

## Is the linear relationship between the slope and intercept observed in field emission S-K plots an artifact?

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The performance of field emitters is usually analyzed by linear fitting of a Fowler-Nordheim plot. It has sometimes been observed that the fitted slopes and intercepts show a strong correlation, but no convincing explanation has been provided. We propose a simple model showing that this correlation is due to fluctuations in the slope and the fact that the experiments are carried out over a defined range of voltage and current. By performing a meta-analysis, we show that this explanation correctly predicts the results of other groups in this field.

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## I. INTRODUCTION

Field emission electron sources<sup>1</sup> are the subject of intensive research aimed at developing better microwave amplifiers, electron microscope guns, field-emitter displays and X-ray sources. A simple and widespread method for analyzing the properties of a field emitter is to use Fowler-Nordheim (F-N) theory. It assumes a triangular tunneling barrier and predicts a straight line if the current  $I$  and applied voltage  $V$  are plotted as the logarithm of  $I/V^2$  as a function of  $1/V$ . This plot is called an F-N plot. Although the triangular barrier model is incorrect and leads to an overestimation of the emission area by several orders of magnitude, F-N plots are often used, even if an image charge potential is taken into account to estimate experimental parameters. Using an F-N plot is fairly unreliable and led some theoreticians to propose alternative approaches<sup>2-6</sup> whose experimental success is questionable<sup>7-10</sup>. Among the various methods proposed, an intriguing one has emerged that illustrates the drift of an emitter. It involves performing several I-V measurements and extracting each slope and intercept from the linear fit of the F-N plots<sup>11</sup>. When the slopes are plotted against the corresponding intercepts for different voltage sweeps, an almost linear relationship has often been observed. This representation is commonly referred to as an SK chart or SK plot either because seppen and katamuki are the Japanese words for intercept and slope, respectively<sup>12</sup> or because it was named after the original researchers, Sasaki and Kaneko<sup>13</sup>.

It is tempting to consider the linear relationship in the SK plot as a valid alternative or complementary approach to the analysis of the F-N plot<sup>14,15</sup>. However, the origin of this linear relationship is rather dubious. The group at the origin of the SK plot<sup>16</sup> claimed that "From the location in the SK chart, we can estimate the relative difference in the work function and emission area or even the length of the carbon nanotubes responsible for electron emission" or that "the slope of linear distribution of the SK plots is known to be closely related to the work function". It has also been suggested that the linearity in the SK plot is evidence of a hidden relationship between the enhancement factor, the emission area or the work function<sup>11,17</sup>. However, it is difficult, if not impossible at present, to design an independent experiment in which the work function, the emission area and the enhancement factor of the emitter are well characterized<sup>10</sup>. We therefore doubt that it is feasible to obtain convincing proof of the usefulness of the SK trace for extracting the physical parameters of a field emitter.

To add to the confusion, the same group also stated that "The empirical relation between the slope and intercept is not well understood yet because the behavior of the F-N characteristics on

this chart does not coincide with the theoretical prediction"<sup>18</sup>. Numerical simulations for arrays of tips have shown that varying the number of emitters or their physical parameters according to a log-normal distribution can reproduce the linear behavior observed in SK plots<sup>19</sup>. Another group considered "This plot is a rather sophisticated theoretic problem"<sup>20</sup>. The sophistication of the problem may have escaped us, because in what follows, a simple explanation will be proposed. This does not preclude a more in-depth theoretical analysis with for instance emission models that evaluate current over an entire surface<sup>7,21-23</sup>. Our explanation indicates that the SK plot doesn't arise from the physics of field emission but from a mathematical property of the linear regression theory.

