Single crystal CVD diamond membranes for betavoltaic cell

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Diamond Sensors Laboratory CEA-LIST

ADAMAS

Advanced Diamond Assemblies

Talk outline

Introduction

Motivation + Why Using Diamond

Diamond scCVD PIM Membranes Fabrication

Basic Diamond Betacell Parameters with an SEM

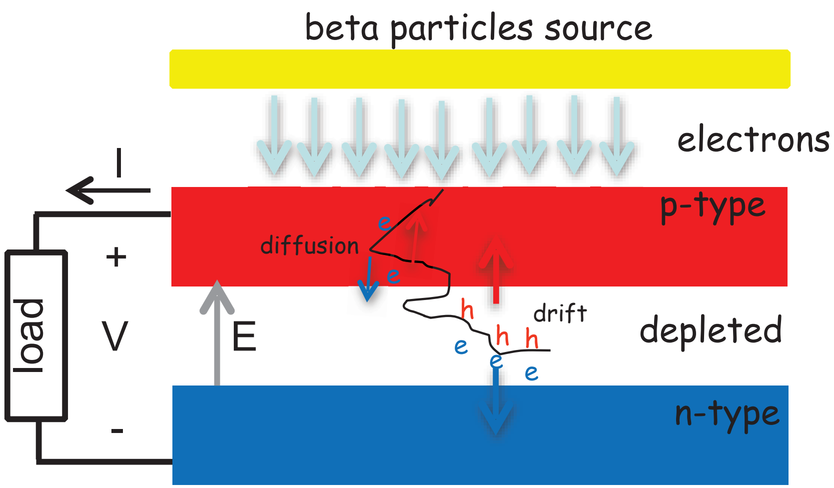
Summary and Outlook

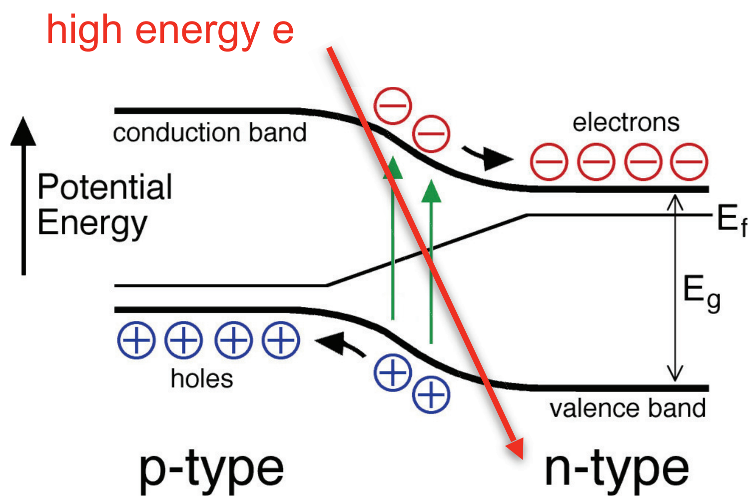
What is betavoltaics?

Similar principle as a solar cell

Generation of power by coupling a beta source to a semiconductor junction device

Separation and drift of the charges carriers in built in potential (typ. p-n junction or Schottky)





- low energy beta emitters are used
- short range of few microns

- interaction with inner shells electrons
- ionisation along the e track
- ionisation cascades (multiplication factor)

Why beta particles (electrons)?

some key parameters of betavoltaic

→ high energy density **[~ 131400 mWh/g; AA Li 300 mWh/g]**

→ long lifetime (no recharging, refuelling) **[≥ 20 years]**

→ low power **[nW to μW; AA Li ~1mW]**

→ can function in harsh remote environments (subsea, space)

→ if low E < 300 keV, no radiation damage, easy to shield

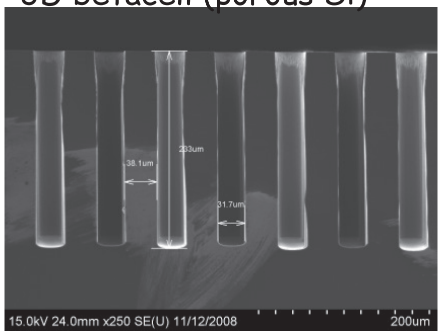
→ onboard MEMS/NEMS power sources **[microbatteries]**

'popular' betavoltaic emitters

Element	Half-Life (yr)	Specific Power (mW/g)	Avg. Beta E (keV)
$_{1}H^{3}$	12.3	0.9	5.7
$_{81}Tl^{204}$	3.78	670	255
$_{28}Ni^{63}$	100.1	6	17.43
$_{61}Pm^{147}$	2.62	345	224.14
$_{38}Sr^{90}$	29.1	921	200

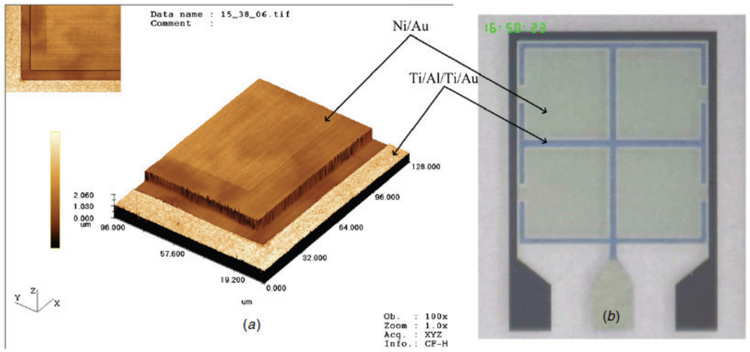
today's betavoltaics is based on micro and nano-technology

3D betacell (porous Si)



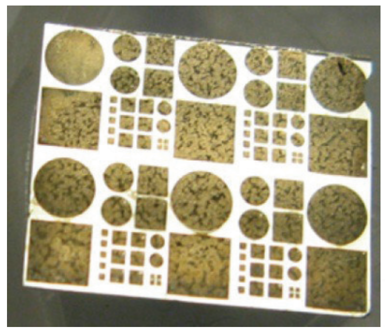
W. Sun et al.
Adv. Mater. 2005, 17, 1230-1233

GaN thinfilm betacell



Z. Cheng, J. Micromech. Microeng. 22 **2012** 074011

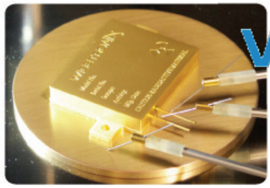
SiC micro-betacells



S. Tin, A. Lal,
Proceedings of PowerMEMS **2009**

…betavoltaic goes industrial

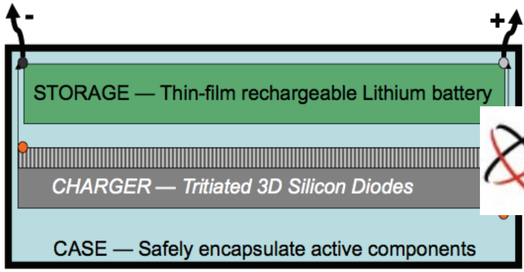
Startups (mainly US based)



WIDETRONIX

SiC T, Ni-63

most probably GaN, T



BetaBatt™

3D Si+Li ,T

Qynergy
ENGAGE ENERGIZE EMPOWER

SiC
Kr-85

Targeted markets: military, medical, space, oil-gas ind., environmental

Micro-power Applications: defense microelectronic eg. non-volatile memory(encryption), RF-ID tag, MEMS/NEMS, sensors, space probes, implants etc.

