## Neutral bremsstrahlung in two-phase Ar electroluminescence: further studies and possible applications

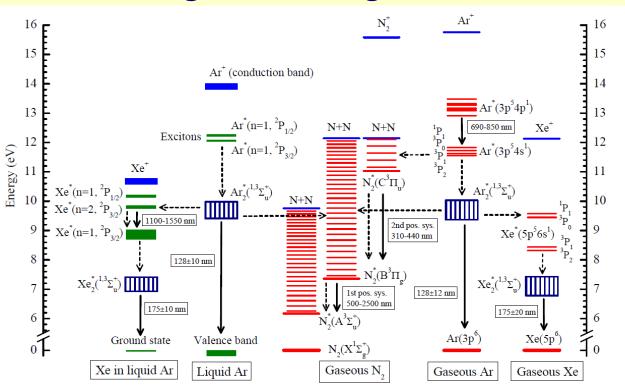
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Basic idea: to detect scintillation light from noble elements in the non-VUV, i.e. in the near UV, visible and near IR

One way to do it is doping Ar with Xe and  $N_2$ , to shift the VUV light of Ar to longer wavelength.



See "Photon emission and atomic collision processes in two-phase argon doped with xenon and nitrogen": the most complete compilation over past 50 years (A. Buzulutskov, EPL 117 (2017) 39002).

# Another way is to use a concurrent scintillation mechanism, namely that of neutral bremsstrahlung (NBrS)

Until recently, it was believed that proportional electroluminescence (EL) in pure noble gases was fully due to VUV emission of noble gas excimers produced in atomic collisions with excited atoms.

On the other hand, our recent experiments have revealed an additional mechanism of proportional EL, namely that of bremsstrahlung of drifting electrons scattered on neutral atoms (so-called neutral bremsstrahlung, NBrS).

### Ordinary EL mechanism

$$e^- + Ar \rightarrow e^- + Ar^*$$
,  
 $Ar^* + 2Ar \rightarrow Ar_2^* + Ar$ ,  
 $Ar_2^* \rightarrow 2Ar + h\nu$ .

### Neutral bremsstrahlung EL mechanism

$$e^{-} + A \rightarrow e^{-} + A + h\nu$$
,  
 $e^{-} + A \rightarrow e^{-} + A^{*} + h\nu$ .



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## Revealing neutral bremsstrahlung in two-phase argon electroluminescence



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#### ABSTRACT

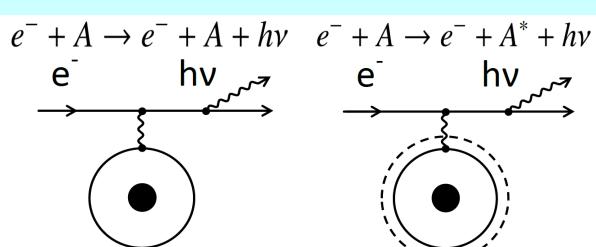
Proportional electroluminescence (EL) in noble gases has long been used in two-phase detectors for dark matter search, to record ionization signals induced by particle scattering in the noble-gas liquid (S2 signals). Until recently, it was believed that proportional electroluminescence was fully due to VUV emission of noble gas excimers produced in atomic collisions with excited atoms, the latter being in turn produced by drifting electrons. In this work we consider an additional mechanism of proportional electroluminescence, namely that of bremsstrahlung of drifting electrons scattered on neutral atoms (so-called neutral bremsstrahlung); it is systemically studied here both theoretically and experimentally. In particular, the absolute EL yield has for the first time been measured in pure gaseous argon in the two-phase mode, using a dedicated two-phase detector with EL gap optically read out by cryogenic PMTs and SiPMs. We show that the neutral bremsstrahlung effect can explain two intriguing observations in EL radiation: that of the substantial contribution of the non-VUV spectral component, extending from the UV to NIR, and that of the photon emission at lower electric fields, below the Ar excitation threshold. Possible applications of neutral bremsstrahlung effect in two-phase dark matter detectors are discussed.

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# Part 1: Theory of NBrS electroluminescence

### Types of bremsstrahlung

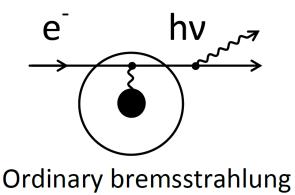


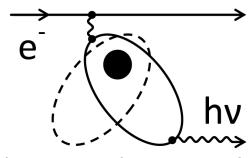
Neutral bremsstrahlung produced by slow (~10 eV) electrons when they are scattered (elastically inelastically) neutral on atoms.

Neutral bremsstrahlung in elastic scattering in inelastic scattering

Neutral bremsstrahlung

At such electron energies, the contribution of ordinary bremsstrahlung (produced in the Coulomb field of a nucleus) and polarization bremsstrahlung (produced by atoms due to their time-dependent polarization) is negligible.





Polarization bremsstrahlung

### Neutral bremsstrahlung: theoretical predictions

The differential cross section for NBrS photon emission is expressed via elastic cross section ( $\sigma_{el}$ ) of electron-atom scattering:

$$\left(\frac{d\sigma}{dv}\right)_{NBrS,el} = \frac{8}{3} \frac{r_e}{c} \frac{1}{hv} \left(\frac{E - hv}{E}\right)^{1/2} \times \\ \times \left[(E - hv) \sigma_{el}(E) + E \sigma_{el}(E - hv)\right]_{10^{-15}}$$

$$e^- + A \rightarrow e^- + A + hv$$

$$e^- hv$$

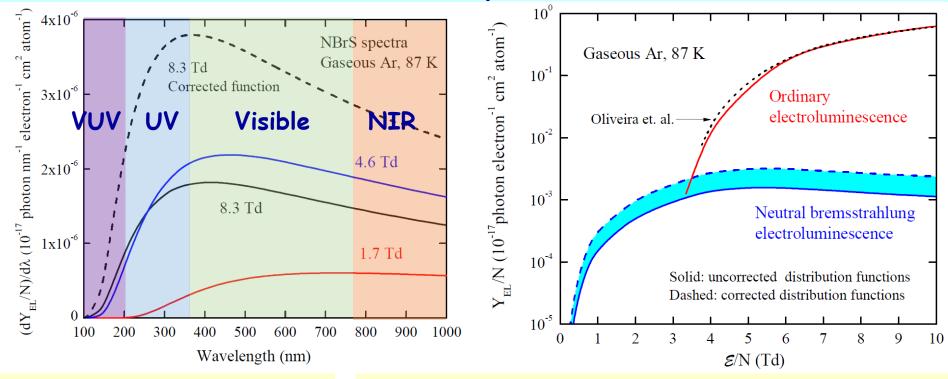
$$in elastic scattering
$$in elastic scattering$$
Neutral bremsstrahlung in elastic scattering
$$10^{-18}$$

$$0.01 \quad 0.1 \quad 1 \quad 10 \quad 1000$$
Electron energy (eV)$$

Using this cross section and electron energy distribution functions, we calculated the spectra of NBrS emission at different reduced electric fields E/N (expressed in Td).

