Penetration of external field into regular and random arrays of nanotubes: Implications for field emission

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We develop an analytical theory of polarization of a vertically aligned array of carbon nanotubes (NTs) in external electric field. Such arrays are commonly utilized in field-emission devices, due to the known electrostatic effect of strong field enhancement near the tip of an individual NT. Small ratio of the NT radius to the separation between neighboring NTs allows to obtain asymptotically exact solution for the distribution of induced charge density along the NT axes. For a regular array, this solution allows to trace the suppression of the field penetration with increasing the density of NTs in the array. We demonstrate that for a random array, fluctuations in the NT density terminate the applicability of our result at distances from the NT tips much larger than the field penetration depth, where the induced charge density is already exponentially small. Our prime conclusion is that, due to collective screening of the external field by the array, the field-emission current decreases drastically for dense arrays compared to an individual NT. We argue that the reason why the strong field emission, described by the Fowler-Nordheim law and observed in realistic arrays, is the strong dispersion in heights of the constituting NTs.

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I. INTRODUCTION

First report of field emission from carbon NTs had appeared a decade ago. It was followed by a demonstration that arrays of NTs can be patterned into emitting and non-emitting regions. Since then, the field emission properties of carbon NTs command a steady interest from researchers worldwide. The uniqueness of these properties originates from geometry of a NT. Namely, due to a small NT radius, r, the electric field applied between the substrate (cathode), on which the NTs are grown, and the anode is enhanced by a large factor $\beta \gg 1$ near the nanotube tip. Such an enhancement translates into high probability of electron tunnelling toward the anode, leading to desirable low turn-on voltage for field emission. This property, combined with high emission current density, made possible a successful fabrication of the row-column matrix-addressable flat panel display based on carbon NTs. Geometrical characteristics of individual NTs utilized in the first displays were highly dispersed. Further advances in fabrication allowed to achieve excellent vertical alignment and high homogeneity in the lengths and radii of NTs. Geometrical characteristics of individual NTs utilized in the first displays were highly dispersed. Geometrical characteristics of individual NTs utilized in the first displays were highly dispersed. Geometrical characteristics of individual NTs utilized in the first displays were highly dispersed. Geometrical characteristics of individual NTs utilized in the first displays were highly dispersed. Geometrical characteristics of individual NTs utilized in the first displays were highly dispersed. Geometrical characteristics of individual NTs utilized in the first displays were highly dispersed. Geometrical characteristics of individual NTs utilized in the first displays were highly dispersed. 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electric field, $F$, directed along their axes, see Fig. 1. It might seem, that, if $d$ exceeds the tunnelling length for field emission from each of the NTs, then both NTs emit independently. This, however, is not the case. The reason is that the enhancement of the electric field near the tip of each NT is governed by the charge density, induced by the external field. For two parallel NTs, the induced charge density per NT is approximately two times smaller than for an isolated NT. As a result, the field enhancement near the tip of each NT becomes smaller due to the presence of the neighbor.

Compared to a pair of neighboring NTs, the suppression of the field enhancement becomes much more pronounced in the NT array. On the qualitative level, this conclusion follows from the fact that each NT in the array interacts with $\sim (h/d)^2 \gg 1$ neighbors. As we will demonstrate below, external field simply does not penetrate into the sufficiently dense array. On the intuitive level, this trend has been previously understood. In particular, in Ref. 23 numerical simulations illustrating the suppression of the field enhancement for three parallel NTs with decreasing distance between them were reported. However, full understanding of this suppression in the array with arbitrary ratio $d/h$ requires an analytical theory. Such a theory is developed in the present paper. In particular we demonstrate that

1) penetration of the external field into the array is described by a simple function, $\sinh(z/a)$, where the penetration depth, $a$, is much smaller than $h$ for a dense array, but still much bigger than $d$;

2) distributions of induced charge density in a regular and completely random NT arrays are approximately the same;

3) with regard to the field emission, the enhancement of external field for the array, as compared to the individual NT, is suppressed by the factor $\approx h/a$.

The reason why the electrostatic problem, in which the variables cannot be separated, allows an asymptotic analytical solution is the presence of large parameters $(h/r) \gg 1$ and $(d/r) \gg 1$. As it was demonstrated in Ref. 31, for a single NT, the relation $h \gg r$ allows one to find analytically the distribution of induced charges in external field. Here the approach of Ref. 31 is generalized to the NT array.

The paper is organized as follows. In Sect. II we review the Thomas-Fermi description of polarization of a single NT in external field. In Sect. III we generalize the Thomas-Fermi equation to the NT array. In Sections IV-V we analyze this equation for a regular array, and find its asymptotic [in the parameter $(d/r) \gg 1$] solution. Robustness of this solution with respect to randomness in the NT positions is demonstrated in Sect. VI. In Sect. VII we apply to the obtained solution for distribution of the induced charge density to calculate the field emission current from the array. Relation of our theory to experiment is addressed in Sect. VIII.