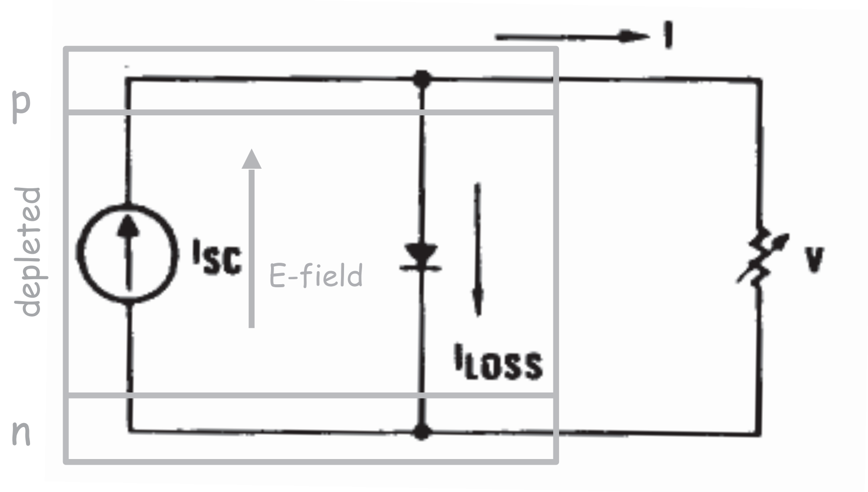
.. but still low efficiency < 10%, R&D ongoing…

BATGAN (French ANR project) betacell based on thin films GaN, started in 2012 (36 months)

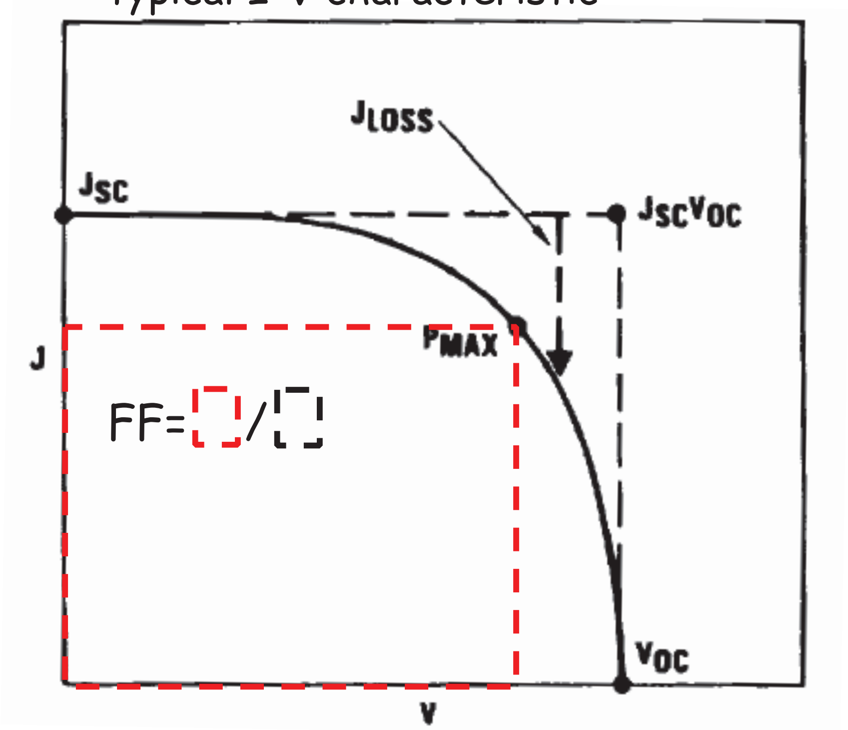
(Diamond Sensors Laboratory (J.de Sanoit) task: Ni-63 electrodeposition)

Some basic parameters of a betacell

equivalent circuit model



typical I-V characteristic



power: $P_{max} = J_{sc} \times V_{oc} \times FF$

overall efficiency

$\eta = (P_{max}/P_{in}) \times 100\%$

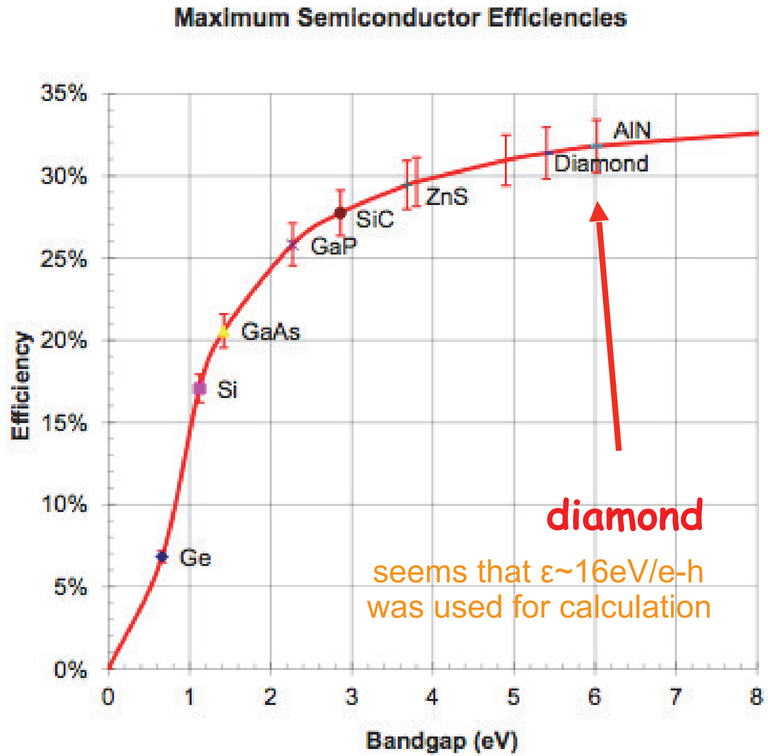
$\eta = \eta_\beta \times \eta_c \times \eta_s$

$\eta_\beta = N_\beta / N_0$ source

$\eta_c = (1-r) \times CCE$ semiconductor + source

$\eta_s = [V_{oc} \times FF / \varepsilon]$ and $\eta_s \leq E_g/\varepsilon$ semiconductor

Why diamond?



frequently seen graph in 'betapapers'

large band gap - low leakage current (I_{loss})

- high conversion efficiency (high E_g /ε)

low Z - negligible back-scattering for betas

high density - low beta range —> [thinner layers]

high mobility both e-h - diffusion lenght, less recombination (CCE~1)

b-doped scCVD entrance electrode:

- robust, corrosion hard→ direct electrodeposition of radionuclides possible [improved coupling]

radiation hard: Sr-90; alphavoltaic possible(?)

high quality scCVD commercially available **(CCE~1 @ mm)**

Diamond betavoltaic literature (up to date):

D.M Trucchi, E. Cappelli., P. Ascarelli, Proc.13. It. conf. on senors and microsystems, 2009

recent work of V.S Bormashov, Troitsk, last slide....

