tag{2.2}$$

Near the base (i. e. for $z \ll L$), the surface is asymptotic to an ellipsoid stretched along the z -axis,

$$\omega^2(x^2 + y^2) + z^2 = L^2, \quad \omega^2 \approx \frac{1}{4} \exp \left\{ \frac{1 + 2\rho(1 - \frac{1}{2}\nu)}{\rho} - \frac{1}{\rho} \lim_{z \rightarrow 0} \frac{1}{z} \sum_{n \neq n_0} \rho_n X_n(0, 0, z) \right\}. \tag{2.3}$$

Formulas (2.2) and (2.3) are accurate within small corrections $O(\sqrt{x^2 + y^2}/L) + O((z - L)/L)$ and $O(\sqrt{x^2 + y^2}/L) + O(z/L)$, respectively.

For applications, it is important to maximize the emission current in terms of the tube distances. When we change the positions (x_n, y_n) of the charged segments, then, naturally, the surface $z = g(x, y)$ changes. Nevertheless, we can choose the parameters ρ_n in such a way that the tubes still have the same “structure” for all considered positions, in the sense that they retain the same lengths

L_n , apex curvatures κ_n , and parameters ν_n . Since the tubes have the asymptotic form stated in Proposition 2, we can find such values $\rho_n(L_n, \kappa_n, \nu_n, x_n, y_n)$ from the system

$$u(x_m, y_m, L_m + \frac{1}{2\kappa_m}) = 0, \quad m = 1, 2, \dots, N.$$

This is a linear system with respect to the ρ_n s,

$$\sum_{n=1}^N \rho_n X_n \left(x_m, y_m, L_m + \frac{1}{2\kappa_m} \right) = L_m + \frac{1}{2\kappa_m}, \quad m = 1, 2, \dots, N, \quad (2.4)$$

if $X_n(x_m, y_m, L_m + \frac{1}{2\kappa_m})$ is the potential of the n th tube at the apex of m th tube.

Let us rewrite this system in matrix form as $\mathbf{H}\vec{\rho} = \vec{l}$, for the vectors $\vec{\rho} = (\rho_n)_{n=1}^N$, $\vec{l} = (L_m + \frac{1}{2\kappa_m})_{m=1}^N$, and the matrix $\mathbf{H} = (X_n(x_m, y_m, L_m + \frac{1}{2\kappa_m}))_{m,n=1,2,\dots,N}$. We denote the diagonal part of \mathbf{H} by $\mathbf{A} := (\delta_{mn} X_n(x_m, y_m, L_m + \frac{1}{2\kappa_m}))_{m,n=1,2,\dots,N}$ and set $\mathbf{B} := \mathbf{H} - \mathbf{A}$. Using the asymptotics of X , one can show that

$$\|\mathbf{A}\| \sim L \frac{L}{R}, \quad \|\mathbf{B}\| \sim L \frac{L^3}{S^3},$$

where L , R , and S are the averages of tube lengths, radii, and distances, respectively. Thus, the following proposition holds.

Proposition 3. *Let the numbers $L_n, \kappa_n, \nu_n, x_n, y_n$ for the tube array under consideration be such that the aspect ratio is high (i.e., $1/\kappa_n \sim R_n \ll L_n$) and that the density of tubes in the array is medium or low (i.e., the average distance between tubes, S , and the average length, L , are of the same order, $S \sim L$)¹. Then the matrix $\mathbf{H} = \mathbf{A} + \mathbf{B}$ can be inverted by the Neumann series and, in view of (2.4), the “effective charge” vector, $\vec{\rho} = \{\rho_n\}$, is given by the asymptotic formula*

$$\vec{\rho} = \left(1 - \mathbf{A}^{-1}\mathbf{B} + O\left(\frac{R L^2}{S^3}\right)^2 \right) \mathbf{A}^{-1}\vec{l}. \quad (2.5)$$

Thus, the effective charges have the same order as the aspect ratio, $\rho_n \sim R/L$. The potential $u(x, y, z)$ defined by (2.1) with these ρ_n approximates the potential of an array of nanotubes with tube lengths L_n , apex curvatures κ_n (or tube diameters $D_n = 2/\kappa_n$), and tube positions (x_n, y_n) . The supplementary parameters ν_n can be used to fit the emission properties of real tube arrays.