II. SINGLE NT

Denote with $\rho(z)$ the linear charge density on the NT surface at a distance $z < h$ from the substrate. The Thomas-Fermi equation for $\rho(z)$ reads\(^{31}\)

$$eFz = \frac{1}{g} \rho(z) + \frac{1}{e} \int_0^h dz' S_0(z, z') \rho(z'),$$  \hspace{1cm} (1)

where the kernel, $S_0(z, z')$, is defined as

$$S_0(z, z') = \Phi(z - z') - \Phi(z + z').$$  \hspace{1cm} (2)

Here the function $\Phi(z)$ is the interaction potential between two points on the NT surface, separated vertically by $z$. It represents an azimuthal average of the Coulomb potential

$$\Phi(x) = \frac{e}{\pi} \int_0^\pi d\alpha \left[ x^2 + 4r^2 \sin^2(\alpha/2) \right]^{1/2},$$  \hspace{1cm} (3)

where $r$ is the NT radius. The second term in Eq. 2 accounts for the image charges. The meaning of Eq. (1) is the following. The lhs is the bare potential. The potential, acting on a given electron at the NT surface represents the sum of this potential and of the potential, created by the induced charges. This resulting potential defines the local value of the Fermi energy, which, in turn, fixes the local value of the Fermi momentum. This Fermi momentum, on the other hand, is linearly proportional to the charge density in one dimension. This is a standard reasoning behind the Thomas-Fermi description. Within this description Eq. (1) is nothing but the condition that the electrochemical potential remains constant along the NT.

The prime simplification that allows analytical solution of Eq. (1) is that, with logarithmic accuracy, $\rho(z')$ in the integrand in the rhs of Eq. (1) can be replaced by $\rho(z)$. Upon this replacement, we have

$$\frac{1}{e} \int_0^h dz' S_0(z, z') \approx \ln \frac{h^2}{r^2} - \frac{\ln h^2}{z^2} + \ln \left( \frac{1 - z/h}{1 + z/h} \right),$$  \hspace{1cm} (4)

where we assumed that $(h - z) \gg r$. With the same logarithmic accuracy, for $z \gg r$, the rhs of Eq. (4) can be replaced by $2L_h$, where $L_h = \ln(h/r)$. Then we obtain the following analytical solution for the induced charge density:\(^{31}\)

$$\rho(z) \approx \frac{gFz}{1 + 2gL_h},$$  \hspace{1cm} (5)

where we have introduced a dimensionless interaction parameter $g = 2Ne^2/\pi \hbar v_0$. The above result for $\rho(z)$ is approximate, in the sense, that the numerical factor in the argument of a large logarithm, $L_h \gg 1$, is not specified. Equation (4) represents the solution of Eq. (1), which satisfies the obvious condition $\rho(0) = 0$. An improved analytical description yielding the result coinciding with
Eq. (5) in the limit of large $L_h$ was recently reported in Ref. 32.

The remarkable property of the solution (5) is that the NT with poor “metallicity”, $g < 1$, eventually becomes metallic as the length, $h$, of NT is increased. This is not the case if the NT is located parallel to the conducting gate at distance $D \ll h$. In the latter case, one has to replace $L_h$ by $\ln(D/r)$.

III. THOMAS-FERMI EQUATION FOR AN ARRAY

Consider an array of parallel NTs located at points, $R_i$, on the substrate, see Fig. 1. To set the Thomas-Fermi equation for a given NT, $i$, in the array, one has to take into account that the external potential, leading to the charge separation, contains, in addition to $eFz$, the potentials created by charges induced on all other NTs. Then the generalized Eq. (1) reads

$$eFz = \frac{1}{g} \rho_i(z) + \int_0^h dz' \sum_j \rho_j(z') S(z, z'; R_i - R_j),$$

(6)

where the kernel, $S$, is given by

$$S(z, z'; R) = \frac{1}{\sqrt{(z-z')^2 + |R|^2}} - \frac{1}{\sqrt{(z+z')^2 + |R|^2}}.$$  

(7)

It is convenient to rewrite Eq. (6) in the “continuous” form by introducing the position-dependent density, $\rho(z, R)$, and the local concentration of NTs

$$N(R) = \sum_i \delta(R - R_i).$$  

(8)

In the new notations Eq. (6) takes the form

$$eFz = \frac{1}{g} \rho(z, R) + \int dR' N(R') \int_0^h dz' S(z, z'; R - R') \rho(z', R').$$  

(9)

The Thomas-Fermi equation in the form (9) is convenient for further analysis. This is because, as we will see below, the distributions, $\rho_i(z)$, are almost the same for all $i$ even for completely random array.

IV. REGULAR ARRAY

Consider a regular array in the form of a square lattice with a lattice constant, $d$. Then the coordinates of $R_i$ in Eq. (5) are $\{nd, md\}$ with integer $m$ and $n$. Obviously, for the regular array the induced charge density, $\rho(z)$, is the same for all NTs, so that Eq. (9) acquires the form

$$eFz = \left(1 + \frac{2gL_d}{g}\right) \rho(z) + \int_0^h dz' S_{ext}(z, z') \rho(z'),$$  

(10)

where

$$S_{ext}(z, z') = \sum_{\{m,n\} \neq \{0;0\}} \left[ \frac{1}{\sqrt{(z-z')^2 + (m^2 + n^2)d^2}} - \frac{1}{\sqrt{(z+z')^2 + (m^2 + n^2)d^2}} \right].$$  

(11)

Here we have isolated the self-action, $m = n = 0$, of a NT. For a single NT this self-action is described by a large logarithm $L_h$. In the array, however, the interaction is screened at distances $|z - z'| \gtrsim d$ by the neighboring NTs. To account for this screening, the logarithm, $L_h$, in Eq. (10) is replaced by $L_d = \ln(d/r)$.

