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Contributions to the theory of Scandate cathode emission

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Outline:

1. Introduction: Semiconducting features of Scandate cathodes

2. High resolution characterization results [1,2]

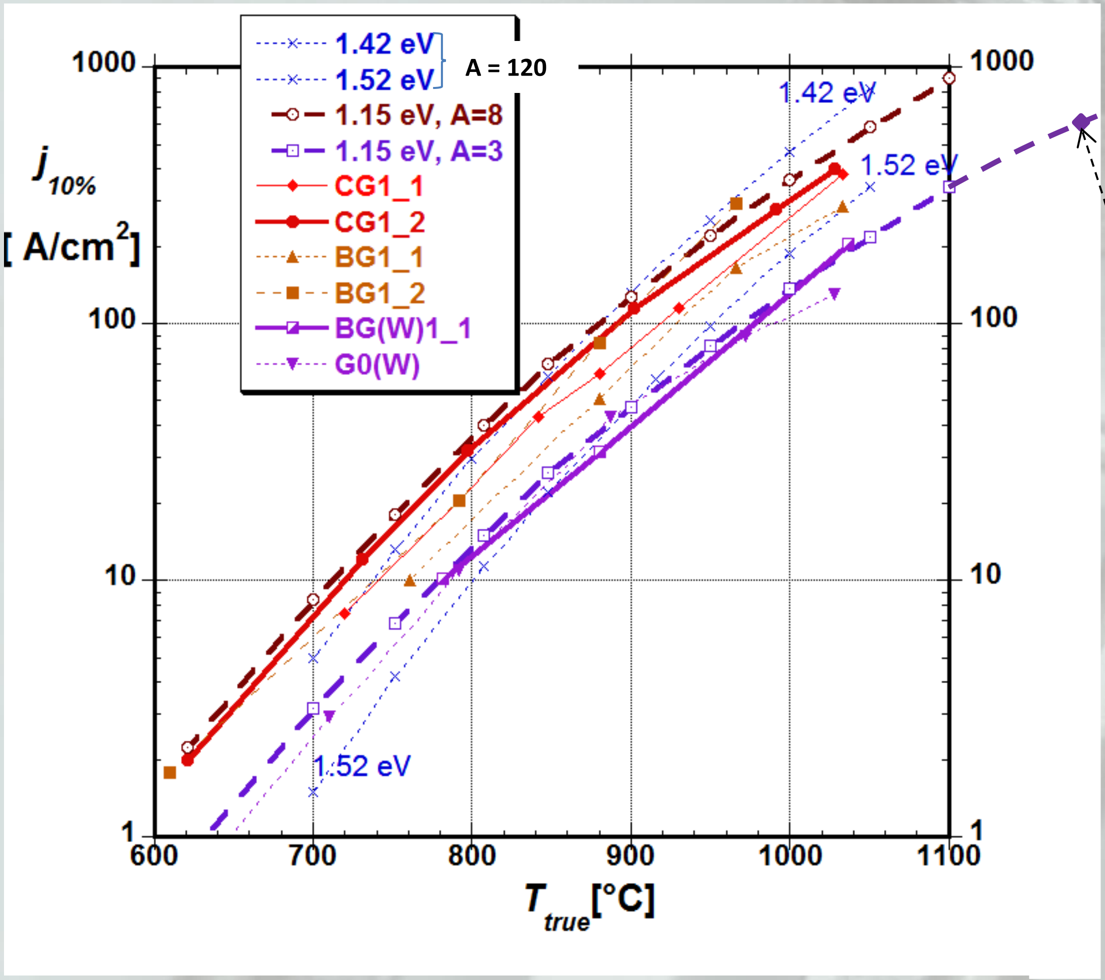
3. Current semiconductor theory of oxide and Scandate Cathodes

4. Emission explained by localized emissive oxide nanoparticles (or µm-sized spots/pores) and the beamlet effect (Gaertner, [8])

5. Thin film semiconductor emission by strong band bending and short Debye length

6. Conclusions and outlook

1a. Semiconducting features of Scandate cathodes: Low WF, low A_R



Saturation emission current density $j_s \approx j_{10\%}$ versus true temperature of LAD top-layer Scandate cathodes (see [1,3])

Red and orange curves: LAD with Re (on W)