1 Td corresponds to electric field of 0.87 kV/cm at 87 K.

## Neutral bremsstrahlung vs ordinary electroluminescence: theoretical predictions



Calculated spectra of NBrS emission.

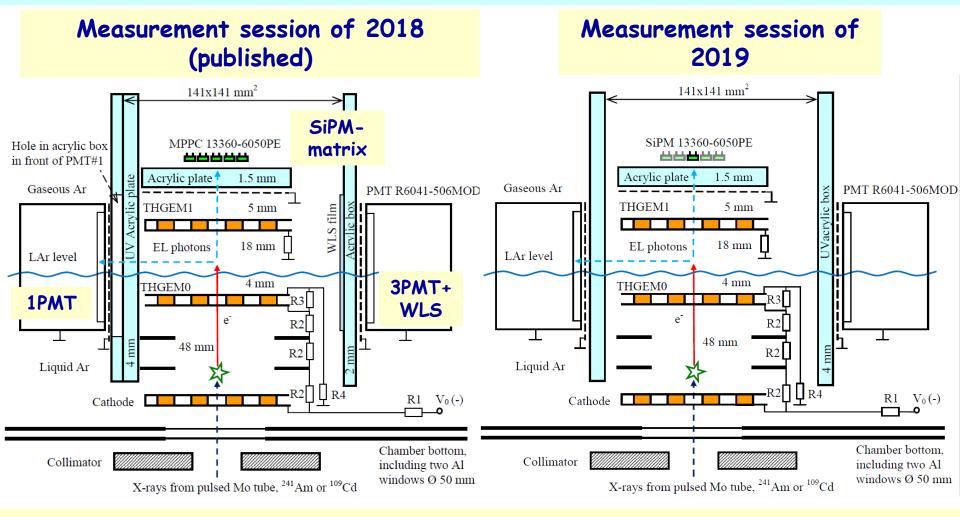
Reduced ordinary EL yield and that of neutral bremsstrahlung at 0-1000 nm in gaseous Ar as a function of the reduced electric field.

### Summarizing, the theory of NBrS EL predicts:

- 1. electroluminescence below the Ar excitation threshold (~4 Td), in the UV, visible and NIR regions;
- 2. appreciable non-VUV component above the Ar excitation threshold, extending from the UV to NIR.

## Part 2: NBrS electroluminescence: experiment vs theory

### Experimental setup



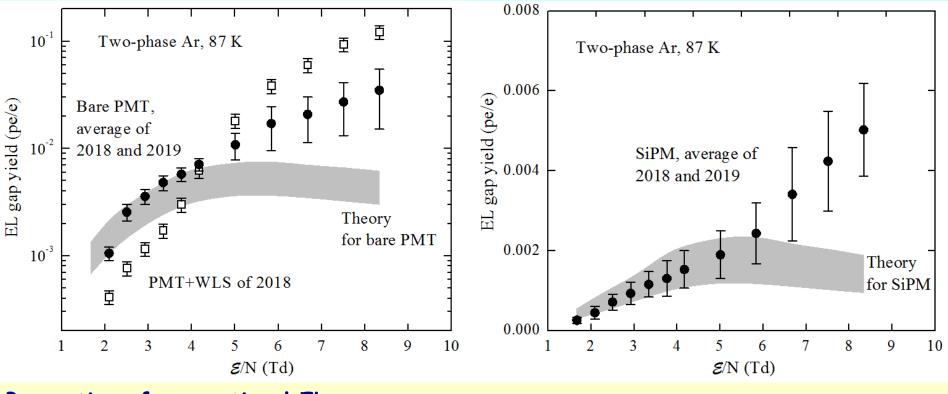
Bare PMT: 300-650 nm (via direct recording)

PMT+WLS: 100-650 nm (at <400 nm via re-emission in WLS, at >400 nm via

direct recording)

SiPM: 400-1000 nm (via direct recording)

### EL gap yield for PMT and SiPM

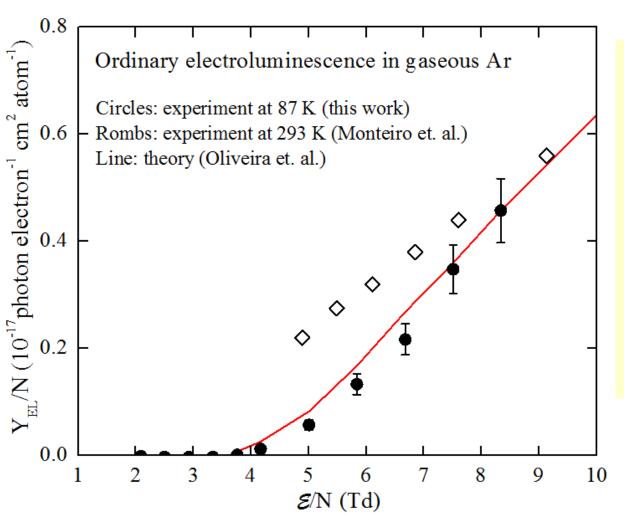


### Properties of proportional EL:

- 1) there is a noticeable contribution of the non-VUV spectral component in EL radiation, extending from the UV to NIR.
- 2) there is a photon emission at lower electric fields, below the Ar excitation threshold (at 4 Td) where the non-VUV component fully dominates.

