

# ***Effects of Nitrogen and Oxygen contamination in Liquid Argon***

***Roberto Acciarri***

***Università degli Studi dell'Aquila, Italy***

***INFN-Laboratori Nazionali del Gran Sasso, Italy***

***on behalf of WArP Collaboration***

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# WARP Collaboration



## WARP Collaboration

R.Acciarri<sup>a</sup>, M.Antonello<sup>b,a</sup>, B. Baibussinov<sup>c</sup>, M.Baldo-Ceolin<sup>d</sup>, P.Benetti<sup>e</sup>, F.Calaprice<sup>g</sup>,  
E.Calligarich<sup>f</sup>, M.Cambiaghi<sup>e</sup>, N.Canci<sup>a</sup>, F.Carbonara<sup>h</sup>, F.Cavanna<sup>a</sup>, S. Centro<sup>d</sup>, A.G.Cocco<sup>i</sup>,  
F.Di Pompeo<sup>a,b</sup>, G.Fiorillo<sup>h</sup>, C.Galbiati<sup>g</sup>, V. Gallo<sup>i</sup>, L.Grandi<sup>b,a</sup>, G. Meng<sup>c</sup>, I.Modena<sup>a</sup>,  
C.Montanari<sup>f</sup>, O.Palamara<sup>b</sup>, L.Pandola<sup>b</sup>, F. Pietropaolo<sup>c</sup>, G.L.Raselli<sup>f</sup>, M.Roncadelli<sup>f</sup>,  
M.Rossella<sup>f</sup>, C.Rubbia<sup>b</sup>, E.Segreto<sup>b</sup>, A.M.Szelc<sup>j,a</sup>, F.Tortorici<sup>b</sup>, S. Ventura<sup>c</sup>, C.Vignoli<sup>f</sup>

<sup>a</sup> *Università dell'Aquila e INFN, L'Aquila, Italy*

<sup>b</sup> *INFN - Laboratori Nazionali del Gran Sasso, Assergi, Italy*

<sup>c</sup> *INFN - Sezione di Padova, Padova, Italy*

<sup>d</sup> *Università di Padova e INFN, Padova, Italy*

<sup>e</sup> *Università di Pavia e INFN, Pavia, Italy*

<sup>f</sup> *INFN - Sezione di Pavia, Pavia, Italy*

<sup>g</sup> *Princeton University - Princeton, New Jersey, USA*

<sup>h</sup> *INFN - Sezione di Napoli, Napoli, Italy*

<sup>i</sup> *Università di Napoli e INFN, Napoli, Italy*

<sup>j</sup> *IFJ PAN, Krakow, Poland*

# Background

- Dark Matter (DM) search is of primary interest in the present astroparticle physics scenario. Direct detection of DM with noble gases liquified as target medium is one of the most promising line of development in experimental technology.
- The two-phase (liquid-gas) technology developed by WArP is based on the simultaneous detection of both signals produced by ionization events in liquid Argon (LAr): free electron charge and scintillation light.
- Ionization charge quenching by electro-negative impurities (mainly Oxygen) present at residual level in commercial grade Argon has been widely studied. Purification technique is well mastered and normally employed in experimental applications.
- Effects of impurities on scintillation light yield are less precisely explored. Quenching of the light yield in  $N_2$  and  $O_2$ -contaminated LAr has been found. This effect is important because it imposes limitations on the collection of light and deteriorates the capabilities of the detector to discriminate background events (electrons recoils from  $\gamma$  events) from the signal (Ar-recoils potentially induced by WIMP interactions).

# Scintillation light in Liquid Argon

- ✓ The interactions of ionizing particles in LAr cause the formation of both electron-hole (ion) pairs  $e^- - Ar^+$  and excited atoms  $Ar^*$  ( $Ar^*/Ar^+ \sim 0.21$ ).
- ✓ Both states lead to the formation (with times of the order of tens of picoseconds) of the excited dimer  $Ar_2^*$  ( $^1\Sigma_u$  singlet state and  $^3\Sigma_u$  triplet state).
- ✓ The de-excitation of the excited dimer:  $Ar_2^* \rightarrow 2Ar + \gamma$  produces a Vacuum Ultra Violet (VUV) photon with  $\lambda = 128$  nm ( $\sigma \sim 3$  nm).

## Time dependence of pure LAr scintillation light emission

$$\mathcal{L}(t) = \frac{A_S}{\tau_S} \exp\left(\frac{-t}{\tau_S}\right) + \frac{A_T}{\tau_T} \exp\left(\frac{-t}{\tau_T}\right)$$

+ possible intermediate component with  $\tau_i \sim 40$  ns

$$\tau_S (^1\Sigma_U) = 2 \div 7 \text{ ns}$$

$$\tau_T (^3\Sigma_U) = 110 \div 170 \text{ ns}$$

$$\int \mathcal{L}(t) dt = A_S + A_T = 1$$

$A_S/A_T$  depends on ionizing radiation (PSD discrimination)

# Time dependence of X<sub>2</sub>-contaminated LAr scintillation light emission

The quenching process can be sketched as:



Where X can represent either Nitrogen or Oxygen molecule.

This **non-radiative collisional reaction**, in competition with de-excitation process leading to VUV emission, brings to a reduction of Ar<sub>2</sub><sup>\*</sup>.

$$I'(t) = \frac{A'_S}{\tau'_S} \exp\left(\frac{-t}{\tau'_S}\right) + \frac{A'_T}{\tau'_T} \exp\left(\frac{-t}{\tau'_T}\right)$$

$$\frac{1}{\tau'_j} ([X_{\gamma}]) = \frac{1}{\tau_j} + k[X_{\gamma}]$$

$$A'_j ([X_{\gamma}]) = \frac{A_j}{1 + \tau_j k [X_{\gamma}]}$$

**k**: quenching process  
rate constant

**[X<sub>2</sub>]**: contaminant  
concentration

$$\int I'(t) dt = A'_S + A'_T \leq I$$

$$Q_F = A'_S + A'_T$$

$$I \leq Q_F \leq I$$

# *Sketch of the work*

Our work was dedicated to a systematic study of the effects of Oxygen and Nitrogen contamination in LAr.

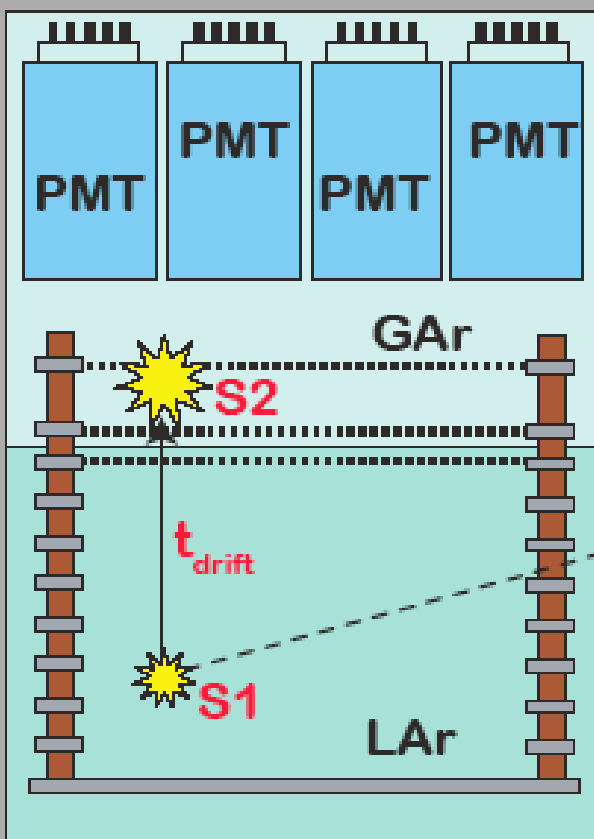
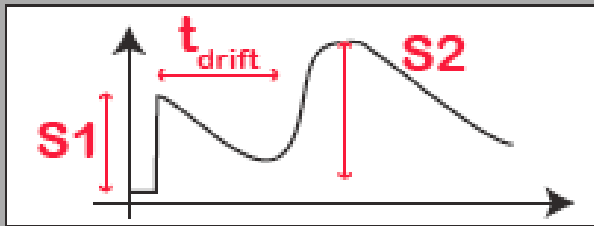
In particular:

- to determine the overall quenching factor ( $Q_F$ ) affecting the LAr scintillation light yield at various  $O_2$  and  $N_2$  concentrations;
- to verify (or rule out) the onset of an intermediate scintillation component;
  - to determine the effective lifetimes  $\tau_j'$  as a function of the  $O_2$  and  $N_2$  contamination, as well as the quenched amplitudes  $A_j'$ ;
- to extract the main characteristics of the scintillation light emission in pure LAr and the value of the rate constant  $k$  of the quenching process associated to the presence of either  $O_2$  or  $N_2$  contaminant.