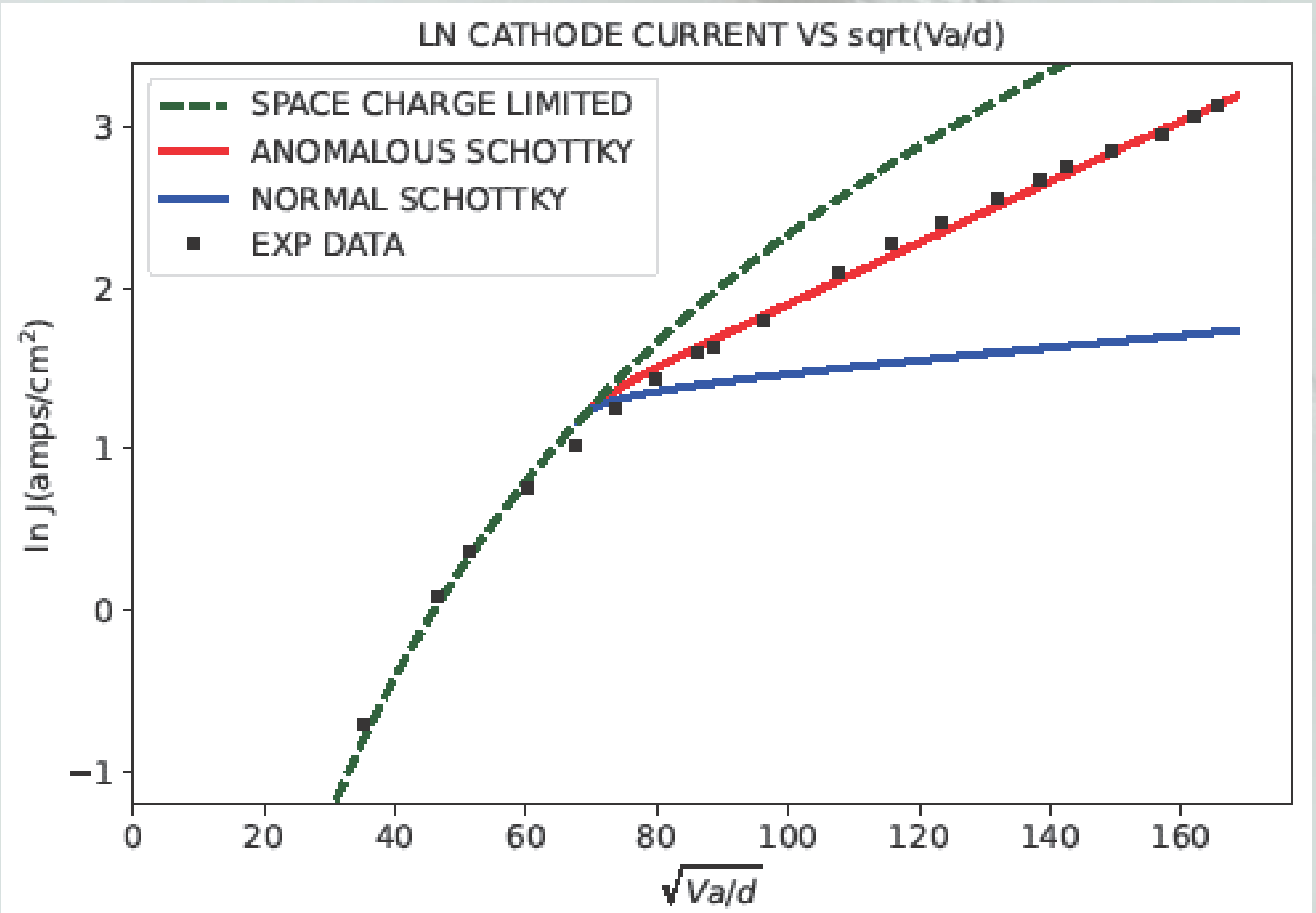
Violet curves: LAD with W

The violet (check) data point outside the scale is the top result of Shengyi Yin from 2020, compatible with W base and A_R = 3 at about 1190 °C true temperature

A general feature of all Ba-scandate cathodes is the **work function** of about **1.15 eV;** the **Richardson constant A_R is varying between 3 and 8 A/(cm²K²)**. For comparison j_s curves for work functions of 1,52 eV and 1,42 eV with A = 120,4 is also shown.

This is similar to oxide cathodes, which also exhibit low WF and low A_R

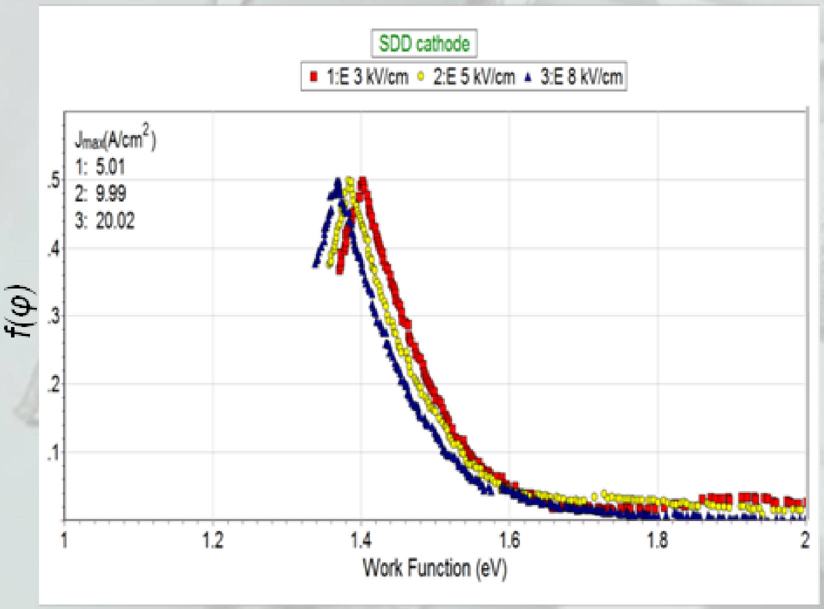
1b. Semiconducting features of Scandate cathodes: Anomalous Schottky Plot