Above the threshold, the theory quickly diverges from experiment. This discrepancy was explained by the effect of Feshbach resonance: see [DeMunari et al. Lett. Nuov. Cim. 2 (1971) 68], [Dyachkov et al. Sov. Phys. JETP 38 (1974) 697], [Buzulutskov et al. Astroparticle Physics 103 (2018) 29]

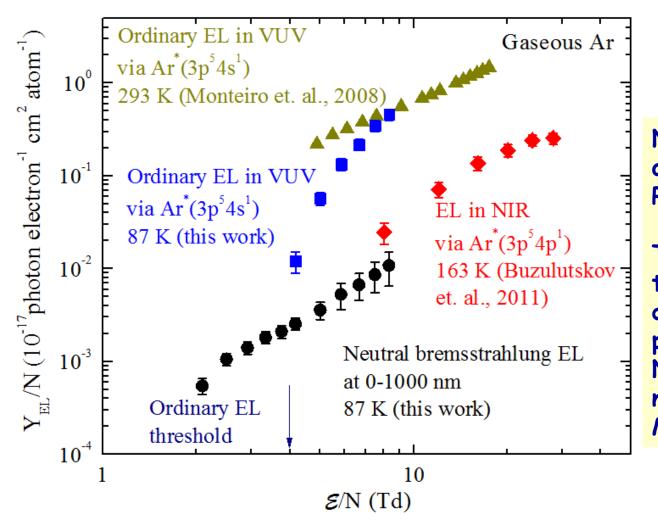
### Reduced EL yield in the VUV (ordinary EL)



Reduced EL yield for ordinary (VUV) electroluminescence, obtained using NBrS paradigm, was compared to the yields at room T, obtained experimentally [Monteiro et al, Phys. Lett. B 668 (2008) 167] and theoretically [Oliveira et al, Phys. Lett. B 703 (2011) 217].

This figure demonstrates a convincing agreement between the theory and our experiment, the latter using NBrS paradigm in proportional EL.

## Summary of experimental EL yield in gaseous Ar (our results are presented using NBrS paradigm)



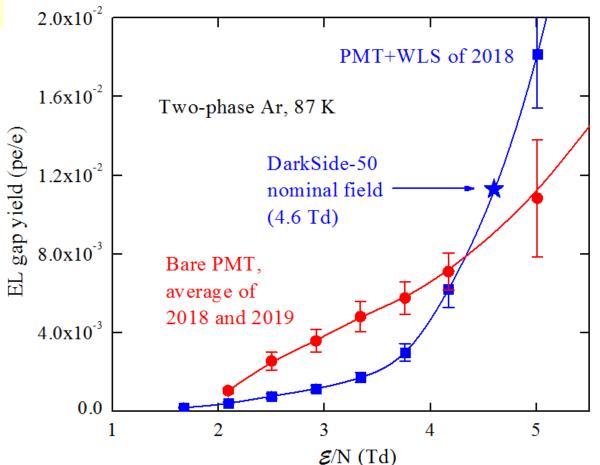
NBrS yields were obtained by averaging PMT and SiPM data.

The discrepancy between the our and Monteiro data at lower fields is presumably due to the NBrS effect, which was not taken into account by Monteiro.

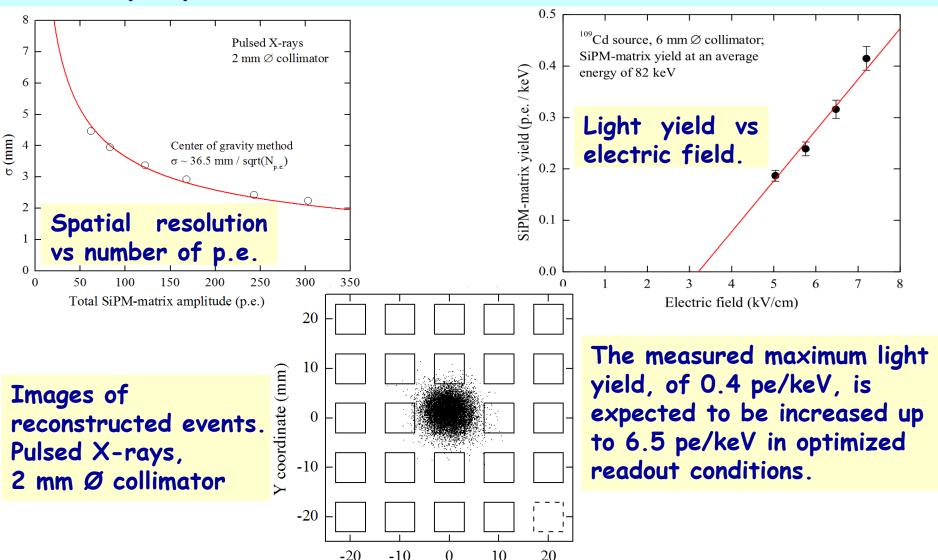
# Part 3: Possible applications of NBrS effect

## Direct optical readout of two-phase Ar TPCs using NBrS electroluminescence

The important result of this work is that at moderate electric fields in the EL gap, below 5 Td, the amplitude of the S2 signal from the bare PMT is comparable with that of the PMT with WLS, in the absence of optical contact between the WLS and the PMT. This observation paves the way for direct readout of S2 signals in two-phase dark matter detectors using PMTs and SiPM-matrices.

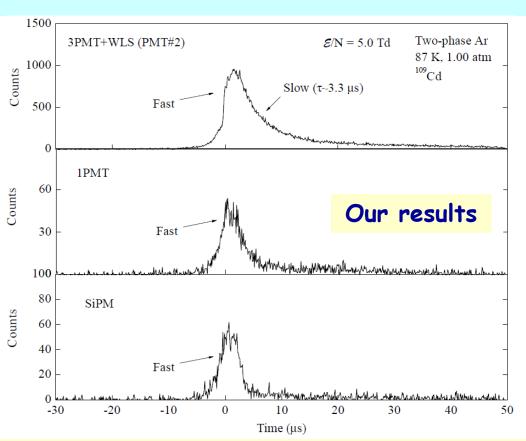


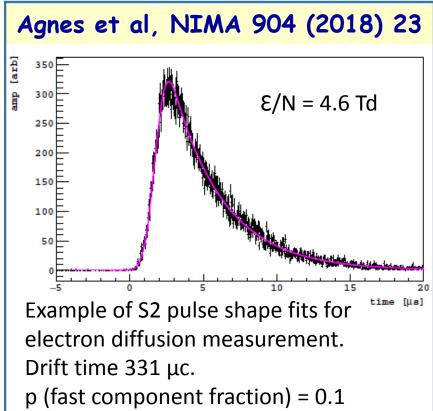
# SiPM-matrix readout of a two-phase Ar TPC using electroluminescence in the visible and NIR range (in preparation, to be submitted to NIM A)



X coordinate (mm)

### Pulse shape: fast component fraction in S2 signal

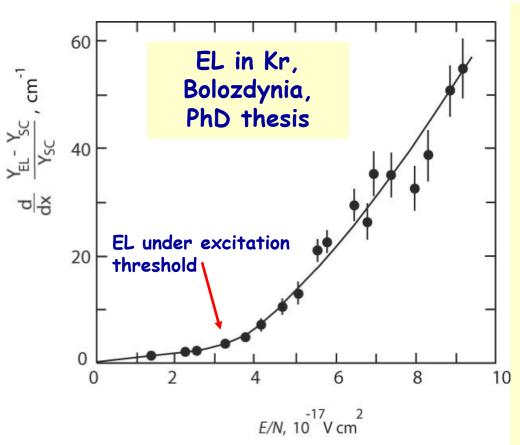




In DarkSide-50 the fast component fraction was taken p=10% (using ordinary EL approach). According to our data and using NBrS paradigm, this fraction is predicted to be substantially larger: of about 50%.