It is apparent that both terms in the sum (11) diverge due to the contributions from large $m$, $n$. However, the divergent parts in both terms cancel each other. Physically, this reflects the screening by the image charges (see Fig. 1). Replacing the sums over $m$ and $n$ in Eq. (11) by integrals, we obtain the following expression for the kernel $S_{ext}(z, z')$

$$S_{ext}(z, z') = 2\pi N_0 \left[ z + z' - |z - z'| \right] = 4\pi N_0 \left[ z' \Theta(z - z') + z \Theta(z' - z) \right],$$  

(12)

where $\Theta(x)$ is the step-function. Note that both steps, replacing $L_h$ in Eq. (10) by $L_d$ and replacing the sums in Eq. (11) by integrals are by no means obvious and require justification. This justification is provided in the next Section. In the present Section we demonstrate that the integral equation (10) with the kernel (12) can be solved analytically. Upon taking the derivative from both sides of Eq. (10), we obtain

$$eF = \left(1 + \frac{2gL_d}{g}\right) \frac{d\rho}{dz} + 4\pi N_0 \int z dz' \rho(z').$$  

(13)

It is now easy to see that, within a factor, the first term in Eq. (13) is the second derivative of the second term. Thus, Eq. (13) can be viewed as a second-order differential equation with respect to $\int z dz' \rho(z')$. The solution of this equation, satisfying the condition $\rho(0) = 0$, has the form

$$\rho(z) = \rho_0 \sinh(z/a),$$  

(14)

where $\rho_0$ is defined as

$$\rho_0 = \frac{eFga}{(1 + 2gL_d) \cosh(h/a)},$$  

(15)
and the length, \(a\), is given by

\[
a = \frac{1}{2} \sqrt{\frac{1 + 2gL_d}{\pi gN_0}}.
\]

The above expression for the induced charge density constitutes the main result of the present paper. It is seen from Eq. (14) that \(a\) plays the role of the penetration depth of the external field into the array. In the limit of very low density, \(N_0 = d^{-2} \ll 1/h^2\), Eqs. (14)–(16) reproduce the result Eq. (6) for a single NT, as it could be expected, since the mutual influence of NTs, separated by a distance \(\gtrsim h\) is negligible.

It also follows from Eqs. (13)–(16) that in the limit of large \(N_0\), such that \(a \ll h\), the induced charge density is concentrated near the NT’s tips and falls off towards the substrate exponentially, as \(\exp\{-\sqrt{h^2 - a^2}/a\}\). This weak penetration of external field into the array is a consequence of the collective screening. Indeed, in terms of screening properties, the array of a high density can be viewed as a homogeneous metallic plate. Our crucial observation, however, is that the penetration depth exceeds parametrically the lattice constant, \(d\), both for large and small values of the interaction parameter, \(g\).

By virtue of the relation \(a/d > 1\), there are many NTs within the penetrations depth. This, in turn, suggests that Eqs. (14)–(16) apply to the random array with average areal concentration of NTs \(N_0 = d^{-2}\).

### V. DERIVATION

In the previous section the derivation was based on two intuitive assumptions. Namely, we have assumed the self-action of a NT in the array is described by \(L_d\) instead of \(L_h\) for an isolated NT, and that the sum over \(m\) and \(n\) in Eq. (11) can be replaced by the integral. To justify these assumptions, below we calculate the sum Eq. (11) more accurately. In order to do so, we employ the following (obvious) identity. Consider a two-dimensional vector, \(\mathbf{b}\), with projections \(b_x, b_y\). Then, for arbitrary \(x\), we have

\[
\frac{1}{\sqrt{x^2 + |\mathbf{b}|^2}} = \frac{1}{2\pi} \int \frac{dq_x dq_y}{\sqrt{q_x^2 + q_y^2}} \exp(-\sqrt{q_x^2 + q_y^2} x) \exp(iq_x b_x + i q_y b_y).
\]

To use this identity in Eq. (11), we set \(b_x = nd, b_y = md\). Then the summation over \(m\) and \(n\) can be readily performed, yielding the sum of \(\delta\)-functions, i.e.

\[
\left(\frac{2\pi}{d}\right)^2 \sum_{p, l} \delta\left(q_x - \frac{2\pi p}{d}\right) \delta\left(q_y - \frac{2\pi l}{d}\right).
\]

where \(p\) and \(l\) assume all integer values. After that the rhs in Eq. (19) acquires the form

\[
\int_0^h dz' \rho(z') S(z, z'),
\]

where \(S(z, z')\) is given by

\[
S(z, z') = \frac{2\pi}{d^2} (z + z' - |z - z'|) + \sum_{p, l \neq 0} \frac{1}{d^2 \sqrt{p^2 + l^2}} \left\{ \exp\left(-\frac{2\pi}{d} \sqrt{p^2 + l^2} \sqrt{(z - z')^2}\right) \right. \left. - \exp\left(-\frac{2\pi}{d} \sqrt{p^2 + l^2} \sqrt{(z + z')^2}\right) \right\}.
\]

The first term in (20) describes the continuous limit and comes from \(p = l = 0\) in Eq. (18). It coincides with the kernel, \(S_{ext}(z, z')\), defined by Eq. (12). The remaining sum over \(p\) and \(l\) recovers the kernel \(S_0\) in Eq. (11). The easiest way to see this is to replace the sums over \(p\) and \(l\) by corresponding integrals, which would immediately yield \(S_0(z, z')\). However, such a replacement is justified only when the large number of terms contribute to the sum. This is the case only when the condition \(|z - z'| \lesssim d\) is met. For \(|z - z'| \gtrsim d\) the sum over \(p\) and \(l\) in Eq. (20) is dominated by the terms \(p = 0, l \pm 1\) and \(l = 0, p \pm 1\), and is exponentially decaying function of \(|z - z'|\). This suggests that \(S_0(z, z')\) should be substituted into Eq. (19), in which the integration over \(z'\) should be restricted to the interval \(|z - z'| \lesssim d\). Within this interval, \(\rho(z')\) in the integrand of Eq. (19) can be replaced by \(\rho(z)\). The remaining integral yields \(2L_d = 2\ln(d/r)\), similarly to Eq. (41) with \(h\) replaced by \(d\). The product \(2L_d \rho(z)\) is nothing but the first term in the rhs of Eq. (9).