Two detectors have been used for this purpose: the WArP 2.3 lt prototype and a small (0.7 lt) dedicated detector, coupled with a system for the injection of controlled amounts of gaseous contaminants.

# WArP 2.3 It prototype

Integrated light signal



➤ Particle hits Ar atom, causing ionization and excitation.

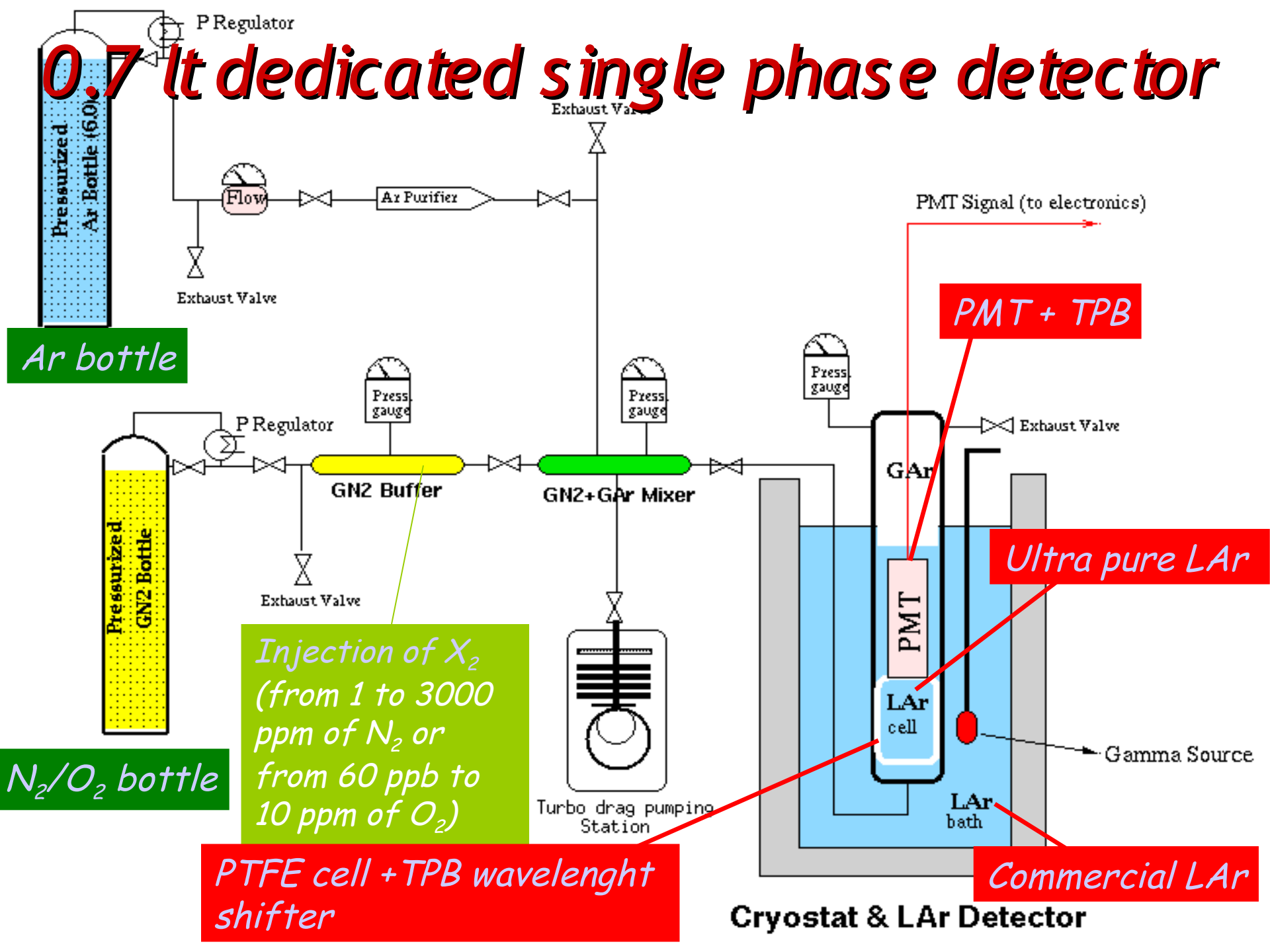
➤ Partial recombination of electron-ion pairs produces scintillation S1 in LAr.

➤ Remaining electrons from ionization are drifted by the constant field to the gas-liquid interface.

➤ Electrons are extracted from liquid by E2 and accelerated in Gas to produce scintillation S2.

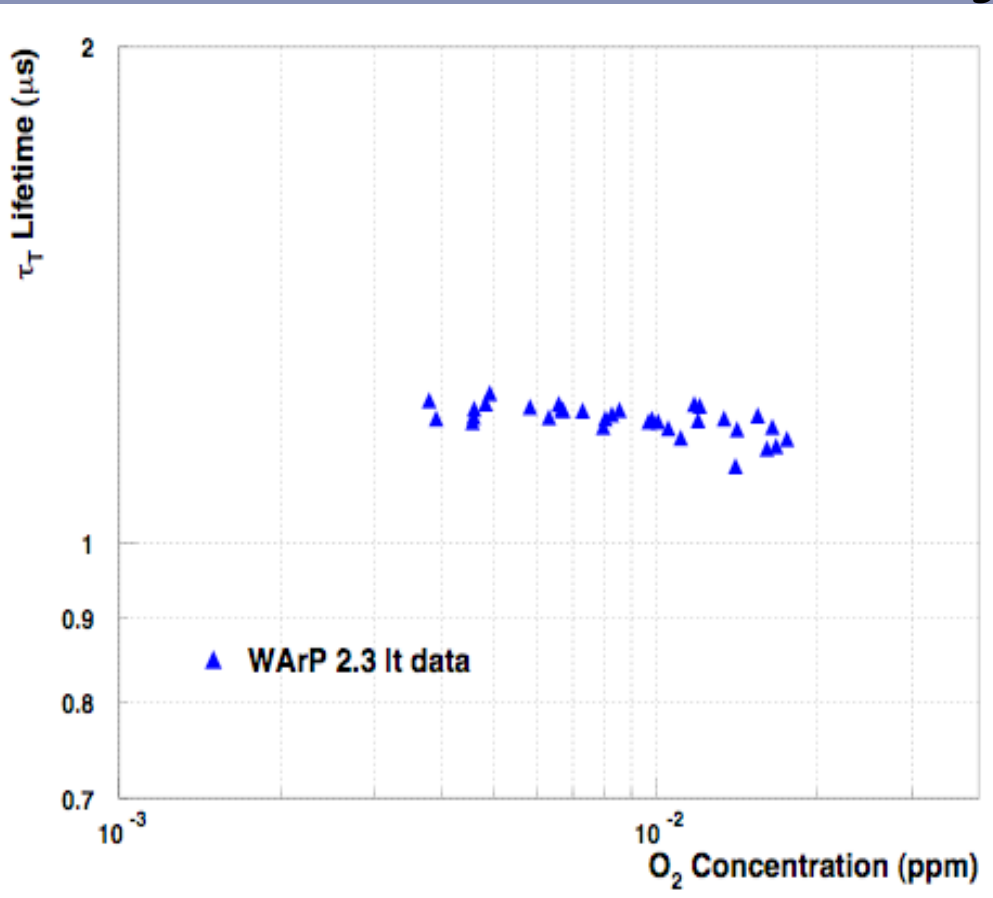
➤ Anode signal from each PMT is integrated (shaping time 120  $\mu$ s) and sent to a 10 bit Flash ADC with 100 MHz sampling frequency.