This enhancement can affect the determination of the quantities using the fast component, such as the diffusion coefficients in liquid Ar or z-coordinate fiducialization.

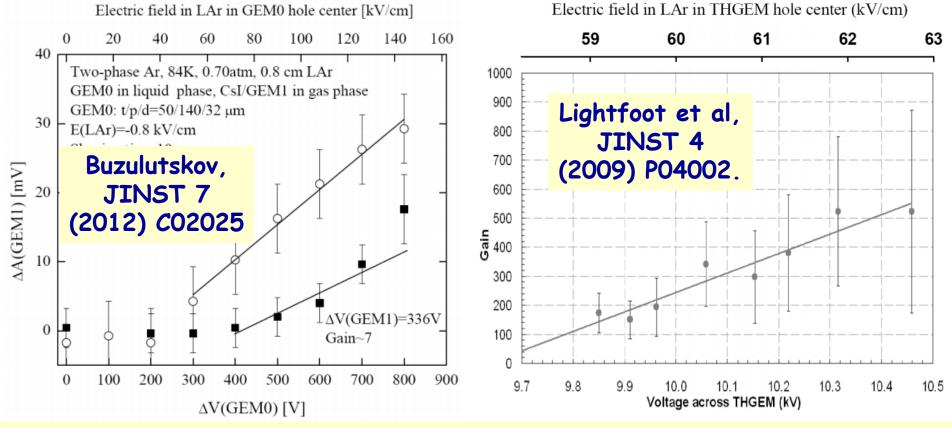
### Universal character of NBrS electroluminescence



NBrS electroluminescence should be present in all noble gases, including He, Ne, Kr, Ar and Xe. That is why we assume that NBrS electroluminescence is present in S2 signals of two-phase Xe detectors, such as LUX, PandaX, Xenon100 and RED100.

Presumably it has not been yet observed due to the fact that the S2 signal in Xe is recorded directly using PMTs with quartz windows (i.e. without WLS reemission losses), resulting in that the NBrS signal is difficult to observe at the background of a strong main signal.

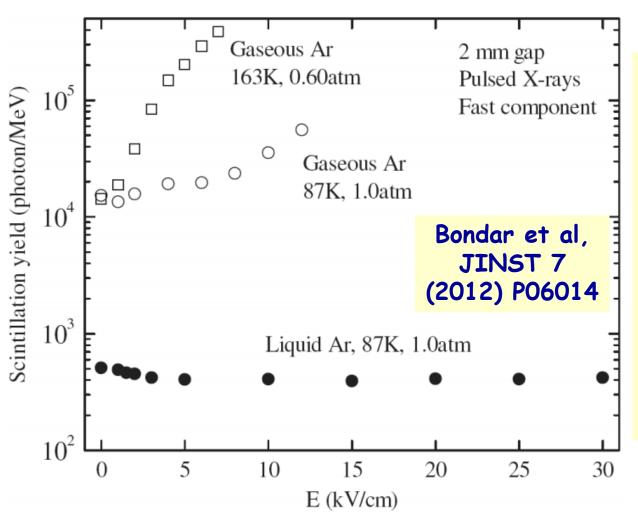
## NBrS electroluminescence in noble liquids (in immersed GEM-like structures)?



The NBrS effect can be responsible for proportional EL observed in liquid Ar and Xe using immersed GEM-like structures.

The reduced electric fields in the center of GEM or THGEM holes used in liquid Ar, of 0.3 - 0.7 Td. For such reduced electric fields, the theory predicts that NBrS electroluminescence already exists. It also predicts the linear dependence of the EL yield observed in experiments.

### NBrS primary scintillation in noble liquids?



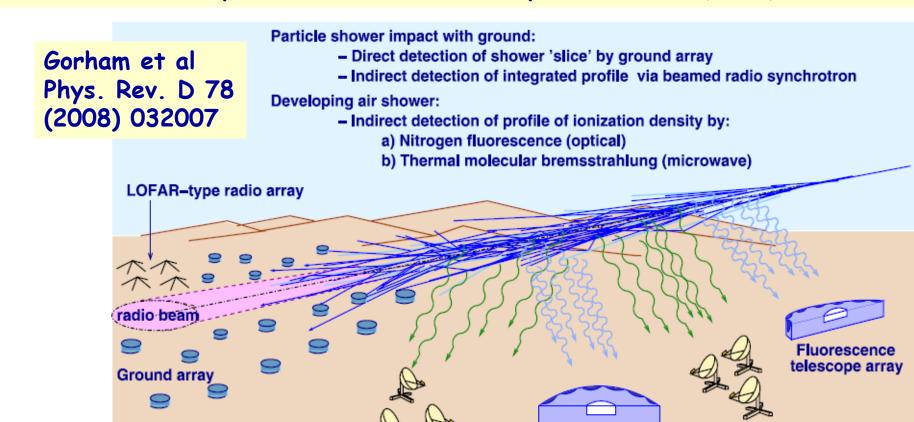
The weak primary scintillations in liquid Ar in the visible and NIR observed earlier, range, be explained might by neutral bremsstrahlung of ionization the primary electrons.

NBrS primary scintillation in liquid Ar is currently under further study in our laboratory.