## II. METHODS

In a standard Fowler-Nordheim plot field emission I-V data are fitted to a straight line to extract the slope  $A$  and intercept  $B$  such that :

$$\log \frac{I}{V^2} = \frac{A}{V} + B \quad (1)$$

where  $I$  is the emission current,  $V$  is the applied voltage difference between the anode and cathode and  $\log$  is the natural logarithm. Then experimentalists often try to extract experimental parameters such as the enhancement factor, the emission area or the work function from  $A$  and  $B$ . Such an estimation is not very reliable<sup>4,9,10</sup>, especially for small radius emitters like carbon nanotubes. However, it is less common to try to reproduce such an I-V curve to estimate the variation and thus the reliability of the extracted slope and intercepts. Moreover, although the idea of an FN plot was proposed after the FN theory with a triangular barrier, the extraction of  $A$  and  $B$  makes no assumptions about the model that will be used to interpret the data. The model can include an image charge potential, the voltage dependence of the emission area or the enhancement factor, the replacement of the smooth flat surface hypothesis by a proper atomistic structure, the field and temperature dependence of the work function, the calculation of the electron density beyond the Sommerfeld free electron model. An SK plot analysis is independent of the choice of the model. In this article, we do not question the fact that by plotting  $A(t)$  versus  $B(t)$ , where  $t$  is the time between successive voltage sweeps, a convincing linear relationship can appear on an SK plot. Such behavior has been observed in a large variety of samples from single emitter to cathode arrays, encompassing metallic emitters, Spindt cathodes or carbon nanotubes. It was even measured in an experiment that lasted more than 20 years<sup>24</sup>.

The sample used in this article is a <111> tungsten tip. Its initial radius observed in a scanning electron microscope is equal to 15 nm. It was fabricated by electrochemical etching of a tungsten wire with a diameter of 125  $\mu\text{m}$ . Field emission experiments were performed in an ultra-high vacuum chamber with a base pressure of  $3 \times 10^{-10}$  Torr. The tip was degassed, several times, for 30 seconds, with a resisting loop at a temperature of 1700 K. Tip blunting occurs due to these multiple heatings. A quadrupole was placed in front of the tip at a 1 mm distance. The current was measured with a coupled microchannel plate MCP/phosphor screen system connected to a homemade current amplifier. The MCP/phosphor can amplify very low currents and allows visualization of the emission area. We performed the measurements with DC field emission current in the fA to pA range.

### III. RESULTS

Fig.1 shows such an SK plot for a standard W tip field emitter measured in our ultra-high vacuum chamber. It was obtained from 5 successive upward and downward voltage sweeps. The coefficients of determination  $R^2$  corresponding to the 10 independent F-N plots were very good with values ranging between 0.9993 and 0.9999. The I-V curves and fits can be seen in the Zenodo repository<sup>25</sup>. As previously observed by other groups, a fairly linear decrease is obtained when the negative F-N slope is plotted as a function of the F-N intercept. The linear fit of this SK plot has a coefficient of determination  $R^2 = 0.9855$ . As it is a little confusing to talk about the slope and intercept of one graph in relation to the slope and intercept of another graph, we will call the coefficients extracted from the linear fit of an SK plot meta-slope and meta-intercept. The meta-slope is  $-1693 \pm 72$  V and the meta-intercept is  $-68578 \pm 1212$  V. Each data point was labeled with a number corresponding to the order in which the measurements were taken. As consecutive measurements jump randomly on the linear curve, it seems that no clear trend emerges regarding the evolution of the emitter. Furthermore, the meta-slope and meta-intercept are not universal values as they differ when the overall emission characteristics are different as can be seen in the literature.