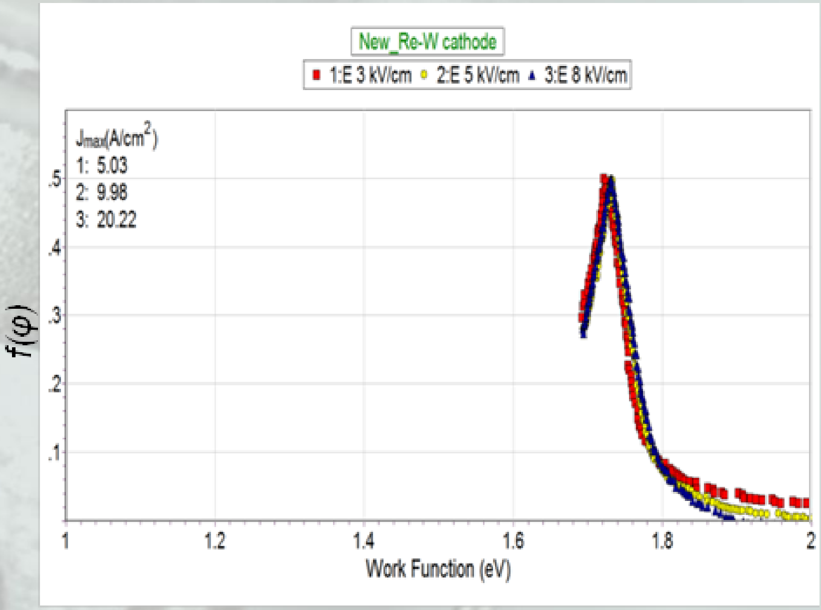


Schottky Plot N5401K1, from [1], d_{ca} = 513 µm, T = 1259 K

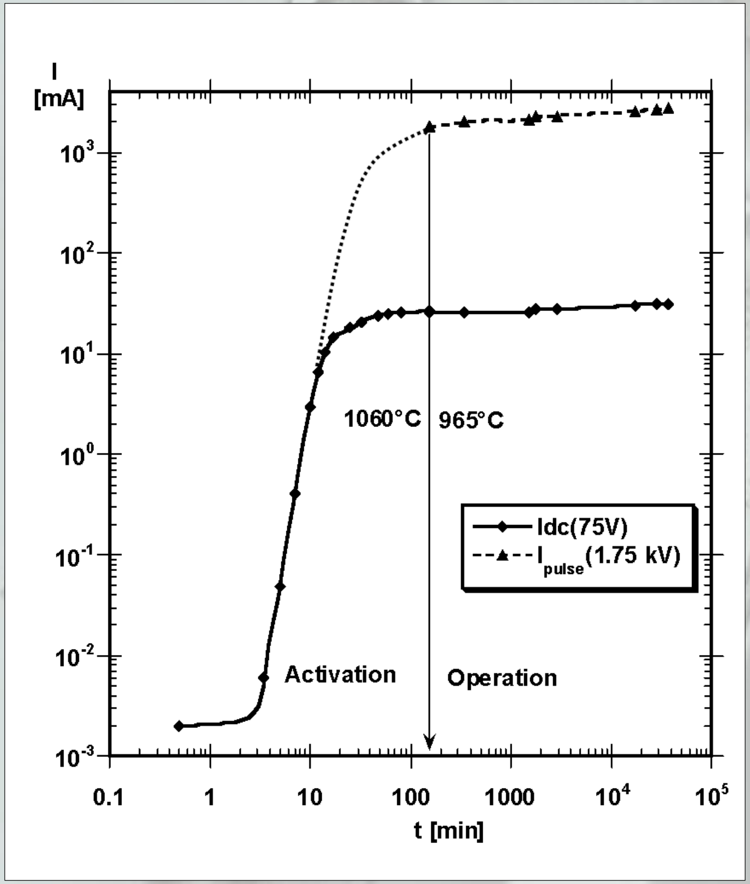
1c. Semiconducting features of Scandate cathodes:

The peak work function in an PWFD or Miram Plot derived from roll-off or underheating curves is dependent on the electric field (plot left), typical for a semiconductor, which is not the case for Ba dispenser (I) cathodes (right side)

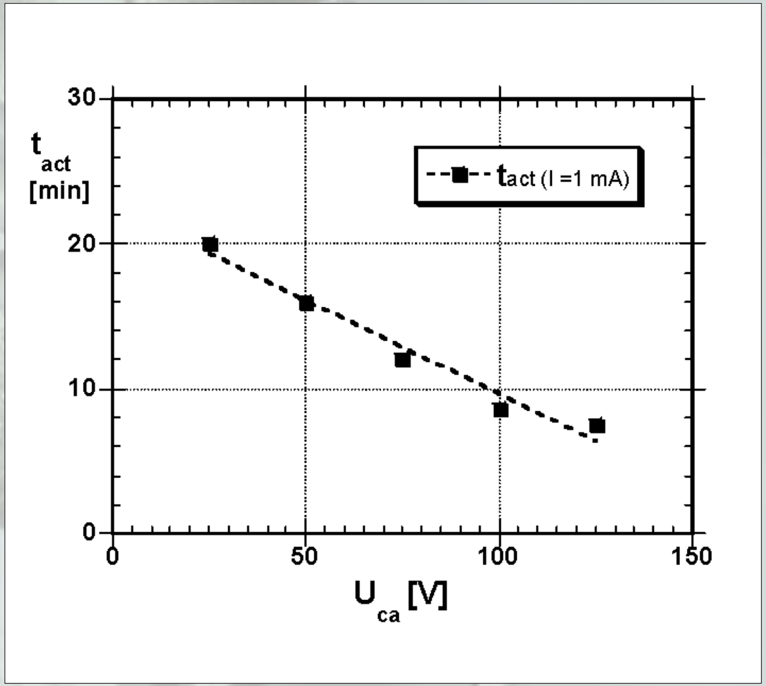




1d. Semiconducting features of Scandate cathodes: Faster activation with increasing electric field



Activation and operation of LAD scandate cathode at an average temperature of about 1060 $^{o}C_{Mo\text{-}Br.}$ with constant electric field of 75 V/ 0.25 mm. Shown is current in log scale, dc or pulsed, versus time in log scale [3]. At t=27 days: j_{dc}(75V)= 3.8 A/cm^2 ; j_{pulse}(1,75kV) = 320 A/cm^2.



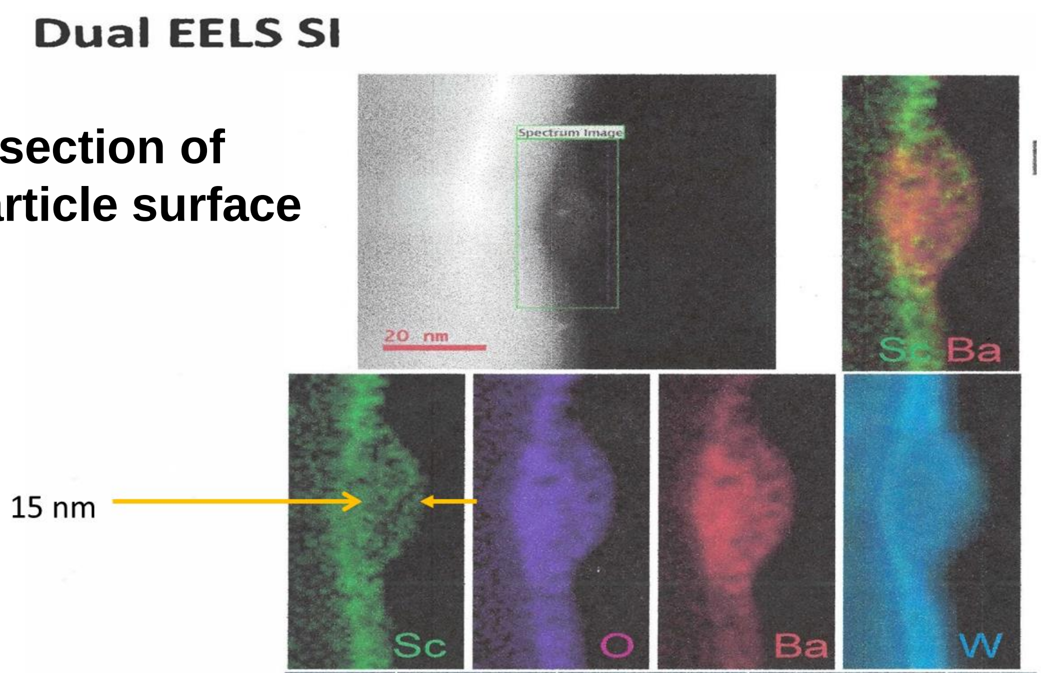
Activation time (delay) to dc current level of 1 mA at a temperature of about 1060°C for LAD scandate cathodes as a function of applied voltage (d_{CA} = 0.25 mm) [3]