The restriction of the integration interval in Eq. (19) to \(|z - z'| \lesssim d\) for the part of \(S(z, z')\), coming from the second term in Eq. (20), is, in fact, a delicate step. Although this part decays as \(\exp\left(-2\pi|z - z'|/d\right)\) outside this interval, the behavior of \(\rho(z')\) outside this interval is also exponential, namely, it increases as \(\exp(z'/a)\). Therefore, the restriction of the integration interval in (19) is allowed only when the exponent in \(\rho(z')\) is slower, i.e. \(a \lesssim d\). However, we know from Eq. (11) that this is indeed the case.

### VI. FLUCTUATIONS OF INDUCED CHARGE DENSITY IN A RANDOM ARRAY

The conclusion drawn in Sect. IV that the charge distribution Eq. (14) applies not only to regular but also to a random NT array was based on the relation \(a > d\) between the penetration depth and the lattice constant. Thus, this conclusion pertains only to the “body” \(z \sim a\) of the distribution. Since \(\rho(z)\) falls off exponentially away from the NT tip, it might be expected that in the tail
$(h - z) \gg a$ the randomness in NT positions would terminate the applicability of Eq. (14). To verify this fact, one can incorporate the positional disorder into Eq. (19) perturbatively, i.e. to find the correction to the average $\rho(z)$ linear in the fluctuation of the NT density. Then the region of applicability of Eq. (14) to the random array can be established from the condition that the typical disorder-induced correction is smaller than the average. This program is carried out below.

In a random array, fluctuations in the areal concentration of NTs, $\delta \tilde{N}(R) = \tilde{N}(R) - \langle \tilde{N}(R) \rangle$, lead to fluctuations in the distribution of the induced charge density $\delta \rho(z, R)$. We linearize Eq. (11) and in the first order over the fluctuations obtain

$$\hat{H} \{\delta \rho(z, R)\} = \mathcal{F}(z, R),$$

where the integral operator, $\hat{H}$, is defined as

$$\hat{H} \{ f(z, R) \} = 4\pi N_0 a^2 \left[ f(z, R) + \frac{1}{4\pi a^2} \int dR' \int_0^h dz' S_{ext}(z, z'; R - R') f(z', R') \right].$$

The rhs in Eq. (21) is the potential created by the fluctuation, $\delta \tilde{N}(R)$, of the density of NTs with unperturbed charge distribution, Eq. (11). As follows from Eq. (9), this potential is given by

$$\mathcal{F}(z, R) = - \int dR' \delta \tilde{N}(R') \int_0^h dz' S(z, z'; R - R') \rho(z').$$

The fact that the kernel of the integral operator, $\hat{H}$, depends on the difference $(R - R')$ suggests transformation from $\delta \tilde{N}(R)$ and $\delta \rho(R, z)$ to the Fourier harmonics $\delta \tilde{N}(q)$ and $\delta \rho(z, q)$, where $q$ is the in-plane wave vector. Upon the Fourier transform, Eq. (21) assumes the form

$$4\pi N_0 a^2 \delta \rho(z, q) + \frac{2\pi N_0}{q} \int_0^h dz' \delta \rho(z', q) \times \left\{ \exp[-|z - z'|q] - \exp[-(z + z')q] \right\} = \mathcal{F}(z, q),$$

where the rhs is proportional to $\delta \tilde{N}(q)$

$$\mathcal{F}(z, q) = \frac{4\pi a \rho_0}{q(a^2 q^2 - 1)} \left\{ \sinh(qz) \exp\{-qh\} \right.$$

$$\left. \times \left[ \cosh(h/a) + qa \sinh(h/a) \right] - aq \sinh(z/a) \right\} \delta \tilde{N}(q).$$

The structure of the kernel in Eq. (24) is similar to that in the unperturbed equation Eq. (10). It appears that, due to this similarity, Eq. (24) can be solved analytically in the same way as Eq. (10). Namely, upon taking the second derivative from both sides, Eq. (24) reduces to the following second-order differential equation with $z$-independent coefficients

$$\delta \rho''(z, q) - \gamma_q \delta \rho(z, q) = \frac{1}{4\pi N_0 a^2} \left[ \mathcal{F}''(z, q) - q^2 \mathcal{F}(z, q) \right],$$

where $\gamma_q$ is defined as

$$\gamma_q^2 = q^2 + \frac{1}{a^2}.$$  

Note that the rhs of Eq. (26) can be cast in the following simple form

$$\mathcal{F}''(z, q) - q^2 \mathcal{F}(z, q) = 4\pi \delta \tilde{N}(q) \rho_0 \sinh(qh/a).$$

It can be now seen from Eq. (28) that $\rho_0 \delta \tilde{N}(q) \sinh(qh/a)$ is a particular solution of the differential equation (26). However, to find the solution of the original integral equation (24), one has to complement the particular solution with the solution of the homogeneous equation, i.e. to write

$$\delta \rho(z, q) = \left[ \chi_q \sinh(\gamma_q z) + \lambda_q \sinh(z/a) \right] \delta \tilde{N}(q)$$

$$= P(z, q) \delta \tilde{N}(q),$$

and find the constants $\chi_q$ and $\lambda_q$ by substituting Eq. (29) into Eq. (24). This yields

$$\chi_q = \frac{2\rho_0}{N_0 a^2 q^2} \left[ \frac{\cosh(h/a) + qa \sinh(h/a)}{\gamma_q \cosh(\gamma_q h) + q \sinh(\gamma_q h)} \right],$$

$$\lambda_q = -\frac{\rho_0}{N_0 a^2 q^2}.$$  

Note that $\lambda_q$ diverges at small $q$. However, the full solution Eq. (29) remains finite in the limit $q \to 0$. It also satisfies the obvious condition $\delta \rho(0, q) = 0$.