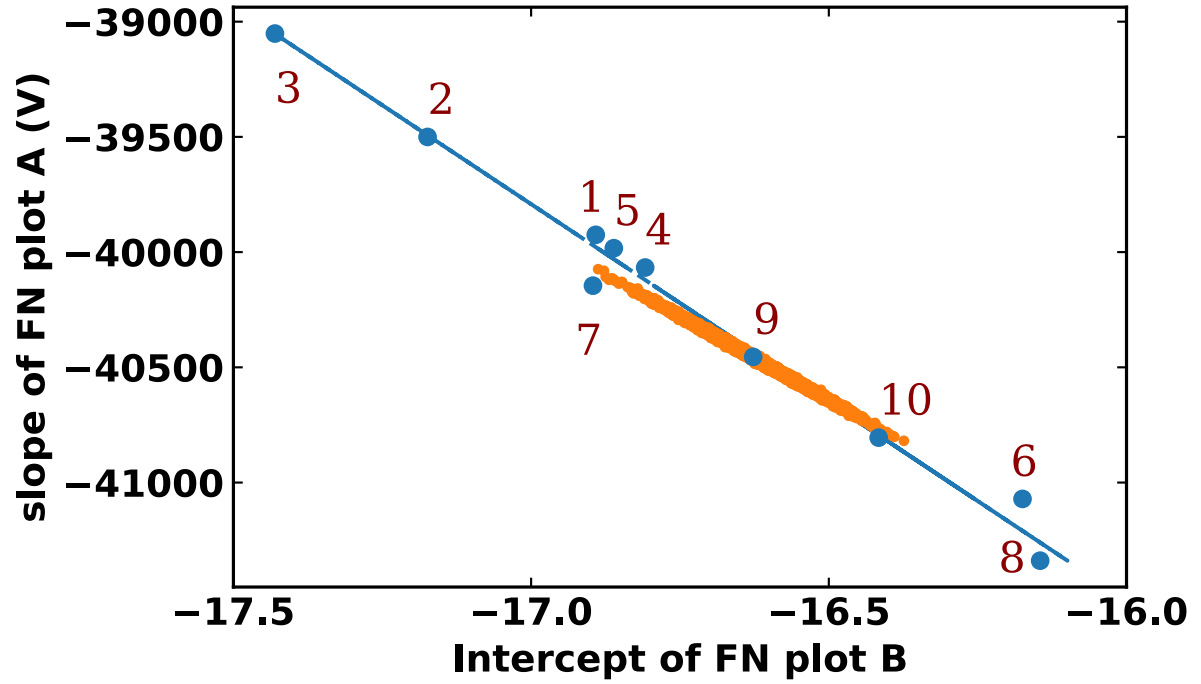


FIG. 1. Evolution of the F-N slope as a function of the F-N intercept in a standard SK plot representation for a W field emitter. The number close to each data point corresponds to the chronological order of the measurements. The solid line is a fit of the data point. The cloud of points around the data point number 9 is from numerical simulations

#### IV. DISCUSSION

Two conclusions can be drawn from these facts: i) there is a correlation between the F-N slope and the F-N intercept, ii) this correlation depends somehow on the "average" properties of the emitter. A closer look at Eq. 1 suggests that loosely speaking, on average the F-N relationship can be expressed as :

$$\langle A(t) \rangle_t = V_c \log \frac{I_c}{V_c^2} - V_c \langle B(t) \rangle_t \quad (2)$$

where  $t$  is the time where the I-V curves has been performed,  $\langle A(t) \rangle_t$  and  $\langle B(t) \rangle_t$  are the average F-N slope and intercept,  $V_c$  and  $I_c$  are the voltage and current corresponding to the abscissa and the ordinate of the middle of the data range in the F-N plot. Moreover, the fluctuations of the F-N slope and intercept can be expressed as :