2.2. Field Enhancement Factor and Emission Current

As shown by numerous experiments (see, e.g., the review [2] and the bibliography therein), the current-voltage characteristic of tube arrays is often well described by the Fowler–Nordheim formula (in appropriate voltage ranges). It reads

$$I = AaF^2 e^{-B/F}, \quad F = \beta \frac{U}{d},$$

for some field enhancement (amplification) factor β and the effective emission area a , which are special for each particular array, and A, B are physical constants that depend on the work function.

Even though we consider arrays of tubes, the derivatives of the potential u decay so fast that, near the apex of a tube, they depend mainly on the parameters of this particular tube (i.e., of L, κ, ρ , and ν). The very important “screening effect” (see, e.g., [1, 7]) is taken into account through the “effective charges” $\rho_n(L_n, \kappa_n, \nu_n, x_n, y_n)$ as defined by system (2.4), for any given tube data

¹This is to be distinguished from “high density” arrays, where $S \sim R$.

$L_n, \kappa_n, \nu_n, x_n, y_n$. To find the derivatives of u near the tube with index n_0 in the array, one uses its tube data $(L_{n_0}, \kappa_{n_0}, \rho_{n_0}, \nu_{n_0})$ and the effective charge $\rho_{n_0}(L_n, \kappa_n, \nu_n, x_n, y_n)$ from (2.5) in the same asymptotic formula as for a single tube. (Asymptotic formulas for the derivatives at the apex were presented in the first part of this work, [3].) Then we calculate the emission current from the integral in (1.2), evaluating it by the Laplace method. Finally, suppressing the index n_0 again, we obtain for the field enhancement factor $\beta = \frac{\partial}{\partial z} u(x, y, L + \frac{1}{2\kappa})$ and the emission current I , respectively,

$$I \approx \frac{2\pi A}{(2\kappa)^2} \frac{\beta F_0}{B} (\beta F_0)^2 e^{-\frac{B}{\beta F_0}}, \quad (2.6)$$

$$\beta \approx 2\kappa L \rho(1 + \nu), \quad F_0 = \frac{U}{d}, \quad (2.7)$$

where $\rho = \rho_{n_0}$ is the effective charge described above. We have omitted error terms of order $O(R/L)$ and $O(R/S)$ in formulas (2.6) and (2.7), respectively, arising from the asymptotic calculation of the u -derivatives, as well as a term $O(F/B)$ arising from applying the Laplace method (for the relevant error estimates, see, e.g., [8]).

The total current is the sum of the currents arising from all tubes in the array $I = \sum I_n$.

3. CALCULATIONS WITH THE PROPOSED MODEL

3.1. Approximation Algorithm

We now describe the approximating procedure.

First of all, we choose the parameters $L_n, x_n, y_n, \kappa_n, \nu_n$ in a sensible way. When studying tube arrays manufactured by the same method, we may often assume that the tubes have very similar properties. In the framework of the proposed model, we then may set all tube parameters to be equal to their averages, $L_n = L, \kappa_n = \kappa$, and $\nu_n = \nu$. The tube length L and the diameter $2R$, as well as the tube positions (x_n, y_n) in the array, are usually known, perhaps with some dispersion, as a result of the production method. One can produce arrays with tubes placed for example in a square lattice with coordinates (x_n^0, y_n^0) , but the tubes can bend, and hence, their heads deviate from their initial positions, changing the coordinates to (x_n, y_n) . Such an array can effectively be approximated by an array with vertical tubes placed at the positions (x_n, y_n) . So we choose some positions (x_n^0, y_n^0) and define κ and ν for these positions using an experimentally obtained current-voltage characteristic. Then we consider arrays with random positions (x_n, y_n) , with averages (x_n^0, y_n^0) , and calculate the current for these random arrays. In random arrays, L_n, κ_n , and ν_n can also be random with averages L, κ , and ν .

As soon as the parameters L_n, x_n, y_n, κ_n , and ν_n are defined, the “effective charges” ρ_n are then again calculated from system (2.4). This can be done either by the iteration method leading to formula (2.5) or by any other computational method.² Finally, the enhancement factor β and the emission current I can be calculated by formulas (2.6) and (2.7), respectively. We can also find the emission current density $J = I/a_{\text{ar}}$ of an array of tubes, which is the emission current I of the array over its area a_{ar} .

We conclude by noting that our model can be used to study the dependence of the emission current on the tube distance. In particular, one can determine the optimal tube distance (generally depending on the applied voltage) and study the influence of fluctuations in the geometrical parameters on the emission properties. Examples of such studies are presented below.