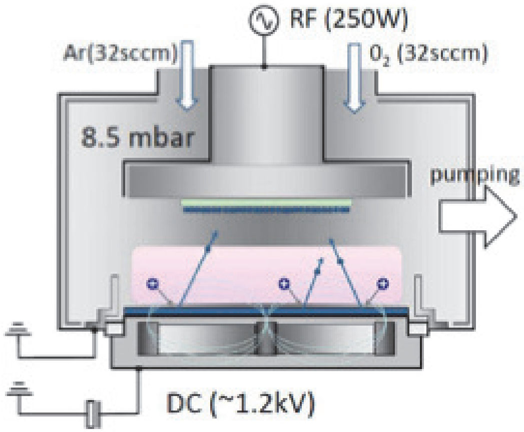
diamond as converting material can improve overall efficiency of betacells

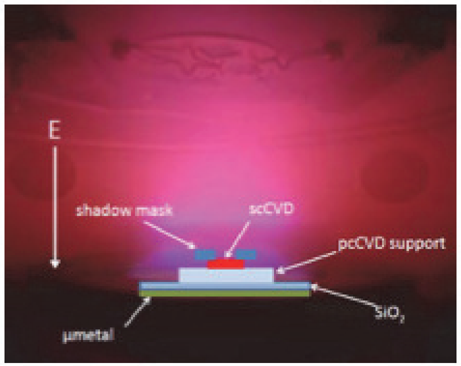
scCVD diamond self-supported membranes

Processing → **Characterisation** → **Devices**

Processing

Ar/O2 plasma deep etching





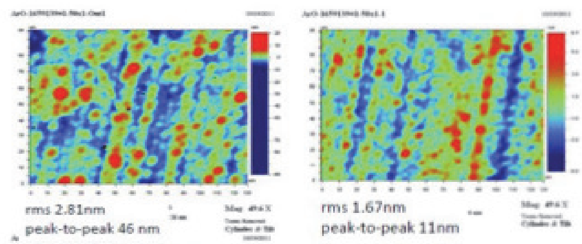
max. rate 1.5 µm/h

starting material:
e6 scCVD 30-40 µm thick (EG N<1ppb, OG N<1ppm)

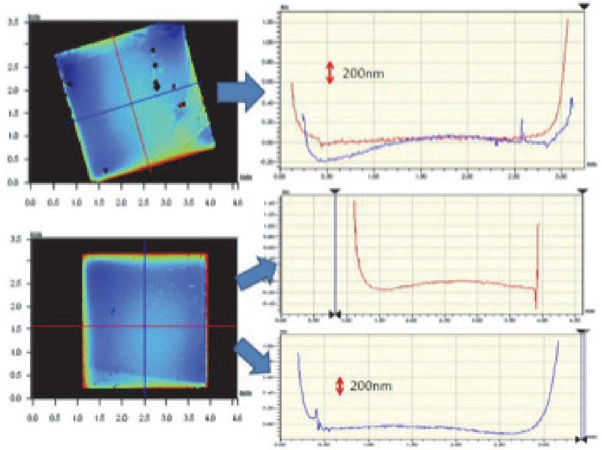
Characterisation

surfaces quality after ~30 µm removal

microscopic roughness after deep etch (INTR)

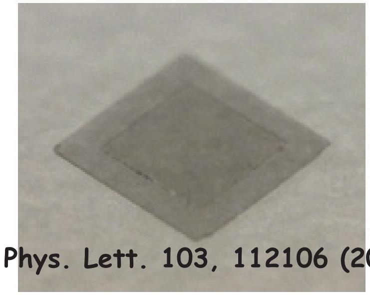


macroscopic roughness after deep etch (INTR)