$$\delta A = -V_c \delta B \quad (3)$$

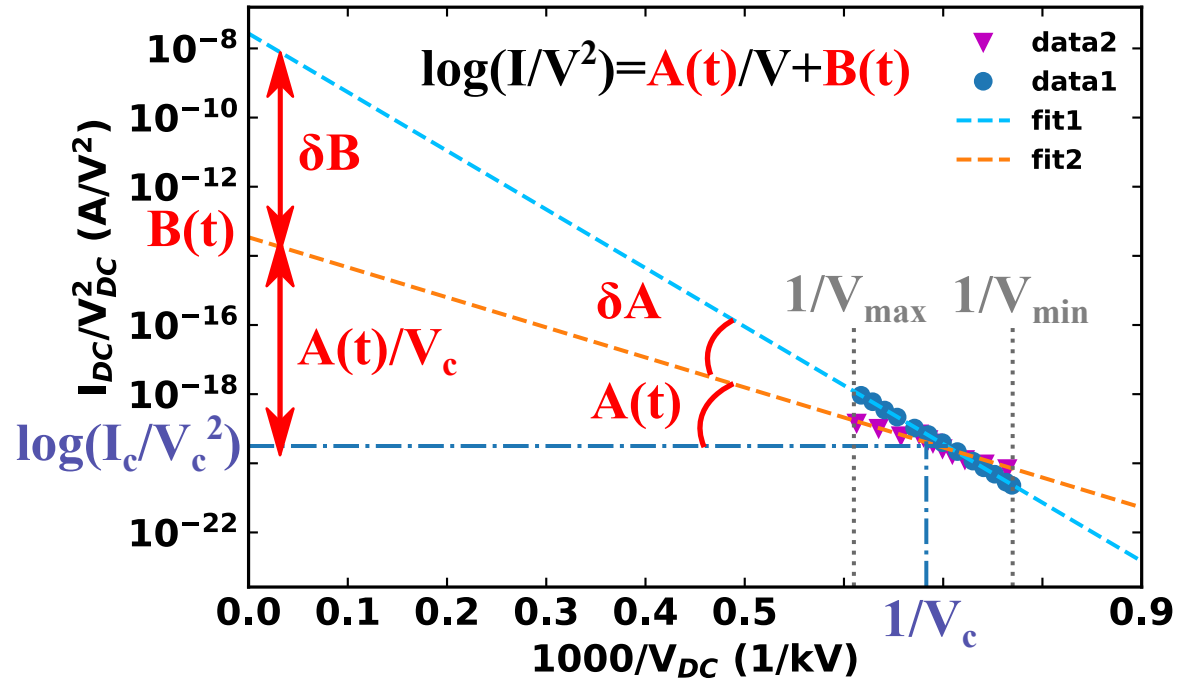


FIG. 2. Illustration of the cantilever effect of an F-N plot proposed in the text. data1 corresponds to the 3<sup>rd</sup> I-V curve with fit1 its linear fit. data2 and fit2 are fictitious data points for illustration purposes.

As long as the fluctuations of  $V_c$  and  $I_c$  can be neglected. Then

$$A(t) = \langle A(t) \rangle_t + \delta A = V_c \log \frac{I_c}{V_c^2} - V_c (\langle B(t) \rangle_t + \delta B(t)) = V_c \log \frac{I_c}{V_c^2} - V_c B(t) \quad (4)$$

For the readers skeptical about this not-so-mathematical derivation, this relationship also has a geometrical interpretation (see Fig.2). Fluctuations of the F-N intercept depend both on fluctuations of the coordinates of the tangent point to the F-N curve ( $1/V_c, \log \frac{I_c}{V_c^2}$ ) and on fluctuations of the F-N slope  $A(t)/V_c$ . If the intercept is far from the tangent point, a cantilever effect amplifies the contribution from slope fluctuations. If the coordinates of the tangent point do not fluctuate too much, the contribution from slope fluctuations dominates. Fig.2 illustrates this cantilever effect. The points labelled "data1" are taken from experiments but the points corresponding to "data2" have been artificially separated from the "data1" points to make the fluctuations more apparent.

Eq. 4 can also be better derived from statistical analysis. This analysis is not specific to field emission and is valid for any case where a linear regression is used. Firstly, if there is a linear relationship between the slope and intercept of an F-N plot, then :

$$A(t) = p' + q'B(t) + \epsilon'_t \quad (5)$$



where  $p'$  and  $q'$  are constant and  $\varepsilon'_t$  is a noise of zero mean and constant standard deviation independent of  $t$ .  $p'$  is the meta-intercept and  $q'$  is the metaslope of the SK plot. Mathematically, the formula are simpler and exact if the roles of  $A(t)$  and  $B(t)$  are swapped and if we plot a "KS" chart instead of a SK chart. So a linear regression of

$$B(t) = p + qA(t) + \varepsilon_t \quad (6)$$

where  $p$  and  $q$  are constant and  $\varepsilon_t$  is a Gaussian noise of zero mean and constant standard deviation independent of  $t$  gives

$$q = \frac{\langle A(t)B(t) \rangle_t - \langle A(t) \rangle_t \langle B(t) \rangle_t}{\langle A(t)^2 \rangle_t - \langle A(t) \rangle_t^2} \quad (7)$$