3.2. The Screening Effect and Computations for Random Arrays

We describe first some verifications of our model. As $\kappa \sim 1/R$, the field enhancement factor predicted by our model is of the order of the tube aspect ratio L/R , which is in good agreement with other calculations and also with experiments. We compare our prediction of the field enhancement factor as function of the aspect ratio with the formulas obtained in [9–12] in Fig. 2. Here we take $\kappa = 2.2/R, \nu = 0$.

²We have used Wolfram Mathematica that takes about 1 minute to perform the computations for an array of 30×30 tubes size for which a matrix of 900×900 has to be inverted.

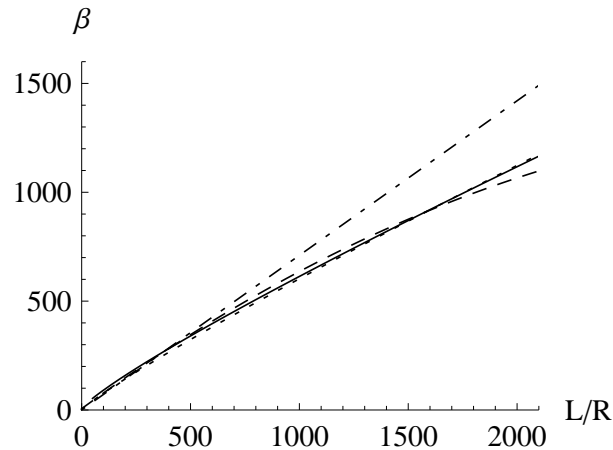


Fig. 2. Comparison of the enhancement factor β as function of the aspect ratio L/R (see [12]): (a) linear dependence (dot-dashed); the dependence as defined by the empiric formulas (b) $\beta = 1.2(2.15 + L/R)^{0.9}$ [9,10] (dotted) and (c) $\beta = 5.93 + 0.73 L/R - 0.0001(L/R)^2$ [11] (dashed); (d) the dependence predicted by our model (solid).

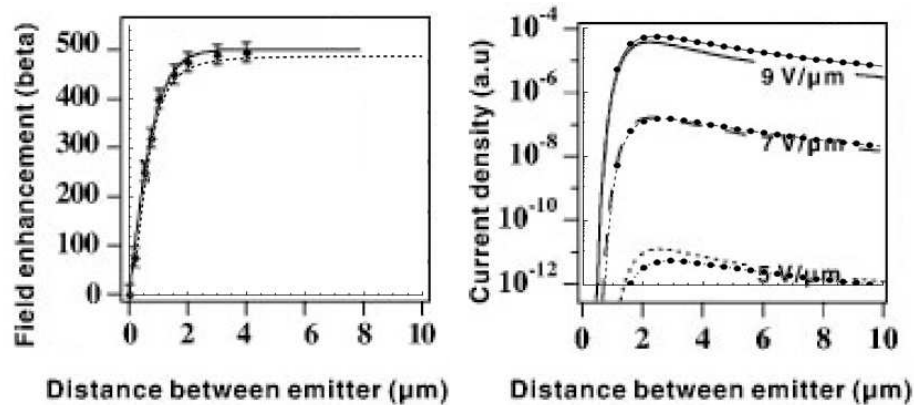


Fig. 3. Comparison of the results from [1] with the calculations by our model. Left: dependence of the enhancement factor β on the tube distance S computed by [1] (solid) and by our model (dashed); right: dependence of the emission current density J on S computed by [1] (lines without dots) and by our model (with dots).

Next we compare the predictions of our model for the screening effect with those given in [1], where computations for a regular array (i.e., a periodic square lattice of identical tubes) are presented. The dependence of the enhancement factor β and the emission current density J on the tube distance S for different biases is given in Fig. 3. Here $L = 1 \mu\text{m}$, $R = 2 \text{ nm}$, and we take $\kappa = 3/R$, $\nu = 5$ (which gives $\rho = 0.06, 0.15, 0.22$ for $S = 1, 2, 5 \mu\text{m}$, respectively).