# 0.7 lt dedicated single phase detector



# Measurements with WArP prototype: $O_2$ contamination

*From WArP data is possible to determine both the absolute  $[O_2]$  and the slow component decay time ( $\tau_T$ ) (determined by fitting the shape of background electron signals).*



➤ Variation on  $[O_2]$  is due to activation of the recirculation/purification system

➤  $\tau_T$  is almost constant in the range below  $\approx 20$  ppb

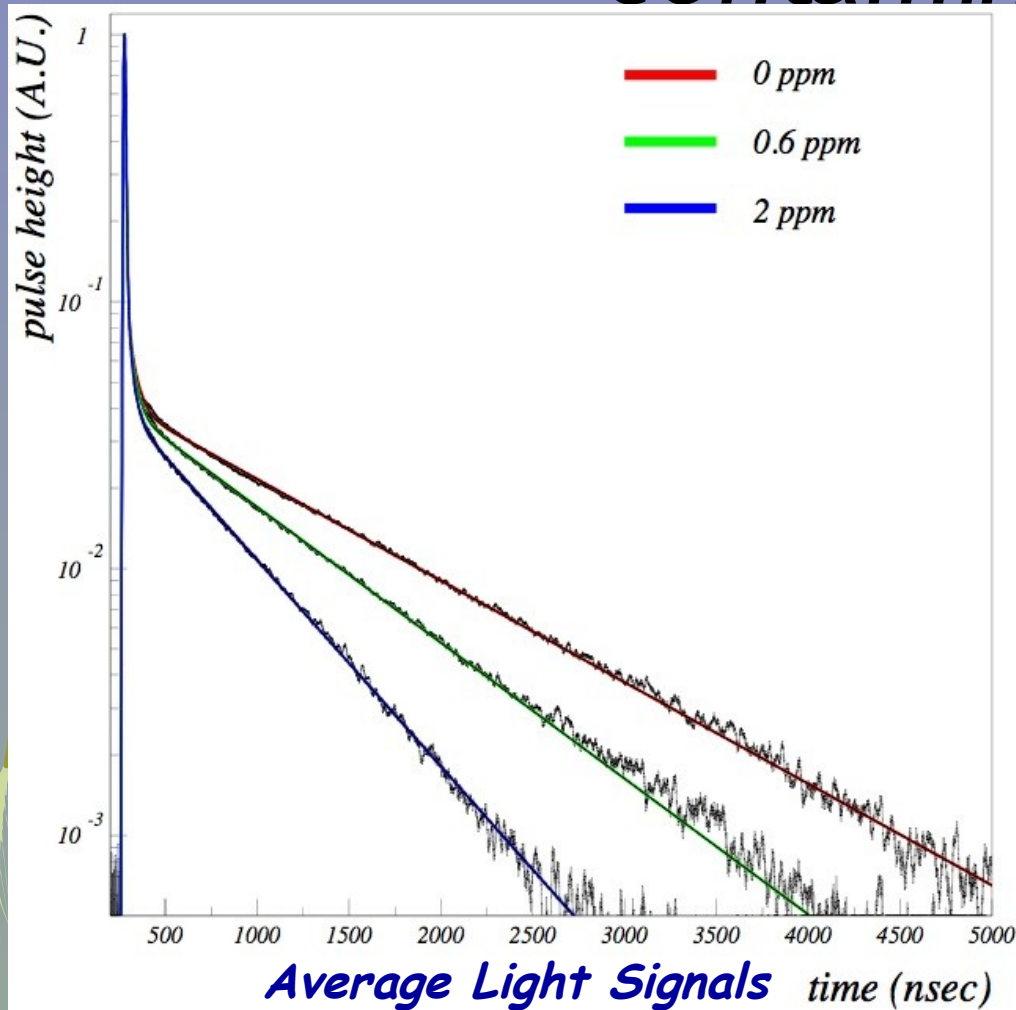


Scintillation emission is roughly unaffected by the quenching process at this low contamination

# Test with 0.7 lt detector: O<sub>2</sub>

Wfm recording of PMT signal (1 ns sampling time) allows for a detailed study of the LAr scintillation: individual components, relative amplitude and decay time.

## contamination



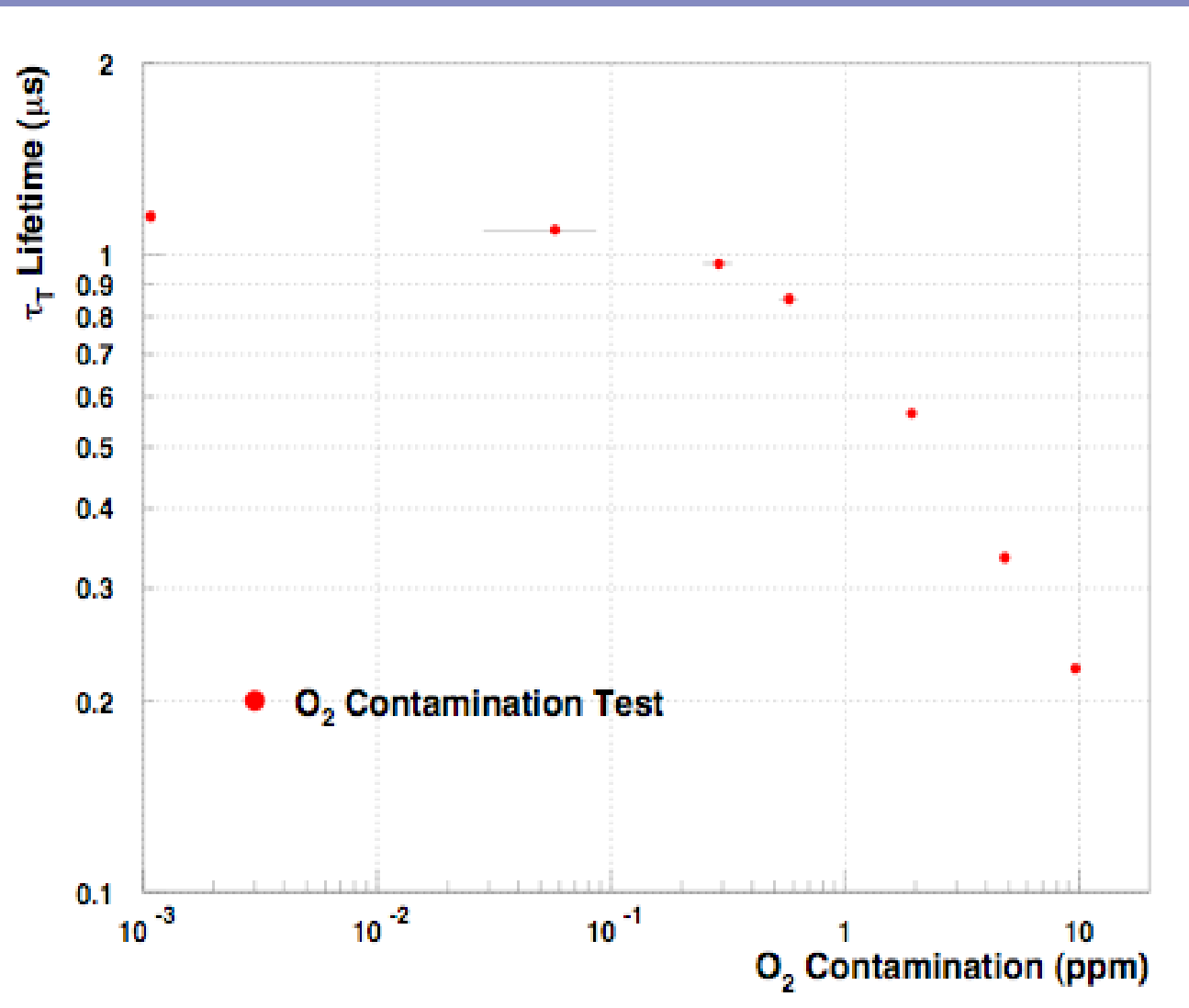
➤ Argon was progressively contaminated with increasing quantities of O<sub>2</sub> (0 ppm, 0.06 ppm, 0.3 ppm, 0.6 ppm, 2ppm, 5 ppm, 10 ppm).

➤ Single waveforms produced by a <sup>60</sup>Co  $\gamma$  source have been acquired at each level of contamination.

➤ Average signal  $V(t)$  is calculated (100k wfms) for each [O<sub>2</sub>].

➤ By means of a deconvolution procedure the true light signal  $S(t)$  is obtained from  $V(t)$ .