In our experiments  $t$  takes  $N = 10$  different discrete values  $t_i$  and the average can be explicitly written, for instance for  $A(t)$ :

$$\langle A(t) \rangle_t = \frac{1}{N} \sum_{i=1}^N A(t_i) \quad (8)$$

It can be noticed that in Eq. 7 the numerator correspond to the covariance of  $A(t)$  and  $B(t)$ , and the denominator is the variance of  $A(t)$ . As  $A(t)$  and  $B(t)$  are also given by regression theory, their variance and covariance can be estimated thanks to the covariance matrix. For an I-V curve performed at a time  $t_i$ ,  $n = 33$  current data points  $I_{i,j}$  are measured for each applied  $V_{i,j}$  and according to Eq. 1 we have :

$$\log \frac{I_{i,j}}{V_{i,j}^2} = \frac{A(t_i)}{V_{i,j}} + B(t_i) + \varepsilon_{i,j} \quad (9)$$

where  $\varepsilon_{i,j}$  is a Gaussian noise of zero mean and constant standard deviation  $\sigma$  independent of  $i$  and  $j$ . Then the covariant matrix of the linear fit is given by :

$$\begin{bmatrix} \text{var}(B(t_i)) & \text{covar}(A(t_i)B(t_i)) \\ \text{covar}(A(t_i)B(t_i)) & \text{var}(A(t_i)) \end{bmatrix} = \frac{\sigma}{n(\langle \frac{1}{V_{i,j}^2} \rangle_j - \langle \frac{1}{V_{i,j}} \rangle_j^2)} \begin{bmatrix} \langle \frac{1}{V_{i,j}^2} \rangle_j & - \langle \frac{1}{V_{i,j}} \rangle_j \\ - \langle \frac{1}{V_{i,j}} \rangle_j & 1 \end{bmatrix} \quad (10)$$

where  $cov$  is the covariance of two variables,  $var$  is the variance,  $\langle \frac{1}{V_{i,j}} \rangle_j$  is the average over the different values of the voltage in a single run:

$$\langle \frac{1}{V_{i,j}} \rangle_j = \frac{1}{n} \sum_{j=1}^n \frac{1}{V_{i,j}} \quad (11)$$

This latter term is independent of the run  $i$  as the voltage steps where the same for each run and is by definition equal to the term  $1/V_c$  introduced above. Then the correlation of the slope and intercept estimates is given by the non-diagonal terms of the variance-covariance matrix :

$$\text{cov}(A(t), B(t)) = -\text{var}(A(t)) \frac{1}{V_c} \quad (12)$$



Thus, from Eq. 7,  $q$  is equal to  $-\frac{1}{V_c}$  as claimed in Eq. 2. Furthermore, Eq. 12 clearly shows that even without any physical correlation, the slope and intercept are statistically correlated, if the inverse of the applied voltages are not well distributed around zero. In a field emission experiment, it is impossible to cancel this correlation, as it would require applying infinite voltages. For our data, the normalized correlation of the slope and intercept estimates is :

$$\left\| \frac{\text{cov}(A(t), B(t))}{\sqrt{\text{var}(A(t))\text{var}(B(t))}} \right\| = \frac{\langle \frac{1}{V_{i,j}} \rangle_j}{\sqrt{\langle \frac{1}{V_{i,j}^2} \rangle_j}} \approx 0.9979 \quad (13)$$

This value is close to one indicating that any fluctuation in the determination of the slope will have a strong impact on the intercept.

Secondly,  $p$  can be obtained by averaging Eq. 6 :

$$\langle B(t) \rangle_t = p + q \langle A(t) \rangle_t \quad (14)$$

and Eq. 9:

$$\langle \log \frac{I_{i,j}}{V_{i,j}^2} \rangle_j = \frac{A(t_i)}{V_c} + B(t_i) \quad (15)$$

The term on the left hand side can be considered as independent of the I-V run if the emitter is not too unstable and equal to  $\log \frac{I_c}{V_c^2}$ . Then Eq. 15 can also be averaged to give :

$$\log \frac{I_c}{V_c^2} = \langle A(t) \rangle_t \frac{1}{V_c} + \langle B(t) \rangle_t \quad (16)$$

and thus  $p = \log \frac{I_c}{V_c^2}$  as expected.

