Обнаружение дополнительного механизма электролюминесценции в двухфазных детекторах темной материи: тормозное излучение электронов на атомах

Revealing additional mechanism of proportional electroluminescence in two-phase dark matter detectors: neutral bremsstrahlung

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> Экспериментальный семинар ИЯФ 23/03/2018

Group of cryogenic and avalanche detectors for rare-event experiments

Group of Cryogenic and Avalanche Detectors is combined from Lab 3-3 (BINP) and Lab of Cosmology and Elementary Particles (NSU)

The group is operated in the frame of BINP and NSU research programs:

A. Bondar (head of Lab 3-3), A. Buzulutskov (leader of group), A. Dolgov (head of LCECh), A. Chegodaev, E. Frolov, V. Nosov, V. Oleynikov, T. Shakirova, L. Shekhtman, E. Shemyakina, R. Snopkov, A. Sokolov

We collaborate with S. Polosatkin on neutron scattering system and with V. Parkhomchuk and A. Petrozhitski on low-pressure TPC for ion ID.

We are members of DarkSide-20k collaboration

Outline

- 1. The problem of proportional electroluminescence (EL) in two-phase Ar
- 2. Neutral bremsstrahlung (NBrS): history and theory
- 3. Experimental setup
- 4. EL yields: experiment vs theory
- 5. Applications of NBrS EL
- 6. Conclusions

Section 1. The problem of proportional EL in two-phase Ar



Two-phase dark matter detectors: principles of operation



Photon emission and collisional processes in gaseous and liquid Ar doped with Xe and N_2



Proportional EL in GAr: ordinary EL – strong emission in VUV

Strong ordinary EL in the VUV (128 nm) due to excimers, via $Ar^*(3p^54s^1)$ excited states, at >4 Td:

8

9

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

 $\begin{array}{l} \operatorname{Ar}^*(3p^54s^1) + 2\operatorname{Ar} \to \operatorname{Ar}_2^*({}^{1,3}\Sigma_u^+) + \operatorname{Ar} \\ \operatorname{Ar}_2^*({}^{1,3}\Sigma_u^+) \to 2\operatorname{Ar} + h\nu \left(\operatorname{VUV}\right) \end{array}$





Satisfactory agreement between the microscopic theory and experiment

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 $E / p_{293K} [Vcm^{-1}Torr^{-1}]$

Proportional EL in GAr: EL in presence of N2 (~1%) – strong emission in near UV, competing to ordinary EL in the VUV

In the presence of N2 dopant, EL in the near UV due to excitation transfer from Ar to N2 and N2 emission of 2nd Positive System:

$$\begin{aligned} \operatorname{Ar}^{*}(3p^{5}4s^{1}) + \operatorname{N}_{2} \to \operatorname{Ar} + \operatorname{N}_{2}^{*}(C^{3}\Pi_{u}), \\ \operatorname{N}_{2}^{*}(C^{3}\Pi_{u}) \to \operatorname{N}_{2}^{*}(B^{3}\Pi_{g}) + h\nu. \end{aligned}$$

$$\overset{\operatorname{Ar}^{*}(3p^{5}4s^{1}) + \operatorname{N}_{2} \to \operatorname{Ar} + \operatorname{N}_{2}^{*}(C) \qquad k_{6} \sim 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} \\ k_{6} = 3.6 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} \\ k_{6} \geq 6.5 \times 10^{-9} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \end{aligned}$$

$$\overset{\operatorname{300 K}}{\operatorname{Ar}^{*}(44,46]} \\ \overset{\operatorname{300 K}}{\operatorname{300 K}} [44,46] \\ \overset{\operatorname{300 K}}{\operatorname{59}} 2.4 \, \mu \operatorname{s} \\ \overset{\operatorname{K}_{6} \geq 1.5 \times 10^{-9} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{6} \sim 2.15 \times 10^{-9} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{7} \sim 10^{-9} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \sim 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{7} \sim 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \sim 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \sim 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \sim 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \sim 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \sim 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \sim 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?) \\ \overset{\operatorname{K}_{8} \simeq 1.5 \times 10^{-11} \operatorname{cm}^{3} \operatorname{s}^{-1} (?)$$

The problem of proportional EL in two-phase Ar

 The problem of proportional EL identified in our works
 EPL, 112 (2015) 19001

 EPL, 117 (2017) 39002
 J. Instrum. 12 (2017) C05016
 , in two-phase Ar+N2 (10-50ppm):

 observation of non-VUV component (in the UV and visible range), in addition to that of VUV, of not that small intenstity;
 at such small N2 contents the appearance of the non-VUV emission could not be explained in a simple model of excitation transfer from Ar to N2



The problem of proportional EL in two-phase Ar

In this work, we partially resolved the problem: we have studied proportional electroluminescence in pure gaseous Ar, without additives, in the two-phase mode.