Equations (24) - (31) allow one to quantify the effect of disorder in the NT positions on the distribution of induced charge. The most interesting case is $h \gg a$, when this distribution is determined by collective screening involving many NTs. In this limit Eq. (29) can be simplified by replacing $\sinh(h/a)$ and $\cosh(h/a)$ by $\exp(h/a)$ and introducing $z_1 = (h - z) \ll h$. Then $h$ drops out from $z_1$-dependent part of $P(q)$ in Eq. (29), and we obtain

$$P(z_1, q) = \frac{\rho_0 \exp(h/a)}{N_0 a^2} \left[ \frac{\exp(-\gamma_q z_1) - \exp(-z_1/a)}{q^2} - \frac{\exp(-\gamma_q z_1)}{(q + \gamma_q)(\gamma_q + 1/a)} \right].$$

The form (32) is very convenient to study the effect of disorder in the “tail”, i.e. at large $z_1$. Indeed, assuming Gaussian fluctuations in $\delta \tilde{N}(R)$, so that

$$\langle \delta \tilde{N}(q_1) \delta \tilde{N}(q_2) \rangle = 2\pi N_0 \delta(q_1 - q_2),$$  

$$P(z_1, q) = \frac{\rho_0 \exp(h/a)}{N_0 a^2} \left[ \frac{\exp(-\gamma_q z_1) - \exp(-z_1/a)}{q^2} - \frac{\exp(-\gamma_q z_1)}{(q + \gamma_q)(\gamma_q + 1/a)} \right].$$

The form (32) is very convenient to study the effect of disorder in the “tail”, i.e. at large $z_1$. Indeed, assuming Gaussian fluctuations in $\delta \tilde{N}(R)$, so that
the variance of random fluctuations in the induced charge density can be expressed as follows

\[
\langle \delta \rho(z_1)^2 \rangle = \frac{1}{4} \int d\mathbf{R} \langle \delta \rho(z_1, \mathbf{R})^2 \rangle = \frac{N_0}{2\pi} \int dq P(z_1, q)^2. \tag{34}
\]

Here \( A \) is the normalization area. It is now seen that the \( q \)-dependence of \( P(z_1, q) \) is dominated by the first term in Eq. (32). The reason for this is the following. As was explained in the beginning of this Section, the applicability of Eq. (13), obtained for the regular array, is expected to be terminated in the random array at “depths” \( z_1 \) that are \( \geq a \). At these depths the average field is strongly suppressed. On the other hand, for \( z_1 \geq a \) one can use the expansion \( \gamma_q = \sqrt{q^2 + 1/a^2} \approx 1 + \frac{q^2}{2} a^2 \). This, in turn, suggests that characteristic values of the wave vector, \( q \), are \( q \leq 1/(a z_1)^{1/2} \leq 1/a \). Then the typical ratio of the second and the first terms in (32) is \( q^2 a^2 \ll 1 \). It also follows from the expansion of \( \gamma_q \) that the main exponents in \( \delta \rho(z_1) \) and in the average \( \rho(z_1) \) are the same. Upon neglecting the second term in Eq. (32), the \( q \)-dependence of \( P(z_1, q) \) acquires the form \( P(z_1, q) \propto \exp(-a q^2 z_1^2/2) / q^2 \). Then the \( q \)-integration in Eq. (34) can be easily performed. We will present the final result as the ratio of variance, \( \langle \delta \rho(h - z_1)^2 \rangle / \langle \rho(h - z_1)^2 \rangle \), and the square of average charge density

\[
\frac{\langle \delta \rho(h - z_1)^2 \rangle}{\langle \rho(h - z_1)^2 \rangle} = \frac{\ln 2}{2} \left( \frac{z_1}{N_0 a^3} \right). \tag{35}
\]

The above result offers the quantitative answer to the question about fluctuations of the induced charge density due to the randomness in the NT positions. In particular, it can be concluded from Eq. (35) that the disorder-induced fluctuations in the charge density are negligible, if \( z_1 \leq N_0 a^3 \). Since this value is much bigger than \( a \), Eq. (35) confirms our earlier claim that Eq. (14) applies not only for regular, but also for the random array. However, this applicability is limited by the distance \( z_1 \leq N_0 a^3 \). For larger \( z_1 \) the variance exceeds the average suggesting that the charge density strongly fluctuates within the plane \( z_1 = \text{const} \). Note, however, that these fluctuations are smooth with characteristic scale \( (z_1 a)^{1/2} \), which is much smaller than \( z_1 \), but much bigger than the penetration depth, \( a \).

As a final remark of this Section, we point out that the lower is the density of the random array the bigger is the depth, \( z_1 \), down to which Eq. (14) applies, as it follows from Eq. (35). However, the magnitude of the decay of the charge density, \( \rho(h - z_1)/\rho(h) \), is governed by the ratio \( z_1/a \). For \( z_1 = N_0 a^3 \), this ratio depends on the density of the array only weakly (logarithmically).