Besides the above semiconductor features scandate cathodes also exhibit **features of metal matrix Ba dispenser cathodes** :

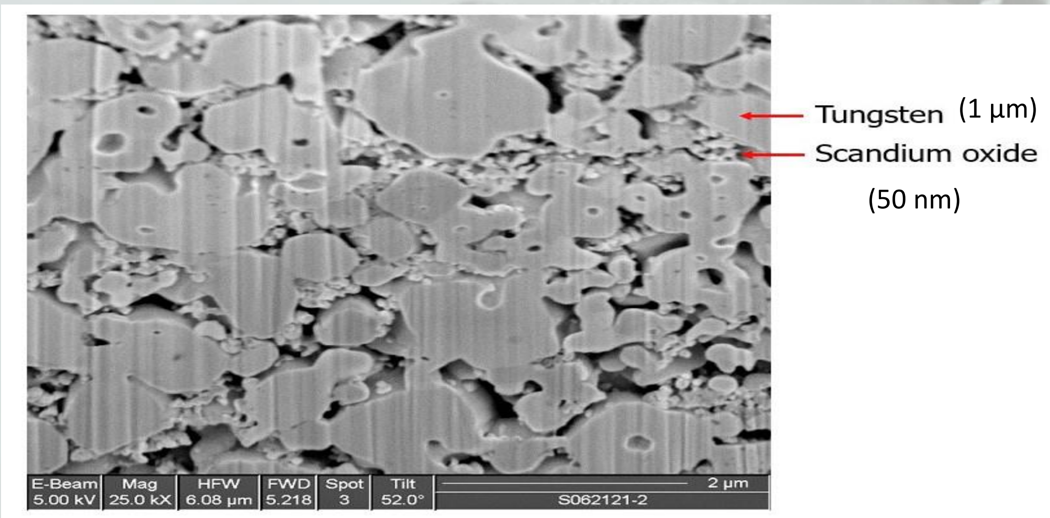
- they are insensitive to duty factor = no conductivity limitation as for oxide cathodes
- only very thin semiconducting layer or semiconducting particles on the cathode surface

2. High resolution characterization results [1,2]

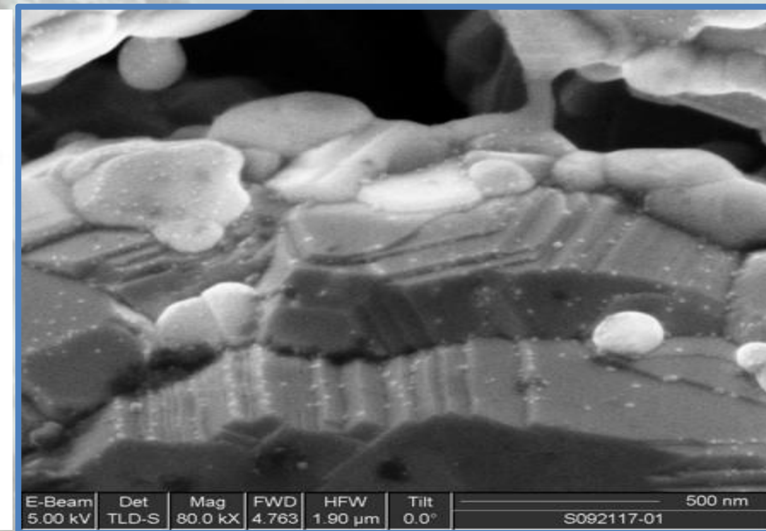
TEM Cross-section of tungsten particle surface



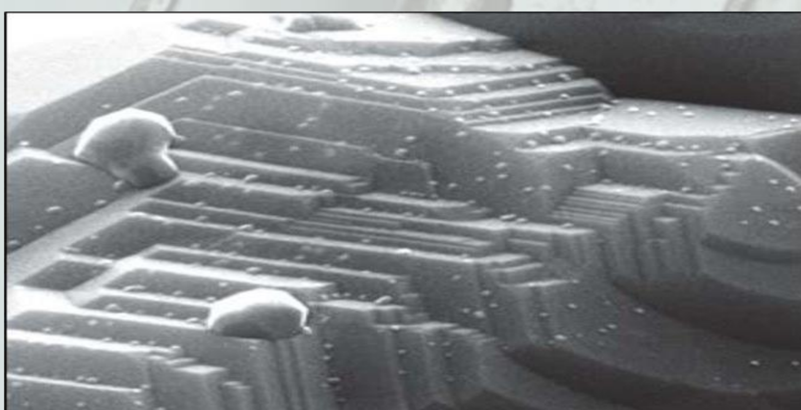
2.b High resolution characterization results [1,2]