Surprisingly, we again observed a non-VUV component in EL radiation. Moreover, this component was observed not only at higher electric fields, but also at lower fields, below the Ar excitation threshold.

These unexpected observations made us recall the idea of an additional EL mechanism in two-phase Ar, namely that of neutral bremsstrahlung (NBrS), that acts concurrently with the ordinary mechanism.

Section 2. Neutral bremsstrahlung (NBrS): history and theory

NBrS history

NBrS is produced by slow electrons when they are scattered on neutral atoms, at electron energies of the order of 1–10 eV [Firsov and Chibisov 1961, Kasyanov and Starostin 1965, Dalgarno and Lane 1966]. The NBrS effect was used to explain continuous emission spectra in a weakly ionized plasma.

More interesting is that back in 1970, the NBrS mechanism was proposed as an explanation for proportional electroluminescence in xenon [Butikov et al. 1970]. In the subsequent work [DeMunari and Mambriani 1971], it was stated that in Xe the EL rate agrees, in order of magnitude, with the calculated NBrS rate of light production.

However this statement was refuted in [Dias at al. 1986]: "... mechanisms based on the direct excitation of the noble gas atoms by the electrons can fully account for the secondary light production. There is no need to consider other processes like ... neutral bremsstrahlung".

Since then bremsstrahlung in connection with electroluminescence was almost forgotten. It was mentioned only once as a possible explanation of the underthreshold electroluminescence in gaseous krypton in book [Barabash and Bolozdynya 1993], later reproduced in books [Aprile et al 2006] and [Bolozdynya 2010].

NBrS history

In the present work we show that at least for Ar the NBrS mechanism is definitely needed to explain the properties of proportional electroluminescence: we developed the theory of NBrS electroluminescence that for the first time quantitatively described the experiment at lower fields, below the excitation threshold, and that have chances to do it at higher fields, above the threshold.

What has changed since the 70s, that has allowed us to develop a quantitative theory? \rightarrow Just one thing: correct calculations of the electron energy distributions functions using Boltzmann equation solver (free software).

Accordingly, this work might be considered as a revival of the idea of NBrS electroluminescence, on a new higher level.

NBrS EL theory



Ordinary bremsstrahlung



Polarization bremsstrahlung



Neutral bremsstrahlung in elastic scattering e hv

Neutral bremsstrahlung in inelastic scattering $e^- + A \rightarrow e^- + A + h\nu$, $e^- + A \rightarrow e^- + A^* + h\nu$.

NBrS EL theory: basic equations

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

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

in photon/(s nm electron),

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

in (photon cm^2)/(electron atom)

$$\frac{d(Y_{EL}/N)_{NBrS}}{d\lambda} = \int_{h\nu}^{\infty} \frac{\upsilon_e}{\upsilon_d} \frac{d\sigma}{d\nu} \frac{d\nu}{d\lambda} f(E) dE$$

in (photon cm²)/(electron atom nm),

NBrS EL theory: basic equations

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

Electron energy distribution function normalization: two ways

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

Ordinary electroluminescence, involving excimers

NBrS EL theory: cross-sections



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

NBrS EL theory: photon emission spectra



Ar and Xe.

E/N is expressed in Td. 1 Td = 10^{-17} V cm² atom⁻¹, corresponding to ~0.87 kV/cm in gaseous Ar at 87 K.

NBrS spectra are rather flat.

Inelastic collision contribution can be neglected



NBrS EL theory: EL yield field dependence



V,

8

E/N (Td)

10

0

2

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

EL yield theory: NBrS EL vs ordinary EL



Summarizing, the theory of NBrS EL predicts:

1) electroluminescence below the Ar excitation threshold, in the UV, visible and NIR regions;

2) appreciable non-VUV component above the Ar excitation threshold, extending from the UV to NIR.

