Triplet helium molecule detection by laser-induced fluorescence

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The signal in the neutron EDM experiment is the result of neutron absorption on ^{3}He:

$$n + {}^3He \longrightarrow p + {}^3H + 764 \text{ keV}$$

The ionization and excitation of helium atoms results in the rapid formation of Helium molecules, which radiatively decay, emitting EUV scintillation light. This scintillation chiefly occurson three different times scales, corresponding to three different physical processes:

1) Prompt scintillation (time $<$ 20 ns)
- Radiative decay of singlet helium molecules

2) Afterpulsing (varies in intensity as roughly 1/time)
- Results from destructive interaction of triplet molecules with each other (Penning ionization). Products of this can result in singlet molecule formation, which then radiatively decay. This component is more intense for neutron absorption events than for gammas.

3) Phosphorescence (exponential decay with 13 second lifetime)
- Triplet molecules radiatively decay (unless they hit a wall first)
- This component is less intense for neutrons than for gammas

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Radiative decay of the metastable He_2 $a\,^3\Sigma_u^+$ molecule in liquid helium

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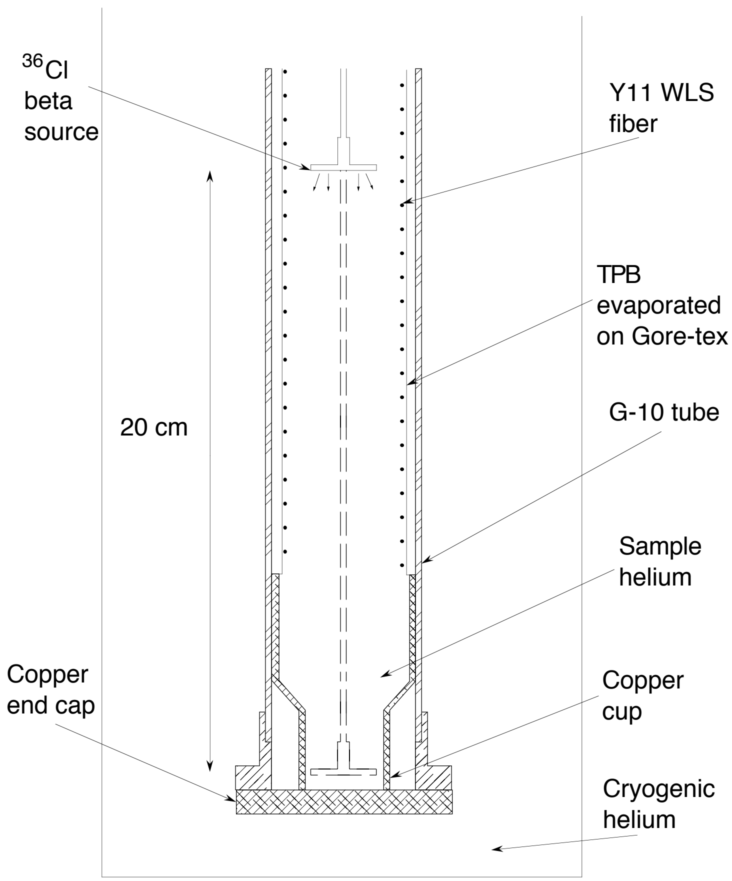
(Received 27 July 1998)



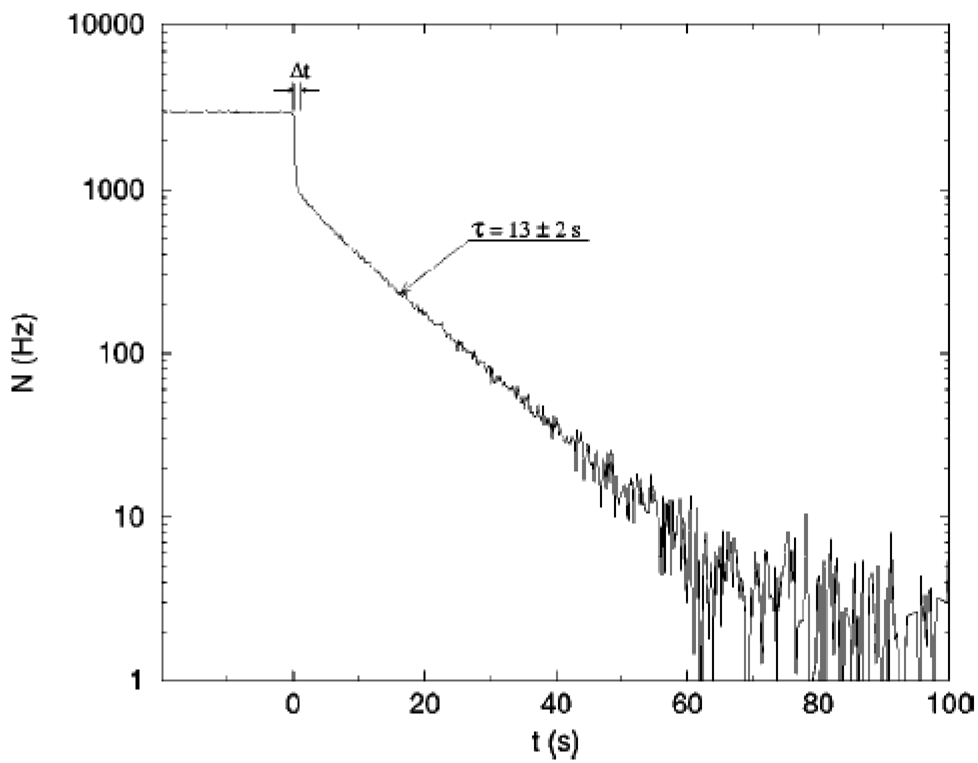
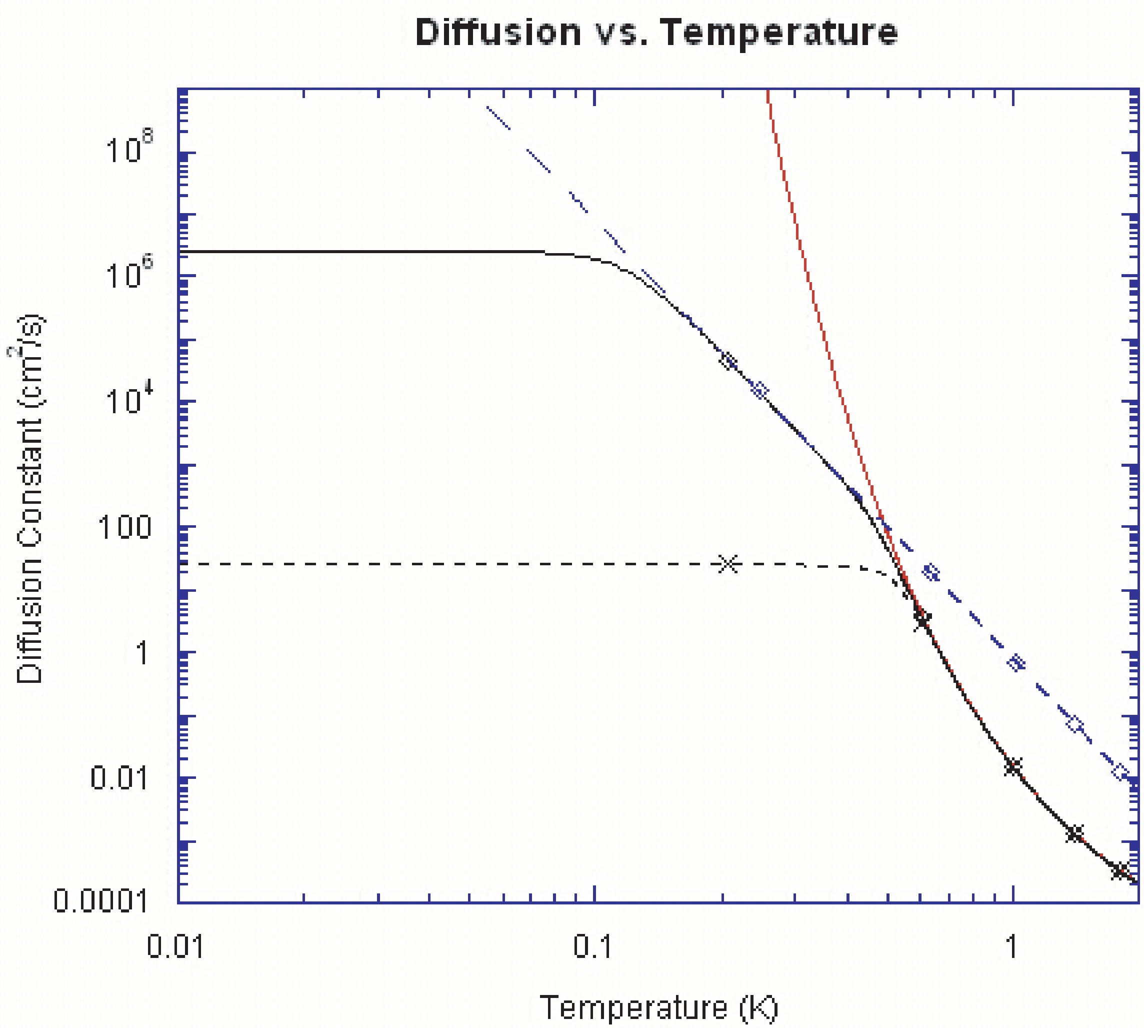


FIG. 2. Count rate N of detected $He_2(a\,^3\Sigma_u^+)$ decays versus time. A ^{36}Cl β source is placed in the center of the detection region and then removed in a time $\Delta t < 1$ s. This measurement was performed at a temperature of 1.8 K and resulted in a measured decay rate τ of 13 ± 2 s.