## Slow component decay time vs $O_2$ concentration

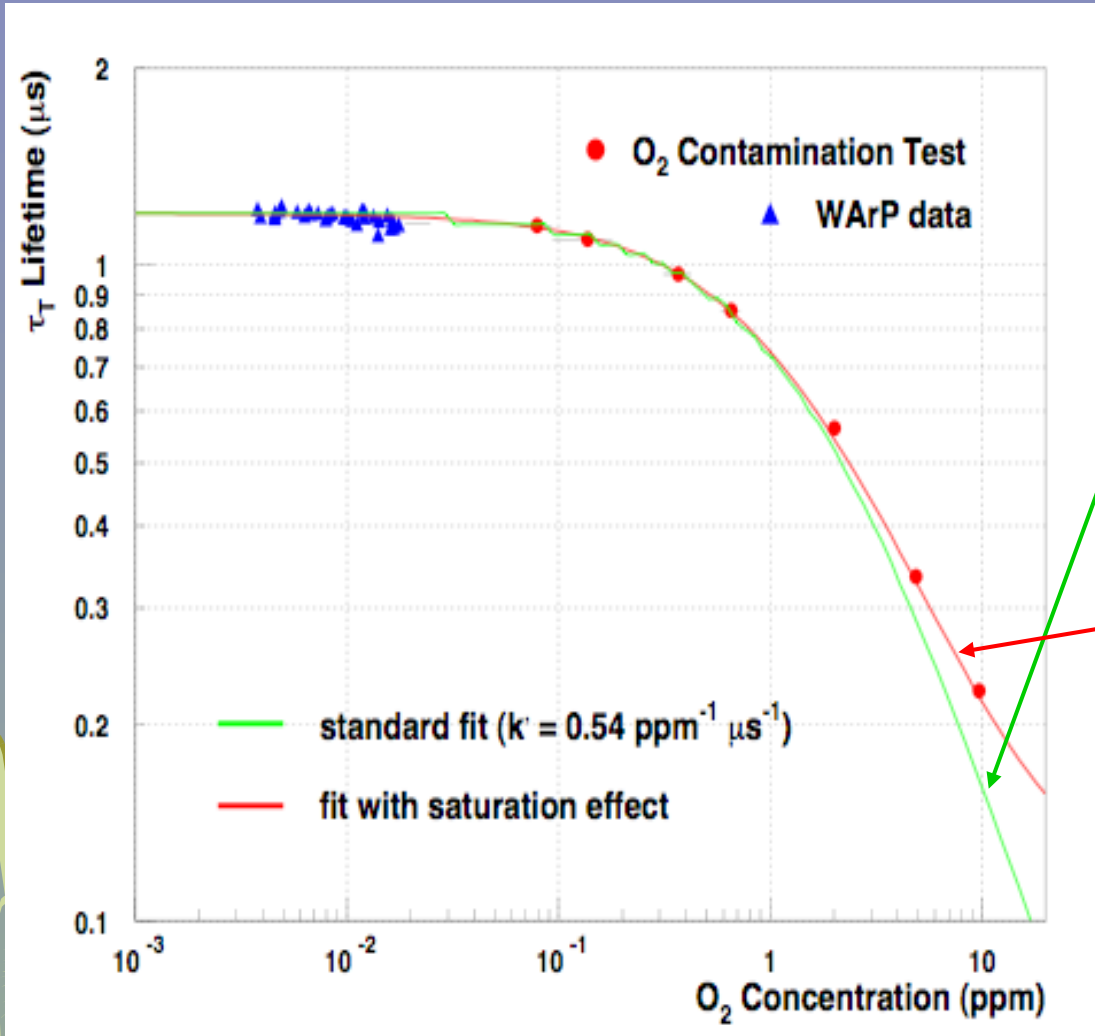


➤ Because of the quenching process,  $\tau_T$  clearly decreases as the  $[O_2]$  exceeds the  $\sim 100$  ppb level.

➤  $\tau_S$  is instead almost unaffected even at largest contaminations.

## Slow component decay time vs $O_2$ concentration: combined results

$\tau_T$  characterizing the scintillation signal shape in LAr has been measured over a wide range of  $O_2$  concentrations with WArP prototype and 0.7 lt detector.



$$\frac{1}{\tau'_T} = \frac{1}{\tau_T} + k' [O_2]$$

Fit with saturation effect:

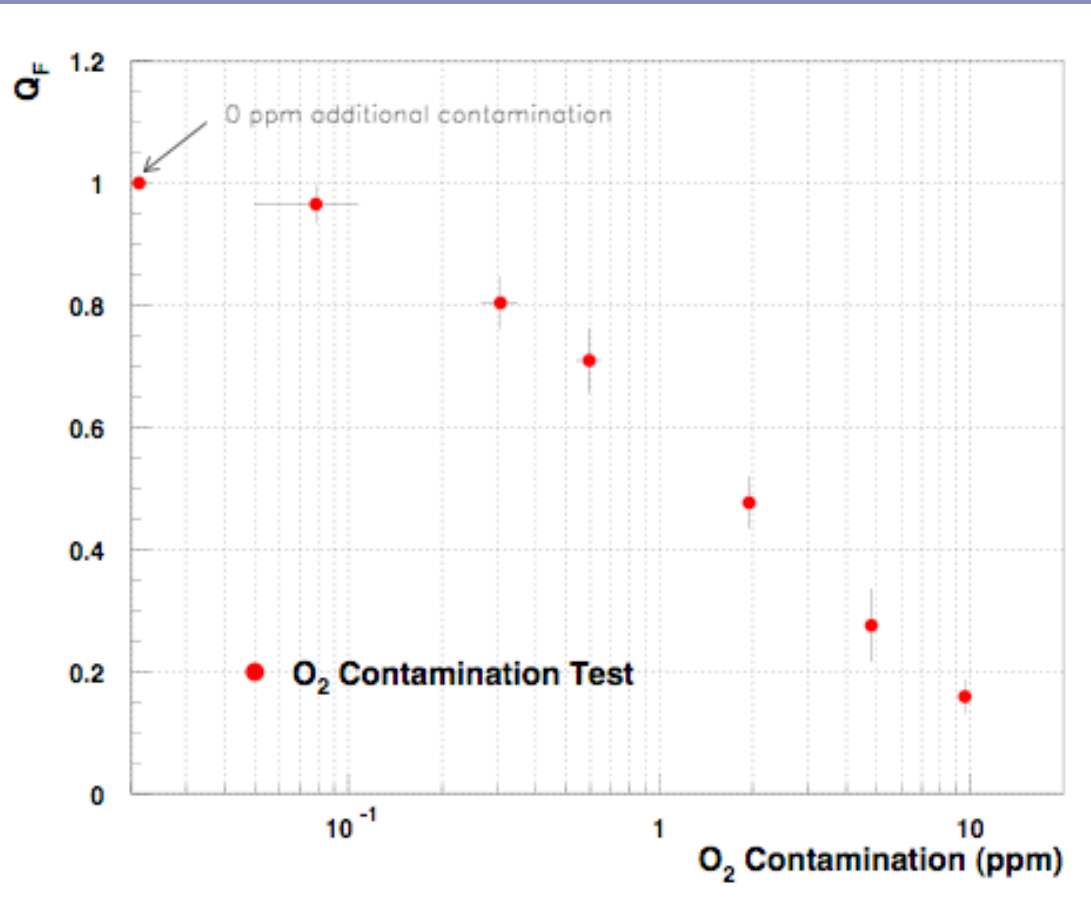
$$\frac{1}{\tau'_T} = \frac{1}{\tau_T} + k' \beta \left( 1 - e^{-[O_2]/\beta} \right)$$

$\beta$  represents the concentration scale where saturation becomes effective (C.L.  $\approx 90\%$ )

## Signal Amplitude Analysis

The signal amplitude, obtained by single waveform integration, is proportional to the energy deposited by  $^{60}\text{Co}$  source  $\gamma$ .

Pulse amplitude spectra (from Compton scattering) are obtained for each run at different  $[\text{O}_2]$  value.



