

A benign, low Z electron capture agent for negative ion TPCs

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Abstract

We have identified nitromethane (CH_3NO_2) as an effective electron capture agent for negative ion TPCs (NITPCs). We present drift velocity and longitudinal diffusion measurements for negative ion gas mixtures using nitromethane as the capture agent. Not only is nitromethane substantially more benign than the only other identified capture agent, CS_2 , but its low atomic number will enable the use of the NITPC as a photoelectric X-ray polarimeter in the 1-10 keV band.

Key words: , Dark Matter, negative ion, X-ray polarimeter, diffusion, ion mobility
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1 Introduction

The negative ion time projection chamber (NITPC) achieves high spatial resolution by transporting charge in the form of negative ions, rather than electrons, thereby reducing diffusion to the thermal limit in both the transverse and longitudinal drift directions[1]. This provides the highest 3-D space-point resolution attainable for long drift distances, particularly where operation at low pressures is required to make very low energy tracks long enough to measure (for example, dark matter WIMP recoil atoms). In relatively low rate

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25 environments, the NITPC also has the advantage that the ion drift velocities
26 are about two orders-of-magnitude less than those of electrons. This allows
27 the use of low-bandwidth electronics that results in lower noise and power
28 consumption.

29 The key to reducing diffusion to the thermodynamic lower limit (Equation
30 1 below) is to prevent net energy transfer from the drift field to the drift-
31 ing primary ionization[2]. Such “heating” of the drifting charge occurs when
32 electrons are drifted at reduced fields $E/p > \sim 10\text{-}50$ V/m·Torr. Heating is
33 reflected in an increase of the parameter ϵ in Equation 1 to values higher than
34 $\frac{3}{2}kT$, resulting in increased diffusion. For electron drift, gas additives with
35 large integrated cross sections for inelastic scattering of electrons, such as hy-
36 drocarbons and CO_2 , provide channels for dissipation and are known [2,3] to
37 prevent heating of drifting electrons up to moderate values of the reduced field
38 (~ 50 V/m·Torr).

39 In contrast, ions drifting in even a high field are very efficiently thermally
40 coupled to the room-temperature gas, since the ion mass is comparable to
41 that of the gas molecules. Then, even elastic scattering produces substantial
42 energy transfer from the drifting ion to the bulk gas. Ion diffusion has been
43 measured in a number of CS_2 mixtures suitable for TPC operation[1,4,5] and
44 was found to follow the “thermal, low field” limiting behavior[2]:

$$45 \quad \sigma_D = \sqrt{\frac{4\epsilon L}{3eE}} \quad (1)$$

46 Here σ_D is the rms diffusion spread for carriers with charge e after drifting
47 through a distance L in electric field E with average (thermal) energy ϵ . The
48 parameter ϵ (the average thermal energy of a drifting charge carrier) remained
49 approximately equal to the room-temperature value, up to reduced drift fields
50 as high as $\sim 2.5 \times 10^3$ V/m·Torr.

51 Many chemical vapors at STP will capture thermal electrons to form negative
52 ions[6]. However, to maintain good track resolution the agent must have
53 a capture mean free path of a hundred microns or less at a relatively low
54 partial pressure. Since gas gain is desired at the TPC endcaps, the agent
55 must also relinquish its electrons in the high, strongly inhomogeneous field of
56 the gain structures, allowing Townsend avalanches to form. In all previous
57 work, CS_2 has been used as the only capture agent fulfilling these requirements
58 reasonably well.

59 While the NITPC has been successfully implemented with CS_2 as the capture
60 agent in dark matter observatories[7–9], the utility of the NITPC would be ex-
61 panded by the identification of new capture agents. CS_2 has a combination of
62 low flash point (-30° C), high vapor pressure (400 Torr) and low explosive mix-

63 ture limit in air (1.3%) which require great care in handling the material[10].
64 Nitromethane[11] has a flash point far above room temperature and a vapor
65 pressure less than a tenth that of CS₂. The comparative health effects of the
66 two materials are indicated by the respective ACGIH Threshold Limit Values
67 of 1 ppm for CS₂ vs. 20 ppm for nitromethane. CS₂ also may form sulfide
68 deposits under conditions of high rate or discharge. More importantly, new
69 applications for the NITPC would be enabled by capture agents with different
70 functional properties. For example, a single-ion counting NITPC with high
71 energy resolution for double beta decay searches has been proposed[12] that
72 would benefit from a capture agent with higher ion mobility than CS₂. An-
73 other example is photoelectric X-ray polarimetry below 10 keV with a NITPC,
74 which requires capture agents with lower atomic number (see below).