The diffusion constant is strongly temperature-dependent.



At 250 mK, diffusion constant is expected to be > 10,000 $cm^2\,s^{-1}$

For t = (5 cm)^2/Diffusion constant) , this gives t < 2.5 ms

Why try to detect the triplet molecules?

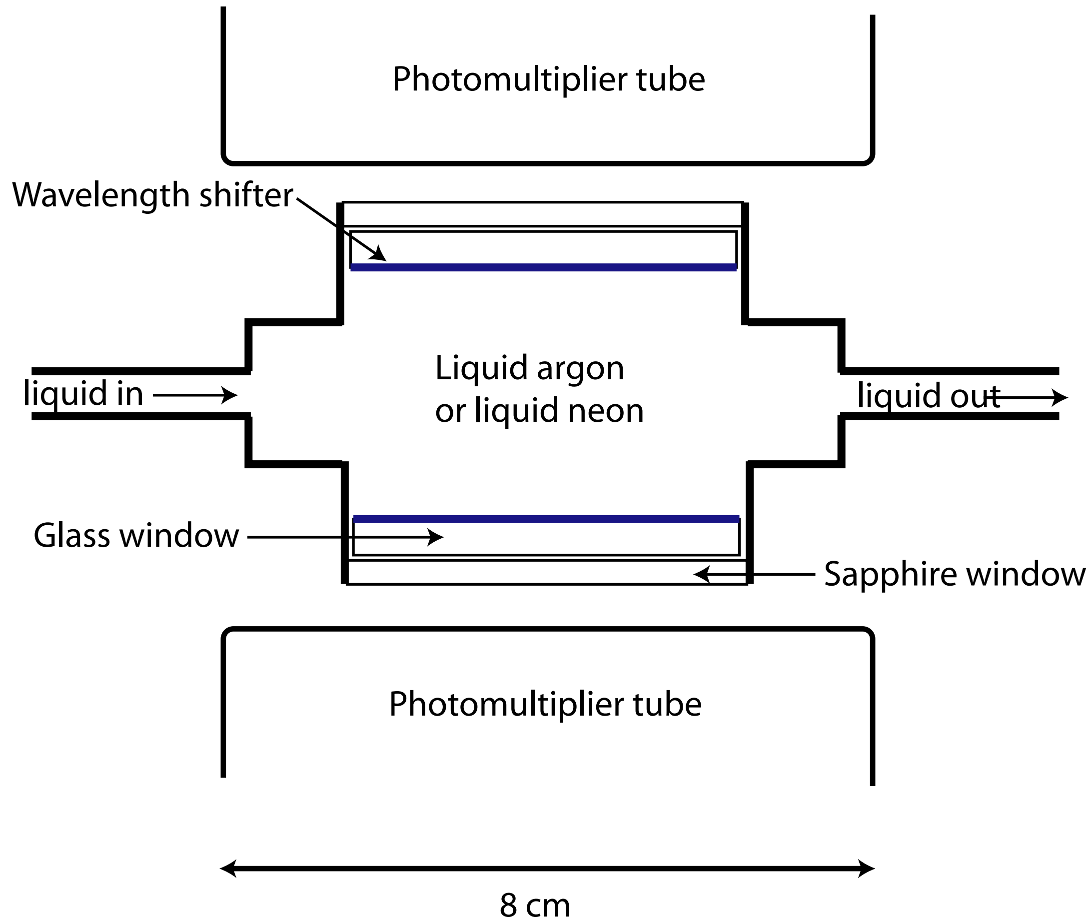
1) More signal:

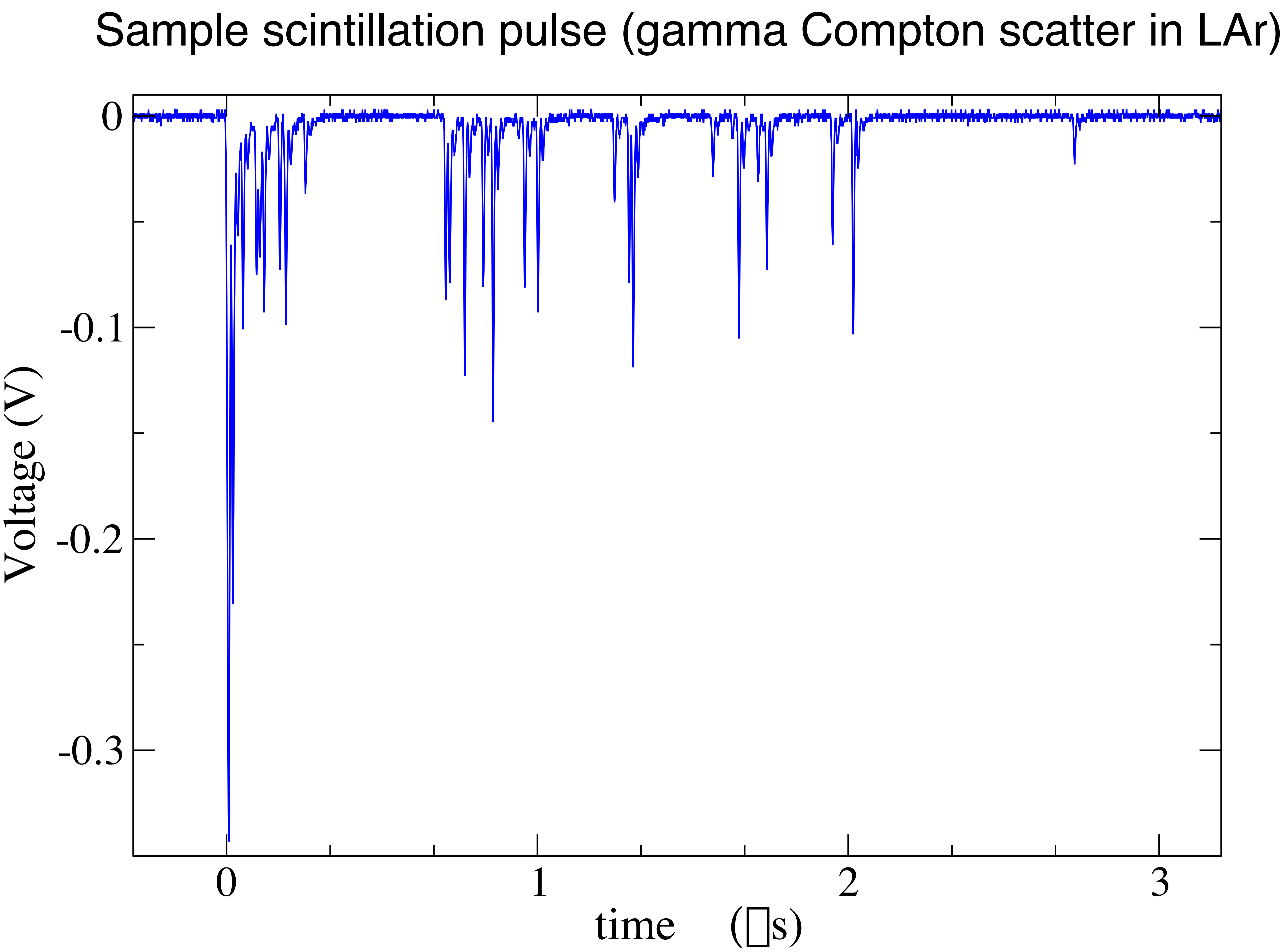
Past experience (on neutron trapping experiment) would imply that every photoelectron is valuable for digging out of activation and luminescence backgrounds.