# Part 4: Applications of NBrS effect already used in practice

## Detection of ultrahigh-energy cosmic rays using NBrS ("molecular bremsstrahlung") in the radio-frequency range

NBrS is supposed to be used in practice to develop a detection technique for ultrahigh-energy cosmic rays. Here the NBrS radiation in the radio-frequency range is emitted by primary ionization electrons left after the passage of the showers in the atmosphere [AlSamarai et al Phys. Rev. D 93 (2016) 052004]

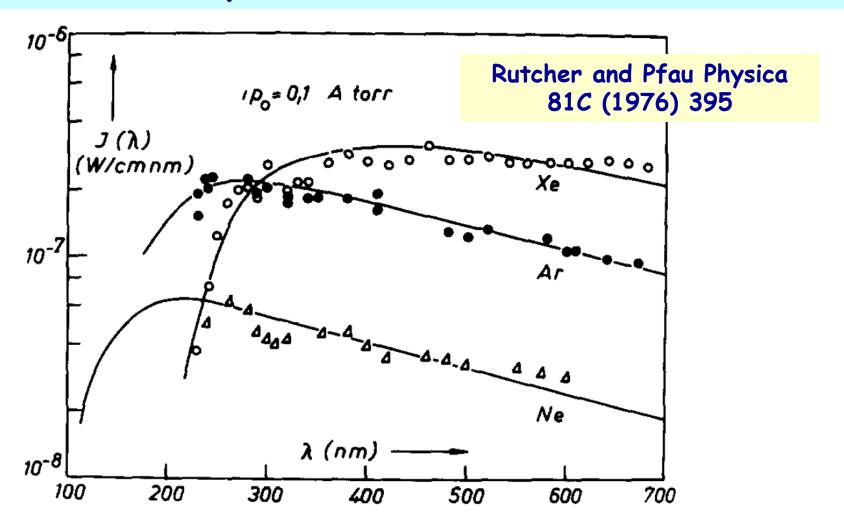


AMBER array

AMBER array

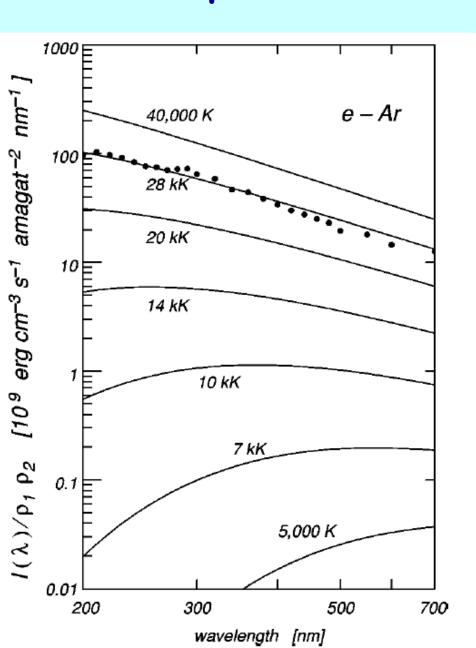
Fluorescence telescope array

## Continuous emission spectra in a weakly ionized plasma due to NBrS



The NBrS effect was used to explain continuous emission spectra in a weakly ionized plasma in Ne, Ar and Xe, in glow and radio-frequency discharges.

### Emission spectra in sonoluminescence due to NBrS



Frommhold Phys. Rev. E 58 (1998) 1889

The NBrS effect was used to explain sonoluminescence spectra of the bubbles in liquid Ar.

A measured sonoluminescence spectrum of Ar bubbles is also shown (dots).

## Summary

A new mechanism of proportional electroluminescence (EL) in two-phase Ar has been revealed, namely that of neutral bremsstrahlung (NBrS), that quantitatively describes the photon emission below the Ar excitation threshold and non-VUV component above the threshold.

This paves the way for direct readout of S2 signals in two-phase TPCs, using PMT and SiPM matrices. In addition, it predicts the enhanced contribution of the fast component to S2 signal, which can affect the correct determination of diffusion coefficients and z-coordinate fiducialization in liquid Ar.

The NBrS effect has a universal character: it should be present in all noble and molecular gases. It may also explain the non-VUV components observed earlier in various light emission processes, in particular the primary and secondary scintillations in noble liquids in the visible and NIR range.

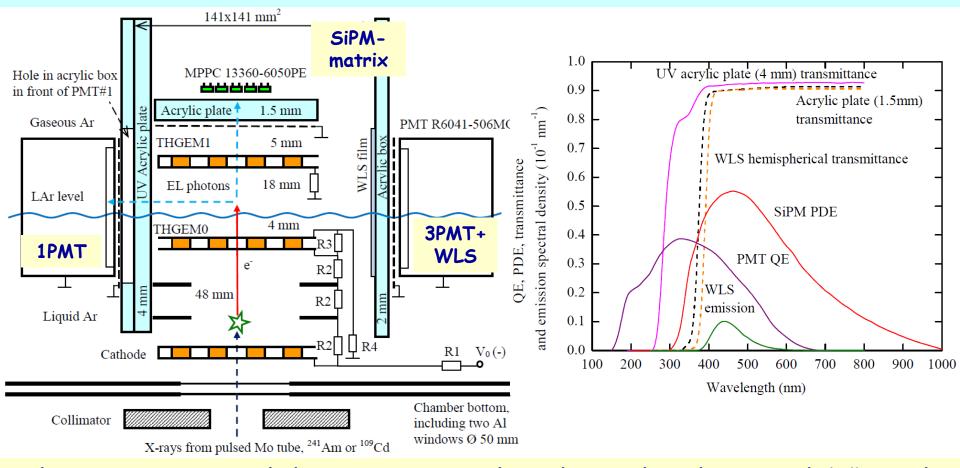
## Backup slides

### Our global objective and current activity

Development of liquid Ar detectors of ultimate sensitivity for dark matter search and coherent neutrino-nucleus scattering experiments and their energy calibration.

- Our group is currently conducting researches in the following directions, in the frame of Laboratory of Cosmology and Elementary Particles (NSU and BINP) and in the frame of DarkSide experiment:
- Measurement of electroluminescence (EL) yields in twophase Ar using a 9-liter detector.
- Problem of Ar doping with Xe and N2.
- Measurement of ionization yields of nuclear recoils in liquid Ar using neutron scattering technique.
- Development of new readout technique in two-phase Ar detectors using SiPM-matrices.

### Experimental setup



Taking into account light propagation through acrylic plates and WLS, the detectors were sensitive in the following wavelength regions:

1PMT (bare PMT): 300-650 nm (via direct recording)

3PMT+WLS: 100-650 nm (at <400 nm via re-emission in WLS, at >400 nm via

direct recording)

SiPM-matrix: 400-1000 nm (via direct recording)

### NBrS EL theory: basic equations

$$\int_{0}^{\infty} f(E) dE = 1 \qquad \int_{0}^{\infty} E^{1/2} f'(E) dE = 1$$

Electron energy distribution function normalization: two ways

Electron energy distribution functions was calculated using Boltzmann equation solver BOLSIG+ (free software)

$$\begin{array}{c} e^- + Ar \rightarrow e^- + Ar^* \; , \\ Ar^* + 2Ar \rightarrow Ar_2^* + Ar \; , \\ Ar_2^* \rightarrow 2Ar + hv \; . \end{array} \qquad \left(\frac{Y_{EL}}{N}\right)_{excimer} = \int\limits_{E_{exc}}^{\infty} \frac{\upsilon_e}{\upsilon_d} \sigma_{exc}(E) f(E) \; dE \\ \end{array}$$