**VII. IMPLICATIONS FOR FIELD EMISSION**

**A. Single NT**

It is commonly accepted that the field emission current, \( J \), from the NT tip is described by the Fowler-Nordheim law

\[
\left| \ln(J(F)/J_0) \right| = 4 \sqrt{2 \pi m W^2/3 e \hbar F^2}, \tag{36}
\]

where \( J_0 \) is the prefactor, \( m \) is the electron mass, \( W \) is the work function, which, in principle, is dependent on the tip geometry\(^{26,37,38} \). Parameter \( \beta \) is the field enhancement factor. Various applications of the field emission from NTs are based on the fact that \( \beta \) is large as a result of the NT geometry, more specifically, due to the large ratio \( h/r \). The expression for the enhancement factor routinely used in the fitting the experimental \( I-V \) curves\(^{39} \) is \( \beta = Ch/r \), where \( C \sim 1 \) depends on specific geometry of the tip. Within the Thomas-Fermi description of the induced charge distribution, outlined in Sect. II, the expression for field at a distance, \( z_1 \), from the NT tip is given by the derivative of the potential, \( \phi(z_1) \), created by the induced charges

\[
F_{\text{ind}}(z_1) = \frac{d\phi(z_1)}{dz_1} = \frac{d}{dz_1} \int_0^h dz \rho(z) S_0(z, z_1), \tag{37}
\]

where \( \rho(z) \) is given by Eq. (5). Then the evaluation of the integral (37) yields

\[
\frac{F_{\text{ind}}(z_1)}{F} = \left( \frac{\hbar}{2L_h r} \right) \min\{1, r/z_1\}, \tag{38}
\]

where we had assumed \( F_{\text{ind}} \gg F \). It is seen from Eq. (38) that the enhancement factor indeed has the conventional form, \( \beta = Ch/r \), with \( C \approx (2 L_h)^{-1} \) for \( z_1 \leq r \), but it falls off with increasing \( z_1 \). This suggests that for low enough applied fields, when the electron tunnelling length \( \sim W/F_{\text{ind}} \) exceeds \( r \), the \( I-V \) characteristics deviates from the Fowler-Nordheim law. In order to estimate this deviation, we substitute

\[
\phi(z_1) = \frac{F_h}{2L_h} \left\{ \frac{z_1}{r} \Theta(r - z_1) + \left[ 1 - \ln(r/z_1) \right] \Theta(z_1 - r) \right\} \tag{39}
\]

into the tunnelling action

\[
\left| \ln(J(F)/J_0) \right| = \frac{2 \sqrt{2 m}}{e \hbar} \int_0^{z_t} dz_1 \sqrt{W - e \phi(z_1)}, \tag{40}
\]

where \( z_t \) is the turning point at which the expression under the square root is zero. In (40) we had neglected the bare potential \( eF z_1 \). It is now convenient to measure the electric field in terms of \( F_0 \), defined as \( F_0 = W/e \beta r = \)
2WΛz/eh. The integral in Eq. (10) can be reduced to the error function, erf (x), after which Eq. (10) acquires the form

\[ \ln(J(F)/J_0) = \frac{4\sqrt{2mW^3}}{3\hbar \beta F_0} G(F_0/F) , \] (41)

where the dimensionless function \( G(\tau) \) is defined as

\[ G(\tau) = \tau, \quad \tau < 1; \]
\[ G(\tau) = \tau - \left( \tau + \frac{1}{2} \right) \left( 1 - \frac{1}{\tau} \right)^{1/2} + \frac{3}{4} \sqrt{\frac{\pi}{\tau}} \exp(-1) \text{erf} \left( \sqrt{\tau-1} \right), \quad \tau > 1. \] (42)

The plot of the function \( G(\tau) \) is shown in Fig. 2. Strictly speaking, the Fowler-Nordheim region, corresponds to \( \tau < 1 \), where the slope of \( G(\tau) \) is identically unity. However, \( G(\tau) \) can be linearized around \( \tau > 1 \), where the slope is larger. For example, at \( \tau = 2 \) the slope is \( \approx 2 \). This can be interpreted as a two-times reduction of the enhancement factor, \( \beta \), in Eq. (41). A significant reduction of the enhancement factor (e.g., 30 times) occurs around \( \tau \approx 5 \). It should be noted, that, since Eqs. (41) and (42) were derived neglecting the bare potential, their applicability is limited by \( \tau < \tau_{\text{max}} \), where \( \tau_{\text{max}} \) corresponds to the applied field \( F = F_0/\tau_{\text{max}} \), for which the turning point, \( z_1 \), in the tunnelling action Eq. (40) reaches the value \( W/F \). The latter condition can be rewritten in the form \( (\tau_{\text{max}} - 1) = \ln(h\tau_{\text{max}}/r) \), yielding \( \tau_{\text{max}} \approx \ln(h/r) = \Lambda_0 \). For \( \tau > \tau_{\text{max}} \), i.e. for applied fields \( F > F_0/\tau_{\text{max}} \), the I-V characteristics is given by the Fowler-Nordheim law \( \phi^\text{FN} \) with \( \beta = 1 \).

Overall, Fig. 2 indicates that, for low enough applied fields there are significant deviations from the Fowler-Nordheim law in the I-V characteristics of an individual NT. For such fields the linearity of the Fowler-Nordheim plots shows only within very narrow interval of \( F \). On the experimental side, there are reports, e.g., Ref. 11, where applicability of the Fowler-Nordheim law was demonstrated within a rather wide (exceeding 3 times) interval of change of \( F \). In other reported measurements, see, e.g., Ref. 11, linearity of \( \ln J \) vs. \( 1/F \) holds only within a limited (less than 2 times) interval of applied fields. Whether or not Eq. (11), derived for a single NT, is suitable to fit experimental results depends crucially on the geometry of the array, as we discuss below.