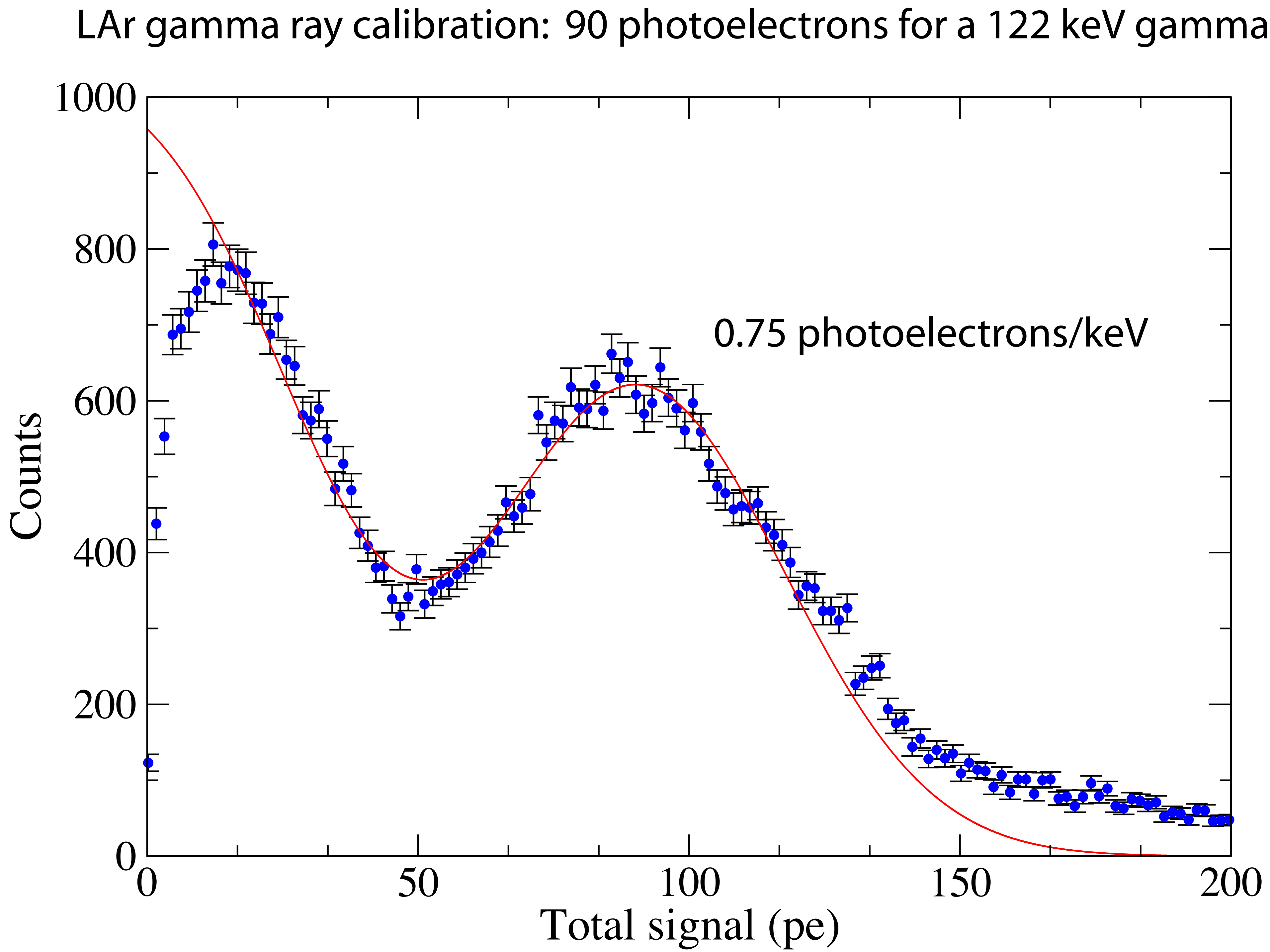
2) Complementary information for gamma rejection:

In liquid neon and liquid argon, the ratio of singlet molecules to triplet molecules is a strong indicator of the ionization density and can be used to discriminate between gammas and heavy ionizers. This is probably because for heavy ionizing particles, many of the triplet molecules are lost through destructive triplet-triplet interactions (Penning ionization). It is likely that this would work in liquid helium as well.

Pico-CLEAN at Yale



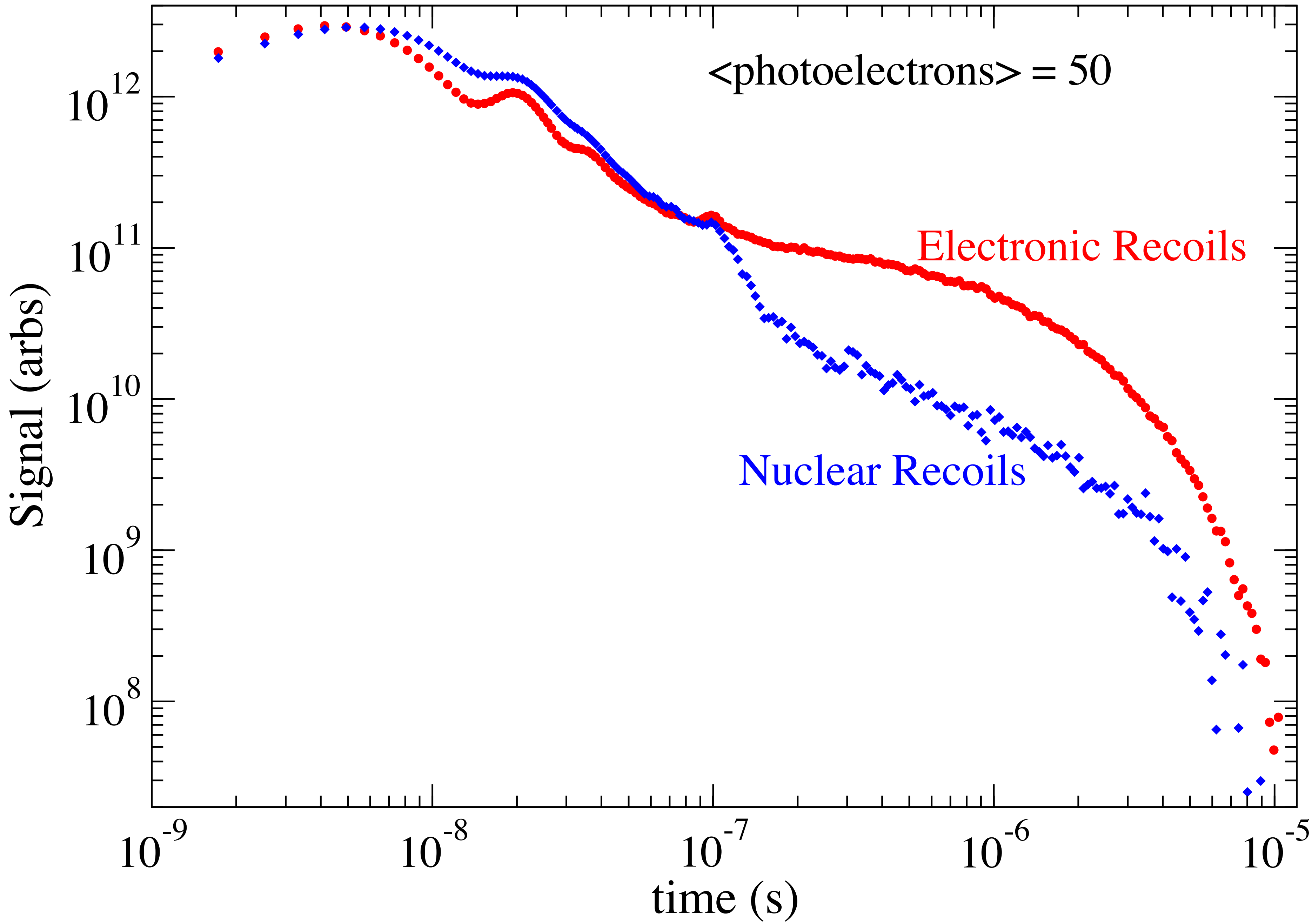

Sample scintillation pulse (gamma Compton scatter in LAr)
0
-0.1
-0.2
-0.3
Voltage (V)
0
1
2
3
time (μs)

