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Thermal runaway and evaporation of metal nano-tips during intense electron emission

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When metal surfaces are subjected to high electric fields, plasma is formed even under ultra high vacuum conditions. This phenomenon, known as vacuum arcing, is a major limiting factor in various modern applications such as particle accelerators, fusion reactors, etc, as practice shows that surface cleaning is not sufficient to prevent it. In this letter we report an intrinsic mechanism of how a metal surface responds to the application of a very high electric field, which leads to plasma build-up. We present multi-scale atomistic simulations that concurrently include field-induced forces, electron emission with finite-size and space-charge effects, Nottingham and Joule heating. This unique approach allowed us to analyze the dynamic evolution of a copper nano-tip in atomic detail. We observed a thermal runaway process that triggers atom evaporation at a rate sufficient to initiate plasma.

Electric discharges in the form of arcs appearing in vacuum, also known as vacuum breakdowns, have been studied thoroughly for more than 70 years both experimentally and theoretically¹⁻⁸. Vacuum breakdown phenomena play a significant role in various technological applications by being either exploitable or highly undesirable. For example, they can be used in ion sources⁹ or for physical vapour deposition⁷. On the other hand, they hinder the function and limit the performance of various vacuum devices that require high electric fields, such as fusion reactors¹⁰, vacuum interrupters¹¹, electron sources⁴ and powerful linear accelerators to be used in particle colliders for new insightful experiments at CERN (CLIC)¹².

Over decades, vacuum arcing strongly attracted the attention of researchers from different fields, since various physical phenomena are involved. However, the mechanisms of plasma onset are still under debate. During an arc, plasma is ignited in the vacuum and burns until the available energy from the power source is consumed. It is well-known since the 1950's^{2,3,7,8} that vacuum arcs appear after intense field electron emission, but the physical mechanism that leads from the latter to plasma ignition is not yet fully understood.

Recent experimental studies show that the improvement of surface and vacuum quality cannot diminish the probability of vacuum arcing¹³. In the same direction, arcs were found to appear on single metal tip cathodes when their current density exceeded a critical value even for well-controlled emission conditions¹. These indicate the existence of an inherent mechanism of surface response that initiates breakdown.

One hypothesis commonly used to explain the plasma build-up from field emission is "explosive emission"^{7,14-16}. According to it, when intensive field emission takes place, there is a critical current density beyond which heating is produced at a rate the emitter cannot dissipate. This leads to heat accumulation and extremely high local temperatures, sufficient to cause in-

stant explosion and plasma formation⁷. However, this hypothesis is based on only phenomenological considerations, since the atomic level insight in such a complex phenomenon as vacuum arcing was not available.

Here we present multi-physics atomistic simulations that reveal a thermal runaway process. The latter is based on the gradual deformation of an emitting tip, due to the field-induced forces. The simulations show that both high current density and sufficiently large tip size are needed to initiate the melting at its apex. After that, the field-induced forces gradually deform the tip, elongating and sharpening its apex in a process similar to the Taylor cone formation in liquid metal ion sources¹⁷⁻¹⁹. This process, in combination with the decrease of the electrical and thermal conductivities at high temperatures, leads to a positive feedback thermal runaway mechanism. Eventually, large fractions of the emitter evaporate in the form of neutral atoms, but also as charged nano-clusters. The total number of evaporated atoms (both isolated and clustered) is compatible with the minimum evaporation rate required to ignite plasma, as found by recent Particle-In-Cell (PIC) calculations^{20,21}.

In order to study the atomic level processes leading to the initiation of plasma, we perform Molecular Dynamics (MD) simulations. However, the thermal runaway process involves various physical phenomena, which classical MD cannot describe implicitly due to its atomistic nature. The electric field interacts with the material, inducing charges and forces on the surface atoms and generating electron emission. The latter heats the nano-tip due to the Nottingham^{22,23} and Joule effects. The combination of the above causes significant changes in the structure and geometry of the material and hence they have to be quantified and included in the calculations.

In this letter we propose a model, which extends the capability of the classical MD method by solving the heat and electrostatic equations concurrently for a dynamically evolving nano-emitter shape. We have updated our HELMOD model²⁴ that couples MD simu-

lations with electric field calculations, and includes the charge-induced forces in the MD interactions. Although this model provided many interesting insights into the interaction of high electric fields with metal surfaces, the rigid grid used to calculate the electric field imposed critical limitations for systems dynamically evolving at high temperatures. To overcome this limitation we used our recently developed model FEMOCS^{25,26} to calculate the electric field distribution around the tip on a flexible mesh, and integrate this information into HELMOD.