Section 3. Experimental setup



Experimental setup



Experimental setup







Experimental setup



- 3 types of photosensors provide 3 different spectral ranges of optical readout of S2:
- in VUV (around 128 nm), using PMT+WLS;
- in near UV and visible (300-600 nm), using mostly bare PMT (+UV acrylic);
- in visible and NIR (400-1000 nm), using SiPM (+acrylic).

Section 4. EL yields: experiment vs theory



EL gap yield



 $Y_{XX} = N_{pe}/q_e$; XX=1PMT, 3PMT+WLS or SiPM

- Electroluminescence below Ar excitation threshold (at 4.0 Td), where non-VUV component fully dominates.
- Substantial contribution of non-VUV component above the threshold.
- Strong response of bare PMT: as large as 40% of PMT+WLS amplitude at higher field, at 8.3 Td. At lower fields, below 4.6 Td (that of DarkSide-50), bare PMT amplitude starts exceeding that of the PMT+WLS

EL gap yield



Non-VUV component is well described by NBrS theory, below Ar excitation threshold.
Above the threshold, the theory quickly diverges from experiment.

Back in 80s, it was suggested how to eliminate such a discrepancy:

- To account for possible electron trapping at Feshbach resonance energies, which leads to enrichment of the high-energy tail of the electron energy distribution function [De'Munari 1971,1984].

- It was theoretically demonstrated that the NBrS yield at resonance is significantly (>3 times) enhanced [Dallacasa and Leonardis 1980].

Relying on these hypotheses, we adopt here the paradigm that all the data on non-VUV component are those induced by NBrS electroluminescence.

Reduced EL yield in the VUV



Reduced EL yield for ordinary (VUV) electroluminescence obtained in this work and compared to the yields at room T obtained experimentally and theoretically [Oliveira 2011]. \rightarrow Convincing agreement between the theory and our experiment, the latter using NBrS EL paradigm on non-VUV component origin. True EL gap yield for ordinary (VUV) electroluminescence from 3PMT+WLS data, where non-VUV component is subtracted using 1PMT and SiPM data, the shapes of emission spectra being provided by NBrS theory.



Summary of experimental EL yield in gaseous Ar



Section 5. Applications of NBrS EL

Resolving the problem of proportional electroluminescence in two-phase Ar: the admixture of N2 to Ar at minor contents (~50 ppm) cannot be responsible for the non-VUV component; most probably it is NBrS responsible for that.

The amplitude of the S2 signal from the bare PMT (without WLS) is comparable with that of the PMT with WLS (in the absence of optical contact between the WLS and the PMT): paves the ways to direct PMT and SiPM-matrices optical readout of S2.

The presence of NBrS component in proportional EL may result in suggesting to analyze the S2 pulse-shape in a new way, in particular in two-phase Ar dark matter detectors.

Due to universal character of NBrS EL effect, it should be present in other noble gases. In particular, NBrS EL should be present in S2 signals of two-phase Xe detectors.

NBrS electroluminescence should be present in avalanche scintillations, which are used in combined THGEM/SiPM and THGEM/CCD multipliers.

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

It is possible that NBrS emission is present also in S1 signals, i.e. in primary scintillations in liquid Ar, in the form of weak scintillations in the visible and NIR range observed earlier by a number of groups.

Summary

An additional mechanism of proportional electroluminescence (EL) in two-phase dark matter detectors, namely that of neutral bremsstrahlung (NBrS), has been studied. It explains the non-VUV spectral component and photon emission below the Ar excitation threshold, thus partially resolving the problem of proportional electroluminescence in two-phase Ar.

The merit of the present work is that it transformed the idea of NBrS electroluminescence from a hypothesis into a quantitative theory. This allowed to correctly determine the EL yield of proportional electroluminescence in pure gaseous Ar, at cryogenic temperature in the two-phase mode.

The main practical application of the NBrS effect is a better understanding of the S2 signal and justification for its direct (without WLS) optical readout using PMTs and SiPM-matrices, which may help to develop two-phase dark matter and neutrino detectors of ultimate sensitivity.

Backup slides



Example of two-phase detector: DarkSide-50



Fig. 2. The DarkSide-50 liquid argon time projection chamber.