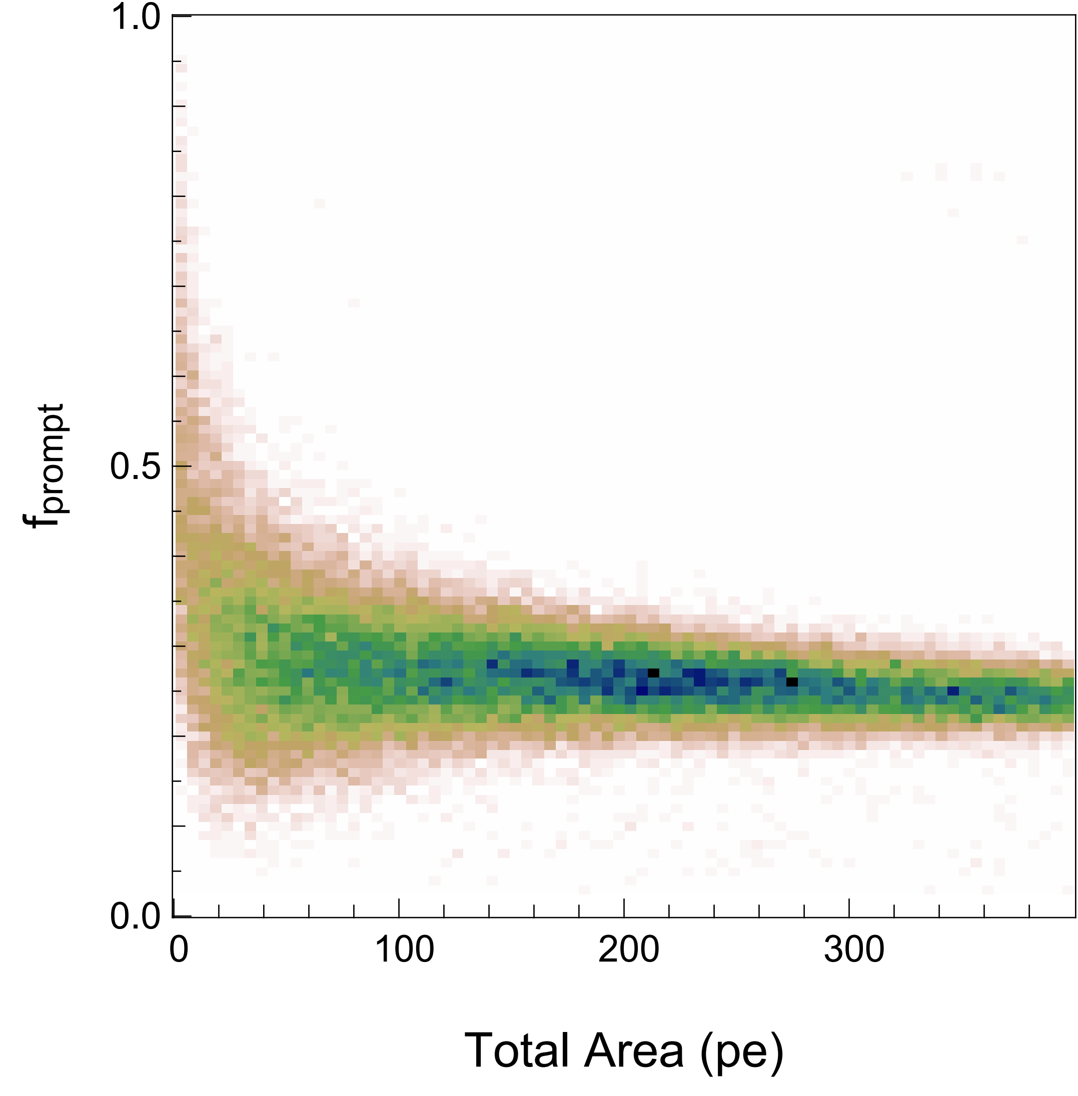


LAr gamma ray calibration: 90 photoelectrons for a 122 keV gamma
0.75 photoelectrons/keV
Counts
Total signal (pe)
0
200
400
600
800
1000
0
50
100
150
200

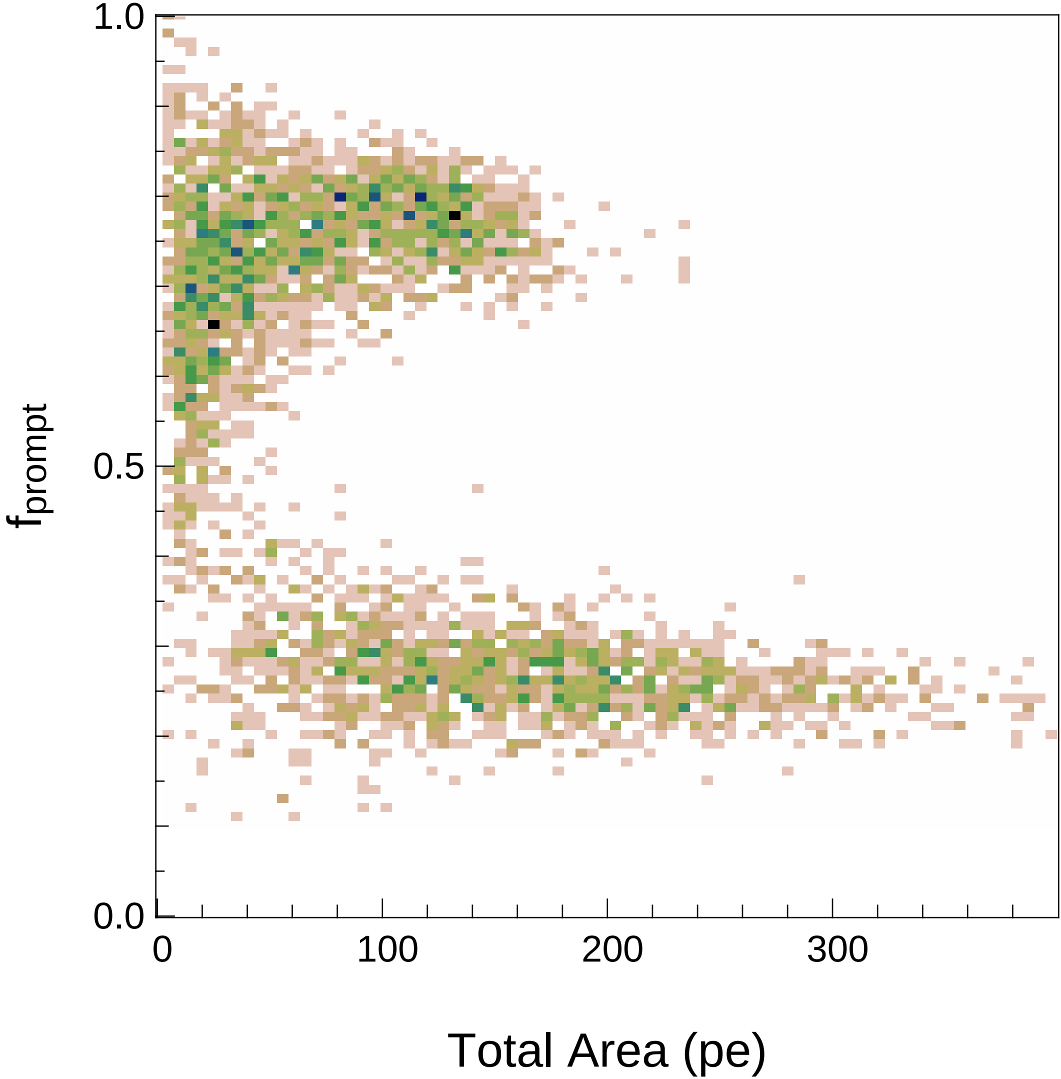
Neutron generator at Yale (2.8 MeV, 10^6 n/s)