FEMOCS solves the Laplace equation $\nabla^2\Phi = 0$ using the Finite Element Method (FEM) on an unstructured mesh. This mesh is generated automatically from the atomic positions in order to follow the varying nano-tip shape. A Dirichlet boundary condition $\Phi = 0$ is then applied on the nano-tip surface. Once the electrostatic potential Φ and field $\vec{E} = -\nabla\Phi$ are obtained, we calculate the induced charge $Q_s = \epsilon_0|\vec{E}|A_s$ on an elementary surface area A_s (see supplementary material section I) of the material-vacuum surface. This surface charge is then distributed to the nearby surface atoms. Given the charge q_i on each atom, we calculate and add to the MD interaction the Lorentz force $\vec{F}_i = \frac{1}{2}q_i\vec{E}_i$ and the Coulomb forces between the charged atoms. Details on the implementation of these calculations are given in the supplementary material section I and ref.²⁴.

Our previous calculations^{27,28} showed that the thermal effects caused by electron emission play an important role in the nanotip evolution under high fields. The classical electron emission equations²⁹⁻³¹ are inadequate to describe combined thermal-field emission³² from nanometrically sharp emitters³³⁻³⁵. However, our recently developed computational tool GETELEC³⁶ provides with the means to consistently and efficiently calculate the electron emission current and Nottingham heat distributions from sharp nano-tips, even at temperatures beyond the melting point. We also note that the local fields and the current densities calculated here are close to the Space Charge (SC) limit; thus the SC effect³⁷ is also included in our model.

Calculating fully the charge density distribution induced by the emitted electrons in three dimensions (3D) is a complicated and computationally expensive calculation³⁸, which is usually done with the PIC method^{39,40}. Nevertheless, Forbes⁴¹ claimed that the standard 1D SC model for field emission⁴² can be used to obtain the reduction of the surface field if a simple correction is introduced to account for the non-planar nature of the emitter. We used this approximation after confirming its validity by comparison to full 3D PIC simulations⁴⁰. Details on our SC model can be found in the supplementary material section II.

Using the results from the SC model we calculate the Joule and Nottingham heat distributions along the tip. For the Joule heating, we also need to obtain the resistivity ρ which depends on the local temperature and the size of the nano-tip. For a given temperature T , we obtain $\rho(T)$ by interpolating tabulated values found in the

literature^{43,44}. $\rho(T)$ is capped at the highest available value of 3500K. Furthermore, the mean free path of the electrons in the material is decreased due to the nanometric size of the nano-tip. To correct our ρ values for this finite-size effect, we use the simulation method and tool of Yarimbiyik et. al.⁴⁵. The latter can calculate the mean free path reduction for a given nano-wire diameter. The value we use for the latter is the mean diameter along the tip for the initial geometry.

In order to obtain the temperature distribution on the emitter we solve the heat diffusion equation⁴⁶. The geometrical structures which we simulate, have one dominant dimension, i.e. their height h is much longer than the lateral dimensions. Hence, in line with our previous publication²⁷, we use a 1D version of the heat equation which is solved using an explicit Euler scheme. Thus we obtain the temperature distribution $T(z, t)$ along the tip for each time step. The heat conductivity κ is calculated from ρ according to the Wiedemann - Franz law⁴⁶. A reduced value for the Lorentz number $L = 2.0 \times 10^{-8} \text{W}\Omega\text{K}^{-2}$ is used due the nanometric tip size⁴⁷. A detailed description and validation of our heat diffusion method is given in the supplementary material, section III.

The obtained temperature distribution $T(z)$ is fed back to the calculations for the next step in two ways. First, it is given as input to the electron emission calculations performed by the GETELEC code for the next timestep. Second, it is used to scale the MD atomistic velocities according to the Berendsen⁴⁸ temperature control scheme with a relaxation time $\tau = 1.5\text{ps}$. The described procedure is repeated for every MD timestep, skipping the full calculation if possible to increase the computational efficiency. If the root-mean-square average of the atomic displacement (compared to the last full-calculation step) is smaller than 0.38\AA , the last calculated solution is reused.

We simulate a conical Cu nanotip built on a $\{100\}$ surface (see figure 1). The cone is terminated with a hemispherical cap of a radius $R = 3\text{nm}$. The tip has a total height $h = 93.1\text{nm}$ and a full aperture angle of 3° . In order to keep the computational time feasible, only the upper half of the tip is fully simulated with MD. According to our estimations, only this part significantly heats providing high kinetic energies to the atoms. The bottom half maintains practically a constant shape and we consider it fixed; the field and temperature calculations are extended to the continuous limit in that region. Thus the total number of simulated atoms is 206000 and the MD simulation box has a size of $11.64 \times 11.64 \times 46.6\text{nm}$ (shown with grey lines in figure 1).

However the electric field and heat are calculated on the whole tip domain, which includes an extension of the MD system shown by the "virtual" particles lying out of the box in figure 1. These points are used to generate the FEM mesh on a $280 \times 280 \times 560\text{nm}$ box. This size is sufficient to assume periodic boundary conditions at the sides and a Neumann boundary condition at the top. Only a small part of this box is shown in the figure.