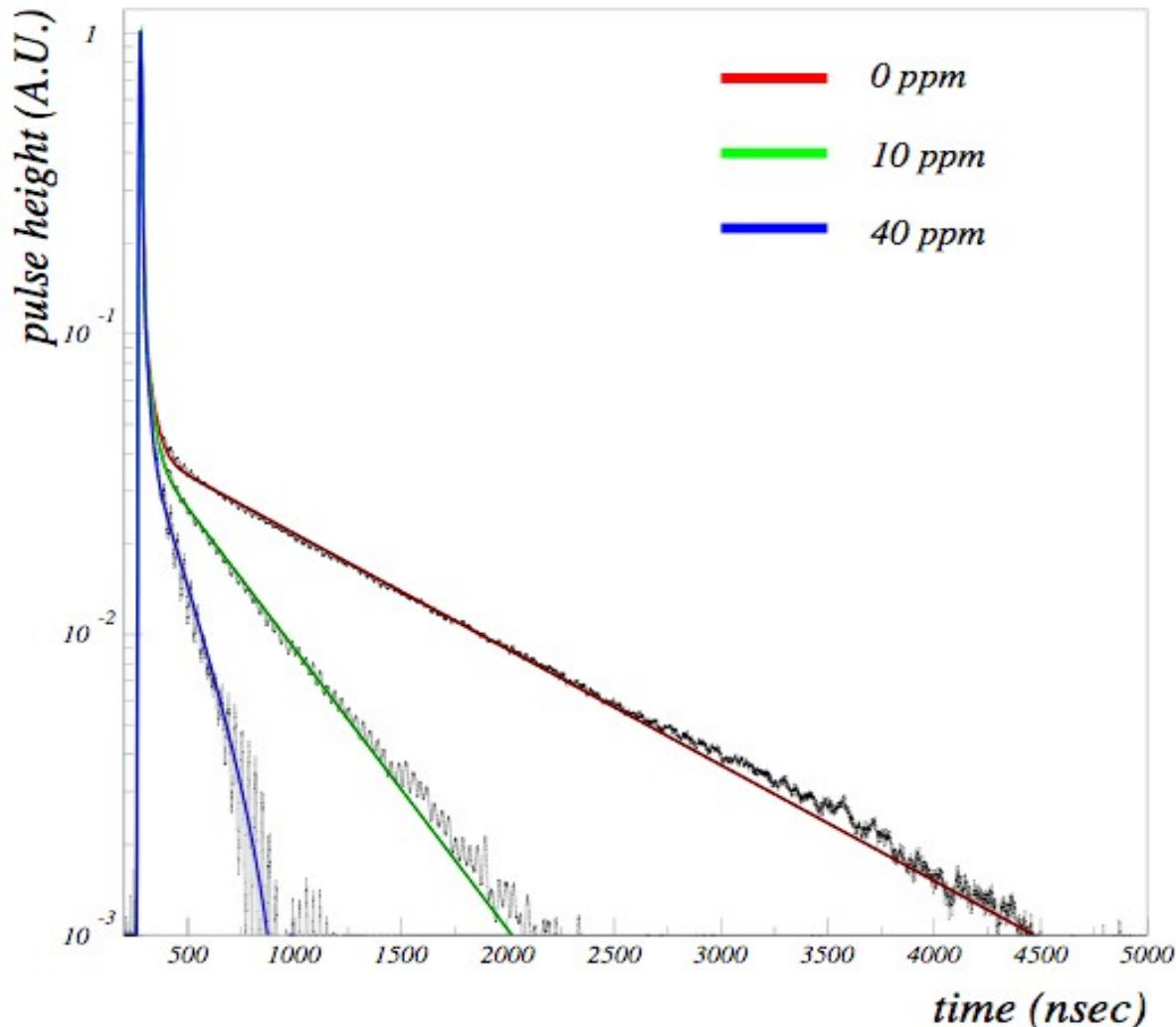
✓ Due to the quenching process, the spectra result down-scaled at increasing  $[\text{O}_2]$ .

✓ Dedicated fitting procedure has been developed to determine the quenching scale factor  $Q_F$  relative to each  $[\text{O}_2]$ .

✓ A sensitive reduction of the scintillation Light Yield is found in the high concentration range ( $[\text{O}_2] \geq 100$  ppb), e.g. the presence of 1 ppm of  $\text{O}_2$  causes a loss of ~40% of the light available.

# Test with 0.7 lt detector: $N_2$

## Average Light Signals



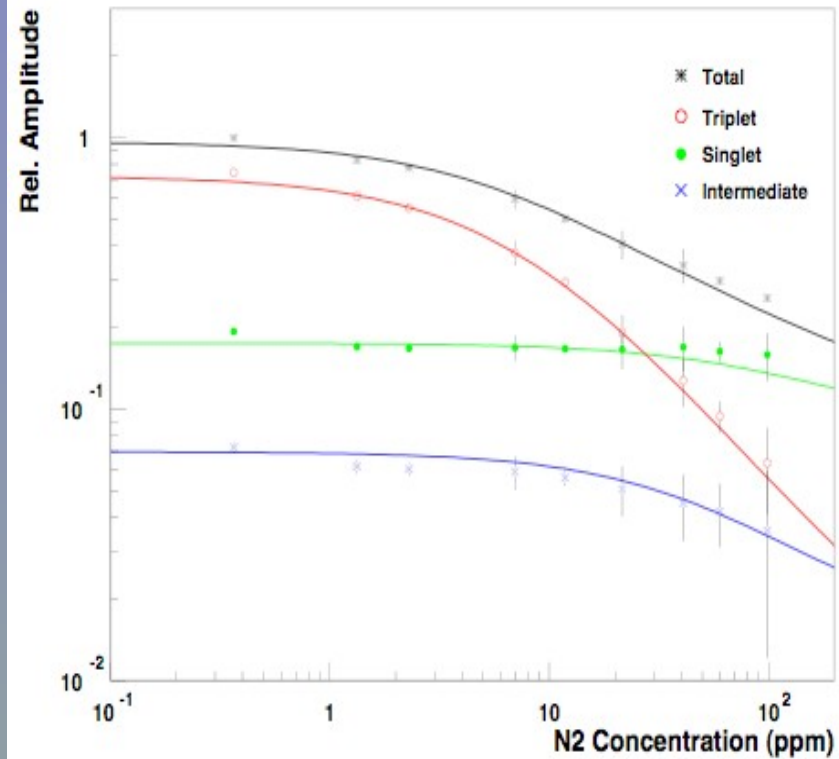
➤ Argon was progressively contaminated with increasing quantities of  $N_2$  (0 ppm, 1 ppm, 2 ppm, 5 ppm, 10 ppm, 20 ppm, 40 ppm, 60 ppm, 100 ppm, 500 ppm, 1000 ppm, 3000 ppm).

➤ Single waveforms produced by a  $^{60}\text{Co}$   $\gamma$  source have been acquired at each level of contamination.

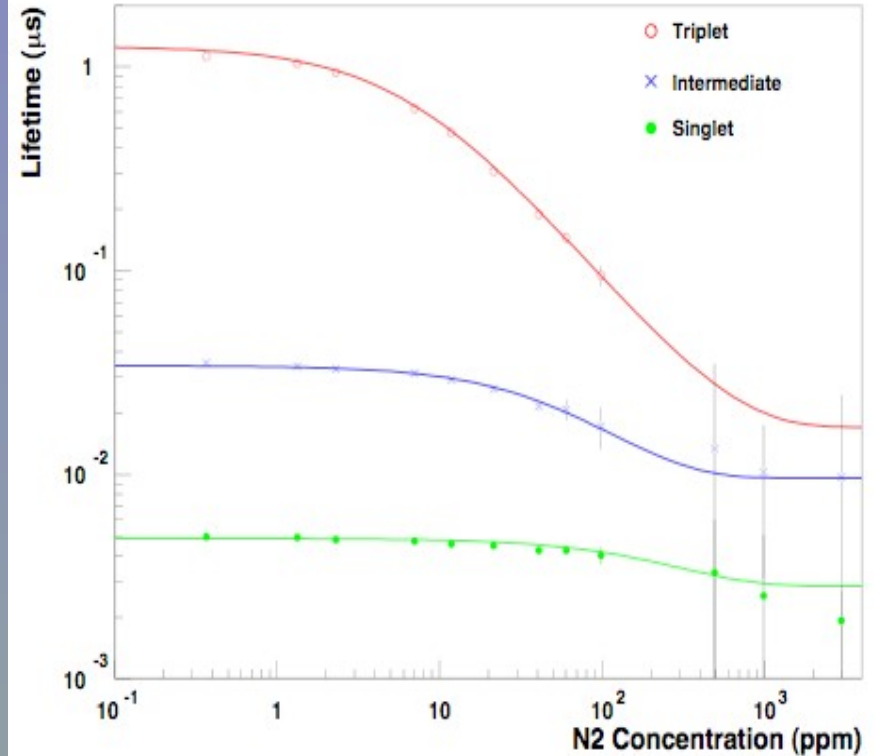
➤ Average signal  $V(t)$  is calculated (100k wfms) for each [ $N_2$ ].