SEM of Sintered Matrix Cross-section



Cathode Surface, Activated Cathode, e beam

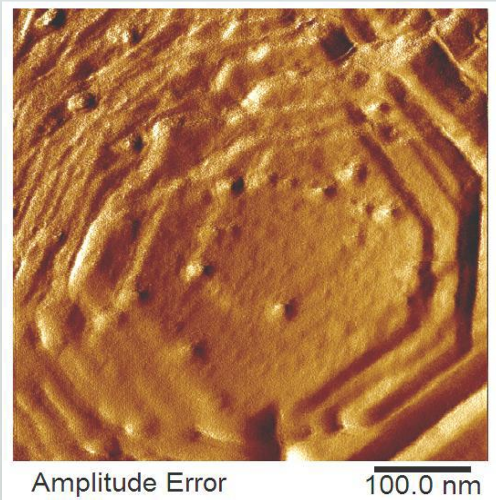


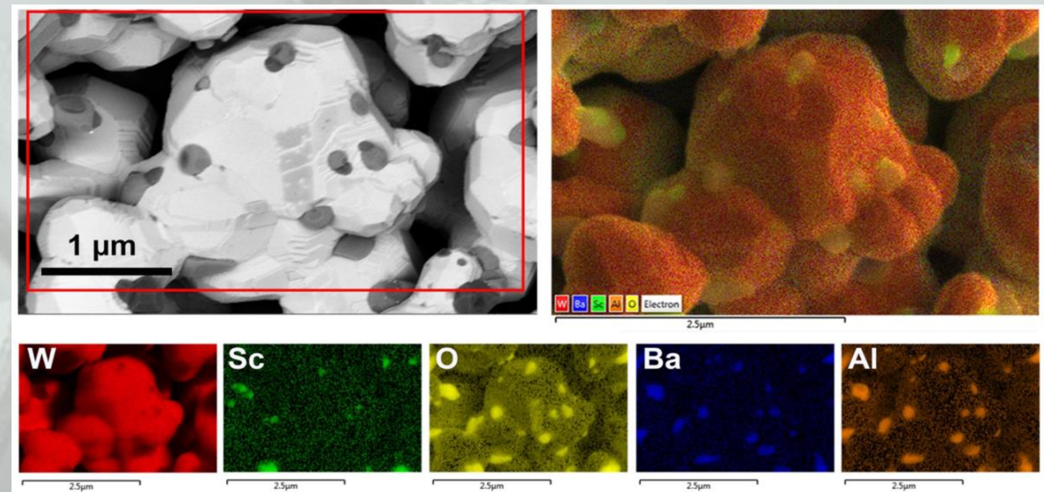
Cathode Surface, BJUT

100 nm

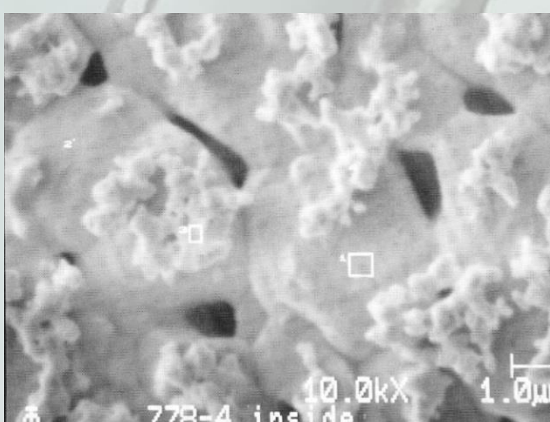
The preparation of SDD scandate cathodes is based on a mixture of **W particles** of about **1 µm** average diameter with **scandia particles** of about **50 nm** average diameter, what can be seen on the SEM micrograph matrix cross section (upper left).
After activation very fine particles of about **20 nm** can be seen on the surface of facetted W grains.
The dual EELS SI (electron energy loss spectra) of a TEM sample of previous slide show an about **6 nm surface layer of Sc, Ba, O** (and W) on W and a **15 nm particle** of the same composition, which is in clear contradiction to a Debye length of 1,5 µm!

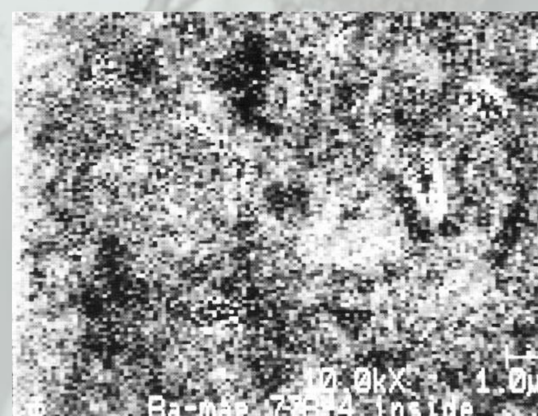
2.c Surface analysis results for Ba scandate dispenser cathodes

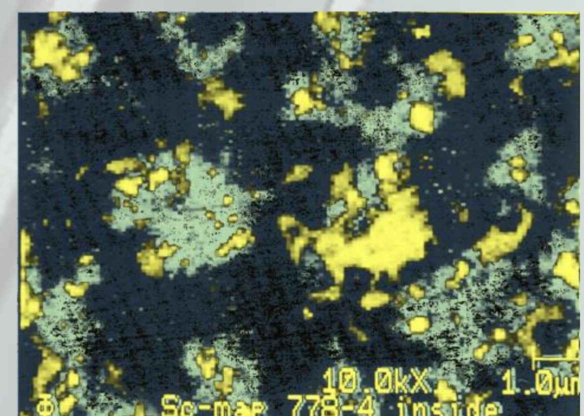




SDD Cathode (Y. Wang et al., J. Balk, B. Vancil et al., [4]); SFM (left) , SEM (center) and SAM micrographs; separate Sc containing nanoparticles!



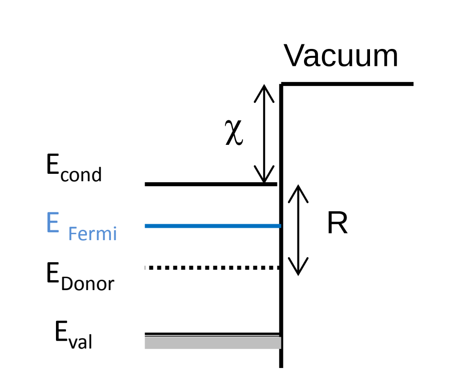




a) SEM micrograph (above) b) Ba SAM mapping c) SAM : Re (green) & Sc (yellow)

of **LAD top layer Scandate cathode** in the central beam emitting region **after ion bombardment**; the cathode was re-activated for 30 min at 1060°C $_{\text{Mo-Br.}}$ and then kept at 965°C $_{\text{Mo-Br.}}$ during SAM [4]. The structures are much larger than on SDD cathodes