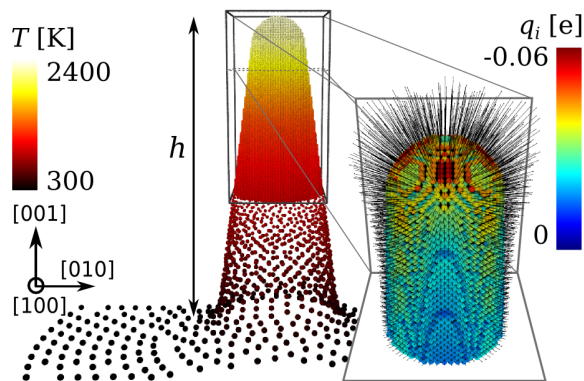


FIG. 1. Simulated tip shape illustrated by the MD atoms (in the box) and the constant "virtual" atoms (out of the box). The color coding represents the steady-state temperature distribution for constant shape. The inset illustrates the atom charges q_i by the color coding and, the corresponding forces \vec{F}_i by the black-line arrows.

For the MD simulations we used two different inter-atomic potentials with a constant timestep $\Delta t = 4.05\text{fs}$. The Sabochick and Lam (SL) EAM potential⁴⁹ and the one from Mishin et. al.⁵⁰. In order to take into account the stochastic nature of the process, we repeated the same simulation 7 times for each potential. Since our tip is isolated and continued to the bottom, no periodic boundaries were applied. The two bottom atomic Xlayers of the MD systems were fixed to obtain a smooth connection to the rest of the tip and avoid translational motion of the whole structure. Finally, the ten bottom layers above the two fixed were controlled to linearly increasing temperatures from 0 to $T(z)$ in order to avoid possible artefacts caused by extreme velocity mismatch.

The inset shows a closer view on the tip apex with the color coding corresponding to the calculated field-induced charges q_i for each atom and the black arrows corresponding to the forces \vec{F}_i .

A macroscopic electric field $E_0 = 0.8\text{GV/m}$, is applied instantly after the MD system is run to relax for 0.5ps. This value for E_0 is relevant to the surface fields used in modern high-gradient accelerating structures^{13,51}. At the initial configuration, the maximum local field at the apex as calculated by the Laplace equation is found to be $E_{max} = 17.9 \pm 0.04\text{GV/m}$. However, such a high field induces an emitted current density with an average value at the bottom of the MD simulation domain (middle of the conical tip) $J_{avg} = 3.57 \pm 0.02 \times 10^{-5}\text{Anm}^{-2}$. This will suppress the fields due to the SC effect by a factor of $\theta = 0.68 \pm 0.0008$ and converge to a final current density of $J_{avg} = 4.6 \pm 0.02 \times 10^{-6}\text{Anm}^{-2}$. For the SC model, we have assumed an applied voltage of $V_0 = 3\text{kV}$ which is then multiplied by a correction factor $\omega = 0.25$ (see section II of the supplementary material).

In this initial configuration, the deposited heat power is very high (reaches $3 \pm 0.1 \times 10^{-5}\text{Wnm}^{-3}$) and is dominated by the Nottingham effect. However, if the system

geometry is kept constant, the heat is rapidly dissipated towards the bulk due to the high thermal conductivity of Cu, and the temperature distribution converges to a steady state after about 233ps, with a maximum value of about $2378 \pm 10\text{K}$ at the apex. Furthermore, even if the applied field and voltage are increased to unrealistic values (double the previous ones), the current density J_{avg} is limited by the SC to a value of $\approx 1.3 \times 10^{-5}\text{Anm}^{-2}$, and the maximum steady-state temperature does not exceed 6200K. All the numerical results reported in the format " $x \pm \delta x$ " correspond to the mean values and standard errors as obtained from 16 different simulation runs.

On the other hand, if we let the whole MD system evolve according to the algorithm described above, we obtain a completely different picture. Figure 2 demonstrates the heat and shape evolution of the nano-tip for one of the simulations with the SL potential. The graph shows the time evolution of the height of the tip. The inset figures (a-g) demonstrate the snapshots of the tip evolution at the time steps designated on the graph respectively. The tip shape initially stays approximately constant ($t < 50\text{ps}$), while its temperature gradually increases. When the temperature exceeds the melting point at the apex, the top atoms become mobile and the field-induced forces can pull the apex atoms upwards, thus sharpening and elongating it. This leads to higher local fields, higher forces and higher emission currents, thus producing more heating. The above process increases further the temperature, thus forming a positive feedback loop and eventually leading to thermal runaway.

After about 200ps the deformed tip has reached more than 3000K. Meanwhile, the field-induced forces cause neck-thinning which becomes more pronounced as the simulation evolves (frame C). This thinning confines the total emitted current in a small cross-sectional area, thus producing a high local current density and Joule heating.