# Overall fit of Amplitudes and Lifetimes

## Relative intensities



## Lifetimes



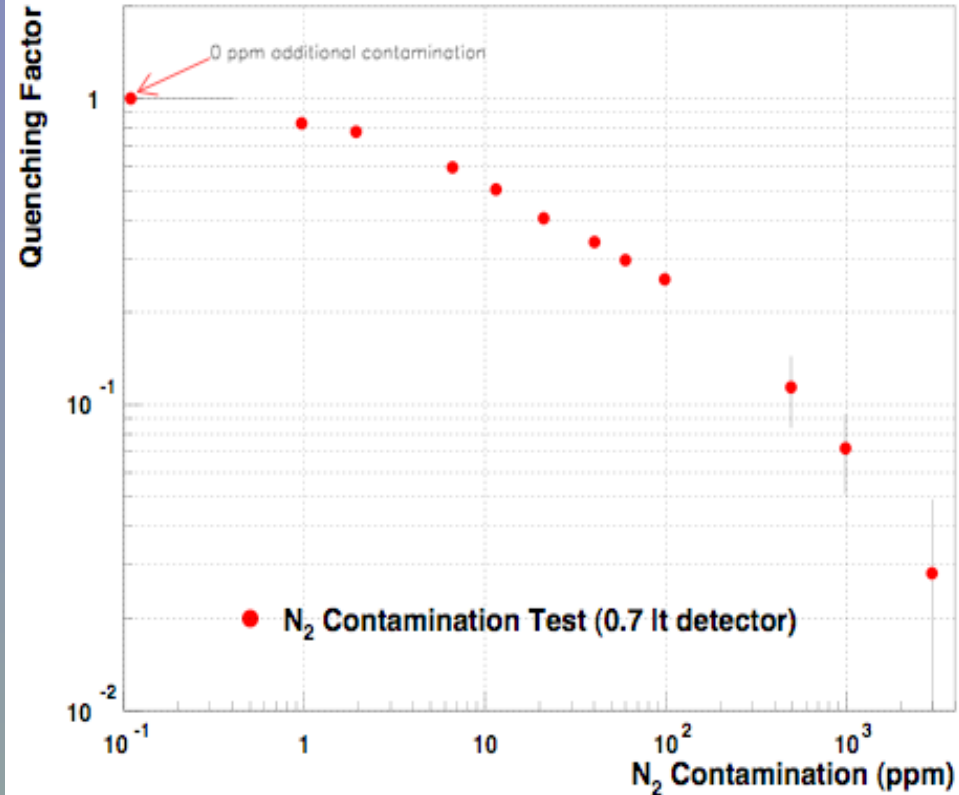
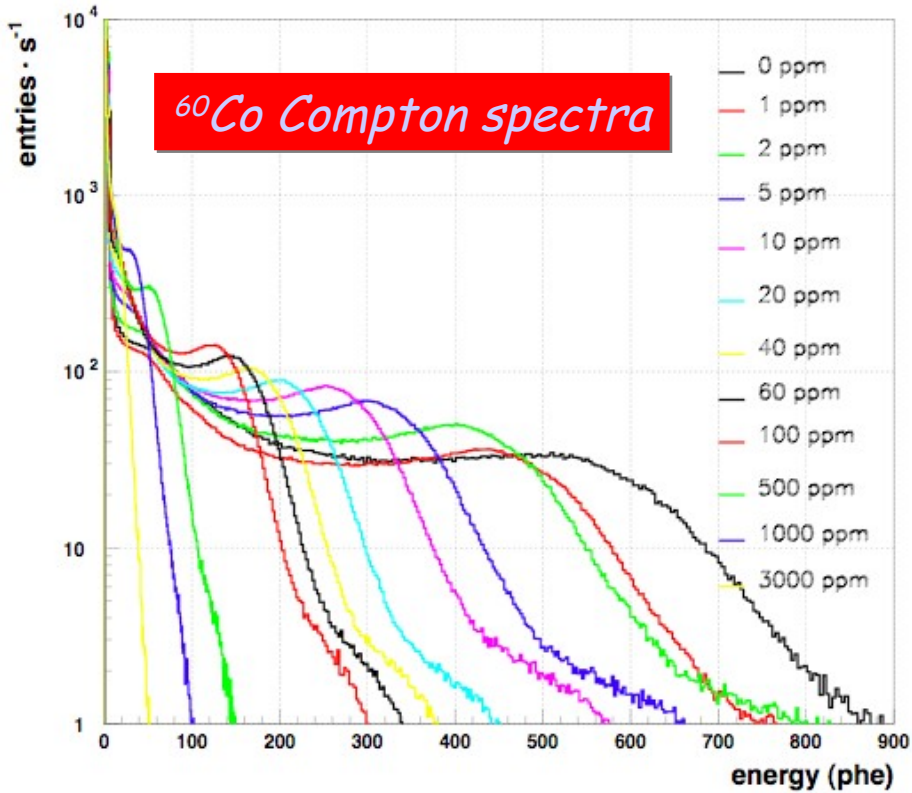
$$A'_j([N_2]) = \frac{A_j}{1 + \tau_j k [N_2]}$$

$$\frac{1}{\tau'_j([N_2])} = \frac{1}{\tau_j} + k [N_2]$$

*Like in the previous case, these equations have been slightly modified to take into account saturation effects at high  $[N_2]$ .*

# Signal Amplitude Analysis

By single waveform integration pulse height spectrum is obtained for each  $[N_2]$ .



*A dedicated procedure allows to fit the uncontaminated (0 ppm) spectrum to the  $N_2$ -contaminated spectra.*

*This allows to determine the overall quenching factor  $Q_F$  as a function of  $[N_2]$ .*

# Analysis Results

	Lifetimes $\tau_i$	Amplitudes $A_i$
Short-Lived Component	$4.9 \pm 0.2$ ns	18.8%
Intermediate Component	$34 \pm 3$ ns	7.4%
Long-Lived Component	$1260 \pm 30$ ns	73.8%

✓  $A_s/A_t = 0.35$  in agreement with available reference data for light ionizing particles.

✓ The rate constant  $k$  of the  $N_2$  quenching process is in agreement with earlier measurements (but with an higher precision than before).

✓ The rate constant  $k'$  of the  $O_2$  quenching process is five times higher than that of  $N_2$  process.

	Rate Constant $k$	Initial Concentration $[X_2]_{in}$
$O_2$	$0.54 \pm 0.03$ $\mu s^{-1} ppm^{-1}$	$65 \pm 15$ ppb
$N_2$	$0.11 \pm 0.05$ $\mu s^{-1} ppm^{-1}$	$0.40 \pm 0.20$ ppm

✓ Initial  $O_2$  and  $N_2$  concentrations are within the specification for ultra-pure Ar.

**Additional fit parameter to take into account initial Ar contamination**

# Conclusions

- Both Nitrogen and Oxygen contaminations have been found to reduce LAr scintillation light. The rate constants of the light quenching process have been found to be  $k(O_2)=0.54 \mu s^{-1}ppm^{-1}$  and  $k(N_2)=0.11 \mu s^{-1}ppm^{-1}$ . This implies that, for example, the presence of 1 ppm of  $N_2$  brings to a  $\approx 20\%$  reduction of the scintillation light, while the same amount of  $O_2$  causes a  $\approx 40\%$  reduction.
- The light signal shape has also been investigated. It results to be well represented by the superposition of three components with exponential decay. The fast and the slow components have decay time constants  $\tau_s=4.9$  ns and  $\tau_T=1260$  ns. An intermediate component is also found to be present (as sometimes reported in literature).
- Residual contents of  $O_2$  in LAr reduce the amount of both charge and light (slow scintillation component) available, while the main effect of residual  $N_2$  in LAr is of reducing the slow component lifetime at increasing concentration. The decrease of the slow component amount worsen the pulse shape discrimination capability of LAr based detectors and therefore makes indispensable the implementation of dedicated methods for removal of the residual  $O_2$  and  $N_2$  content in LAr.