Time dependence of liquid Argon scintillation

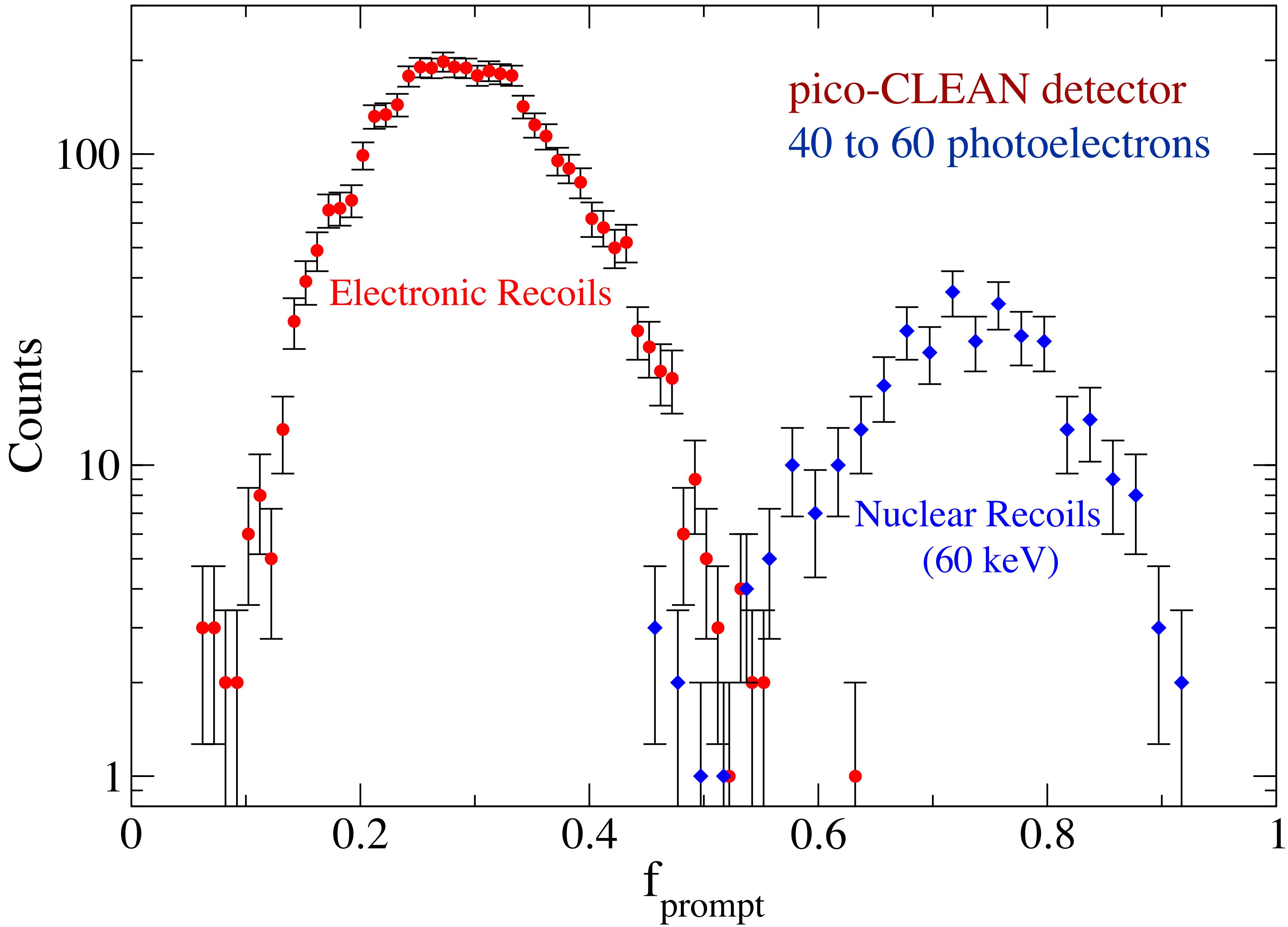


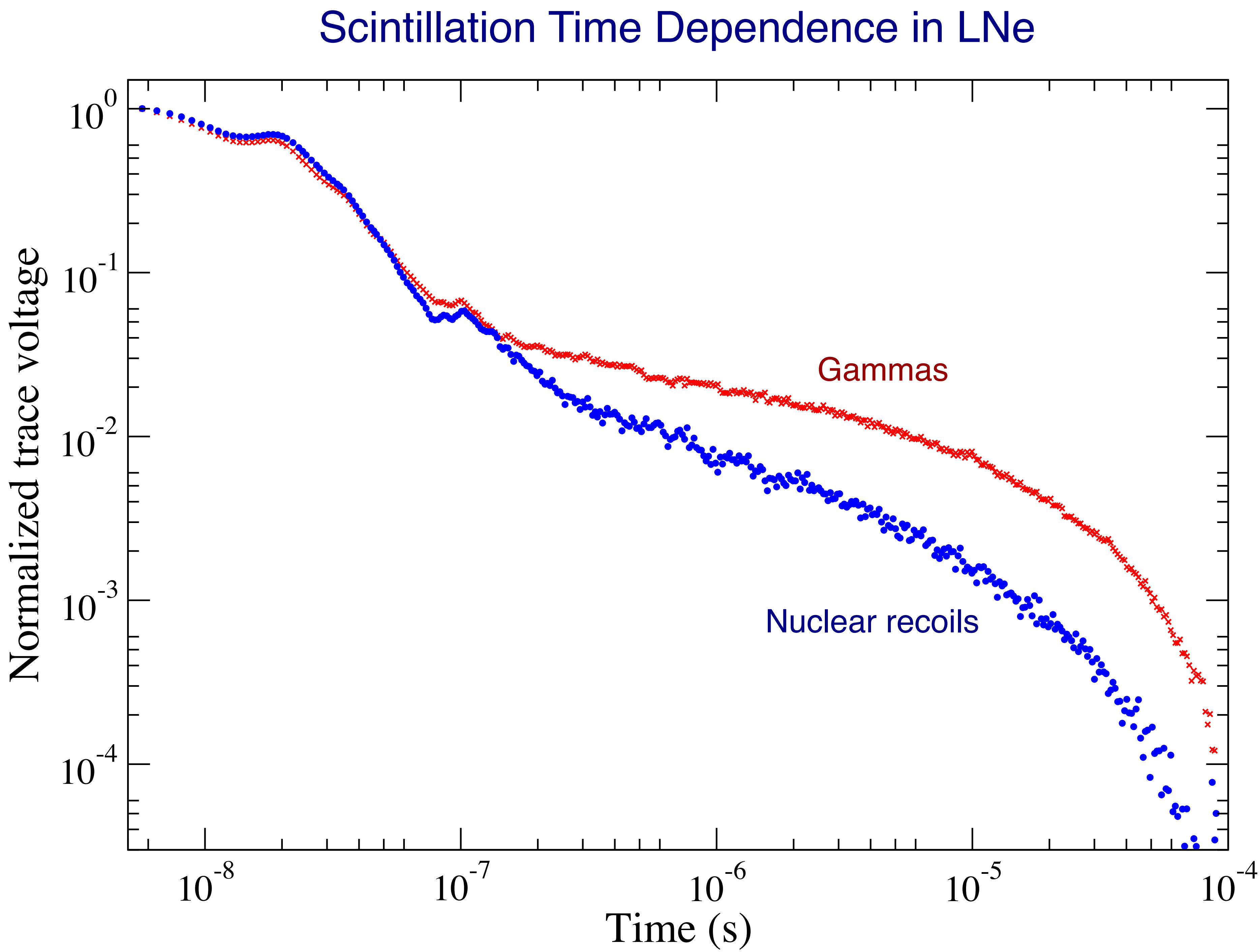


1.0
0.5
0.0
f_{prompt}
0
100
200
300
Total Area (pe)

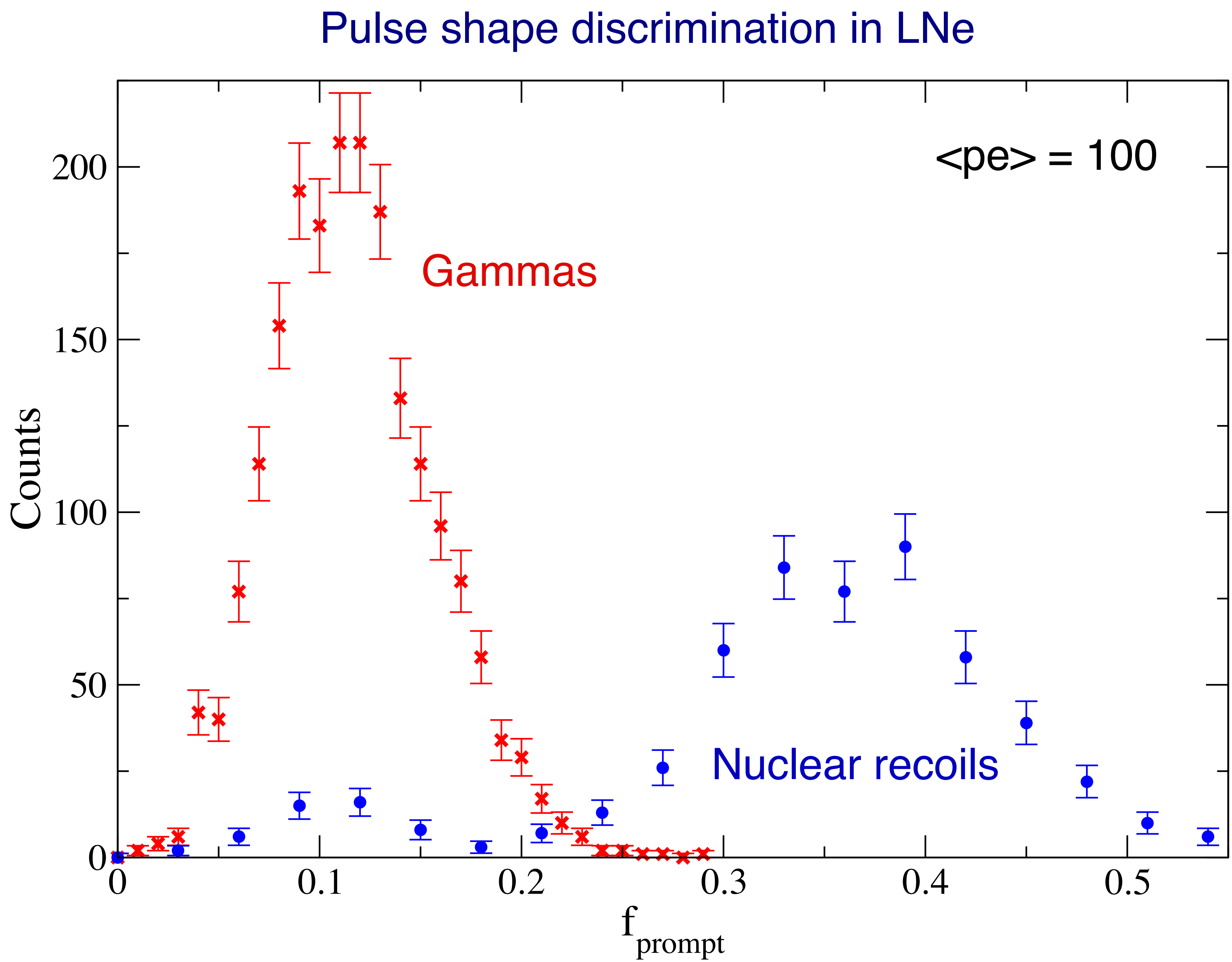

1.0
0.5
0.0
f_{prompt}
0
100
200
300
Total Area (pe)

Pulse shape discrimination in liquid Argon





Scintillation Time Dependence in LNe
Normalized trace voltage
Time (s)
Gammas
Nuclear recoils
10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-8}
10^{-7}
10^{-6}
10^{-5}
10^{-4}


Pulse shape discrimination in LNe
<pe> = 100
Gammas
Nuclear recoils
Counts
f prompt
0
50
100
150
200
0
0.1
0.2
0.3
0.4
0.5

Trace Detection of Metastable Helium Molecules in Superfluid Helium by Laser-Induced Fluorescence

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(Received 10 March 2005; published 8 September 2005)

We describe an approach to detecting ionizing radiation that combines the special properties of superfluid helium with the sensitivity of quantum optics techniques. Ionization in liquid helium results in the copious production of metastable He_2 molecules, which can be detected by laser-induced fluorescence. Each molecule can be probed many times using a cycling transition, resulting in the detection of individual molecules with high signal to noise. This technique could be used to detect neutrinos, weakly interacting massive particles, and ultracold neutrons, and to image superfluid flow in liquid ^4He.