3. Current semiconductor theory of oxide and Scandate Cathodes



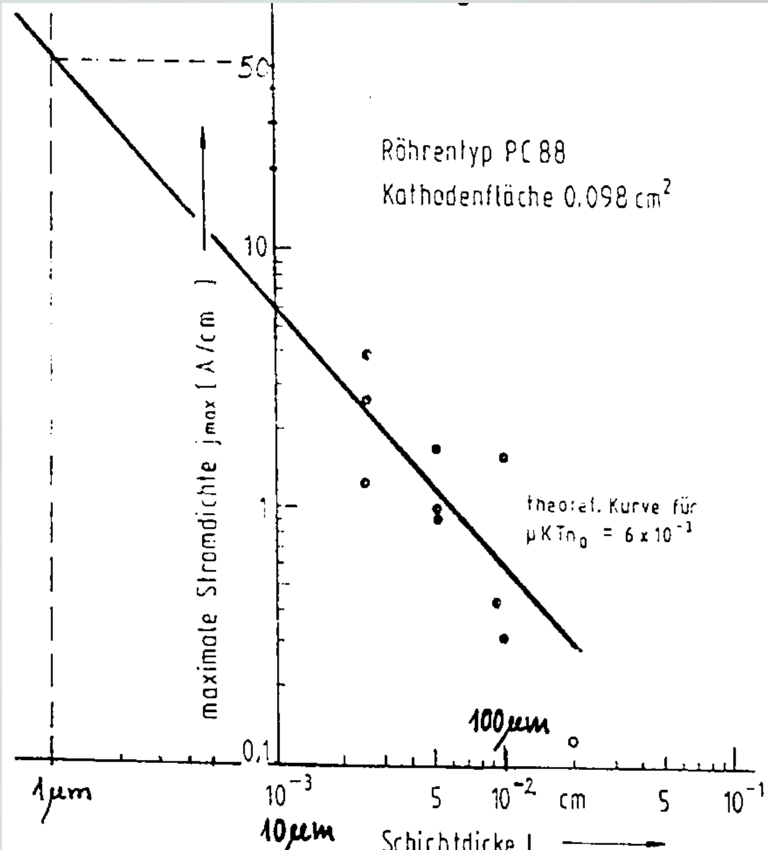
Schematic energy levels of an oxide / semiconductor cathode; E_{val} is the upper edge of the valence band, E_{cond} is the lower edge of the (normally empty) conduction band and R is the energy difference between E_{cond} and the donor level. The **work function Φ** is given by
$\Phi = R/2 + \chi = 1.4$ eV (1), where χ is the electron affinity.
For Ba-oxide cathodes R/2 = 0.7 eV and χ = 0.7 eV

The nature of the mobile **donors** was claimed to be **oxygen vacancies in BaO**. At the surface of the semiconductor **band bending** occurs, depending on charge separation. It is increased by application of an external extracting field.

The screening of the electric field and the band bending curvature is described by the **Debye length $\lambda_D = (\varepsilon \varepsilon_0 kT / (e^2 n_i))^{1/2}$ (2),**
where n_i is the density of ionized donors, T the cathode temperature and $\varepsilon \varepsilon_0$ the dielectric constant of the semiconductor. In a **well activated** oxide cathode e.g. $n_i = 3.9 * 10^{15}$ cm^{-3} and λ_D = 130 nm [9].

It is also expected, that the electropositive Ba monolayer (positive charge at surface) extending over the surface, leads to a subtractive dipole moment lowering the work function.

3b. Current semiconductor theory of oxide and Scandate Cathodes



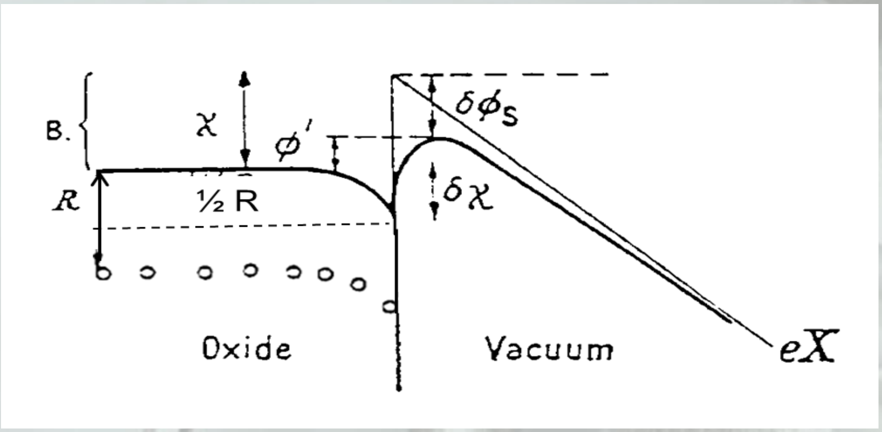
The graph of K. Tischer [10] extrapolated to lower oxide thickness → transition to metallic behaviour!

Since BaO(.SrO.CaO) cathodes are semiconducting, their dc current is limited. There exists a **maximum dc current density**, which **increases with decreasing oxide thickness** d (in the graph: **l)**,

$$j_{max} = kT * n_0 * \mu_- / d \quad (3),$$

where $\mathbf{n_0}$ is the average density of the donors, $\boldsymbol{\mu_-}$ is the mobility of the electrons. Typically oxide cathode coatings have thicknesses of about 75 µm, whereas the Sc and O containing top-layer of scandate cathodes amounts to 20 – 300 nm on the base metal, and hence there the dc current density limitation at d = 300 nm increases to 100 A/cm^2 and still gets higher for thinner layers.

3c. Current semiconductor theory of oxide and Scandate Cathodes



Schematic energy levels in an oxide coating according to Wright and Woods with external field, when interior space-charge/band bending and Schottky effects are both taken into account, and the voltage drop in the coating is neglected. With applied field eX, the work function is modified to $\Phi' = \Phi - \delta\chi - \delta\Phi_s$; the open circles are the donors. Field penetration will tilt the internal levels.

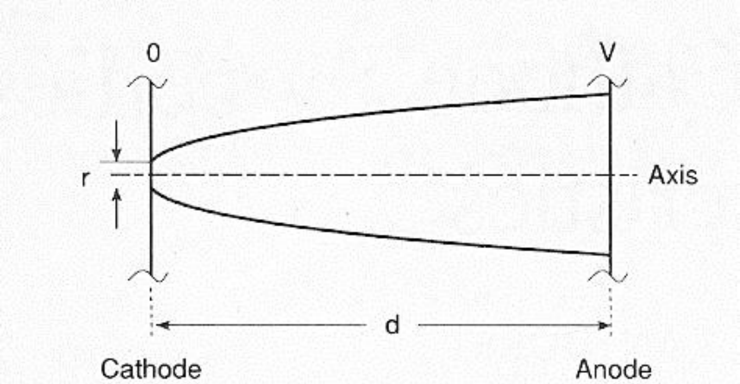
$\delta\chi_0$ **= 2kT* sinh^{-1}[(I_{sat}/σ) / (4(2 ε_{ox} π n_0 kT)^{½})]** at eX = 0 (4a) and

$\delta\chi$ **= - 2kT* sinh^{-1}[X/ (4*(2 ε_{ox} π n_0 kT)^{½})]** for I > I_{sat} and $\delta\chi_0$ < 0.01 eV (4b)

It has to be noted, that the formula (4b) (see Wright [4]) explains the anomalous Schottky effect in in the I/U characteristic of semiconducting cathodes, but does not give a **quantitative model** of the low work function, which is determined at the transition point (zero field) between space charge limited emission and saturation. Yet the Debye length can be used for determination of some of the parameters.