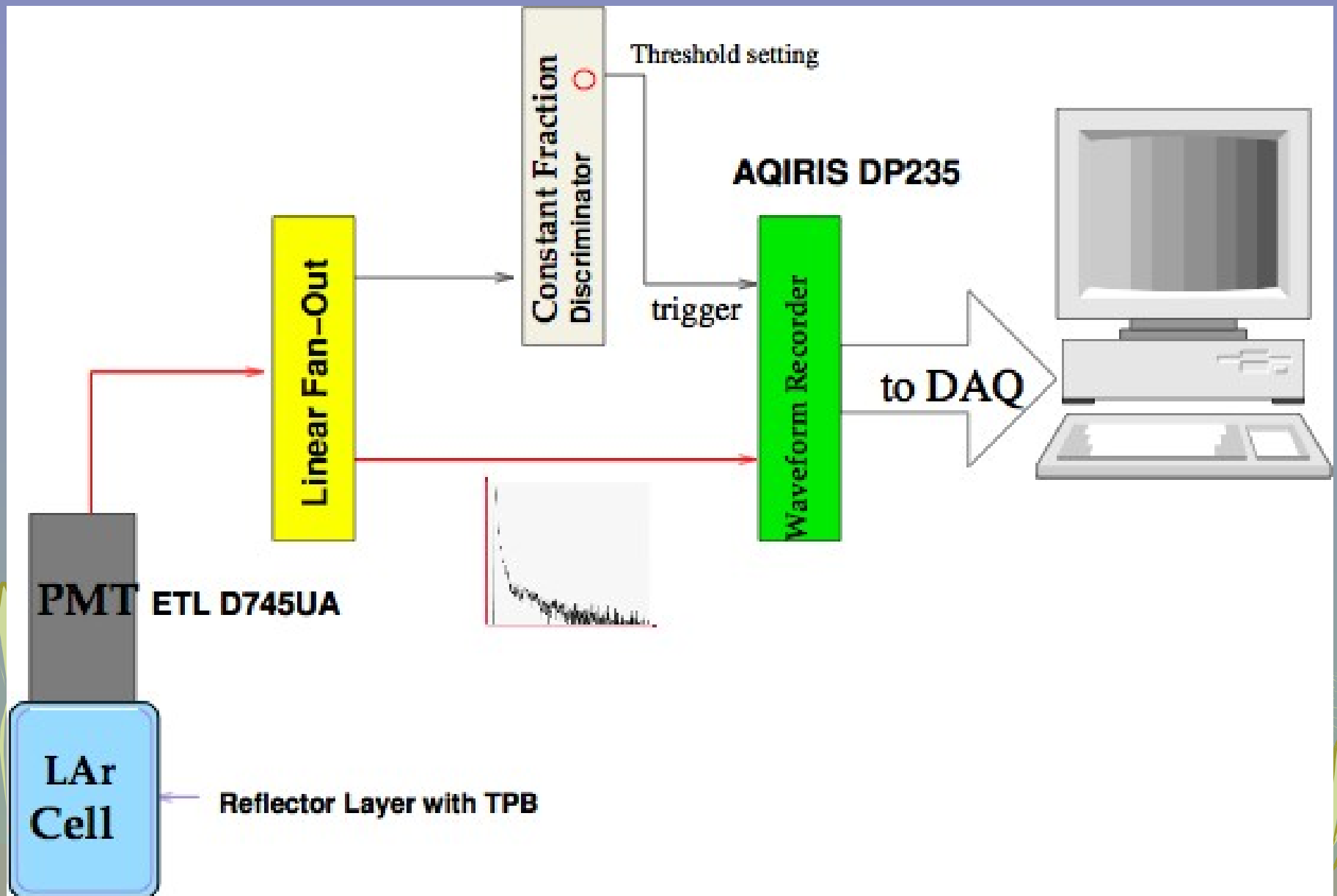
# ***BACKUP SLIDES***

October, 2 2008

IPRD08 - Roberto Acciarri



# 0.7 It detector Data Acquisition



# Measurements with WArP prototype

By the means of WArP prototype it has been possible to describe the behaviour of the long-lived scintillation light lifetime determining the  $[O_2]$  by the electron lifetime measurement.

Electronegative molecules, as  $O_2$ , reduce the full collection of free electrons:



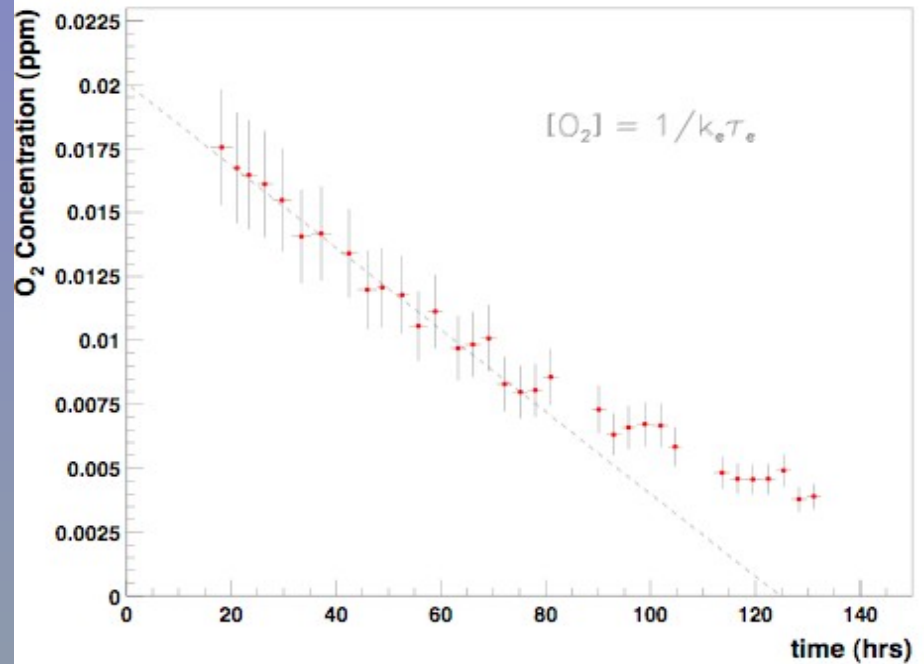
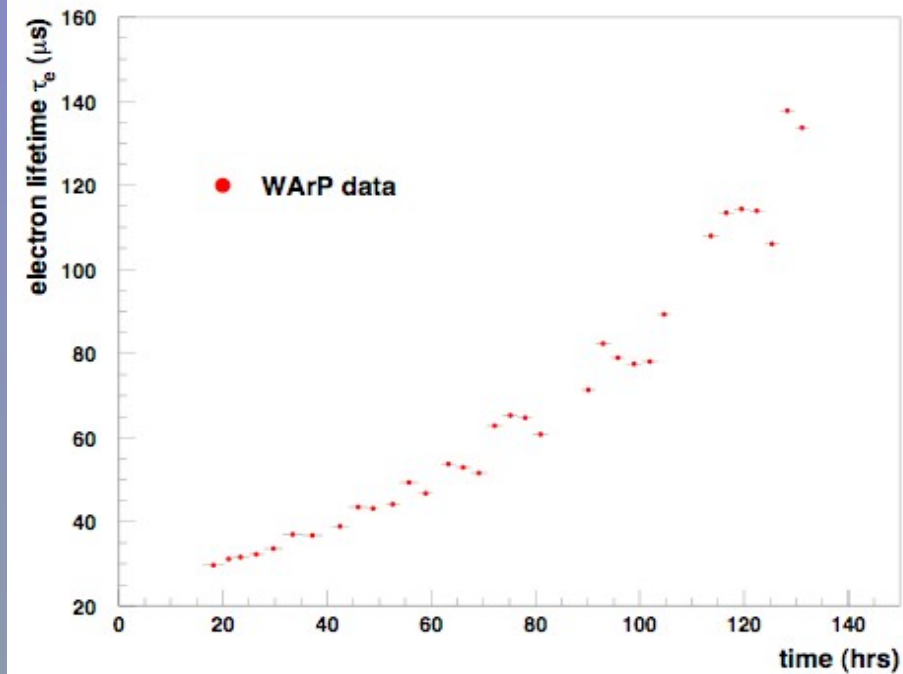
if  $[e^-] \ll [O_2] \Rightarrow [e^-]$  decreases in time as:

$$\frac{d[e^-]}{dt} = -k_e [O_2] [e^-] \Rightarrow [e^- (t)] = [e^- (0)] e^{-\frac{t}{\tau_e}}$$

$$\text{Electron Lifetime } \tau_e = \frac{1}{k_e [O_2]}$$

the value of the rate constant  $k_e$  depends on drift field applied to the active LAr volume