A possible way to measure the triplet signal for the neutron EDM experiment?

Laser-based detection scheme

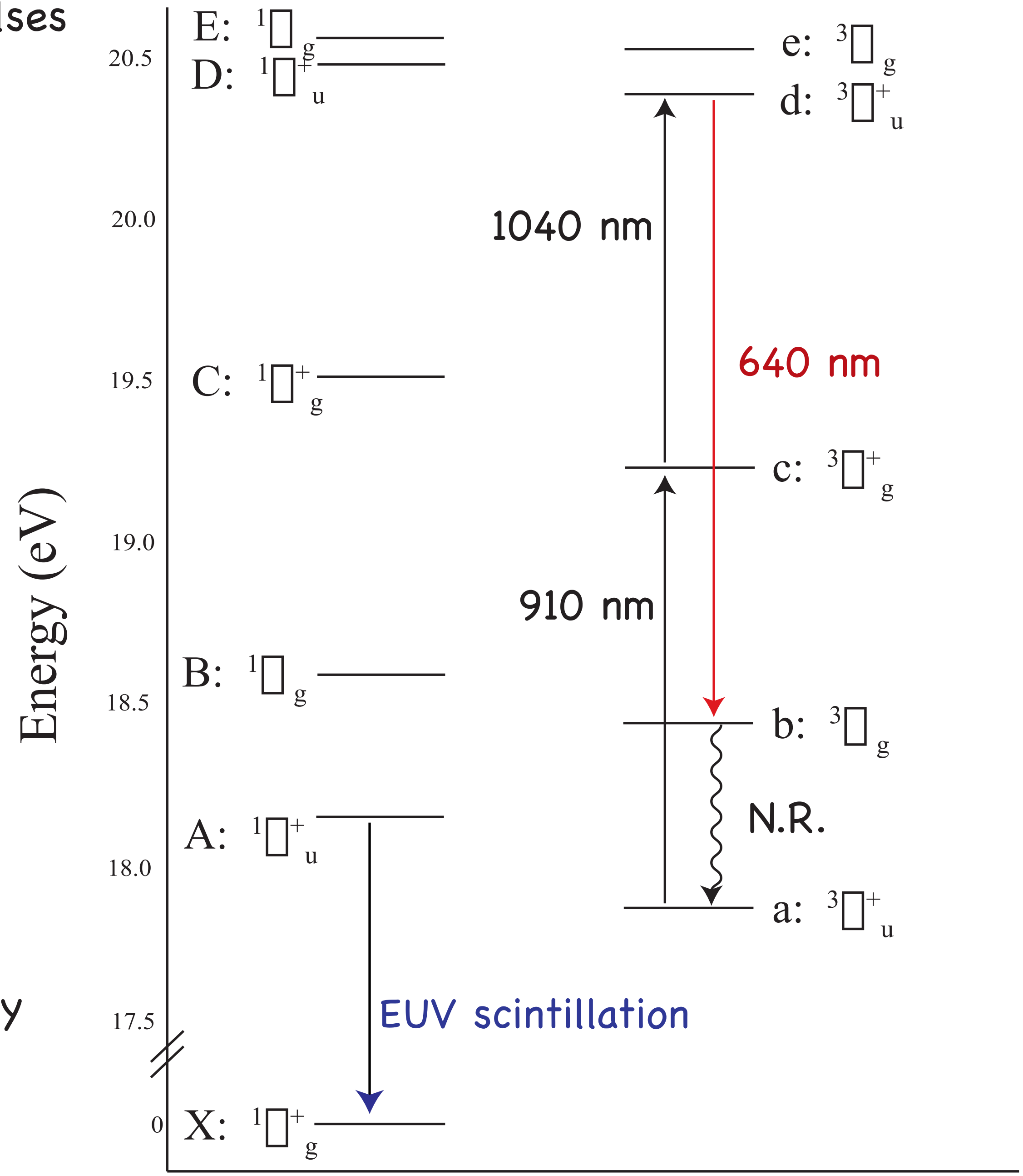
Detection of the scintillation light from A--->X decay would trigger IR laser pulses at 910 nm and 1040 nm.

This will drive the triplet molecules from a--->c and then c--->d.
The molecules will then decay from d--->b with a 90% branching ratio, emitting a 640 nm photon.

The b state will then decay non-radiatively back to the a state.
The molecules can be pumped multiple times, emitting many red photons.

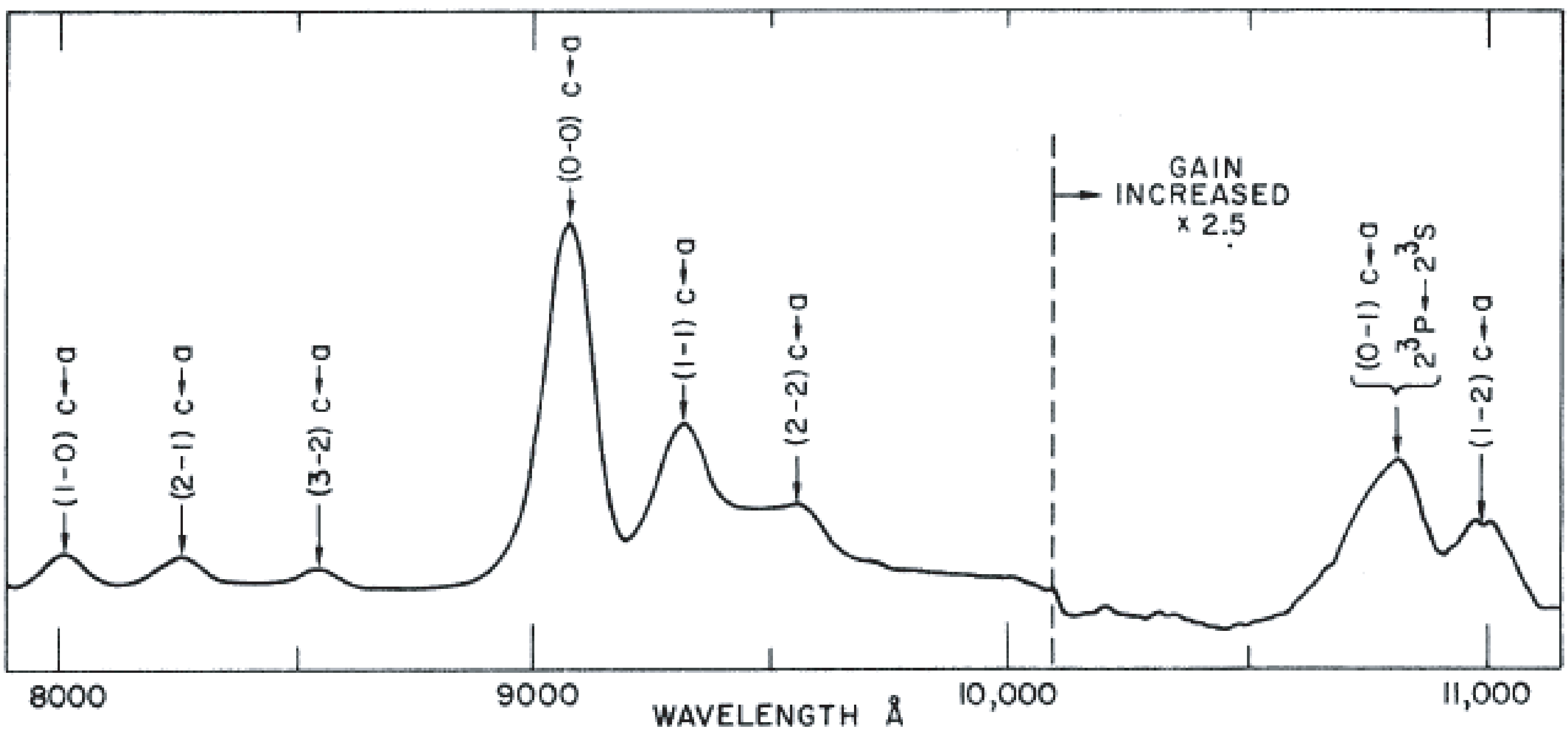
The 640 nm light can be detected with the same photomultipliers used to detect the prompt scintillation light.

By comparing the ratio of prompt light to laser-induced fluorescence, gamma ray backgrounds may be reduced.