B. Array of NTs

As it was mentioned in the Introduction, increasing the density of NTs in the array leads to dramatic suppression of the enhancement factor. To illustrate this point, consider first the array of low density, when the tunnelling length is much smaller than the distance between the neighboring NTs. Then the field created by induced charges near the tip of a given NT can be calculated from Eq. (37), with \( \rho(z) \) given by Eqs. (14), (15). This calculation yields the generalization of the field enhancement factor Eq. (38) to the case of the array of low density

\[ \frac{F_{\text{ind}}(z_1)}{F} = \frac{a}{2r \Lambda_d} \tanh(h/a) \min\{1, r/z_1\}. \] (43)

The above expression recovers the enhancement factor for a single NT in the limit \( a \to \infty \), or equivalently, \( N_0 \to 0 \). For \( a < h \), we conclude that the enhancement factor for the array, compared to the single NT is suppressed as

\[ \frac{\beta(N_0)}{\beta(0)} = \frac{a \Lambda_0}{\hbar \Lambda_d} \left( \frac{\Lambda_0^2}{2\pi \Lambda_d N_0 h^2} \right)^{1/2} \ll 1. \] (44)

We now turn to the high-density array. In such an array, the tunnelling length of an emitted electron can exceed the distance, \( N_0^{-1/2} \), between the neighboring NTs. Then the form of the tunnelling barrier is not anymore given by the potential created by a single NT with charge distribution modified by neighboring NTs. Instead, one has to use a general expression

\[ \phi_A(z_1) = \int_0^h dz \rho_0 \sinh[(h - z)/a] \mathcal{S}(z, z_1), \] (45)

where \( \mathcal{S} \) is the kernel defined by Eq. (20). The first term in the kernel (20) corresponds to the “continuous” limit, and yields a contribution \( Fz_1 \) to \( \phi_A(z_1) \). The second term in Eq. (20) exhibits different behavior for large \( \left| z - z_1 \right| > N_0^{-1/2} \) and small \( \left| z - z_1 \right| < N_0^{-1/2} \) distances. The expression for \( \phi_A(z_1) \) that captures both long, and short-distance behaviors has the form

\[ \phi_A(z_1) = Fz_1 \]
\[ + \frac{Fd}{2\sqrt{2L_d}} \left\{ \frac{a \Theta(z_1 - d)}{2pa + d} \left[ \exp(-2\pi z_1/d) - 1 \right] \right. \]
\[ + \left. \Theta(d - z_1) \ln \left( \frac{d}{z_1} \right) \exp(-z_1/a) \right\}. \] (46)

The long-distance behavior is described by the first term in the square brackets. It represents the correction to the “continuous” first term, \( Fz_1 \), in [10] due to discreteness of the array. Clearly, the field enhancement due to this term is negligible. It is the second term in the square brackets that is responsible for the field enhancement. The physics, captured by this term, is that at distance \( z_1 < N_0^{-1/2} \) the tunnelling electron “sees” not only the NT, from which it was emitted, but also neighboring NTs. This term, however, does not contain the NT radius, \( r \), which was set zero in the derivation of Eq. (40). The dependence on \( r \) can be reinstated in the
same way as for a single NT in Eq. (49), namely

$$\phi_A(z_1) \approx \frac{F d}{2\sqrt{2\xi_d}} \left\{ \Theta(r - z_1) \frac{z_1}{r} + \Theta(z_1 - r)\Theta(d - z_1) \left[ 1 - \ln \left( \frac{r}{z_1} \right) \exp\left(-\frac{|z_1 - r|}{a}\right) \right] \right\}, \quad (47)$$

where we have retained only the part, responsible for the field enhancement. The subsequent calculation of I-V characteristics using Eq. (47) is completely identical to the case of an isolated NT. The result can be presented in the form similar to Eqs. (11), (12)

$$\left[ \ln\left(\frac{J(F)}{J_0}\right) \right] = \frac{4\sqrt{2mW^3}}{3\epsilon h \beta(N_0)} F_A \frac{G_A(F_A/F)}{F}, \quad (48)$$

$$G_A(\tau) = \tau \left[ 1 - \left( 1 - \frac{1}{\tau} \right)^{3/2} \right] + \frac{1}{\sqrt{\tau}} \int_1^{u_r} \left\{ (1 + \ln u_r) \exp\left(-\frac{r}{a}(u_r - 1)\right) \right\} \left\{ (1 + \ln u) \exp\left(-\frac{r}{a}(u - 1)\right) \right\}, \quad (49)$$

where $u_r$, which is related to the turning point, $z_r$, as $u_r = z_r/r$, satisfies the following equation

$$\tau = (1 + \ln u_r) \exp\left(-\frac{r}{a}(u_r - 1)\right). \quad (50)$$

The boundary value of the external field, $F_A$, corresponds to the turning point $z_r = (F_A/F)_r = \tau_A r \geq r$. The field $F_A$ is related to the single NT boundary field, $F_0$, by

$$F_A = \frac{L_d h}{L_h a} F_0. \quad (51)$$

Therefore, the variable, $\tau_A$, defined above, is related to the corresponding single NT variable, $\tau$, as

$$\tau_A = \left[ \frac{2\pi L_d h N_0 h^2}{L_h^2} \right]^{1/2} \tau. \quad (52)$$

The function $G_A(\tau_A)$ is plotted in Fig. 2 together with the function $G(\tau)$. We see that, while the parameter $N_0 h^2$ changes within a wide interval, the function $G(\tau_A)$ remains close to $G(\tau)$. This means that the form of the I-V characteristics for the array is similar to that for a single NT. The difference essentially amounts to rescaling of the characteristic field, $F_0$, by the factor $L_d h / L_h a$. In other words, increasing the density of the array results in suppression of the field emission without the change of the shape of the I-V characteristics. It should be pointed out that this conclusion pertains to the array of randomly positioned, but completely identical NTs. As we will see below, the situation changes dramatically when the heights of NTs are random.