75 Photoelectric polarimeters derive their information by determining the photo-
76 electron emission direction, which is strongly correlated with the electric field
77 vector of the incident photon. The most sensitive photoelectric polarime-
78 ters image the tracks of the photoelectrons with pixels small compared to the
79 photoelectron track length and reconstruct the emission direction on an event-
80 by-event basis. Such polarimeters have been realized with optical avalanche
81 chambers[13,14], micropattern gas detectors with direct pixel readout[15–17],
82 and a micropattern TPC[18].

83 X-ray polarimetry below 10 keV with a NITPC would benefit from low atomic
84 number capture agents for several reasons. First, the correlation between
85 the photoelectron emission direction and photon electric field is strongest for
86 emission from s-orbitals[19], so it is preferable to use gases whose K-shell
87 energies are well below the X-ray energy. Second, the photoelectron is emitted
88 with a kinetic energy that is the difference between the binding energy and the
89 X-ray energy so that lower Z gases result in photoelectrons with larger kinetic
90 energies and longer track lengths. Finally, lower Z gases have less multiple
91 Coulomb scattering, which results in straighter tracks and a better correlation
92 between the photoelectron emission direction and photon electric field.

93 Therefore, motivated by the requirements of low energy X-ray polarimetry and
94 guided by the chemical literature on electron capture, we have explored a num-
95 ber of compounds as alternative NITPC capture agents. We have identified
96 nitromethane (CH₃NO₂) as a suitable capture agent for X-ray polarimetry
97 which is also relatively benign. We present measurements of drift velocity,
98 gas gain and longitudinal diffusion in gases with nitromethane as the capture
99 agent.

100 2 Capture agent screening apparatus

101 Capture agents were screened for negative ion drift velocity, gas gain and
102 longitudinal diffusion at low to moderate fields using a single-wire proportional
103 counter attached to a homogeneous-field drift region. The apparatus was
104 enclosed in a stainless steel bell jar with a simple gas manifold. Drift fields
105 up to 4.0×10^4 V/m were used with this apparatus at reduced pressure.
106 Photoelectrons were generated at an Sn photocathode attached to the drift-
107 cathode. The photocathode was arranged so that it could be cleaned between
108 runs using a glow discharge in pure argon. This was essential for maintaining
109 the photoelectron yield.

110 UV light flashes from an EG&G Flash-Pak[20] were admitted into the bell jar
111 through a quartz window, passed through a hole in the proportional counter
112 wall, and struck the photocathode producing photoelectrons. The standard
113 internal capacitors of the Flash-Pak were augmented with additional HV ca-
114 pacitors to give a stored energy of about 0.2 Joule per pulse. The Flash-Pak
115 was triggered by an external pulser, from which a time-zero signal was also de-
116 rived. The proportional wire signal was read out through an ORTEC 142PC
117 preamp and an ORTEC 579 shaping amplifier. Drift times to the proportional
118 wire and wire-signal time widths were measured with a digital oscilloscope.

119 Drift velocities and gas gain at moderate to high fields (1.0×10^5 - 4.0×10^5
120 V/m) were measured in various gas mixtures with a micropattern gas detector
121 (MPGD) having the same electrode structure as the gas electron multiplier[21],
122 but assembled from two etched stainless steel meshes[18] separated by a 100
123 μm thick teflon spacer. A drift electrode, also of stainless steel, was placed
124 10 mm above the MPGD cathode. The MPGD was operated at a gain of
125 about 3000. The MPGD cathode was instrumented with a charge-sensitive
126 preamplifier followed by a bipolar shaping amplifier with a six microsecond
127 shaping time constant.

128 The MPGD cathode and drift electrode were simultaneously illuminated with
129 a xenon flashlamp (Perkin-Elmer PAX-10), which produced photoelectrons
130 from both surfaces. Photoelectrons from the cathode produced a prompt
131 pulse, while the photoelectrons from the drift electrode produced negative ions
132 and gave a pulse delayed by the drift time across the 10mm drift distance. The
133 drift time was measured by averaging 100 pulses on a digital oscilloscope and
134 taking the difference between the zero-crossing times of the prompt and the
135 delayed pulses.