So the linear relationship in an SK plot can be explained on statistical grounds. This is why numerical simulations in ref. 19 were able to reproduce it. From the extracted F-N parameters of the 9<sup>th</sup> I-V curve, numerical simulations were performed to generate 10,000 I-V curves with a multiplicative Gaussian current noise of a similar amplitude to the experimental conditions. The field emission current was obtained by integrating numerically the Murphy-Good equation<sup>2</sup>. A model with a constant emission area was assumed in this calculation for the sake of simplicity. It might be interesting to repeat this simulations, taking into account the voltage dependence of the emission zone, but we do not expect this will affect our conclusion. This assumption was not made elsewhere in the article. The experimental long-term current drift was not included in the simulations. These curves were then plotted in F-N coordinates and fitted linearly. The resulting slopes and intercepts are represented as a cloud of points in Fig. 1. The shape of this cloud is



consistent with the solid line corresponding to the fit over the 10 experimental I-V curves. It can be noticed that the slope of the cloud is tilted compared to the solid line. The two slopes differ by less than 18 %. A better agreement might be obtained by taking into account the current drift. It would require characterizing the average drift during an I-V curve and adding this drift to the simulations. Since the estimate of the drift current is fairly imprecise, we preferred to keep the modeling simpler.

From this analysis and Eq.2, it can be predicted that in an SK plot, the meta-slope should be equal to  $-V_c$  and the meta-intercept to  $V_c \log \frac{I_c}{V_c^2}$ . For our data on a single tungsten tip, the predicted meta-intercept is -64650 V corresponding to a discrepancy of 6 % compared to the meta-intercept fitted above of -68578 V ( $\pm 2$  % uncertainty). The predicted meta-slope is -1459 V which corresponds to a deviation of 14 % from the meta-slope fitted above of -1693 V ( $\pm 4$  % uncertainty). This simple model can therefore reasonably predict more than 85 % of the meta slope and intercept values. More importantly, an analysis of the published literature on SK plots shows a good agreement between our model and reported data. All results from SK plot papers in which  $V_c$ ,  $I_c$ , the meta slope and intercepts could be reliably extracted have been plotted in Fig.3. It shows that the experimental data are close to the predicted theoretical values over two orders of magnitude. So, it is likely that all the SK plots in the literature have been plots representing the F-N slope as a function of the F-N slope. In some sense, the SK plot is like a Lissajous curve where the most interesting pattern is not a straight line. The main information encoded in an SK plot is the "average" voltage and current ranges explored in the experiment. A deviation from this straight line may indicate a strong modification of the emitter.

This result is not really surprising, as it lies at the heart of the Legendre transform concept where the slope and intercept can replace the coordinates of the crossing point of a curve (i.e. the F-N plot) and its tangent (i.e its fit) without any loss of information (Ref. 33 p.140). Furthermore, this is not the first time that the idea of exploiting the correlation between the slope and intercept of a linear curve has been proposed. For instance, in biology or chemistry, this is known as entropy-enthalpy compensation and most of the reports seem spurious<sup>34-36</sup>.

## V. CONCLUSION

In conclusion, we have demonstrated that in field emission, the linear relationship between the F-N slope and intercept, observed in a so-called SK plot, is probably an artifact. Our experiments