Eventually, the high local temperature in combination with the forces cause detachment of the upper part of the tip, creating an evaporated nano-cluster. The latter is charged (due to partially charged atoms on its surface) and is rapidly accelerated and removed by the field. Although its full behaviour requires further investigation (interaction with the field, collision with the evaporated atoms, emitted electrons, etc), for the purpose of this work we consider it instantly removed and continue the simulation with the remaining tip. All evaporated atoms and clusters are marked and removed by a cluster analysis algorithm⁵² implemented in FEMOCS²⁶, with a distance cut-off equal to the one of the MD potential (4.94Å for SL and 5.5Å for Mishin).

The thermal runaway process is reinitialized in the remaining tip until a new smaller cluster is evaporated. After the detachment of several small nanoclusters (E,F) and numerous atom evaporation events, the process finally stops as the tip blunts and cools down (G). The full animation of the evolution of the tip can be found in the supplementary material. Although the runaway and the evaporation appeared at different times and the

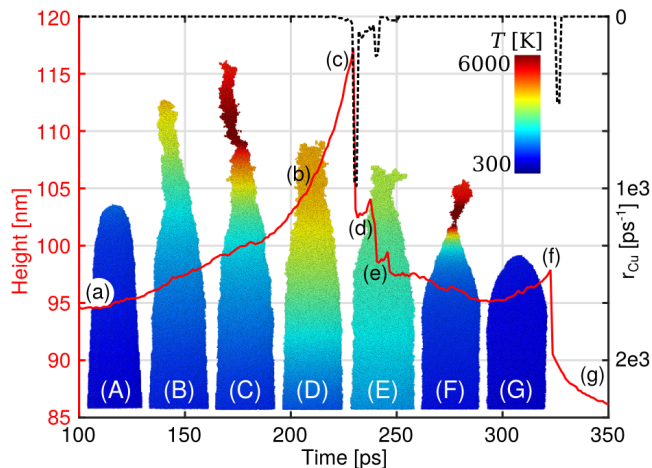


FIG. 2. The tip height (red line, left axis) and the evaporation rate (black dashed line, right axis, inverted upside-down) as a function of time. The inset images demonstrate the shape and the temperature distribution (color coding) in 7 different frames (A-G). The black lower case letters on the height graph demonstrate the time stamp of each frame (a-g).

tips took various shapes in the simulation runs with different random seeds, the steps described above remained qualitatively similar.

An arising question is whether the above process is enough to initiate self-sustaining plasma. In a recent work^{20,21}, PIC simulations showed that plasma can build up in the vicinity of an intensively field emitting spot. This happens if the cathode, emits not only electrons, but also neutral Cu atoms at a rate of at least 0.015 neutrals per emitted electron. However, the physical processes that can lead to the emission of neutrals along with electrons was not fully understood.

We define the average evaporation rate $\langle r_{Cu} \rangle$ as the total number of atoms detached from the tip (isolated or clustered) over the total time interval between the first and the last evaporation events. If we divide this quantity by the average emitted electron current during the same period, we obtain the mean evaporation rate per emitted electron $\langle r_{Cu/e} \rangle = 0.025 \pm 0.003 \text{ atoms/e}$. This rate is strikingly close to the values estimated in²⁰ and even exceeds the reported minimum of $r_{Cu/e} = 0.015 \text{ atoms/e}$ required to ignite plasma.

The consistency of our results with the previous independent simulations that used a different method (PIC) indicates that there is an intrinsic mechanism able to supply neutral atoms sufficient to ignite a vacuum arc near the cathode surface. Our rigorous calculations, based on the state-of-the-art understanding of the electric field-material interactions suggest that an external source of neutral atoms is not necessary to ignite the plasma; the latter can also be fed through self-evaporation of the surface atoms, in a process much less violent than the "explosive emission" conventionally assumed.

An important question is what are the prerequisites

to initiate thermal runaway. The latter is a complicated process that depends on various initial configuration parameters, namely material, geometry, applied field and voltage. A full analysis of all these parameters is out of the scope of this letter, and will be given in a forthcoming publication. However, as a general comment, two are the main contributing factors to the initiation of the thermal runaway: melting and force. This means that the tip height and the current density have to be sufficient to cause melting at the apex region. The minimum height depends on various other parameters that affect the heating, namely specific geometry, applied voltage and conductivities. In general, the current density required to produce such a heating is of the order of 10^{12} A/m^2 , in agreement with the experimental results of Dyke et. al.^{1,2}. Moreover, the importance of tip melting prior to an arc has been observed also experimentally by Batrakov et. al.⁵³.