136 3 Results and Discussion

137 Nitromethane is known to have a large capture cross section for thermal elec-
138 trons[6]. However, at moderate to high drift fields the pure near-saturated
139 vapor at 20 Torr produces both electron-drift and negative-ion-drift signals.
140 This is understandable since a drift field of 5.0×10^4 V/m at 20 Torr is al-
141 ready a reduced field of 2500 V/m·Torr, easily high enough to raise the energy
142 of drifting electrons out of the thermal range before they could be captured.
143 CO₂ was therefore added to the nitromethane vapor. CO₂ is known to be very
144 effective in thermalizing drifting electrons, due to its large inelastic scatter-
145 ing cross sections[3]. Mixtures of 20 Torr nitromethane with 50 Torr or more
146 of CO₂ were found to have satisfactory characteristics as negative ion drift
147 mixtures.

148 Drift velocity and diffusion results for the nitromethane:CO₂ 20:50 mixture
149 are shown in Figures 1 and 2. The drift velocity rises linearly with drift field,
150 with mobility $(4.27 \pm .03) \times 10^{-4}$ m²/V·sec and a small positive intercept
151 $(1.44 \pm .08$ m/s). The diffusion is studied by plotting the square of the pulse
152 width in time as a function of $1/(v_d^2 E_d)$. The pulse width is expected to be the
153 quadrature sum of the amplifier shaping time (10 μsec) and the broadening
154 due to diffusion. If the diffusion is governed by Equation 1, this plot will
155 be linear in the region where diffusion dominates (essentially the entire plot),
156 with slope. Fitting Figure 2 to a straight line gives $T = 313 \pm 25$ K.

157 Drift velocities for higher pressure gas mixtures of nitromethane-CO₂-neon,
158 nitromethane-CO₂-argon and nitromethane-CH₄ were measured up to drift
159 fields of 4.0×10^5 V/m. Results are shown in Figure 3. The linear dependence
160 of drift velocity on drift field persists up to these very high fields. Results of
161 linear fits for the mixtures studied are shown in Table 1. Shown in Figure
162 4 are gas gain curves for higher pressure mixtures of CO₂-neon, and neon-
163 CO₂-nitromethane. Gains as high as several thousands were obtained in all
164 mixtures demonstrating the ability of nitromethane as a feasible, productive
165 detector gas.

166 4 Conclusion

167 A negative ion drift mixture of nitromethane with CO₂ is found to have linear
168 drift velocity vs. field and to exhibit thermal-limit longitudinal diffusion up to
169 drift fields as high as 2.8×10^4 V/m. Higher pressure mixtures with argon,
170 neon, CH₄ and CO₂ also show linear drift velocity up to fields as high as 4.0
171 $\times 10^5$ V/m. All mixtures exhibit stable operation as NITPCs.

172 The introduction of nitromethane as an electron capture agent will enable the
173 use of the NITPC as an X-ray polarimeter in the 1-10 keV band. The low drift
174 velocity of the NITPC greatly eases the difficulty of calibrating drift velocity
175 and substantially reduces power consumption, which is particularly important
176 for satellite-borne astronomical instruments. The low diffusion of the NITPC
177 will also enable large-volume photoelectric polarimeters that could be used,
178 for example, as wide field-of-view instruments for unpredictable astronomical
179 transients, such as gamma-ray bursts, or with rotation modulation collimators
180 for high-resolution imaging polarimetry of solar flares[22].

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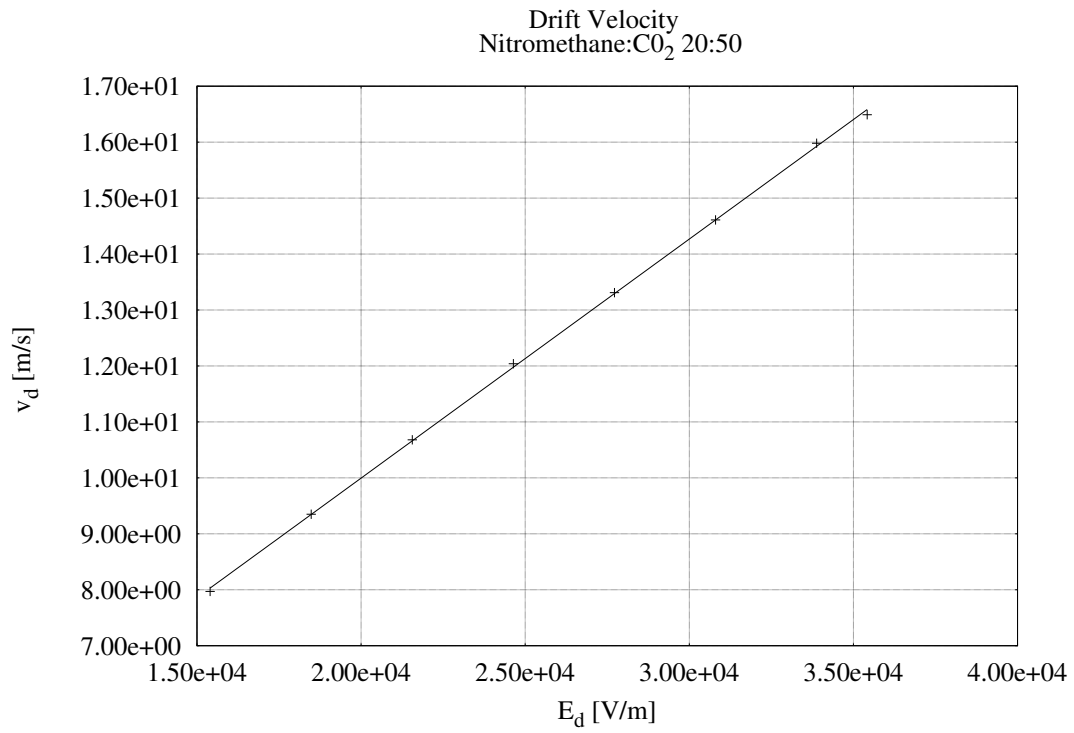