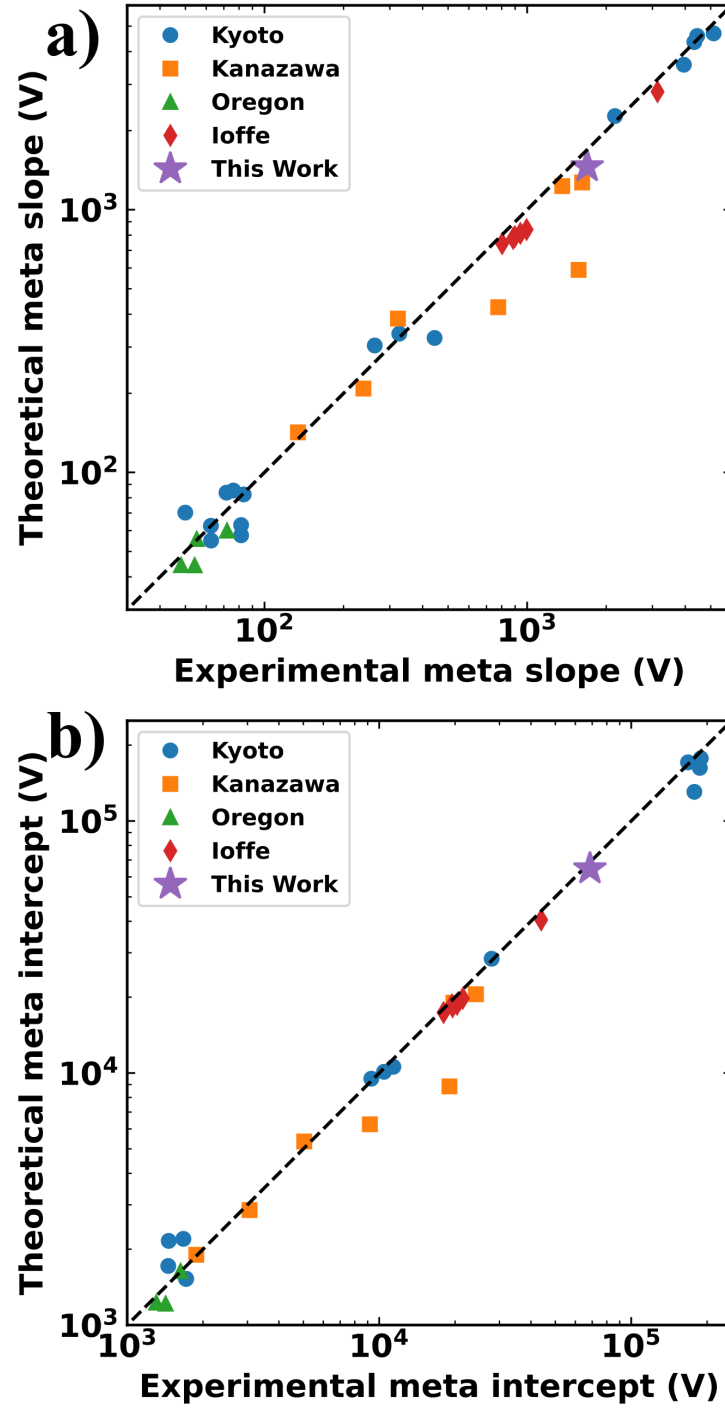


FIG. 3. a) and b) log-log plot of the SK plot meta-slope and meta-intercept as a function of the theoretically predicted slope and intercept. The data from Kyoto University have been extracted from ref. 12, 16, 26–29. The data from Kanazawa Institute of Technology have been extracted from ref. 13. The data from Oregon have been extracted from ref.30 and 31. For this particular set of data, this group analyzed their data with a modified F-N plot. The data from Ioffe Institute have been extracted from ref.20 and 32. The dashed line is a guide for the eye corresponding to the coordinates where the abscissa equals the ordinate.

are in agreement with a statistical interpretation of the origin of the straight line on an SK plot. We have tried to be as exhaustive as possible in analyzing the field emission literature and have shown that the correlation observed by other groups can be explained by our simple model. Since not all data were usable due to the lack of complete information in some articles, it would be interesting if some groups could reanalyze their data and show whether there is an exception to this explanation.

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## CONFLICT OF INTEREST

The authors have no conflicts to disclose.

## DATA AVAILABILITY

The data that support the findings of this study are openly available in Zenodo at <https://doi.org/10.5281/zenodo.11199621>, Ref. 25

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