Absorption spectrum of electron-excited liquid helium

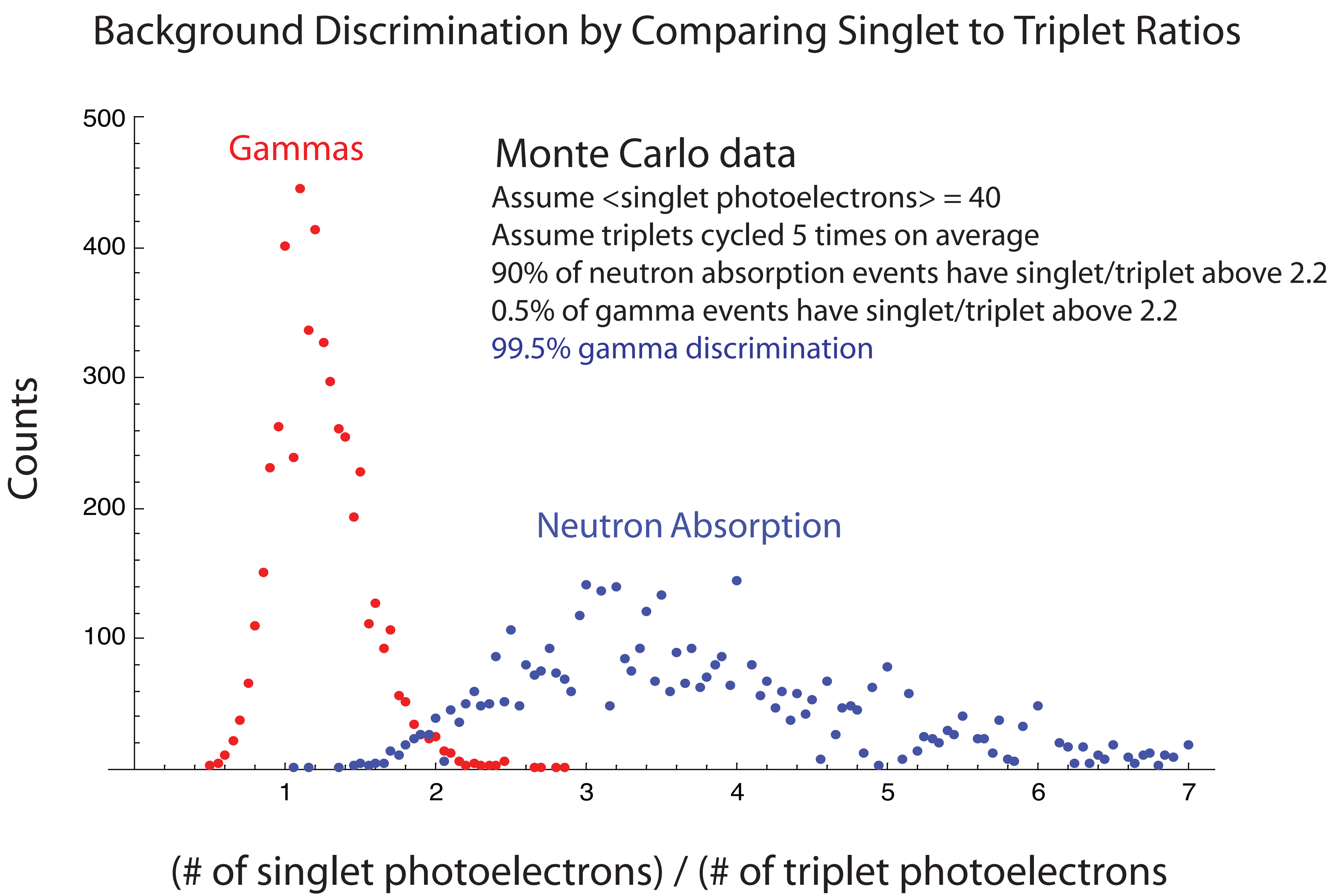
J. C. Hill et al, Phys. Rev. Lett. 26, 1213 (1971).



Strong absorption at 910 nm: c-a transition, 0-0 vibrational

Other vibrational transitions available, including 0-1 at 1070 nm for repumping

(otherwise vibrational relaxation lifetime is 140 ms)


Background Discrimination by Comparing Singlet to Triplet Ratios
Gammas
Monte Carlo data
Assume <singlet photoelectrons> = 40
Assume triplets cycled 5 times on average
90% of neutron absorption events have singlet/triplet above 2.2
0.5% of gamma events have singlet/triplet above 2.2
99.5% gamma discrimination
Neutron Absorption
Counts
500
400
300
200
100
1
2
3
4
5
6
7
(# of singlet photoelectrons) / (# of triplet photoelectrons

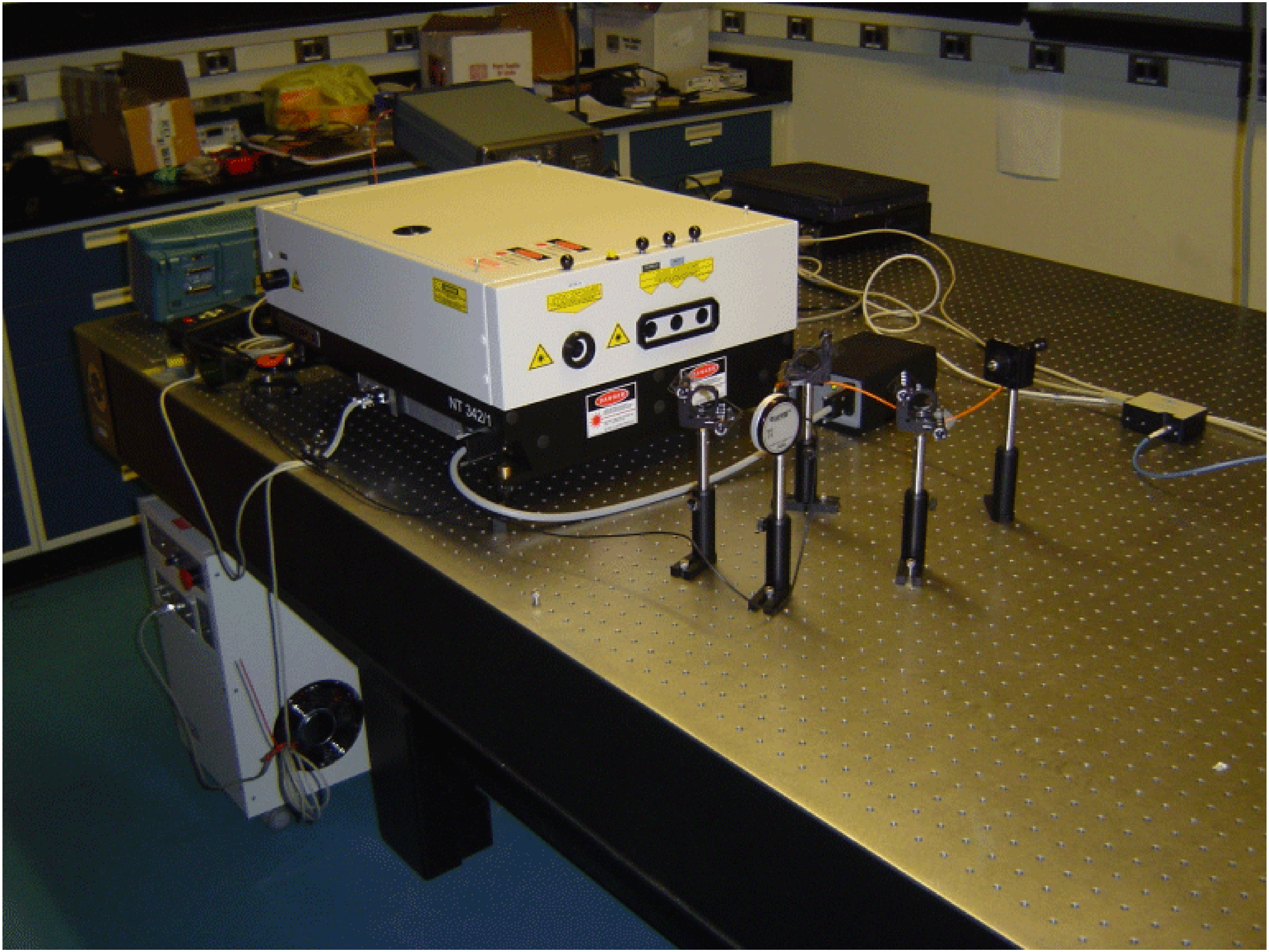
Superfluid helium system fully tested at Yale

Small pumped LHe cell allows laser beam to pass through with T < 1.8 K.

Pulsed OPO laser (10 mJ pulses, tunable from 400 nm to 2400 nm)

A second OPO system will arrive in June.

Photomultipler views cell at 90 degrees, through wavelength filters.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Summary

Detection of triplet helium molecules might be used to enhance the signal from neutron capture in the neutron EDM experiment.