Devices

bare 3x3mm 3 microns thick

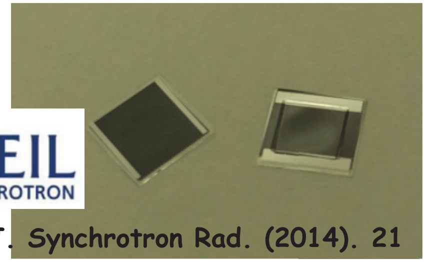


M. Pomorski, Appl. Phys. Lett. 103, 112106 (2013);

diamond membrane HI detector/vacuum window
for living-cell irradiation

V. Grrilj, Appl. Phys. Lett. 103, 243106 (2013)
N. Iwamoto, IEEE Nulr.. 103, vol 60 no.6 (2014)

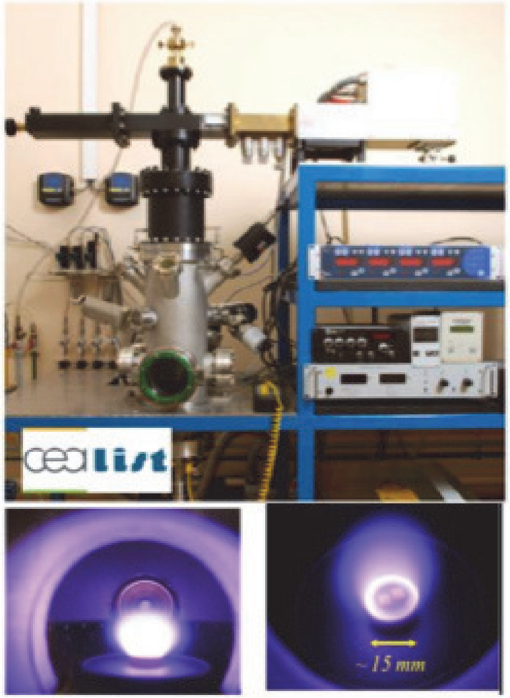
diamond membrane x-ray PSD detectors



K. Desjardins, J. Synchrotron Rad. (2014). 21

scCVD diamond PIM membranes for betacell

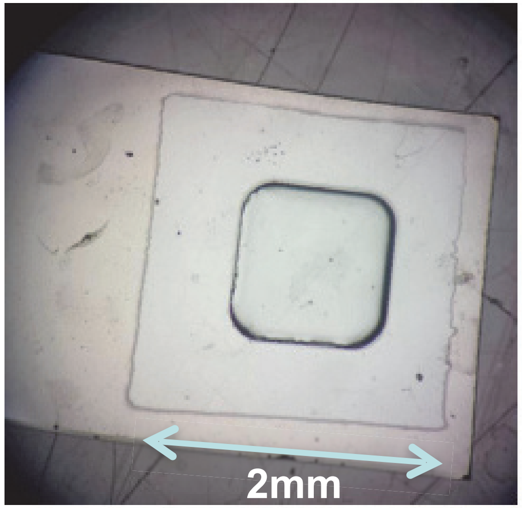
boron p-doped CVD layers



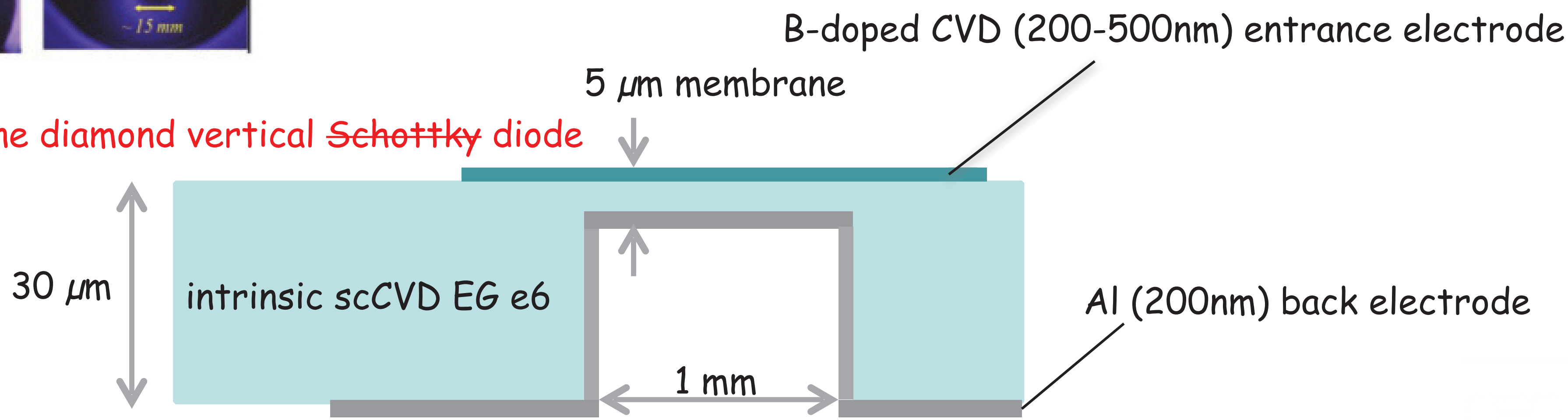
$$[CH4] / [H2] = 0.55\ \%$$
$$(B/C)_{gas} = 2\ \ 10^4\ \text{ppm}$$
$$\text{Power} = 438\ \text{W}$$
$$\text{P} = 120\ \text{mbar}$$
$$\text{T} = 870\ \text{C}$$

surfaces oxidation hot H2SO4+KNO3,
Al back contact by PVD

~5 microns thick PIM membrane



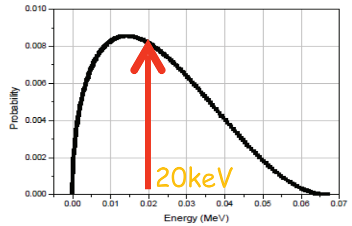
PIM membrane diamond vertical ~~Schottky~~ diode



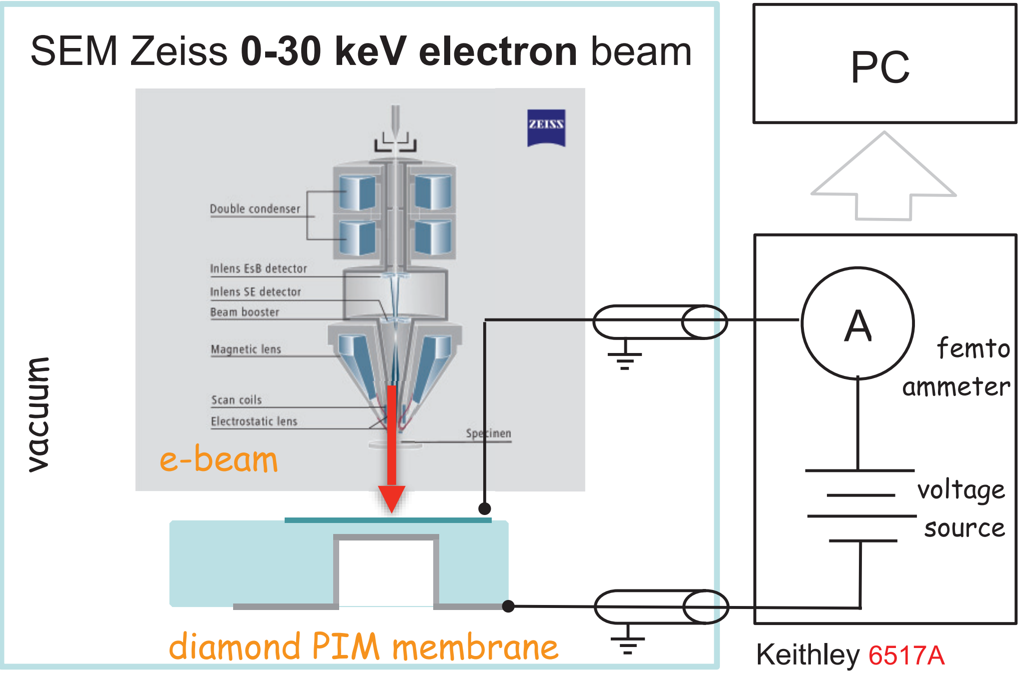
SEM experimental setup – Ni-63 source simulation

Ni-63 beta-spectrum:

Eavg ~ 17.4 keV
Emax ~ 66 keV



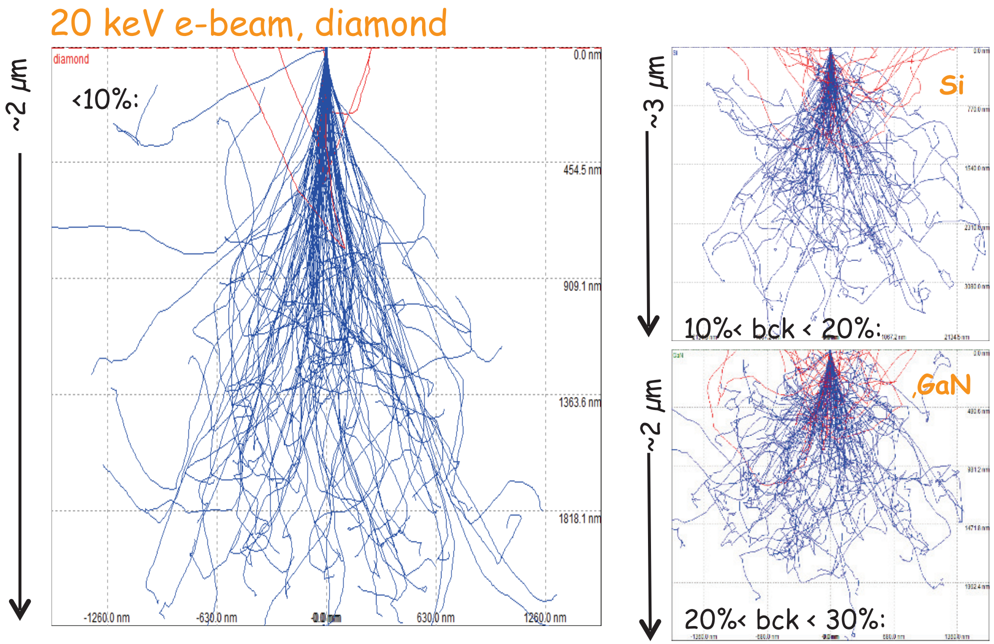
solution: home-made manual electron beam induced current (EBIC)...



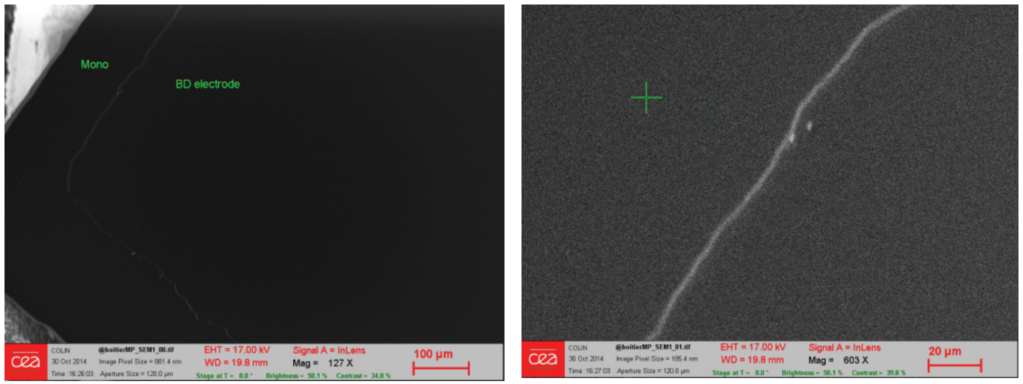
injected power from the SEM:

$$P_{in} = 20\ keV \times 5.2\ nA = 104\ \mu W$$

e-beam interaction 100 e displayed (Casino software)



sample imaging with SEM

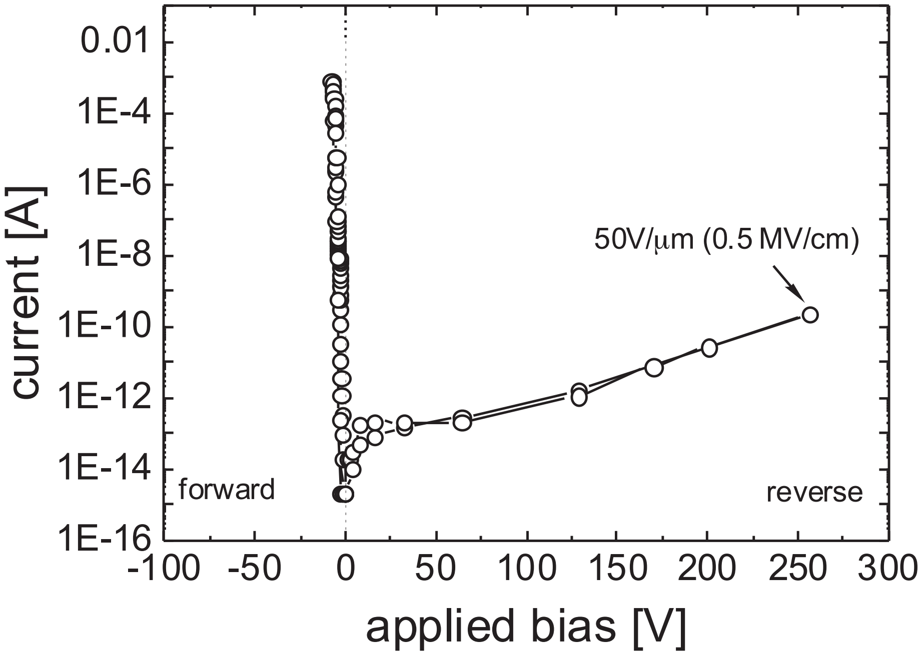


20keV electron beam by SEM

full I-V PIM membrane characteristics

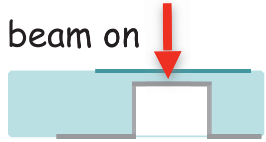
no beam

dark current

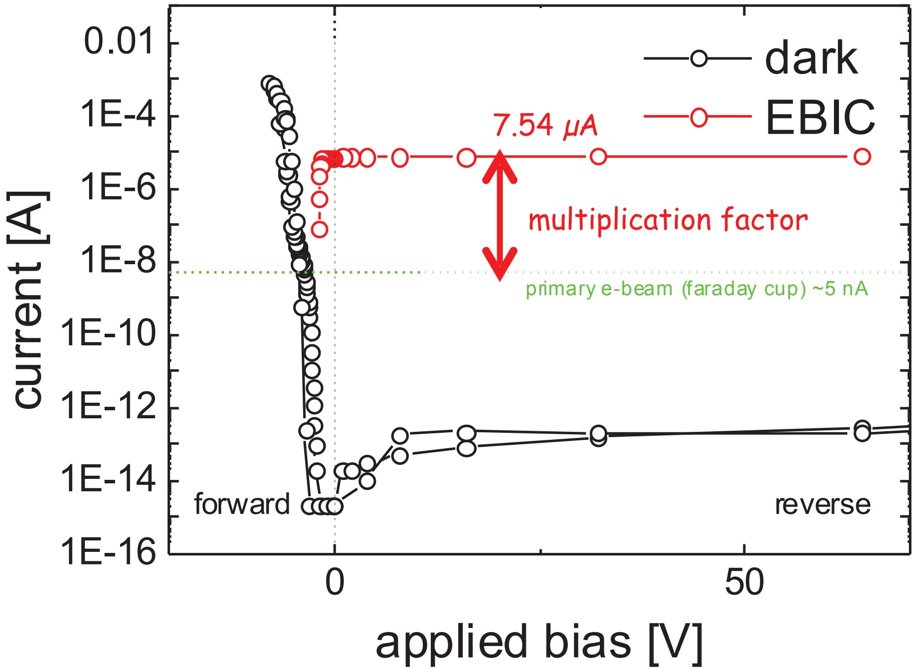


20 keV, 5.2 nA

beam on



EBIC



no breakdown in reverse up to 50V/µm (0.5 MV/cm)