On the other hand, the balance between the field-induced forces and the surface stress plays also a significant role. For the simulations we presented here, a minimum local field at the apex of about 10-12GV/m is necessary. In view of the above, it would be safe to assume that tips with a height of at least several tens of nm are required in the case of Cu. This is in agreement with experimental results⁵⁴, demonstrating that field emission with measured enhancement factors of the order 20-100 always precede vacuum arcs on Cu surfaces. This means that assuming a minimum tip radius of 1nm, the involved tips have heights of several tens of nm.

In conclusion, we have simulated the thermal and shape evolution of intensively electron emitting Cu nanotips by means of a multi-scale atomistic model. Our results reveal a thermal runaway process initiated by the field-induced forces acting on the molten apex of a nanotip. The thermal runaway leads to evaporation of metal fractions in the form of either atoms or nano-clusters, at a rate that exceeds the minimum needed to ignite plasma. This is a self-sufficient process, which does not depend on an external source of neutral atoms. Thus we show that the onset of a vacuum arc in ultra high vacuum is an intrinsic response of a metal surface to the applied high electric field.

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SUPPLEMENTARY MATERIAL

I. CALCULATION OF FIELD-INDUCED CHARGES AND FORCES

In order to calculate the charges and forces on the surface atoms, we use an extension of the Molecular Dynamics – ElectroDynamics (MD–ED) method we developed previously for HELMOD²⁴. We build a rectangular grid on the Molecular Dynamics (MD) simulation box so that every atom belongs to a certain grid cell. Since the atoms are in an FCC crystal (at least when the temperature is still low), there are numerous grid cells (roughly half) that contain no atoms. Furthermore, when the temperatures approach or exceed the melting point there might be some cells (a small percentage) containing more than one atoms, but this does not affect the functionality of our method.

When each atom is assigned to a certain cell, the cells are separated into material and vacuum domains. Figure 3 illustrates this separation. The cubes correspond to the cells of the material domain, for the initial configuration of the simulated tip after a relaxation of 0.5ps. The maroon spheres correspond to the atomic positions. In the inset a closer view of the area in the box is demonstrated, with the smaller grey cubes corresponding to some of the cells of the vacuum domain.

The separation is done in the following way. If a cell contains an atom or has a number of first nearest neighbours containing an atom $N \geq 4$, then it is designated as material (e.g. cell c_1 in the inset). If it does not contain an atom and it has $N \leq 3$ it is designated as vacuum (e.g. cell c_2 in the inset).

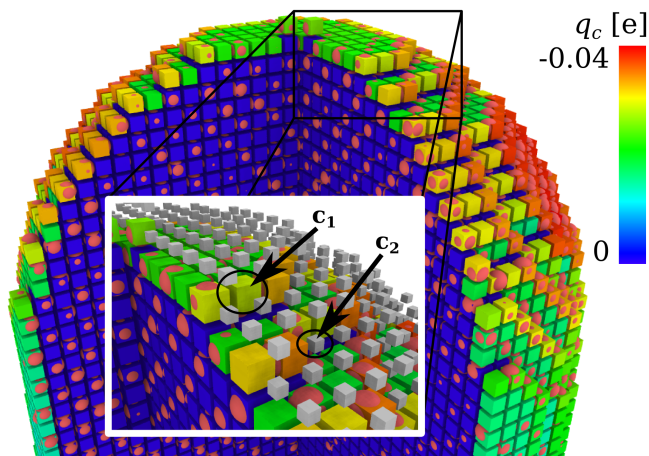


FIG. 3. Atoms on the tip apex (maroon spheres) after a relaxation of 0.5ps along with the corresponding material-designated cubic grid cells. The color coding on the cells corresponds to the charge q_c assigned to each cell. The inset demonstrates a closer view in the box area, with the grey small cubes being vacuum-designated cells.

Then the surface charges are calculated in every cell

face separating a vacuum cell from a material cell according to the Gauss law

$$q_f = \vec{E}_f \cdot \hat{n}_f A_f \quad (1)$$

where q_f is the surface charge on each cubic face, \vec{E}_f is the local electric field in the center of the face, and \hat{n}_f is the unit vector normal to the cubic surface. The electric field distribution has already been calculated by solving the Laplace equation using the recently developed Finite Element Method (FEM) tool FEMOCS²⁶. Then the charge on each surface cubic cell is calculated as

$$q_c = \sum_{f \in MV} q_f \quad (2)$$

where the summation is done over all the faces of the cell that are in contact with a vacuum one. The charges q_c are shown in the color coding of the cells in figure 3. Note that inner cubic cells that have not a common face with vacuum cells have $q_c = 0$.

Since we want to calculate the inter-atomic Coulomb interactions and the field-induced Lorentz forces, the charge on each atom q_i needs to be calculated. For this, all the grid-point charges have to be assigned to atoms while the total charge is conserved. This is done in a very simple way. If a grid point contains one or more atoms, then all its charge q_c is assigned to those atoms (divided equally). In the vast majority of the cases there is only one atom in each grid cell. On the other hand, if a grid cell does not contain any atoms, its charge is distributed equally to the atoms belonging to its first-neighbouring cells.