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Drift field in LAr 200 V/cm Extraction field in LAr 2.8 kV/cm

EL gap field 4.2 kV/cm, at a pressure of 1080 mbar, corresponding to E/N=4.6 Td

E/N is expressed in Td 1 Td = 10⁻¹⁷ V cm² atom⁻¹, corresponding to ~0.87 kV/cm in gaseous Ar at 87 K.

Photon emission and collisional processes in gaseous and liquid Ar doped with Xe and N_2

Table 1: Basic reactions of excited species relevant to the performance in the two-phase mode, namely in Ar in the gas and liquid phase, doped with Xe (1000 ppm in the liquid and 40 ppb in the gas phase) and N₂ (50 ppm in the liquid and 135 ppm in the gas phase), their rate (k) or time (τ) constants reported in the literature and their time constants reduced to given atomic densities at 87 K (τ_{TP}), in particular for Ar to that of 8.63 × 10¹⁹ cm⁻³ and 2.11 × 10²² cm⁻³ in the gas and liquid phase, respectively.

No.	Reaction	$k \mbox{ or } \tau$	T	Reference	τ_{TP}
	Gaseous Ar + Xe (40 ppb) + N ₂ (135 ppm)				
(1)	$\overline{\operatorname{Ar}^*(3p^54s^1) + 2\operatorname{Ar}} \to \\ \operatorname{Ar}^*_2({}^{1,3}\Sigma^+_+) + \operatorname{Ar}$	$k_1 \sim 1 \times 10^{-32} \mathrm{cm}^6 \mathrm{s}^{-1}$	$300{\rm K}$	[44-47]	${\sim}13\mathrm{ns}$
(2)	$\operatorname{Ar}_{2}^{2}(\overset{1,3}{\overset{1}{\overset{1}{\overset{1}{\overset{1}{\overset{1}{\overset{1}}{\overset{1}{\overset{1}{\overset{1}{\overset{1}}{\overset{1}{\overset{1}{\overset{1}{\overset{1}{\overset{1}}{\overset{1}}{\overset{1}}{\overset{1}}{\overset{1}}}}}}}}$	$\tau_2({}^1\Sigma_u^+) = 4.2 \mathrm{ns}$	$300\mathrm{K}$	[48,49]	$4.2\mathrm{ns}$
2.4	2. 0, , , ,	$\tau_2({}^3\Sigma_u^+) = 3.0 - 3.2\mu s$	$300\mathrm{K}$	[12, 47-51]	$3.1\mu{ m s}$
(3)	$\operatorname{Ar}^*(3p^54p^1) \rightarrow$	$\tau_3 = 20{-}40 \mathrm{ns}$	$300\mathrm{K}$	[34, 52, 53]	
	$\operatorname{Ar}^*(3p^54s^1) + h\nu(\operatorname{NIR})$	$\tau_3 < 100 {\rm ns}$	$163\mathrm{K}$	[54-56]	$< 100 \mathrm{ns}$
(4)	$\operatorname{Ar}^*(3p^54s^1) + \operatorname{Xe} \to \operatorname{Ar} + \operatorname{Xe}^*$	$k_4 = (2-3) \times 10^{-10} \mathrm{cm}^3 \mathrm{s}^{-1}$	$300\mathrm{K}$	[13, 57]	$\sim 1\mathrm{ms}$
(5)	$\operatorname{Ar}_{2}^{*}(^{3}\Sigma_{u}^{+}) + \operatorname{Xe} \rightarrow$	$k_5 \sim 5 \times 10^{-10} \mathrm{cm}^3 \mathrm{s}^{-1}$	$300\mathrm{K}$	[12, 13, 58]	$\sim 0.6 \mathrm{ms}$
(α)	$2\text{Ar} + \text{Xe}^{*}({}^{1}P_{1}, {}^{3}P_{0})$	1 1 5 10-11 3 -1	20017	[11.10]	
(6)	$\operatorname{Ar}^{*}(3p^{5}4s^{*}) + \operatorname{N}_{2} \to \operatorname{Ar} + \operatorname{N}_{2}(C)$	$k_6 \sim 1.5 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$	300 K	[44,46]	0.4
		$\kappa_6 = 3.6 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$	300 K	[09]	$2.4 \mu s$
(7)	$A_{n}^{*}(2n54n1) + N$	$k_6 \ge 0.5 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$	87 K	[11]	$\leq 13 \text{ ns}(?)$
(i)	$Ar (3p 4s^{-}) + N_2 \rightarrow Ar + N^* (C B A)$	$k_7 \sim 3 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$	300 K	[44,40] [57,50]	
(8)	$N^*(C) \rightarrow$	$x_7 = 3.0 \times 10^{-10}$ cm s	300 K	[44,46,60]	35 ne
(0)	$N_2^{\circ}(B) + h\nu$ (UV 2nd pos. sys.)	78 = 50 40 113	500 IX	[11,10,00]	55 115
(9)	$N_2(B) \rightarrow$	$\tau_9 \sim 9 \mu s$	$300\mathrm{K}$	[44]	$\sim 9 \mu s$