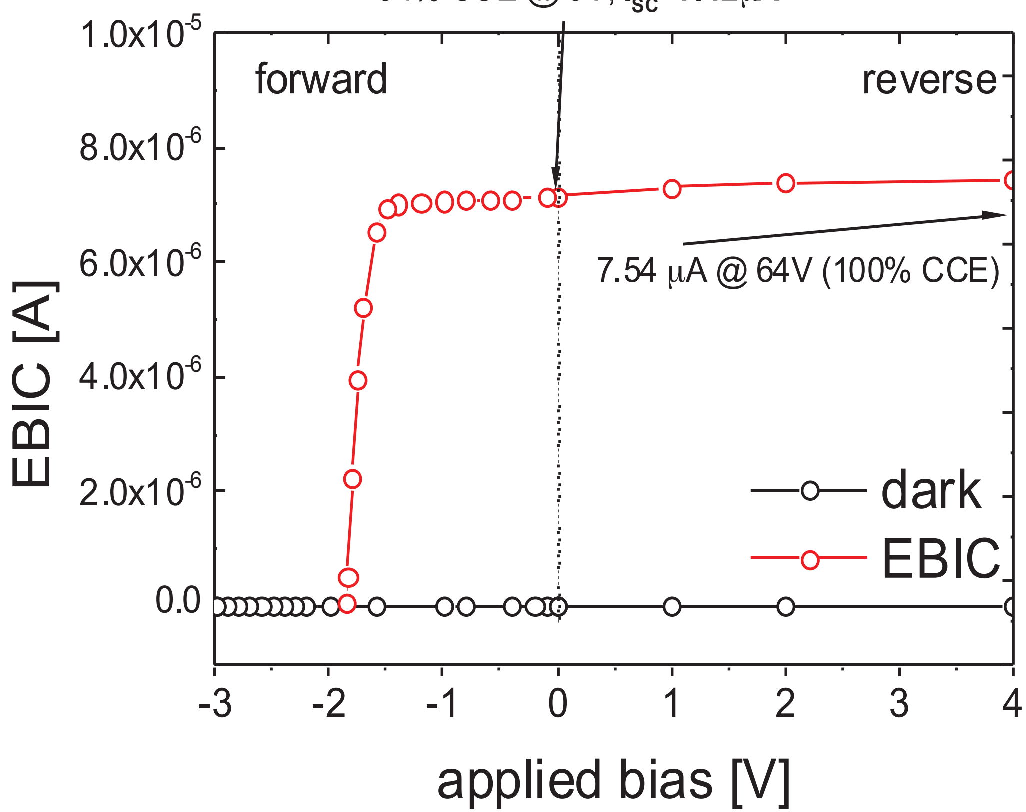
high current density in forward possible
(Keithley current source quickly satuarted)

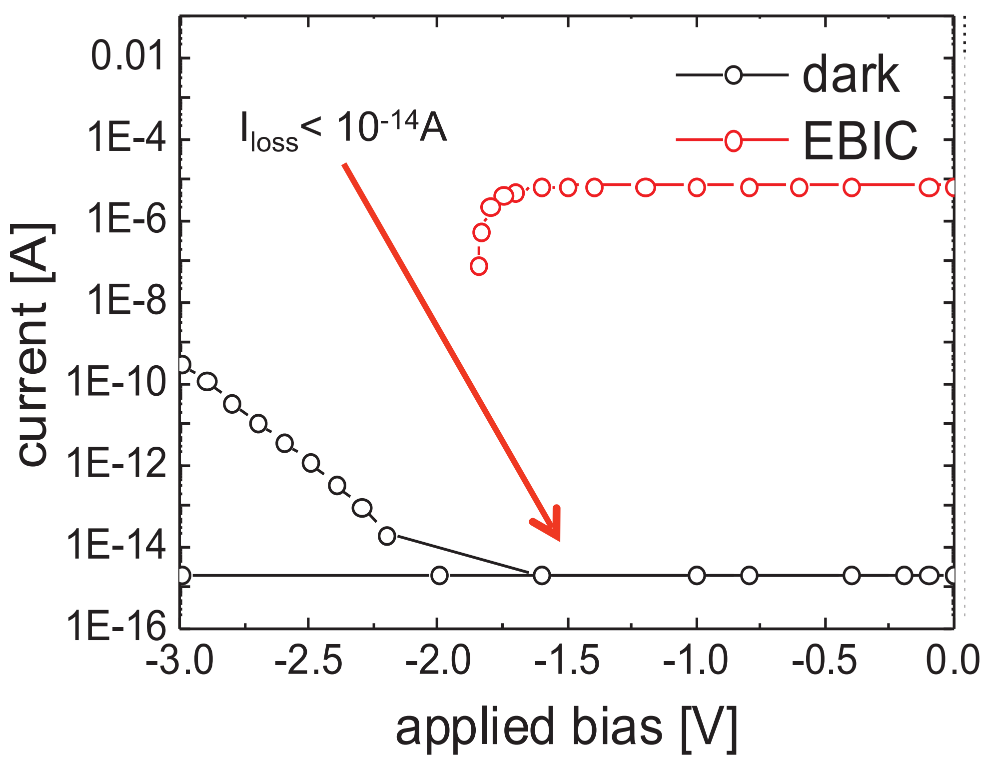
high rectification ratio 10^{10} @ 6V

multiplication factor
≥1450 → avg. ε(e-h) < 13.8 eV/e-h
(from 11.8 -to 16.5 eV/e-h reported)
needs correction for backscattering,
+ p electrode recombination (diffusion lenght)

20keV electron beam by SEM

zoom at photovoltaic mode (…or rather betavoltaic….)