Ordinary electroluminescence, involving excimers

### NBrS EL theory: basic equations

$$\left(\frac{d\sigma}{d\nu}\right)_{NBrS,el} = \frac{8}{3} \frac{r_e}{c} \frac{1}{h\nu} \left(\frac{E - h\nu}{E}\right)^{1/2} \times \left[ (E - h\nu) \,\sigma_{el}(E) + E \,\sigma_{el}(E - h\nu) \right]$$

$$\frac{dI_{ph}(\lambda)}{d\lambda} = \frac{dN_{ph}}{dt N_e dV d\lambda} = N \int_{-\infty}^{\infty} \upsilon_e \frac{d\sigma}{d\nu} \frac{d\nu}{d\lambda} f(E) dE$$

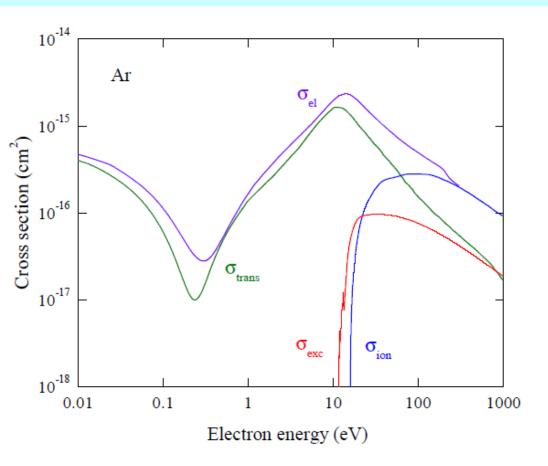
in photon/(s nm electron),

$$\left(\frac{Y_{EL}}{N}\right)_{NBrS} = \frac{dN_{ph}}{dx \ N \ N_e \ dV} = \frac{1}{\upsilon_d \ N} \int_{\lambda_1}^{\lambda_2} \frac{dI_{ph}(\lambda)}{d\lambda} \ d\lambda = \int_{\lambda_1}^{\lambda_2} \int_{h\nu}^{\infty} \frac{\upsilon_e}{\upsilon_d} \frac{d\sigma}{d\nu} \frac{d\nu}{d\lambda} f(E) \ dE \ d\lambda$$

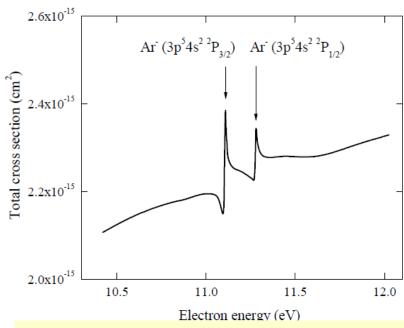
 $\frac{d(Y_{EL}/N)_{NBrS}}{d\lambda} = \int_{-\infty}^{\infty} \frac{\upsilon_e}{\upsilon_d} \frac{d\sigma}{d\nu} \frac{d\nu}{d\lambda} f(E) dE$  in (photon cm<sup>2</sup>)/(electron atom)

in (photon cm<sup>2</sup>)/(electron atom nm),

### NBrS EL theory: cross-sections



Electron scattering cross-sections in Ar obtained from the last version of Magboltz



Experimental cross-section for electron scattering from Ar around Feshbach resonances [Kurokawa 2011]

$$e^{-} + Ar \rightarrow Ar^{-}(3p^{5}4s^{2}) \rightarrow e^{-} + Ar$$

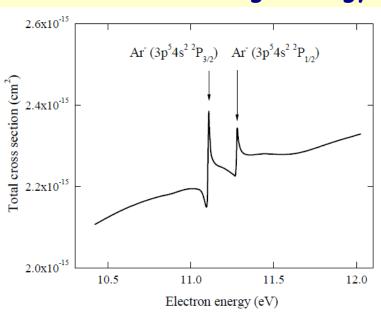
### Hypothesis of electron resonance trapping

### Mechanism of electron resonance trapping:

G.M. De'Munari et al., Electroluminiscence of rare gases and electron bremsstrahlung, Lett. Nuov. Cimento 2 (1971) 68

G.M. De'Munari et al., Effects of molecular gases on Xe electroluminescence and electron resonance trapping, Nuov. Cimento 3 (1984) 963

When the electron, accelerated by the electric field between the collisions, reaches the resonance energy, with high probability it is captured to form a negative ion state Ar-. The electron spends there a certain time, of about 0.5 ps, and then releases at somewhat lower energy, since part of the energy is transferred to the atom. Then the cycle repeats, which finally leads to trapping of a part of the electrons at the resonance energy and thus to the enrichment of the high-energy tail of the electron energy distribution function.

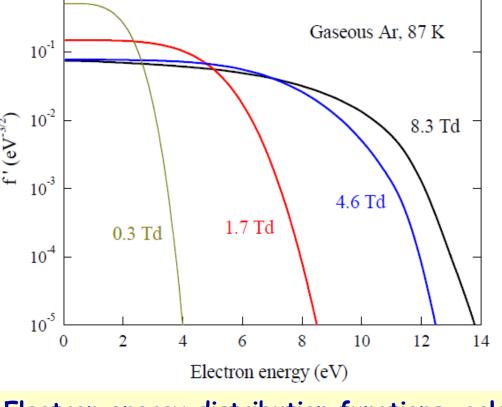


Experimental cross-section for electron scattering from Ar around Feshbach resonances [Kurokawa 2011]

$$e^{-} + Ar \rightarrow Ar^{-}(3p^{5}4s^{2}) \rightarrow e^{-} + Ar$$

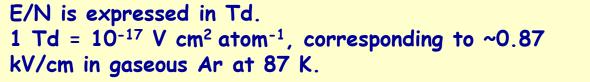
# NBrS EL theory: electron energy distribution functions

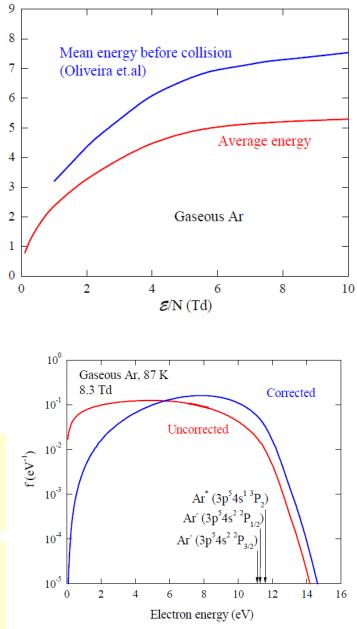
Energy (eV)