It is instructive to know, that Rare earth ions, such as Sc ions Sc^{2+}_{Ba} on Ba-sites can become electron donors according to:
Sc^{2+}_{Ba} → Sc^{3+}_{Ba} + e^- (5), which is also important for scandate cathodes.

4.a Emission explained by localized emissive oxide particles and the beamlet effect (J. Hasker, 1985)



Application to scandate cathodes:
In the highly emitting submicron size patches beamlets originate. The cathode emission consists of a bundle of beamlets or micro-beams.

Model: Planar diode with one circular patch having space-charge limited emission [5];
With current density in the beamlet j_b
and Child-Langmuir current density j_{CH}

→ $j_b = (d_{ca}/r_{sp})^{1/2} * j_{CH}$ (6)

(based on scaling laws for Poisson equation)

Example Ba diffusion from the pores: If λ is the diffusion length → Current
$I = \pi * \lambda^2 * j$ (7)
See also J. Hasker, N. van Hijningen, Appl. Surf. Sci. 24, (1985), 318 – 329 [6]

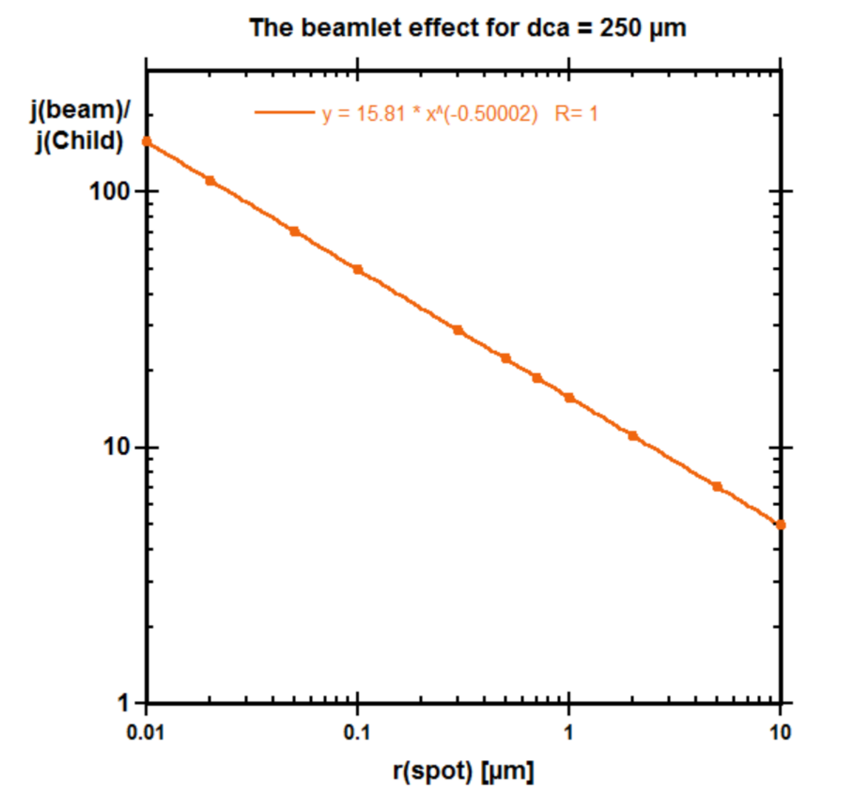
The current density in the beamlet or micro beam at the spot-surface is much higher than expected from the Child-Langmuir space charge law!
E.g. for d_{ca} = 250 µm and r_{sp} = 100 nm
$j_b = 50 * j_{Ch}$
An experimental confirmation of the beamlet effect was obtained by Free and Gibson [7] for I cathodes by measuring the Ba diffusion length from AES Ba maps.

4.b The beamlet effect (J. Hasker) and scaling of Poisson equation

$$\frac{\partial^2 V}{\partial x^2} + \frac{\partial^2 V}{\partial y^2} + \frac{\partial^2 V}{\partial z^2} = -\frac{\rho}{\epsilon_0}, \qquad (8)$$

Eventually a solution of the **Poisson equation** in and around the patch is not available and hence Hasker [6] derived equation (6) as an approximate result from the **scaling laws of Poisson equation**.

Scaling of Poisson equation means, that when multiplying all space dimensions with n, all current densities must be divided by n^2. If all potentials are multiplied with a factor f_p, all current densities must be multiplied by a factor $f_p^{3/2}$, in order to obtain solutions again. From these conditions Hasker derived formula (6), see [5,6].



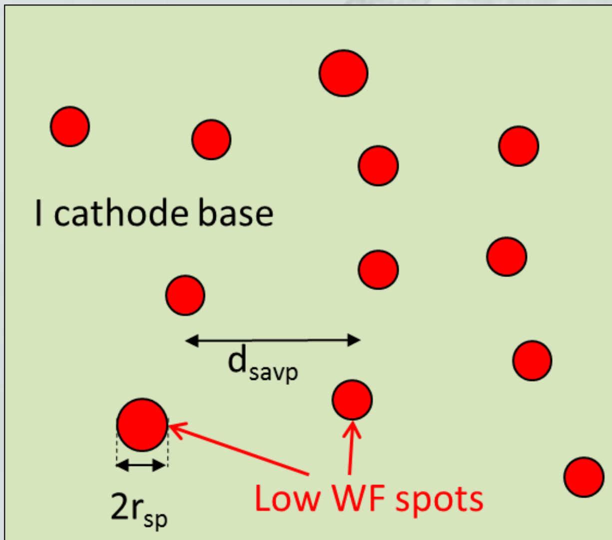
A qualitative explanation of the beamlet effect is, that for small spots space charge in front can expand into the surroundings and hence the space charge barrier is reduced. This also implies a spreading of the beamlet.