Finally, the Lorentz force \vec{F}_i on each atom is calculated by the formula $\vec{F}_i = \frac{1}{2} q_i \vec{E}_i$. The 1/2 term stands for the fact that the atoms are exposed to the electric field only from the one side of the material-vacuum surface. The Coulomb forces are calculated and included in the MD interaction by the same method described in ref.²⁴.

II. THE 1-DIMENSIONAL SPACE CHARGE MODEL

The space charge (SC) plays an important role in the determination of the electric field and the emission current around the emitter, especially in the very high field regime we simulate here. Although a simple model to describe the SC effects in field emission has been developed since the 1950's⁴², it is applicable only for quasi-planar emitters. Nevertheless, it has been proposed by Forbes⁴¹ that the standard 1D model can be used even for nanometric scale emitters, if the applied voltage V is multiplied by a correction factor $\omega < 1$. Here we describe how this approach is developed, implemented in our simulations, and validated versus existing 3D Particle-In-Cell simulations.

According to the standard 1D SC model, the local field on the emitter (uniform in 1D) is found as $F = \theta F_L$,

where θ is the dimensionless field-lowering factor and F_L is the field found by the Laplace equation, i.e by ignoring the SC effects. θ is a function of the the emitted current density J , the local field F and the total applied voltage V which is here an external parameter. J and F are also dependent on θ , which means that a self-consistent set of J, F, θ has to be obtained.

However, for a 3D nanometric emitter, J and F are not well-defined. In a quasi-planar emitter, the values at the apex can be used, since the SC is localized around it in a region much smaller than the radius of the emitter, where these quantities are practically constant. On the contrary, in a nanometric emitter the whole tip surface contributes to the SC, which means that the apex values are no longer representative.

Thus we will define the following representative values F_r, J_r for the field and the current density correspondingly

$$\begin{aligned} J_r &= \frac{\int_{S_h} J dS}{\int_{S_h} dS} \\ F_r &= \frac{\int_{S_h} F J dS}{\int_{S_h} J dS}. \end{aligned} \quad (3)$$

In eq. (3) the surface integrals are performed on the full-width-half-maximum emission surface S_h , i.e the surface where $J > \frac{1}{2} J_{max}$. On this surface, J_r is the mean value of J and F_r is the weighted mean value of F , with the weight being the emitted current density.

Using the representative values, the θ factor is calculated according to equation (13) of reference⁴¹ and then the whole potential and field distributions on the emitter are multiplied by θ . Then J_r, F_r are recalculated according to the new electric field distribution. This procedure is repeated iteratively until self-consistency is reached. Note that the obtained θ is used to multiply the whole electric field distribution, thus affecting not only the electron emission, but the charge and forces calculations described in section I.

In order to validate our 1D space charge model, we compare its results to full Particle-In-Cell (PIC) simulations performed by Uimanov⁴⁰. We reproduced the solution of the Laplace equation for the geometries presented in ref.⁴⁰, and then used our model to obtain the field-lowering factor θ , using a corrected applied voltage ωV . θ is obtained for various values of the applied voltage and for two of the geometries simulated by Uimanov: "FE4" and "FE5". "FE4" is an emitter with 10nm radius and "FE5" with 1nm, thus being the most relevant to our simulated tips that have a 3nm radius.

In figure 4 we see the $F - F_L$ (values at the apex) curves reported in figure 6 of ref.⁴⁰ (markers) along with the results of our model (lines) for the same geometries. The corresponding fitted values for the correction factor ω are shown in the legends. We see that our model is in very good agreement with the full 3D PIC calculation with an error of less than 1%, at a range of F_L up to about 35GV/m.

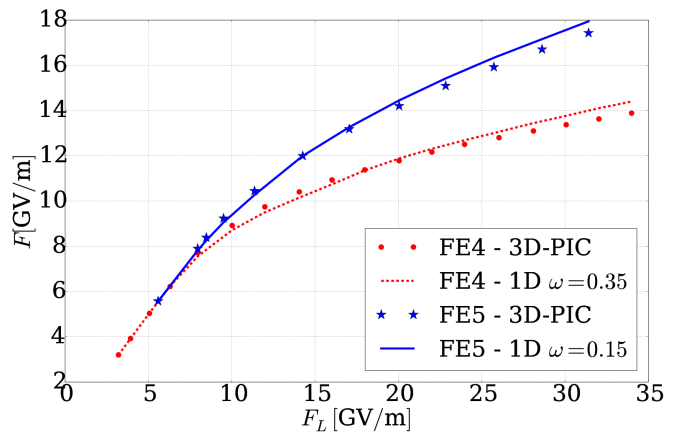


FIG. 4. Local apex field including the SC F versus the one ignoring SC F_L for the two different emitter geometries simulated by Uimanov⁴⁰. Markers correspond to the results of figure 6 in ref.⁴⁰ and the lines to the results obtained by our 1D SC model.