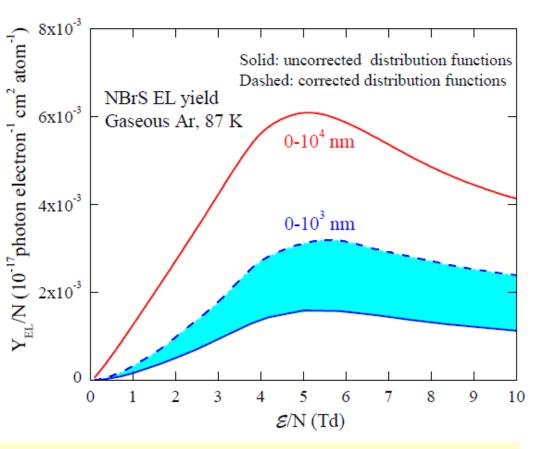
10<sup>0</sup>

Electron energy distribution functions, calculated using Boltzmann equation solver BOLSIG+ (free software)



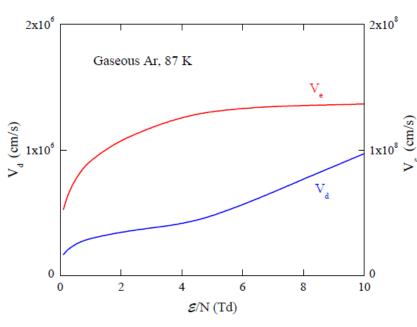


### NBrS EL theory: EL yield field dependence

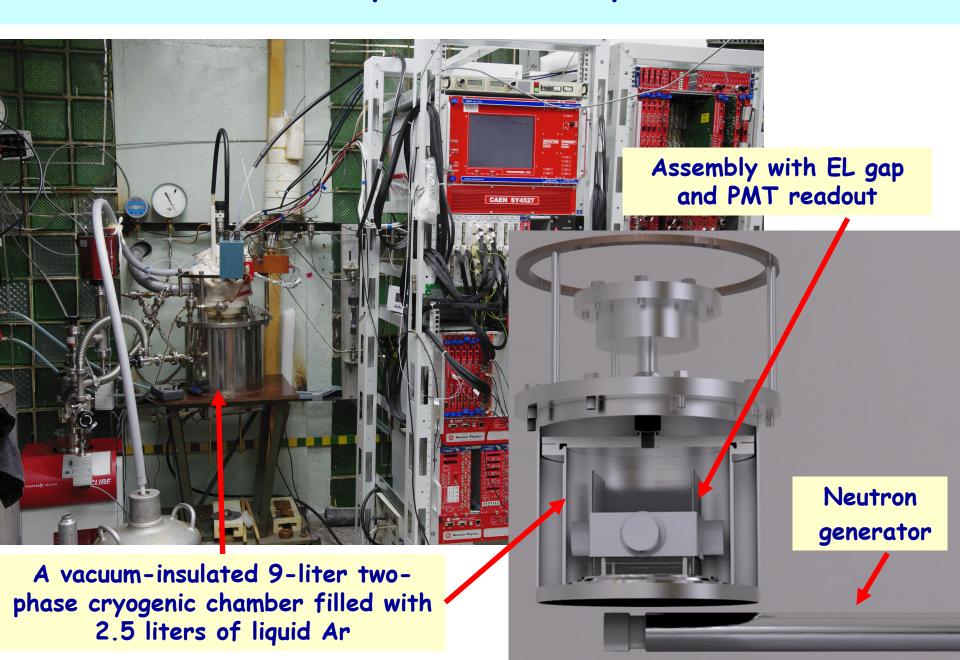


NBrS EL yield in the 0-1000 nm range represents the maximum number of NBrS photons that can ever be detected by existing devices.

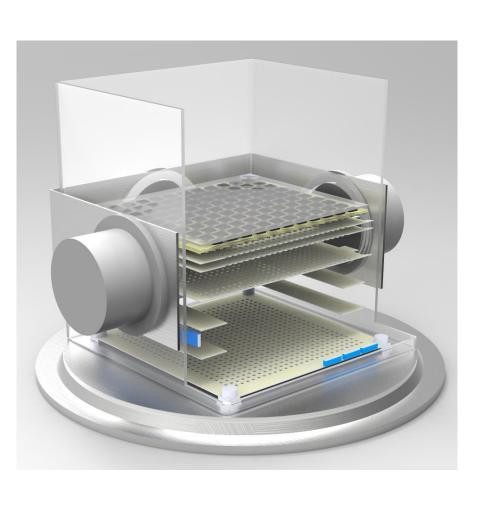
NBrS EL yield first increases, then saturates and even decreases with the field: this reflects  $v_e/v_d$  behavior  $\rightarrow$ 

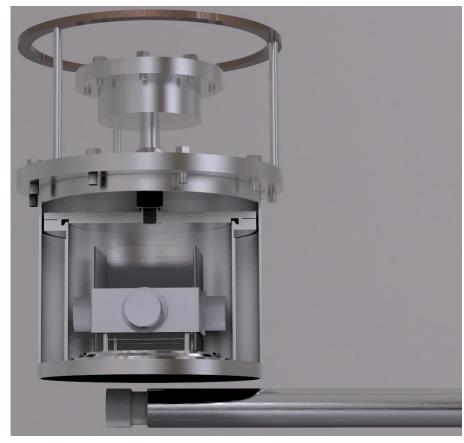


### Experimental setup

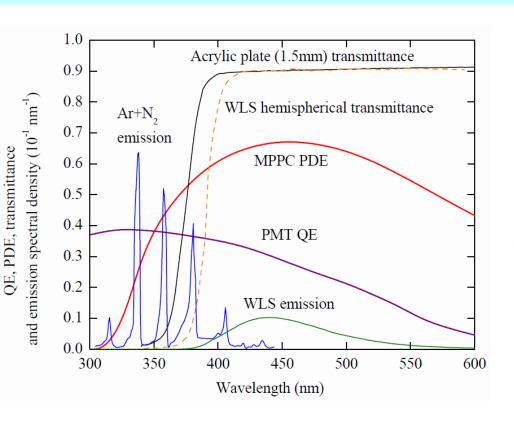


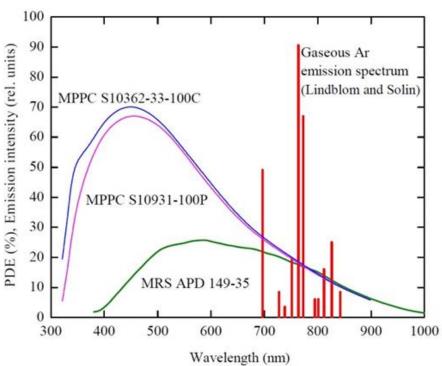
### Chamber 3D - view





### Optical spectra





## There is no rational mechanism other than that of NBrS

Physics of electroluminescence is an exact science: all the reaction rate constants of possible EL mechanisms in the presence of impurities are known, allowing to rule out their effect on electroluminescence at the given impurity content limits.