Fig. 1. Drift velocity v_d vs. drift field E_d for Nitromethane:CO₂ 20:50

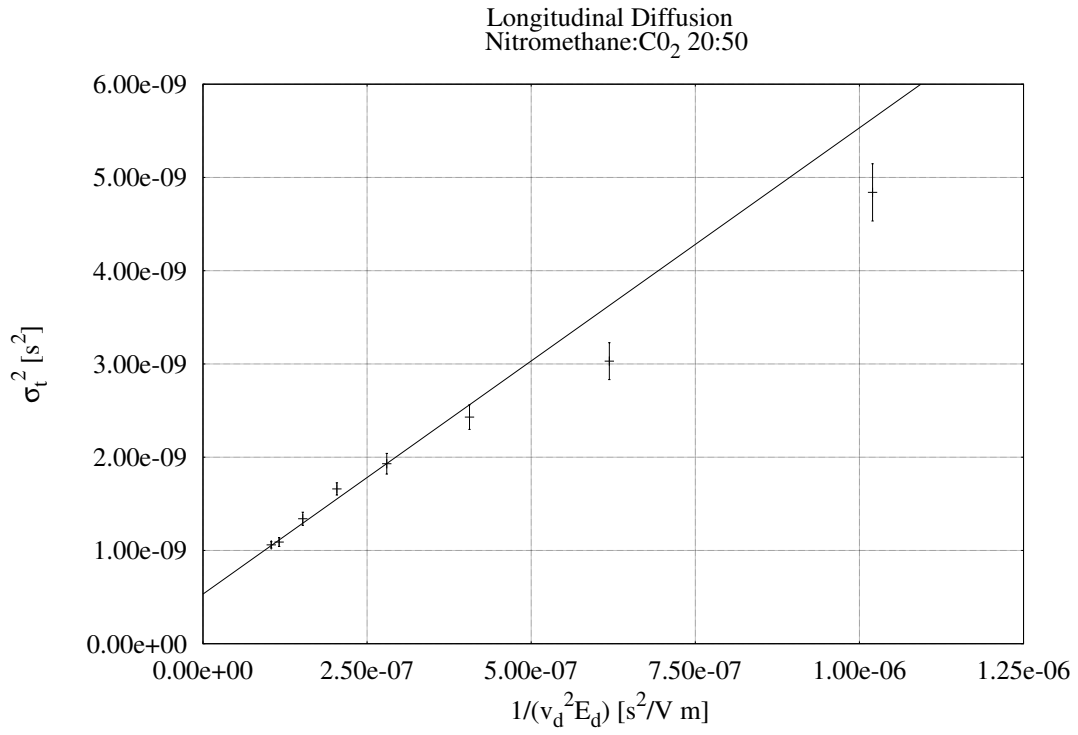


Fig. 2. Longitudinal diffusion data for Nitromethane:CO₂ 20:50, plotted as the square of the pulse time width vs. $1/v_d^2 E_d$ for 80 mm drift.