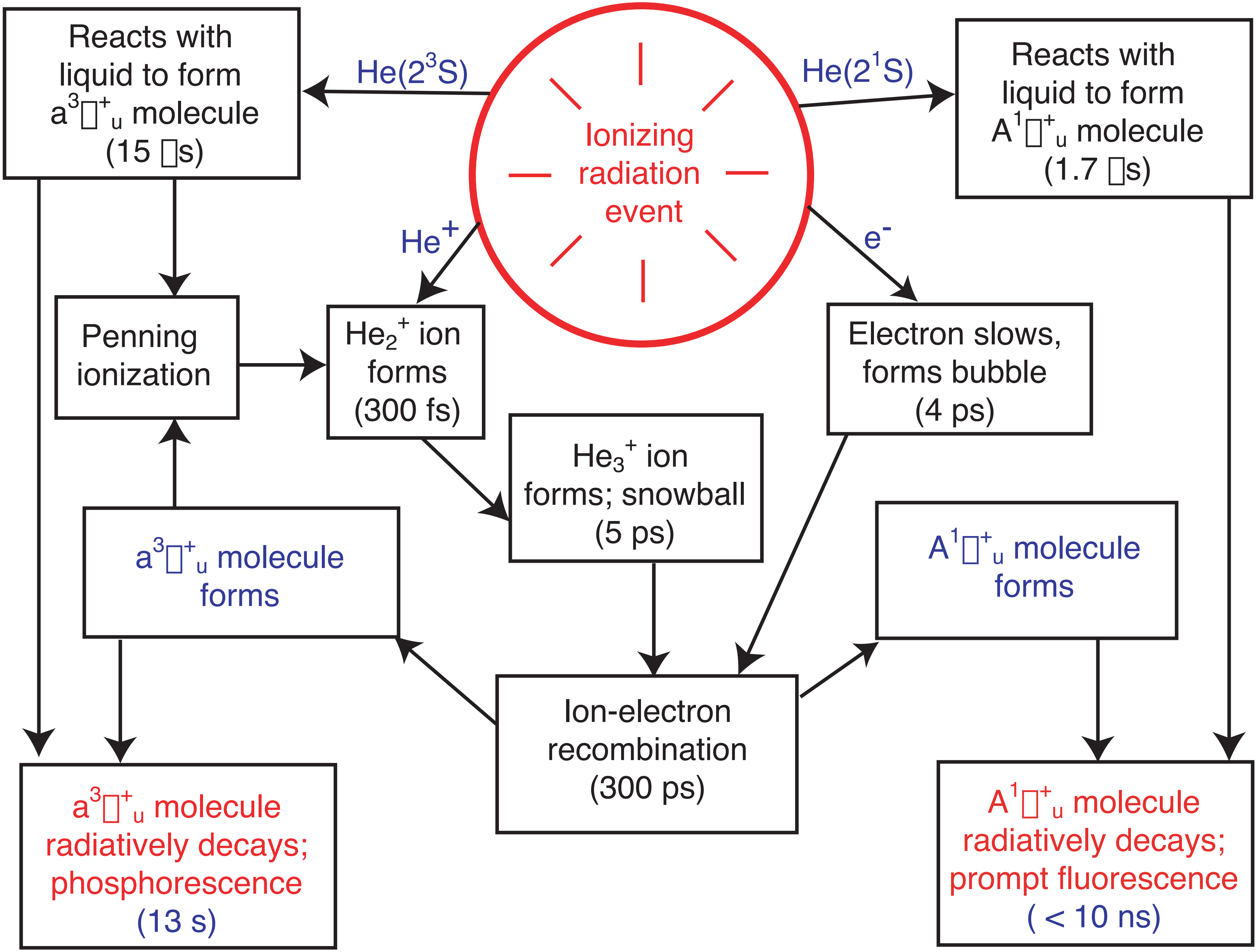
Would require flooding the cell with pulses of 910 and 1040 nm light from a fiber following a scintillation trigger from a candidate neutron absorption event.

Triplet molecules should quench on walls in less than 2.5 ms.

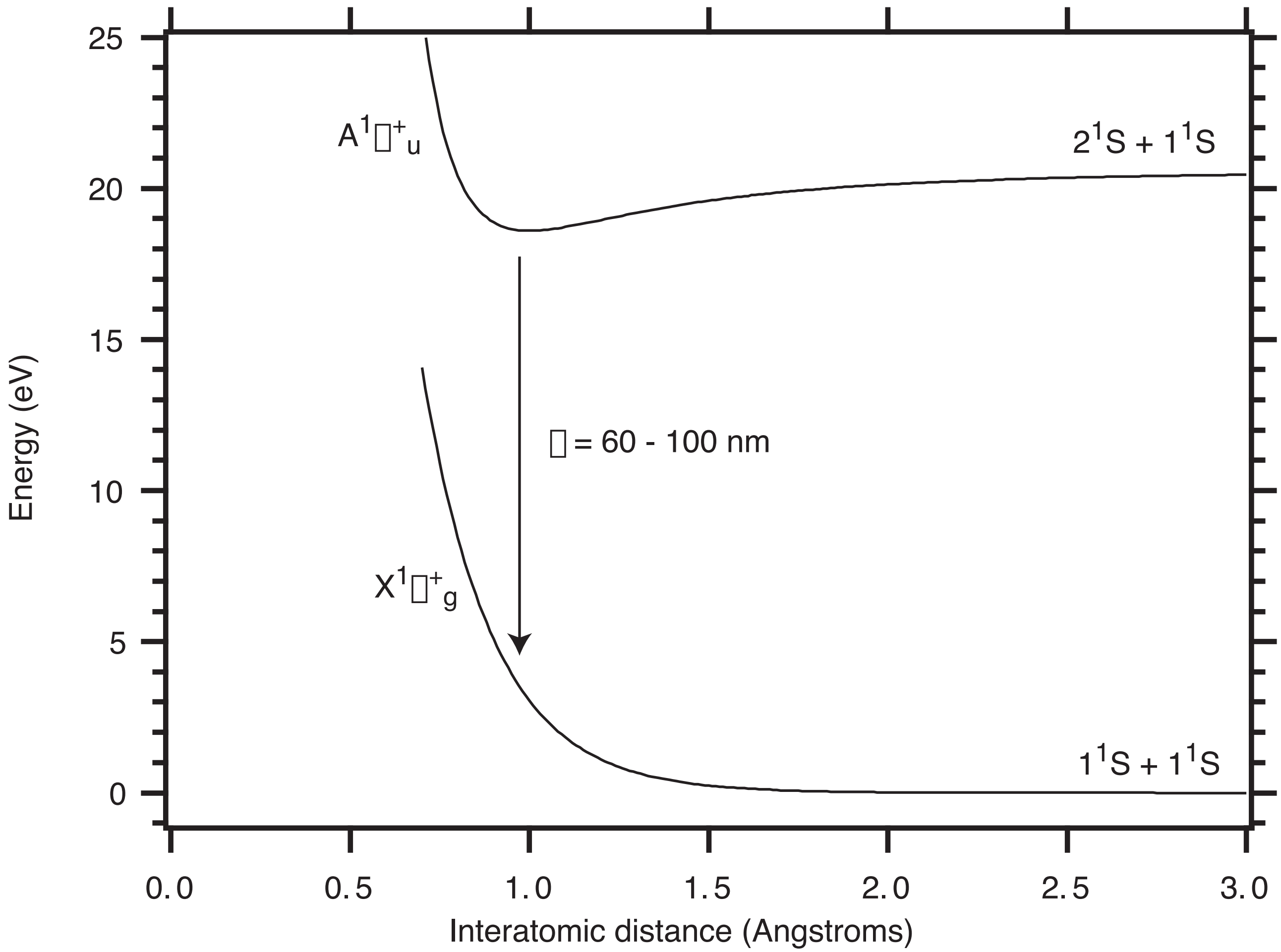
Laser-induced fluorescence would not compromise the "traditional" scintillation detection and could use the same PMTs. Short-pass interference filters would remove any scattered infrared laser light.

This should allow better gamma-neutron discrimination, as recently demonstrated experimentally in liquid neon and liquid argon.

Study of laser-induced fluorescence in superfluid helium is underway at Yale, though no immediate plans to study this at low temperature or with neutron absorption events.



Ionizing radiation event
He(2^{3}S)
He(2^{1}S)
He$^+$
e$^-$
Reacts with liquid to form a^3□^+_u molecule (15 □s)
Reacts with liquid to form A^1□^+_u molecule (1.7 □s)
Penning ionization
He$_2^+$ ion forms (300 fs)
Electron slows, forms bubble (4 ps)
He$_3^+$ ion forms; snowball (5 ps)
a^3□^+_u molecule forms
A^1□^+_u molecule forms
Ion-electron recombination (300 ps)
a^3□^+_u molecule radiatively decays; phosphorescence (13 s)
A^1□^+_u molecule radiatively decays; prompt fluorescence (< 10 ns)



A1□+u
2¹S + 1¹S
X1□+g
1¹S + 1¹S
□ = 60 - 100 nm
Energy (eV)
Interatomic distance (Angstroms)
0
5
10
15
20
25
0.0
0.5
1.0
1.5
2.0
2.5
3.0