	$N_2^*(A) + h\nu$ (NIR, 1st pos. sys.)	5	$119\mathrm{K}$	[38]	
(10)	$N_2^*(C) + Ar \rightarrow N_2^*(B) + Ar$	$k_{10} = 5.6 \times 10^{-13} \mathrm{cm}^3 \mathrm{s}^{-1}$	$300\mathrm{K}$	[44]	$21\mathrm{ns}$
(11)	$N_2^{*}(B) + Ar \rightarrow N_2^{*}(A) + Ar$	$k_{11} = 1.4 \times 10^{-14} \mathrm{cm}^3 \mathrm{s}^{-1}$	$300\mathrm{K}$	[44]	$0.8\mu s$
(12)	$N_2^*(C) + N_2 \to N_2 + N_2^*(B)$	$k_{12} \sim 1 \times 10^{-11} \mathrm{cm}^3 \mathrm{s}^{-1}$	$300\mathrm{K}$	[44, 60]	$\sim 8.6 \mu s$
(13)	$N_2^*(B) + N_2 \rightarrow N_2 + N_2^*(A)$	$k_{13} \sim 1 \times 10^{-11} \mathrm{cm}^3 \mathrm{s}^{-1}$	$300\mathrm{K}$	[44]	${\sim}8.6\mu{ m s}$
(14)	$\operatorname{Ar}_{2}^{*}(^{3}\Sigma_{u}^{+}) + \operatorname{N}_{2} \to 2\operatorname{Ar} + \operatorname{N}_{2}^{*}(B)$	$k_{14} \sim 3.3 \times 10^{-12} \mathrm{cm}^3 \mathrm{s}^{-1}$	$300\mathrm{K}$	[44, 58]	${\sim}26\mu{\rm s}$
	Liquid Ar + Xe (1000 ppm) + N ₂ (50 ppm)				
(15)	$\overline{\operatorname{Ar}^*(n=1,{}^2P_{1/2,3/2})} + \operatorname{Ar} \rightarrow$	$\tau_{15} = 6 \mathrm{ps}$	$87\mathrm{K}$	[25, 61]	6 ps
3 D	$Ar_{2}^{*}(^{1,3}\Sigma_{u}^{+})$				
(16)	$\operatorname{Ar}_{2}^{*}(^{1,3}\Sigma_{u}^{+}) \to 2\operatorname{Ar} + h\nu (\operatorname{VUV})$	$\tau_{16}({}^1\Sigma_u^+) = 7\mathrm{ns}$	$87\mathrm{K}$	[8-10, 62]	$7\mathrm{ns}$
	5 X 3	$\tau_{16}({}^{3}\Sigma_{u}^{+}) = 1.6\mu \mathrm{s}$			$1.6\mu s$
(17)	$\operatorname{Ar}_{2}^{*}(^{1,3}\Sigma_{u}^{+}) + \operatorname{Xe} \rightarrow$	$k_{17}({}^{3}\Sigma_{u}^{+}) \sim$	$87\mathrm{K}$	[17-19]	$\sim 5.3 \mathrm{ns}$
	$2\text{Ar} + \text{Xe}^*(n = 1, 2, {}^2P_{3/2})$	$(0.8-1) \times 10^{-11} \mathrm{cm}^3 \mathrm{s}^{-1}$			100
		$\tau_{17}({}^{3}\Sigma_{u}^{+}) < 90 \mathrm{ns}$	87 K	[18,20]	$< 90 \mathrm{ns}$
(10)	$V_{*}(-1,0,2D) + A_{-} + A_{-}V_{*}^{*}$	$k_{17}(\Sigma_u^+) \sim 3.3 \times 10^{-11} \mathrm{cm^3 s^{-1}}$	87 K	[19]	$\sim 1.4 \mathrm{ns}$
(18) (10)	$Ae^{*}(n = 1, 2, {}^{*}P_{3/2}) + Ar \rightarrow ArAe$	Immediate trapping	87 K	[19]	< 20 mg
(19)	ArXe ⁺ + Xe \rightarrow Ar + Xe ₂ ($^{\circ} \Sigma_{u}^{\circ}$)	$\tau_{19} \leq 20 \text{ hs}$	87 K	[20]	$\leq 20 \text{ ns}$
(20)	$Xe^{(n=1,2,-F_{3/2})+Xe} \rightarrow Xe^{*(1,3\Sigma^+)}$		01 K	[10]	
(21)	$\operatorname{Xe}_{2}^{*}(\overset{1,3}{\Sigma}_{+}^{+}) \rightarrow 2\operatorname{Xe} + h\nu (\mathrm{UV})$	$\tau_{21}({}^{1}\Sigma_{-}^{+}) = 4.3 \mathrm{ns}$	$165\mathrm{K}$	[9,62]	$4.3\mathrm{ns}$
()		$\tau_{21}({}^{3}\Sigma_{u}^{a}) = 22 \mathrm{ns}$	$165\mathrm{K}$	[]	$22\mathrm{ns}$
(22)	$Xe^*(n=2, {}^2P_{3/2}) \rightarrow$	$\tau_{22} < 170 \mathrm{ns}$	$87\mathrm{K}$	[21, 22]	$<\!170\mathrm{ns}$
	$Xe^*(n = 1, {}^2P_{3/2}) + h\nu$ (NIR)			L 7 1	
	Reactions (17)–(21) in total ($\tau_{17} + \tau_1$	9):			$\leq 110 \mathrm{ns}$
		8			
(23)	$\operatorname{Ar}_{2}^{*}(^{3}\Sigma_{u}^{+}) + \operatorname{N}_{2} \to 2\operatorname{Ar} + \operatorname{N}_{2}^{*}(B)$	$k_{23} = 3.8 \times 10^{-12} \mathrm{cm}^3 \mathrm{s}^{-1}$	$87\mathrm{K}$	[24, 25]	$250\mathrm{ns}$
(24)	$\operatorname{ArXe}^* + \operatorname{N}_2 \to \operatorname{Ar} + \operatorname{Xe} + \operatorname{N}_2^*(B, A)$		$87\mathrm{K}$		
(25)	$\operatorname{Xe}_{2}^{*}(^{3}\Sigma_{u}^{+}) + \operatorname{N}_{2} \to 2\operatorname{Xe} + \operatorname{N}_{2}^{*}(B, A)$	-	$87\mathrm{K}$		