The production process of tube arrays often entails nonuniformities which are very hard to capture by numerical approximations. Therefore, it is often assumed that the array is periodic when the Laplace equation is solved numerically, leading to a very time consuming procedure. This is caused by the high aspect ratio of the tubes and the rapid change of the potential and its derivatives near the tube apex. By contrast, the explicit formulas provided by our model admit fast calculations even for random arrays. In Fig. 4, such results for random arrays are presented. We consider arrays with stochastically chosen lengths and positions. The average length and curvature is fixed as in the previous case: $L = 1 \mu\text{m}$, $R = 2 \text{ nm}$, $\kappa = 3/R$, $\nu = 5$. For the average tube distance, we take the optimal values $S = S_{\text{opt}} = 2.9, 2.6, 2.4 \mu\text{m}$ for $U_1 = 5, 7, 9 \text{ V}/\mu\text{m}$, respectively.

Dispersion of positions near the optimal configuration does not have a big effect on the current

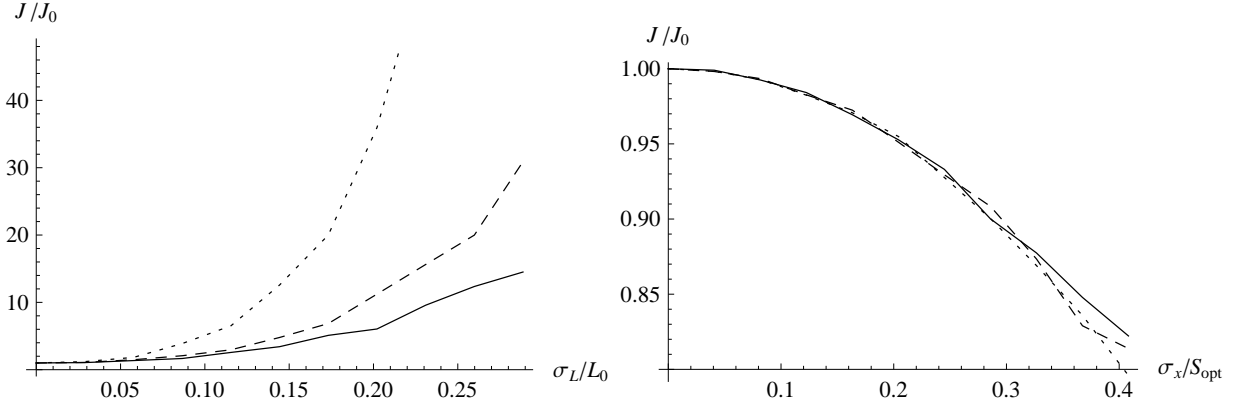


Fig. 4. Influence of dispersion of tube length (left) and tube positions (right) on emission properties at different voltages (5 V/ μm – dotted, 7 V/ μm – dashed, and 9 V/ μm – solid). Tubes positions $r_n = x_n^2 + y_n^2$ are scattered about their optimal positions in square lattice (with $S = S_{\text{opt}}$) that provide maximum current for given applied voltage.

given due to the exponential nature of the tunneling current. Dispersion of lengths has a much greater effect, because it changes the enhancement factor directly. An interesting result is that a higher voltage shows a lesser impact of dispersion of tube lengths on the current; this agrees with the experimental results described in [1].

In addition, our model gives a reasonable description not only of nanotube arrays but also of arrays of “tube-like” emitters, for example, bundles of tubes or the emitters described in [13]. We consider them as a single “tube” with some “effective form” for the purpose of modeling.

APPENDIX. EXPLICIT FORMULA FOR THE POTENTIAL V

The integral in (2.1) can be expressed in terms of elementary functions as follows:

$$\begin{aligned}
 X(x, y, z) &= \int_{-L}^L \frac{(\zeta + \nu\zeta^3/L^2)d\zeta}{\sqrt{r^2 + (z - \zeta)^2}} = \\
 &= \frac{4Lz\left(-1 - \frac{\nu}{3} + \frac{2\nu r^2}{3L^2} - \frac{11\nu z^2}{6L^2}\right)}{\sqrt{r^2 + (z + L)^2} + \sqrt{r^2 + (z - L)^2}} + \frac{5\nu z}{6L} \left(\sqrt{r^2 + (z + L)^2} + \sqrt{r^2 + (z - L)^2}\right) \\
 &\quad + z\left(1 - \frac{3\nu r^2}{2L^2} + \frac{\nu z^2}{L^2}\right) \ln \frac{\sqrt{r^2 + (z + L)^2} + z + L}{\sqrt{r^2 + (z - L)^2} + z - L}, \quad r^2 = x^2 + y^2.
 \end{aligned}$$

This expression leads to the simple asymptotic formulas given in Section 2 (for details, see [3]).

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