4.c The beamlet effect implies an **expansion of the electron beam and hence of the spot by the factor $(d_{ca}/r_{sp})^{1/2}$**. Since from characterization the spot is a particle half sphere, the expansion is not only related to the radial dimension, but also to the beam direction and the particle depth.
This **electrical image expansion** can explain the high Debye length of 1,5 µm in the fit of Caplan as it would occur for a uniformly emitting surface in a diode geometry. From the preceding figure, but replacing **d_{ca}** = 513 µm and for a **spot of r_{sp} = 0.013 µm (values of Vancil et al. [1])** **an expansion factor of 174 is obtained, which means an expansion of the spot to 2.3 µm**. Yet in Caplans paper $\lambda'_D = \lambda_D/\sqrt{2}$ = 1.5 µm was used, and hence his result is λ_D = 2.1 µm, **an apparent Debye length**, which is in good agreement with an apparent thickness of 2.6 µm. That a **real Debye length** in the order of e.g. 15 nm is possible can be seen by inserting the following values in the equation $\lambda_D = (\varepsilon_r \varepsilon_0 kT / (e^2 n_i))^{1/2}$:

n_i density of ionized donors : **n_i** = $3 \cdot 10^{17}$ cm^{-3} (compare Chin et al. [9]), ε_r(Ba-scandate) ≈10 and T = 1260 K → λ_D = 14.1 nm is obtained.

Another possible explanation of the high Debye length especially for **impregnated scandate cathodes as of Yin** could be attributed to **beamlet emission from the larger semiconductor filled pores**, which can play a role at least in the initial phase after activation. A decision between these model contributions can be obtained by **systematic variations of cathode particle sizes, emitting spot sizes and/or pore sizes and evaluation of the emission performance**.

4.d Model of Scandate cathode emission



Idealized map of local emitting spots

One of the problems is the theoretical explanation of the **total emission** and I-U characteristics arising **from** localized distributed **highly emitting patches.**
The **quantification** is determined by several parameters (see beamlet effect), including the **average distance** **$d_{avsp} = r_c/\sqrt{N_{sp}}$ (9)** of the spots. If the average distance is larger than a critical distance, the total current on the anode will be lower than expected from Child law for a uniformly emitting cathode.

This can be calculated from the spot expansion on the anode:
$d_{avsp} \geq r_{sp}\,{}^{*}(d_{ca}/r_{sp})^{1/2} = (d_{ca} * r_{sp})^{1/2}$ (10). The other question is, what happens if neighbouring micro-beams will overlap on the anode. Since the Child law defines the upper limit, the respective beam density on the anode will be reduced accordingly. The other case of increasing spot diameter is finally in a close spaced diode (**$d_{ca} < r_c$**) **limited to** a spot size of **$r_{sp} \leq d_{ca}$** (= no beam) and hence also **the validity of the Hasker formula** for beamlets. It has to be noted, that for this reason the formula (9) given by G. Gaertner in [8] has to be discarded. The beam expansion should of course depend on d_{ca}.

5. Thin film semiconductor emission by strong band bending and short Debye length

The general formulas (4a) and (4b) are also valid for this case, since the condition eVs >> kT was not used for derivation. We regard a uniform W surface coating with about 6 nm of (Sc,Ba,O) layer constituents.

That a **Debye length** in the order of 6 – 12 nm is possible can be seen by inserting the following values in the equation $\lambda_D = (\varepsilon_r \varepsilon_0 kT / (e^2 n_i))^{1/2}$:

$n_i = 1 \cdot 10^{18}$ **cm**$^{-3}$ (compare Chin et al. [9]), ε_r(Ba-scandate) **≈10** and **T = 1100 K →** λ_D **= 7.2 nm**, which means a high ionized donor density.

The knowledge of the relative dielectric constant of the (Ba,Sc,O) particle would be helpful. It was estimated based on the following data:

$\varepsilon_r(Ba_3Sc_4O_9) \approx 10$; $\varepsilon_r(Ba_2Sc_2O_5) \approx 20 - 50$; $\varepsilon_r(BaSc_2O_4) = 13$; $\varepsilon_r(Ba_3Sc_2WO_9) \approx 10$; $\varepsilon_r(BaO) = 34$; $\varepsilon_r(Sc_2O_3) = 14$ (Kwestroo [11]). It should be noted , that from the analysis also W is contained in the emissive particles!

Yet an uniform emission model of current Scandate cathodes with about 6nm surface layer is unlikely: 1. Because the fitted λ_D = 2.1 µm from the anomalous Schottky plot is in clear contradiction, and 2.: Due to the low work function of 1.15 eV and the low Richardson constant: With a uniform surface work function of 1.15 eV enormously high current densities would be achieved. Otherwise the model of Raju and Maloney with oxide thickness thinner than the Debye length (cut-off) could lead to some reduction of emission.

6. Conclusions and Outlook

B. Vancil et al. [1] showed by high resolution characterization, that the Sc rich layer on tungsten is very thin, in the order of 6 nm, with oxide particles in the order of 20 nm. Hence there is a clear contradiction between these very thin films and a Debye length λ_D of 2,1 µm as obtained from a fit of the Schottky I-U characteristic [1,2].

There are three possibilities to explain this discrepancy:

1. **Due to the electrical image expansion of the beamlets originating at distributed Ba-scandate particles (emissive spots) of about 15 – 20 nm diameter by a factor of 100 – 200 the apparent large Debye length can be explained.**
2. **The large Debye length can also be explained by (moderate) beamlet emission from the µm-sized semiconductor filled pores (or larger oxide spots) contributing to high electron emission.**
3. **Thin film uniform semiconductor emission based on the scaling of λ_D, which is decreasing with increased density of ionized donors n_i, can be in the range of the observed layer thickness, but cannot explain the large λ_D of Caplan and the low A_R.**

A decision between these models could be obtained by systematic variations of the relevant parameters.

References:

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[4] D. A. Wright, J. Woods (GEC): “The emission from oxide-coated cathodes in an electric field“, Proc. Phys. Soc. **65B**, 134 – 145 (1952) and D.A. Wright, Nature **164**, (1949), pp 714

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Thank you for your attention !

View over the river Rhine to the „Siebengebirge“ (Seven Mountains)