Mixture	CO ₂ Torr	Neon Torr	Argon Torr	Methane Torr	Mobility [m ² /V·s]
A	510	170	–	–	$8.43 \pm .01 \times 10^{-5}$
B	170	-	510	–	$9.69 \pm .01 \times 10^{-5}$
C	340	340	–	–	$10.5 \pm .01 \times 10^{-5}$
D	170	510	–	–	$13.1 \pm .02 \times 10^{-5}$
E	–	–	–	380	$21.0 \pm .07 \times 10^{-5}$

Table 1

Fitted slopes for ion drift in mixtures of 20 Torr Nitromethane with CO₂, Neon, Argon and Methane.

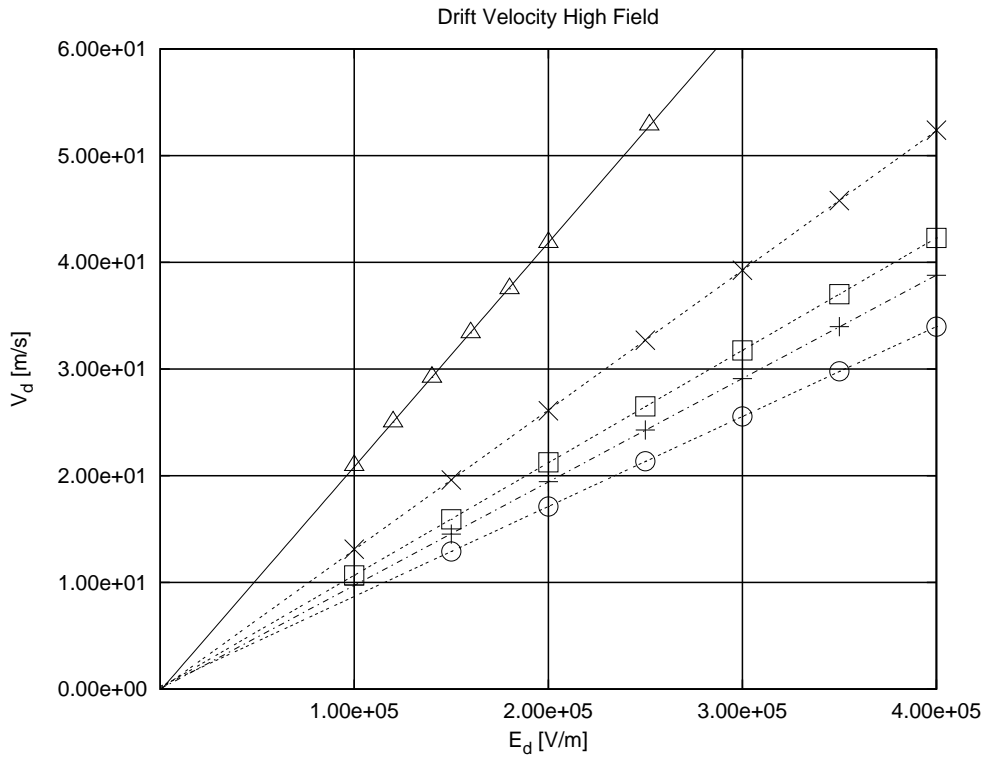


Fig. 3. Drift velocity at high fields in mixtures of Nitromethane and CO₂ and/or Neon, Methane mixtures A-E described in Table 1. Circles: A, Plusses: B, Squares: C, Crosses: D, Triangles: E

Mixture	CO ₂ Torr	Neon Torr	Nitromethane
A	210	490	-
B	170	510	20
C	340	340	20
D	510	170	20