All that was known about proportional electroluminescence before the present study was reflected in [A.Buzulutskov EPL 117 (2017) 39002]

Proportional EL in GAr: ordinary EL - strong emission in VUV

Strong ordinary EL in the VUV (128 nm) due to excimers, via $Ar^*(3p^54s^1)$ excited states, at >4 Td:

$$e^{-} + A \rightarrow e^{-} + A^{*}$$

Ar* $(3p^{5}4s^{1}) + 2$ Ar \rightarrow Ar^{*}₂ $(^{1,3}\Sigma^{+}_{u}) +$ Ar
Ar^{*}₂ $(^{1,3}\Sigma^{+}_{u}) \rightarrow 2$ Ar $+ h\nu$ (VUV)





Proportional EL in GAr: additional EL – weak emission in NIR at higher fields

Weak EL in the NIR via ${\rm Ar}^*(3p^54p^1)$ excited states, at higher electric fields, at >6 Td



EPL, **94** (2011) 52001

NBrS EL theory: electron energy distribution functions



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

E/N is expressed in Td. 1 Td = 10⁻¹⁷ V cm² atom⁻¹, corresponding to ~0.87 kV/cm in gaseous Ar at 87 K.

NBrS EL theory: electron energy distribution functions



kV/cm in gaseous Ar at 87 K.

NBrS EL theory: uncorrected and corrected electron energy distribution functions



Electron energy distribution functions: correction for mean energy before collisions.

 \rightarrow Defines the limits of theoretical uncertainty (factor of 2).

Summary of experimental EL yield in gaseous Ar