In view of the above, the 1D space charge model described above is a good approximation for nanometric size emitters, given that a valid correction factor ω is used. Although we cannot obtain here its value for the specific geometry we simulate, the fitted values obtained above for similar hemisphere-on-a-cone geometries indicate that the used value $\omega = 0.25$ is reasonable for our nano-tip. This value was chosen by linearly interpolating between $\omega = 0.15$ and $\omega = 0.35$, with respect to the logarithm of the emitter radii.

III. SOLUTION OF THE HEAT EQUATION

The heat diffusion equation in 3 dimensions is

$$C_V \frac{\partial T}{\partial t} = \nabla \cdot (\kappa(T) \nabla T) + p \quad (4)$$

where C_V is the volumetric heat capacity, $\kappa(T)$ is the heat conductivity as a function of T and p is the local deposited volumetric heating power density (in W/m^3). The Nottingham heat is expressed as surface density (in W/m^2), but it can also be treated as a volumetric quantity, localized in the surface region with the help of the Dirac δ function. In the following analysis it is considered to be a component of the volumetric density p .

Since the length of the simulated structures is dominant, let us integrate the temperature $T(x, y, z)$ on an infinitesimal "slice" Ω , between z and $z + \delta z$ with width δz , cross-sectional area $A(z)$ and volume $\delta V = A(z) \delta z$

$$C_V \frac{\partial}{\partial t} \int_{\Omega} T dV = \int_{\Omega} \nabla \cdot (\kappa \nabla T) dV + \int_{\Omega} p dV. \quad (5)$$

The integral of the left hand side equals to $\langle T \rangle \delta V$ where $\langle T \rangle$ denotes the mean temperature on Ω . The second term in the right hand side equals to the total heat

$\langle p \rangle \delta V$ deposited on the slice. Using the above notation and Gauss's divergence theorem we can rewrite eq. (5) as

$$C_V \frac{\partial \langle T \rangle}{\partial t} \delta V = \oint_{\partial \Omega} \kappa (\nabla T) \cdot \hat{n} dS + \langle p \rangle \delta V. \quad (6)$$

where \hat{n} denotes the unit vector perpendicular to the surface around the slice $\partial \Omega$. This surface has three components. The two cross-sections $A(z)$ and $A(z + \delta z)$ and the ring around them. For $\delta z \ll 1$ the contribution of the ring is negligible compared to A . Moreover, since the cross-sectional surfaces are along the $x-y$ plane, we have $\hat{n} \cdot \nabla T = \partial T / \partial z$ and the surface integral on the cross-section can be expressed as $A(z) \partial \langle T \rangle / \partial z$. Then eq. (6) yields

$$C_V \frac{\partial \langle T \rangle}{\partial t} A(z) \delta z = A(z + \delta z) \kappa(z + \delta z) \left. \frac{\partial \langle T \rangle}{\partial z} \right|_{z + \delta z} - A(z) \kappa(z) \left. \frac{\partial \langle T \rangle}{\partial z} \right|_z + \langle p \rangle A(z) \delta z. \quad (7)$$

Dividing eq. (7) by $A(z) \delta z$ and considering that the temperature does not vary significantly in the lateral directions, i.e. $\langle T \rangle \approx T$, yields the 1-dimensional (1D) heat equation for a variable cross-section and conductivity wire.

$$C_V \frac{\partial T}{\partial t} = \frac{1}{A} \frac{\partial}{\partial z} \left[A \kappa \frac{\partial T}{\partial z} \right] + \langle p \rangle. \quad (8)$$

The above equation is solved using the Finite Difference Method (FDM) with a simple explicit Euler scheme and a small time step $\Delta t = 0.06$ fs, small enough to avoid numerical instabilities. We divide the tip in N finite slices with width Δz , that coincide with the slices of the rectangular grid shown in figure 3. On each surface cell the current density and the deposited Nottingham heat surface density are calculated by GETELEC, as described in ref.³⁶ (the space charge is also taken into account as described in section II). Then the total current and total Nottingham heating power contributions of each cell (i_c and p_{N_c} correspondingly) are calculated by multiplying the densities with the cell face areas, similarly to the charge calculation described by equations (1), (2) in the previous section. We consider that the emission quantities have the same direction as the electric field of the cell.