**FIG. 2:** Thin solid line is the Fowler-Nordheim law $|\ln(J/J_0)| \propto \tau$, where $\tau = F_0/F$. Thick solid line is the dimensionless current-voltage characteristics $|\ln(J/J_0)|$ vs $\tau = F_A/F$ of an individual NT plotted from Eq. (12). Dashed curves (1), (2), and (3) are the current-voltage characteristics, $|\ln J|$ vs $\tau = F_A/F$ plotted from Eq. (10) for $(h/r) = 10^4$ and dimensionless array densities $N_0 h^2 = 100, 10, 1$, respectively.

**VIII. DISCUSSION AND CONCLUDING REMARKS**

The main result of the present paper is Eq. (14) that describes the crossover of the induced charge density distribution from a single NT to the dense regular array of NTs. We have also demonstrated that Eq. (14) applies to the random array. Disorder in the NT positions terminates the applicability of Eq. (14) only at large distances from the tips, where the charge density had already dropped significantly. Concerning the field emission, our calculations quantify a strong suppression of the emission current in a dense array. The field enhancement factor falls off with the NT density, $N_0$, as $N_0^{−1/2}$ [see Eq. (14)]. This conclusion might seem to contradict the majority of experiments, where high enhancement factors for dense arrays were reported. More precisely, in majority of experiments the dependence of emission current on the NT density is simply not addressed, and the I-V characteristics are interpreted basing on the properties (such as work-function) of individual NTs. In fact, in those few papers where this issue is addressed, the suppression of field emission with increasing the NT density is pointed out on qualitative level. The resolution of this contradiction, in our opinion, lies in the fact that in realistic situations the heights of NTs in the array are widely dispersed. To get an insight how this dispersion in heights affects the field emission, consider a regular array in which one NT is higher than others by $h_1$, which is much larger than the average NT separation, $N_0^{−1/2}$, but much smaller than $h$. Within the interval $0 < z < h$ the distribution of charge in this “sticking out” NT is “enforced” by the neighbors, and is given by Eqs. (14)–(16). However, within the interval $h < z < (h + z_1)$ this NT “sees” the rest of the array as an equipotential
plane. From this observation we immediately conclude that, within the interval \( h < z < (h + z_1) \), the charge density in the sticking out NT is given by Eq. (4) with \( z \) replaced by \((z - h)\). This, in turn, suggests that the enhancement factor of external field in the sticking out NT is high and is equal to \( h_1/2\mathcal{L}_{h_1} r \), as follows from Eq. (45).

The above reasoning suggests that the conjecture that the field emission current is dominated by sparse sticking-out NTs allows to account for the high values of the enhancement factor observed in experiment. We will now demonstrate that this conjecture also allows to explain why the dependence \( \ln(J(F)/J_0) \) follows the Fowler-Nordheim law \( F(z) \) within a wide interval of \( F \), while Eqs. (41) and (42) predict strong deviations from Eq. (43) as \( F \) is decreased.

Obviously, the probability, \( P(h_1) \), to find within the array a NT that sticks out by \( h_1 \) decreases with \( h_1 \). Contribution of NTs with given \( h_1 \) to the emission current is determined by the product

\[
J_{h_1} \propto \exp\left\{ -4/3h^2/2 \right\} \mathcal{G}(F_0/F) P(h_1), \tag{53}
\]

where the first term is the tunneling action, which depends on \( h_1 \) through \( \mathcal{G} = 2W/c\mathcal{L}_{h_1} \); the function \( \mathcal{G} \) is defined by Eq. (42). Since the tunneling action increases rapidly with \( h_1 \), the product \( \mathcal{G} \) has a sharp maximum at a certain optimal \( h_1 \). Therefore, \( \ln(J(F)/J_0) \) is determined by the logarithm of the rhs of Eq. (53) taken at optimal \( h_1 \). The natural choice for \( P(h_1) \) is the Poisson distribution, \( \exp\left(-h_1/H\right) \). We can also use the fact that within the interval \( 2 < \tau < 10 \) the function \( G(\tau) \) can be approximated with high accuracy by the power law

\[
G(\tau) \approx 0.23\tau^{9/2}. \tag{54}
\]

Using this approximation, the optimal \( h_1 \) can be easily found analytically. It is convenient to cast the final result for \( \ln(J(F)/J_0) \) in the following form

\[
\ln(J(F)/J_0) = \left( \frac{2mW^3}{3e\hbar \beta_H F} \right)^{9/11}, \tag{55}
\]

where the “effective enhancement factor” is defined as

\[
\beta_H = 4.8 \frac{H}{r\mathcal{L}_r} \left( \frac{2m^2W}{h^2} \right)^{7/18}. \tag{56}
\]

Firstly, we see from Eq. (55) that the \( I-V \) characteristics is very close to the Fowler-Nordheim law, since the exponent \( 9/11 \) is close to 1. This should be contrasted to the \( I-V \) characteristics of a single NT, for which \( \ln(J(F)/J_0) \propto F^{-9/2} \), as follows from Eq. (54).

Secondly, the effective enhancement factor \( \beta_H \) is large and depends rather weakly on the work function \( h \). Summarizing, the dense array of NTs can exhibit the Fowler-Nordheim field emission provided there is a sufficient spread in the NT heights. In fact, this conclusion is in accord with reported experimental findings. In particular, direct imaging of emission intensity by means of scanning force microscopy reveals that only a tiny portion of NTs in \( 10^{-4} \) or even smaller) contributes to the net current.

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