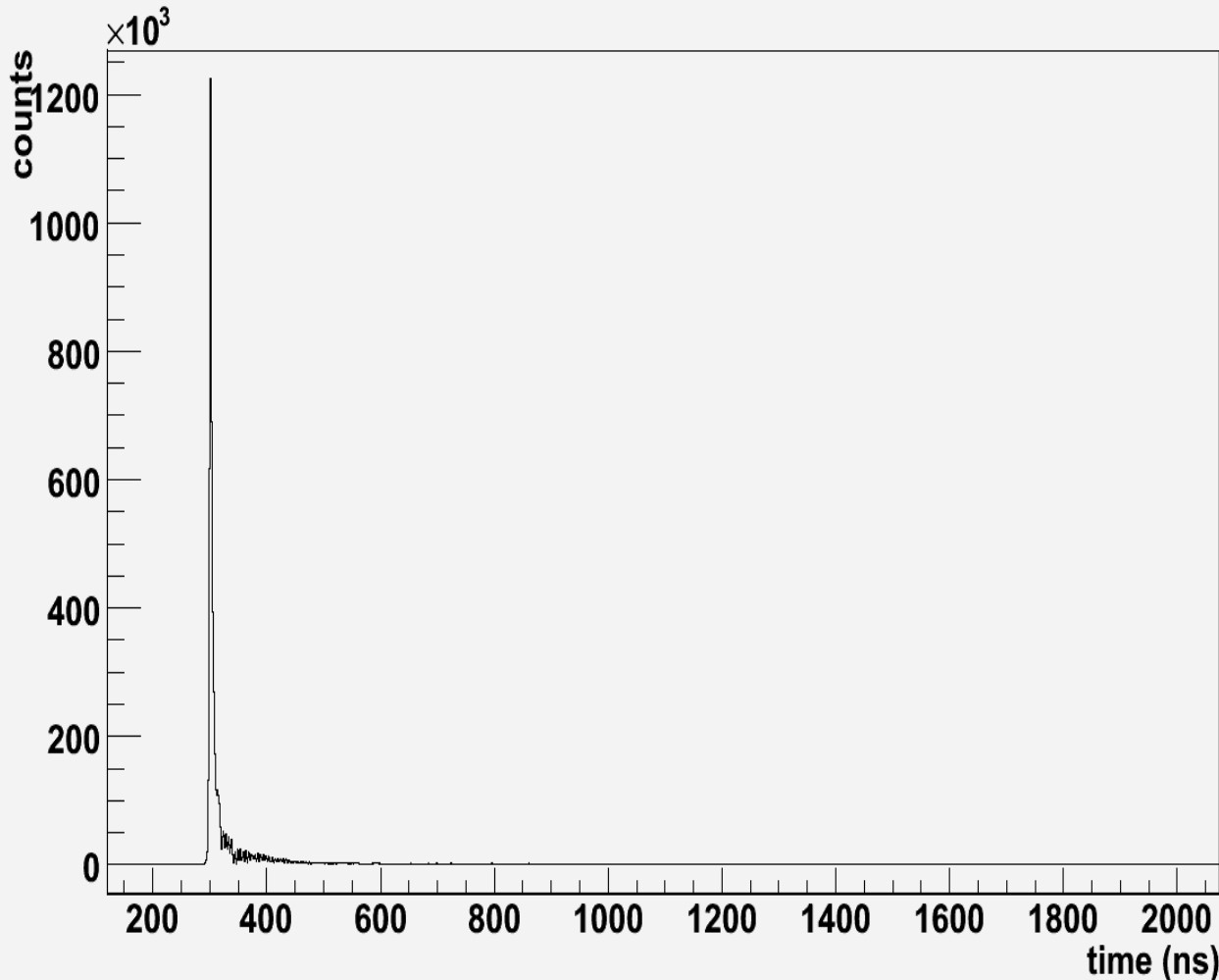
$$k_e = 5.5 \cdot 10^{10} \text{ lt} \cdot \text{mol} \cdot \text{s}^{-1} = 1.9 \text{ ppm}^{-1} \cdot \mu \text{ s}^{-1} \quad @ \quad EF = 1 \text{KV} \cdot \text{cm}^{-1}$$



✓  $\tau_e$  increases up to the limit of sensitivity ( $\geq 200 \mu\text{s}$ ) during measurement performed in successive time segments (of about 3 hrs each).

✓ Extrapolation of initial  $[\text{O}_2]$  of the Ar in the detector up to 20 ppb (coming from  $\text{O}_2$  content in Ar 6.0, outgassing materials and residual leaks).

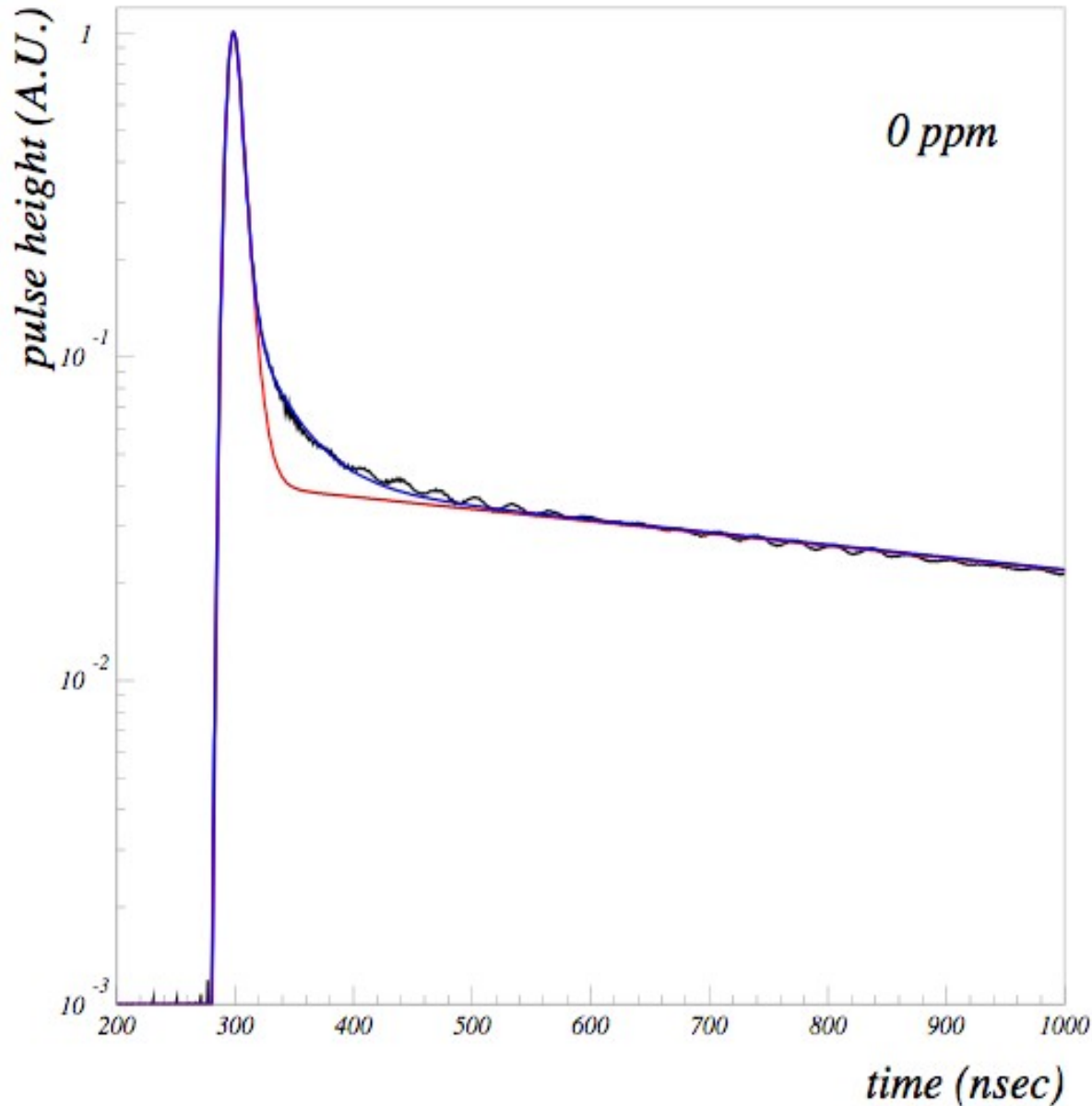
# Read out system response



✓ The average signal can be expressed as:  
 $V(t) = S(t) \otimes R(t)$  where  $S(t)$  is the light signal and  $R(t)$  is the response function of the read-out system.

✓ The signal  $S(t)$  is obtained from  $V(t)$  by a deconvolution procedure using the function  $R(t)$  experimentally determined.

## Evidence for a third component



✓ Attempts to perform a fit of the average wfm with a two component signal failed.

✓ This lead to use a three component model, nicely fitting present data.

✓ Dedicate studies are under way in order to determine the origin of this component.

# Quenching process by residual contaminants

- Residual contaminants (@ ppm level) of  $N_2$ ,  $O_2$ ,  $H_2O$ ,  $CO$  and  $CO_2$  (even in best grade commercial LAr) can lead to a substantial reduction of the scintillation yield.
- Argon purification systems (Oxygen reactants and molecular sieves) are known to reduce  $O_2$ ,  $H_2O$ ,  $CO$  and  $CO_2$  at a negligible level.
- Methods for removing the  $N_2$  residual content are instead less commonly used in experimental applications and the effects of  $N_2$  contamination are rather poorly known. At the same time, the effects of  $O_2$  contamination on LAr scintillation light have been poorly investigated.