The total current flowing through the cross-section of the k -th ($k = 0$ at the apex) slice is cumulative from the apex to the base as dictated by the continuity equation, i.e

$$I_k = I_{k-1} + \sum_{c \in S_k} i_c \quad (9)$$

where S_k denotes the surface cells of the k -th slice. The total heat deposited on the k -th slice is the sum of the Joule and the Nottingham heat components, i.e

$$P_k = \rho_k \frac{I_k^2}{A_k} \Delta z + \sum_{c \in S_k} p_{N_c}. \quad (10)$$

The Euler scheme for equation (8) is $T_k(t + \Delta t) = T_k(t) + \Delta T_k$. Using equation (10) yields

$$\Delta T_k = \frac{\Delta t}{C_V A_k \Delta z} \left[\frac{\kappa_k A_k}{\Delta z} (T_{k+1} + T_{k-1} - 2T_k) + \frac{1}{4\Delta z} (A_{k+1} \kappa_{k+1} - A_{k-1} \kappa_{k-1}) (T_{k+1} - T_{k-1}) + P_k \right] \quad (11)$$

where the subscript denotes the slice number for all quantities. Equation (11) is solved numerically for all slices with a Dirichlet boundary condition at the bottom of the tip, i.e $T_N = 300$ K and a Neumann condition at the apex, i.e $T_{-1} = T_0$. ρ_k and κ_k are updated at each time step according to the current T_k . On the other hand, A_k and P_k are recalculated and updated when either the field distribution is recalculated by FEMOCS (meaning that the atoms have been displaced more than a limit) or $T(z)$ has an RMS difference greater than 1% compared to the last step when A_k and P_k were obtained.

Finally, the resulting temperature distribution $T(z)$ is used to control the temperature of the MD system. The velocities of the atoms residing inside the k -th slice are scaled according to a Berendsen control scheme⁴⁸ with control temperature T_k and relaxation time $\tau = 1.5$ ps. This τ is much smaller than the relaxation time of the heat equation, but also big enough to avoid artefacts appearing in MD when intense velocity scaling.

The above 1D model has the advantage of its simplicity and computational efficiency. Solving the heat equation in 1D adds insignificant CPU time in the whole MD and field calculation. In order to validate the model, we compare its results with our previous 3D Finite Element Method (FEM) model²⁸. That model is used to solve the steady-state heat equation on the same FEM mesh produced by FEMOCS. The emitted current and Nottingham heat are in that case calculated using the standard General Thermal-Field (GTF)³². Only for this comparison, we will ignore the nanometric emitter size effects in electron emission and the space charge effect. Note that originally the FEM model ignored the Nottingham effect. Nevertheless, it was here implemented by adding a Neumann boundary condition to the heat equation, similar to equation (4) of reference²⁸. The boundary condition is

$$\hat{n} \cdot (\kappa \nabla T) = p_N \quad (12)$$

where p_N denotes the surface density of the deposited heating power and \hat{n} is the unit vector normal to the material surface.

Figure 5 shows the comparison between the two models. The lines correspond to the final steady-state (after about 100ps) temperature distribution along the simulated tip, as calculated with the 1D model described above. The markers correspond to the temperature along the vertical axis of symmetry of the tip, as calculated by the 3D FEM model. The calculation was performed for two different tip geometries, taken from two different

frames of the simulation shown in the main text. The calculation for a "regular tip" (solid lines and dots) corresponds to the original regular conical geometry (shown in figure 1a,b in the main text) and the one for "deformed tip" (dashed lines and diamonds) to the deformed shape the tip takes after 208.6ps of simulation (see figure 2 of the main text). We see that the simplified 1D model is in excellent agreement with the 3D FEM calculation, even for the irregular geometry of the "deformed tip". The markers correspond to the mean value of the temperature for a given slice. The deviation of the temperature around that value is smaller than the size of the marker in the figure.

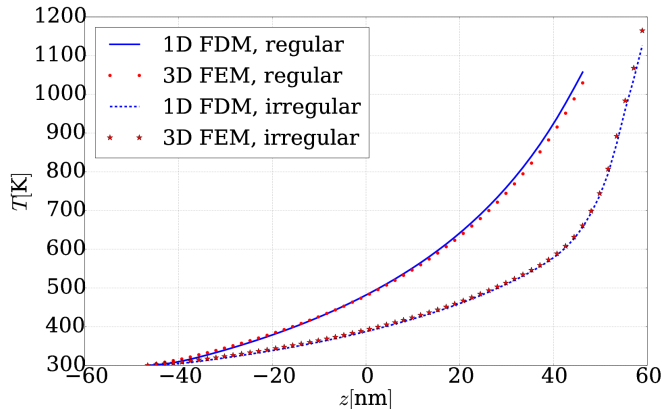


FIG. 5. Steady state temperature distribution as calculated by the 1D FDM (blue lines) and the 3D FEM (red lines) models for two different tip geometries and field values. For the regular conical geometry "Tip 1" (solid lines) the applied field was $E_{appl} = 0.56\text{GV/m}$ and for the oblique geometry "Tip 2" (dashed lines) $E_{appl} = 0.45\text{GV/m}$.

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