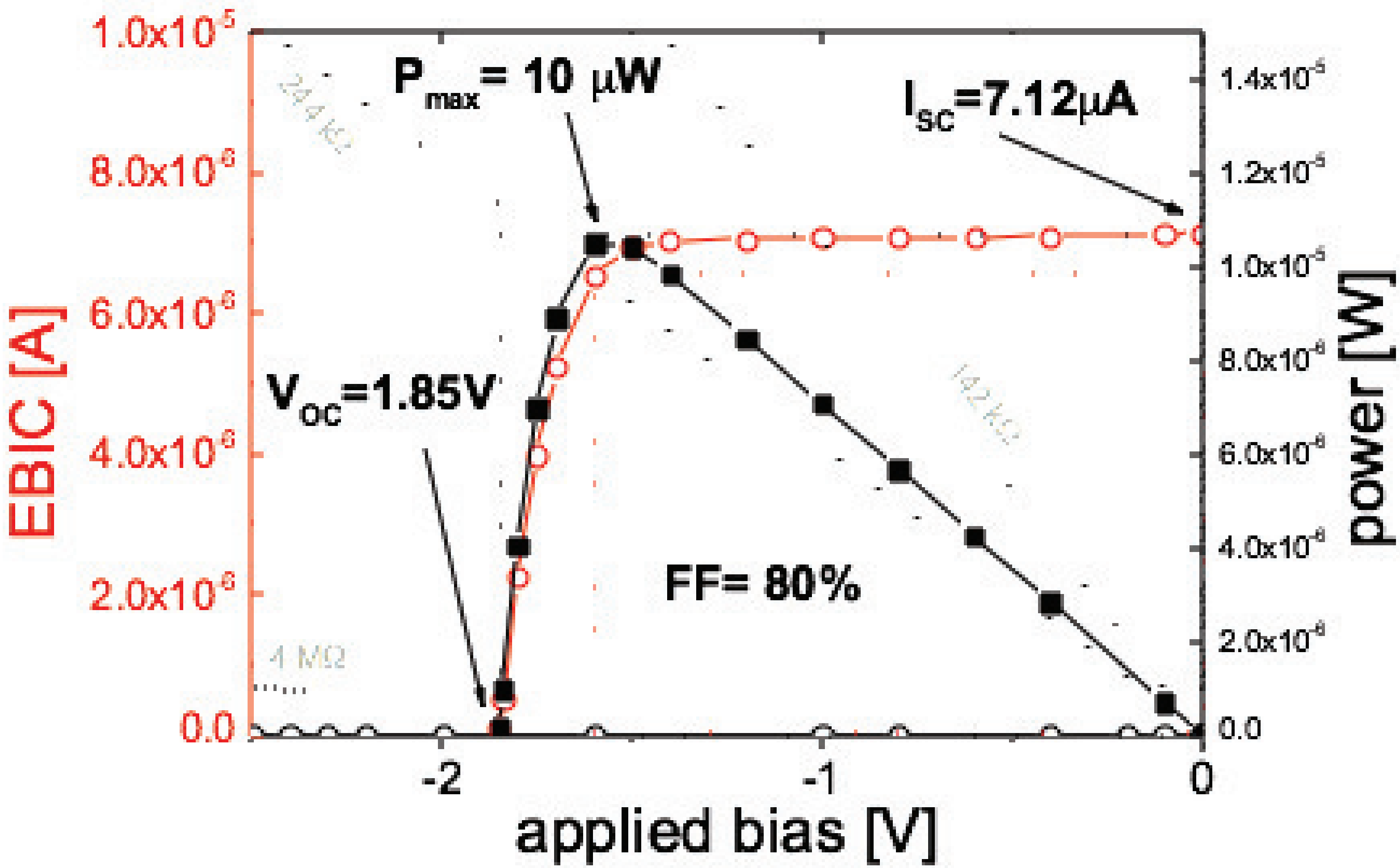


94% charge carriers collected @ 0V

I_{loss} in femtoamp range

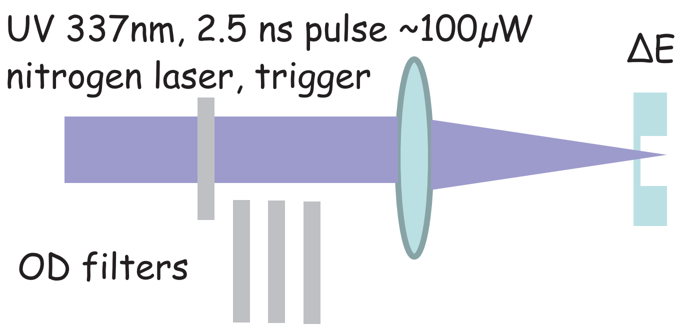
20keV electron beam by SEM

betavoltaic mode → key parameters of scCVD PIM membrane

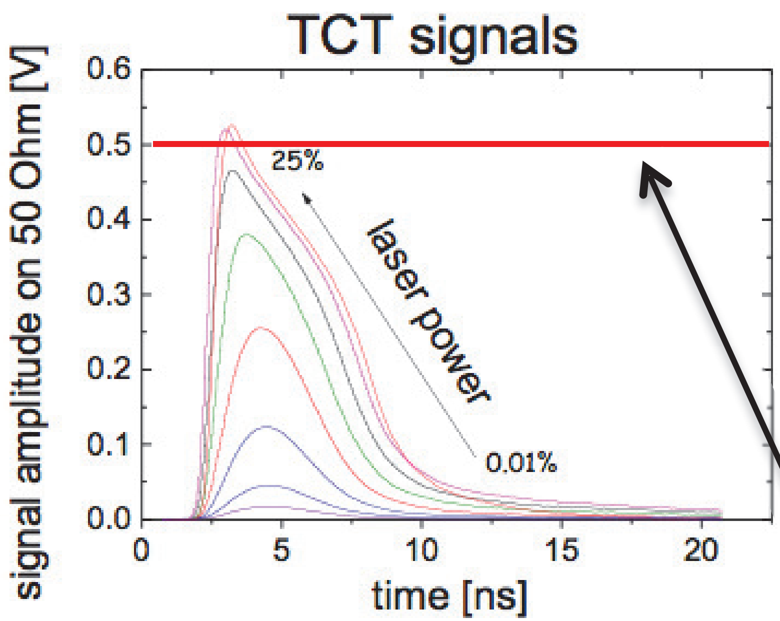


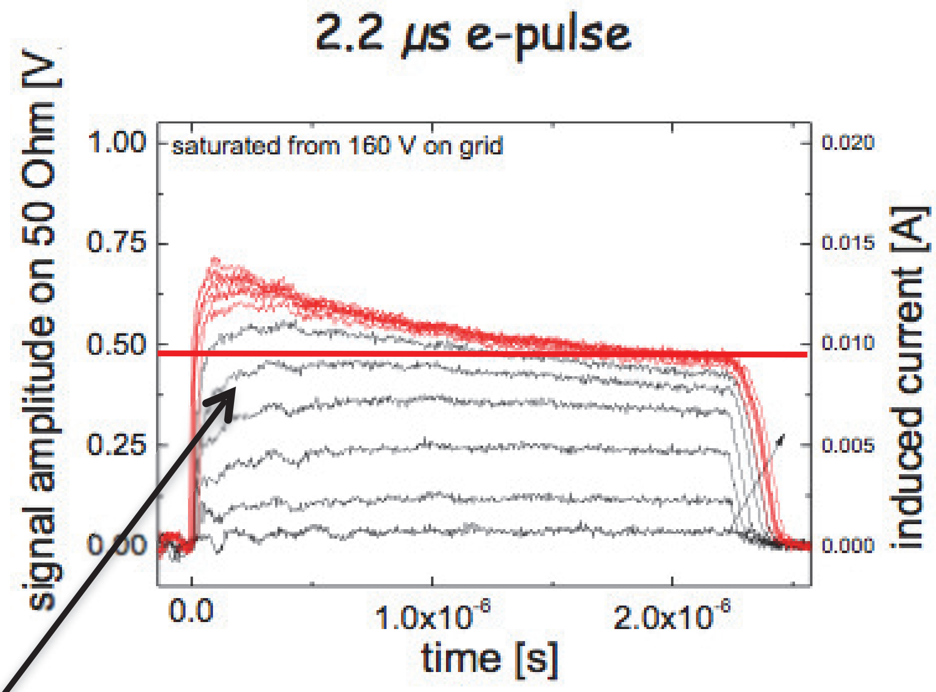
overall efficiency $\eta = (P_{max} / P_{in}) \times 100\% = (10\ \mu W / 104\ \mu W) \times 100\% \sim$ **10%**

Testing the (space charge) limits



semi-DC mode with
intense 4 MeV e-beam (linac)
(membrane @ 0V)





can deliver up to 10 mA (before space charge saturation)
with V =0.5 V —> **~5 mW** (for 4 mm^2)

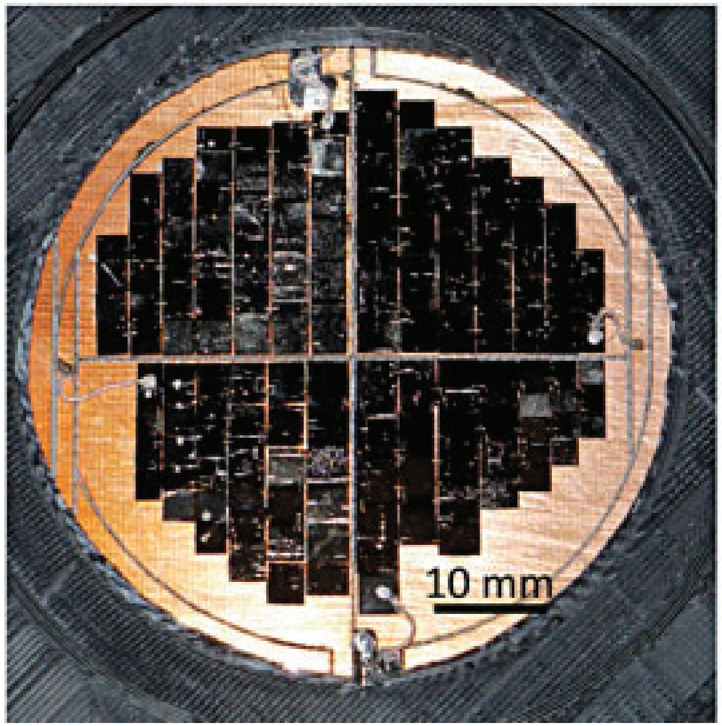
Meanwhile in Russia ….

Phys. Status Solidi A 212, No. 11, 2539–2547 (2015) / DOI 10.1002/pssa.201532214

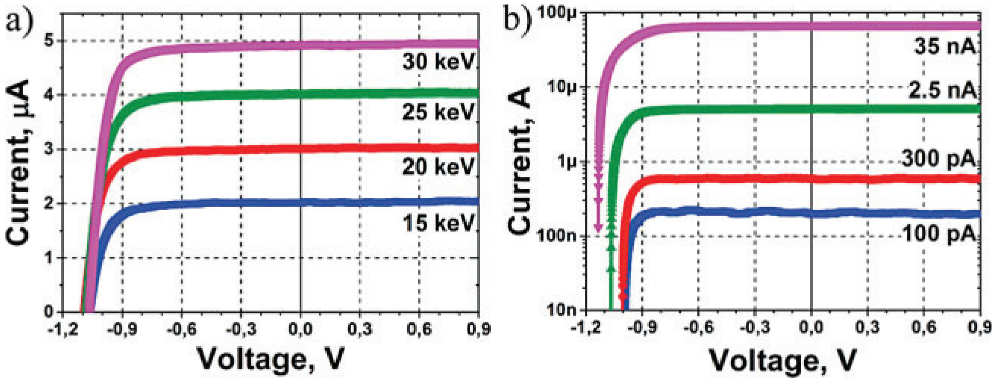
Development of nuclear microbattery prototype based on Schottky barrier diamond diodes

Vitaly Bormashov[*,1], Sergey Troschiev[1], Alexander Volkov[1], Sergey Tarelkin[1,2], Eugeniy Korostylev[3], Anton Golovanov[1,3], Mikhail Kuznetsov[1], Dmitry Teteruk[1], Nikolay Kornilov[1], Sergey Terentiev[1], Sergey Buga[1,3], and Vladimir Blank[1,2,3]

130 scCVD - 'Marinelli type' (HPHT, p+, i, m)