Table 2

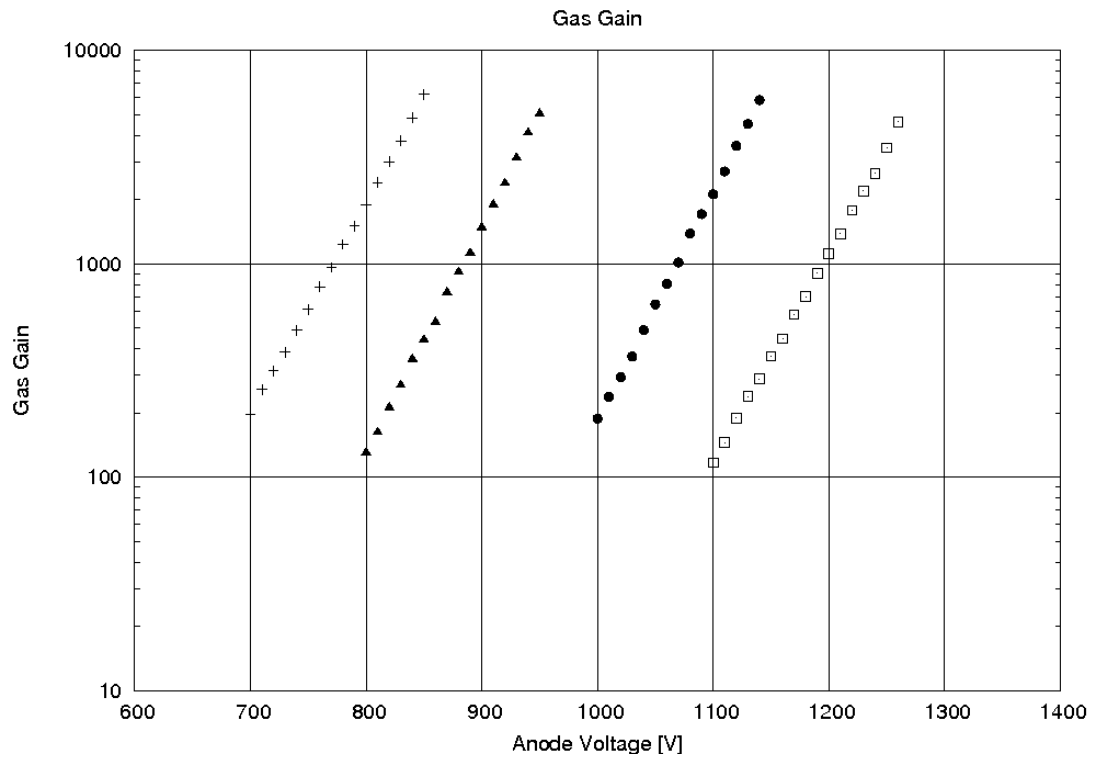


Fig. 4. Gas Gain in mixtures containing Neon and CO and/or Nitromethane A-D described in Table 2. Plusses: A, Triangles: B, Circles: C, Squares: D

183 **References**

- 184 [1] C.J. Martoff et al., Nucl. Inst. and Meth. A 440 (2000) 355.
- 185 [2] W. Blum, L. Rolandi, Particle Detection With Drift Chambers,
186 Springer-Verlag Telos, 1994.
- 187 [3] F. Sauli, “Multiwire Proportional Chambers” in T. Ferbel, ed.,
188 Experimental Techniques in High Energy Physics, Addison-Wesley
189 Publishing, New York, 1987.
- 190 [4] C.J. Martoff, et al., Nucl. Instr. and Meth. A 555 (2005) 55.
- 191 [5] T. Ohnuki, et al., Nucl. Instr. and Meth. A 463 (2001) 142.
- 192 [6] E.C.M. Chen, E.S. Chen, The Electron Capture Detector and the
193 Study of Reactions With Thermal Electrons, John Wiley & Sons,
194 Hoboken, NJ, 2004.
- 195 [7] D.P. Snowden-Ifft, et al., Nucl. Instr. and Meth. A 498 (2003) 164.
- 196 [8] G.J. Alner, et al., Nucl. Instr. and Meth. A 535 (2004) 644.
- 197 [9] T.B. Lawson, et al., Nucl. Inst. and Meth. A 555 (2005) 173.
- 198 [10] Mallinkrodt/J.T. Baker MSDS N5740 for Carbon Disulfide, 2006.
- 199 [11] Mallinkrodt/J.T. Baker MSDS C0957 for Nitromethane, 2006.
- 200 [12] D.R. Nygren, J. Phys. Conf. Ser. 65 (2007) 012003.
- 201 [13] A.A. Austin, B.D. Ramsey, Optical Engineering 32 (1993) 1990.
- 202 [14] H. Sakurai, et al., Nucl. Instr. and Meth. A 525 (2004) 6.
- 203 [15] E. Costa, et al., Nature 411 (2001) 662.
- 204 [16] J.K. Black, et al., Nucl. Instr. and Meth. A 513 (2003) 639.
- 205 [17] R. Bellazzini, et al., Nucl. Instr. and Meth. A 566 (2006) 552.
- 206 [18] J.K. Black, et al., Nucl. Instr. and Meth. A 581 (2007) 755.
- 207 [19] J.W. Cooper, Phys. Rev. A 47 (1993) 1841.
- 208 [20] Model LS-1102-1, EG&G Optoelectronics, Salem, MA.
- 209 [21] F. Sauli, Nucl. Instr. and Meth. A 522 (2004) 93.
- 210 [22] J.K. Black, J. Phys. Conf. Ser. 65 (2007) 012005