In our experiment this limit was below 1 ppm for  $N_2$ , Xe and other non-electronegative impurities in total, while for electronegative (oxygen-equivalent) impurities it was below 5 ppb (corresponding to the electron lifetime in liquid Ar exceeding 100 us).

With such vanishingly small impurity contents, there is only one known mechanism that can explain the EL effect under the Ar excitation threshold, namely that of NBrS.

Finally, regarding possible effect of dissolving of the TPB-based WLS in liquid Ar reported recently, it does not matter for our case since it acts only in the liquid phase. Moreover, it was shown that the WLS films composed of non-saturated TPB in polymer matrix, i.e. similarly to that used in our experiment (1 part of TPB per 3 parts of polystyrene), are resistant to dissolving in liquid Ar.

### Pulse shape: fast component fraction

$$p_{DS-50} = OEL_{fast}/(OEL_{fast} + OEL_{slow}) \sim 0.1$$
  $OEL_{fast}$  - Ordinary EL, fast component (11 ns)  $OEL_{fast}$  - Ordinary EL, slow component (3.2 us)  $OEL_{fast} + OEL_{slow} + k * LY_{without\ WLS} \equiv LY_{with\ WLS}$   $LY_{with\ WLS}$  - Light yield for detector with WLS  $LY_{with\ WLS}$  - Light yield for detector with WLS

 $k \sim \! 0.58$  – Coefficient to account different spectral sensitivity for NBrS for detector with and without WLS

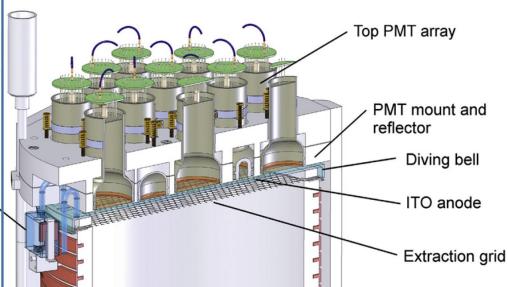
$$p_{new} = \left(OEL_{fast} + k * LY_{without WLS}\right) / \left(OEL_{fast} + OEL_{slow} + k * LY_{without WLS}\right) = \\ \left(OEL_{fast} + k * LY_{without WLS}\right) / LY_{with WLS} = \\ 1 - OEL_{slow} / LY_{with WLS} = \\ 1 - \left[ (1 - p_{DS-50}) / \left(OEL_{fast} + OEL_{slow}\right) \right] / LY_{with WLS} = \\ 1 - \left[ (1 - p_{DS-50}) / \left(LY_{with WLS} - k * LY_{without WLS}\right) \right] / LY_{with WLS} = \\ 1 - \left( 1 - p_{DS-50}\right) / \left( 1 - k * LY_{without WLS} / LY_{with WLS}\right)$$

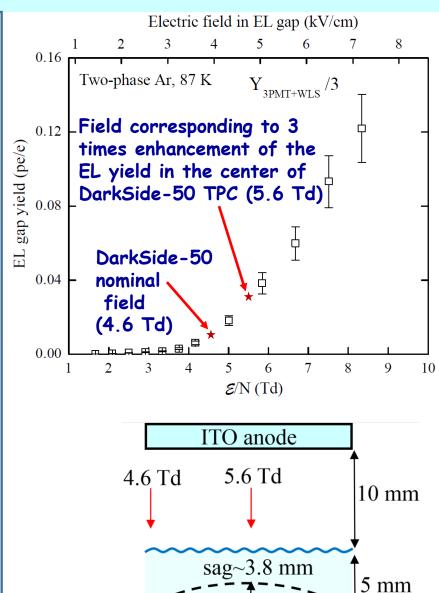
### Estimation of extraction grid sagging in DarkSide-50

Agnes et al, PLB 743 (2015) 456:

"...52 has a strong radial dependence, where events under the central PMT exhibit greater than three times more electroluminescence light than events at the maximum radius."

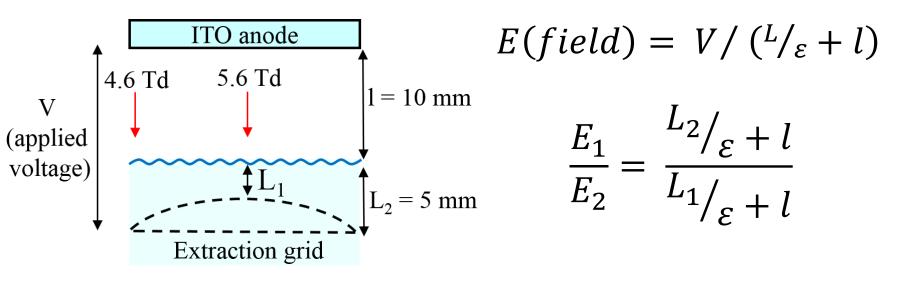
DarkSide-50 TPC operated at nominal EL eclectic field of 4.6 Td.





Extraction grid

### Estimation of extraction grid sagging



$$L_1 = \left[ \left( \frac{L_2}{\varepsilon} + l \right) / \frac{E_1}{E_2} - l \right] \varepsilon$$

$$sag = (L_2 - L_1) \sim 3.8 \ mm$$

### NBrS in avalanche scintillations in noble gases?

So far, direct recording of avalanche scintillations by SiPMs was considered to be provided by NIR emission of higher-level Ar excitation states  $Ar^*(3p^54p^1)$ 

It is not yet clear how large the contribution of **CCD Camera** NBrS electroluminescence is avalanche Sony ICX285AL scintillations, which are used in combined TPB coated THGEM/SiPM and THGEM/CCD multipliers. perspex disc Cryogenic webcam G-APD matrix Extraction grid Anode grid Resistor Source chain actuator Avalanche scintillations in NIR Field shaping Buzulutskov, THGEM1 JINST 7 Proportional scintillations Cathode Gaseous Ar in VUV and NIR (2012) C02025 protection grid Interface grid Hamamatsu Single e Liquid Ar Nuclear recoil Primary scintillations in VUV Mavrokoridis et al, JINST 9 (2014) **PMT** P02006

Radiation (v or WIMP)