SEM



tested as real beta-cell with:

Ni-63,

Promethium-147,

Strontium-90 (high E)

no degradation 1400 h, 4x10^3 rad

Plutonium-238 (alpha)

powers an LED (!), but radiation damaged - Voc goes up

For a single cell:

Voc~1.1V ; FF=74-78%; efficiency ~ 6%@30keV

Summary

- nowadays betavoltaics is being industralized with
R&D ongoing, aiming at efficiency improvement

- diamond is a potential candidate for the high efficiency betacell

- for the first time diamond scCVD PIM (~~Schottky~~) membrane was tested as a converting material for betacell using EBIC

- measured parametrs V_{oc}=1.85, FF=80%, η~10% exceed most of the published parameters from various semiconductors

- only V_{oc} seems to be still limited by the device, I_{sc} approaches intrinsic limits,

Outlook

fabrication of 'real' betacell → deposition of Ni-63 onto diamond membrane

try to improve V_{OC}:

→ play with ~~Schottky~~ barrier (surface preparation, various metals), B-doping, thickness
→ diamond p-n junction (?)

more power:
→ diamond considered as radiation hard → Sr-90, alphavoltaic x 250 J_{max}(?)

geometry and material

→ cheaper OG scCVD, heteroepitaxy, pcCVD
→ 3D devices

... thanks for your attention !!!