Neutron – Alpha irradiation response of superheated emulsion detectors

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1. Introduction

The impact of any particle detector is often dependent on its ability to discriminate between detector-recorded, particle-induced backgrounds. Among the various detector types in present use are superheated liquid devices. These generally consist of either superheated emulsion detectors (SEDs): comprising distributions of micrometric liquid droplets in a gel-like medium (SDDs) or rigid polymer matrix (BDs) [1], or bulk liquid bubble chambers. Both record the acoustic signal associated with the particle-induced bubble nucleation event, as well as other non-particle acoustic backgrounds associated with environmental noise. Due to the thermodynamics of the detector response, particle sensitivity in moderately superheated devices is essentially reduced to high linear energy transfer (LET) radiations; an intrinsic low LET particle insensitivity of better than $10^{-10}$ has been demonstrated by $\gamma$ and electron irradiations [2]. With increasing liquid superheat, this insensitivity decreases and the devices record these events also.

SEDs have been investigated for a number of radiation detection applications including neutron, health, medical, and space physics [1,3] involving neutron, proton, heavy ion, electron and $\gamma$-ray fields. Because of their application to the direct search for astroparticle dark matter, which is critically dependent upon their ability to isolate low energy nuclear recoil events ($< 100 \text{ keV}$) generated by elastic scattering of weakly interacting massive particles from naturally-occurring, low level neutron and $\alpha$ backgrounds in the materials, the question of particle response in superheated liquid devices has come under severe scrutiny and a large part of the advances in their capabilities has emerged from two experiments. In 2008, the PICASSO project using C$_4$F$_{10}$ dispersed in a Gaussian droplet distribution with mean of $<d>$ = 100 ± 25 $\mu$m, high frequency piezo instrumentation, and irradiations effected with AmBe (neutron) and $^{241}$Am and/or $^{226}$Ra doping ($\alpha$), reported a partial separation of the neutron-generated recoil-$\alpha$ acoustic event amplitude (A) distributions [4]. In 2010, the SIMPLE project independently reported [5] a full separation of the two power distributions in irradiations of separate devices, using C$_2$ClF$_5$ with a Gaussian droplet distribution of $<d>$ = 30 ± 7.5 $\mu$m, a low frequency electret microphone, U$_3$O$_8$ $\alpha$-doping and neutron irradiations with either AmBe or epithermal neutrons from the Portuguese Research Reactor (PRR) [6], which was attributed to the difference in proto-bubble formation. In 2011, PICASSO presented “new insights” into the detector response, this time with the same SEDs but based on the recorded event acoustic energy [7] obtained by squaring the waveform of each transducer signal and integrating over its duration, which also identified the difference in recoil-$\alpha$ proto-bubble formation as the determining factor for the separation of the two response distributions. Experimentally, however, only a partial separation between the recoil-$\alpha$ events was again obtained, and only in the case of $^{226}$Ra $\alpha$-calibrations. The difference between the results of the two projects has since remained unresolved [8].
Other researchers have since contributed to the issue of particle response, principally however with respect to neutron-generated nuclear recoil and γ events [9,10]. Das et al. recently reported [11] recoil α response studies using CCl₂F₂, a low frequency condenser microphone, a bimodal Gaussian distribution of small droplet sizes (58% < r1 > ~ 3 ± 1 μm, 42% < r2 > ~ 25 ± 5 μm), and irradiations with neutron (AmBe) and a solid α source (241Am) – the latter which, in contrast to the usual emitter-doping, was positioned outside the SED gel. Analysis of the data via the acoustic signal energy demonstrated an inversion of the recoil - α response amplitudes, accompanied by a significant overlap of the two distributions.

The detectors of each experiment differ in various aspects: superheated liquid and concentration, droplet sizes, gel composition and stiffness, size and volume of detectors, operating temperatures and pressures, instrumentation, signal acquisition and analysis. We here examine the response issue in small (rd < 30 μm) droplet size distribution SEDs, towards providing an improved understanding of the involved mechanics for general use in future SED implementations. Section 2 summarizes the bubble nucleation physics underlying SED performance. Section 3 describes new experiments using single SEDs with several droplet size distributions, and modified gel stiffness, and discusses the results. Section 4 elaborates on the particle-superheated liquid interactions, and introduces a simple model of the SED response which is seen to reproduce well the experimental results. The difference in number of proto-bubbles created by recoil target ion and α’s, the initial energy of the α in the liquid, and the differences in droplet size distributions, are identified as the basis for the particle response. A summary of conclusions is given in Section 5.

2. Irradiation response

The general physics of detector operation, based on the “thermal spike” model of Seitz [12], has been described by various authors [13–21] (and references therein). The process consists of several stages beginning with the incident particle energy deposition in a small (∼3 × 10⁻⁴ μm³) volume of the liquid, creating ionization electrons which generate a localized, high temperature region (the “thermal spike”). The resulting sudden vaporization of the region and its expansion generates a shock wave in the droplet, in which the temperature and pressure within the shock enclosure initially exceed the critical temperature and pressure of the liquid: there is no distinction between liquid and vapor, and no bubble. As the energy is transmitted from the thermalized region to the surrounding medium through shock propagation and heat conduction, the temperature and pressure of the fluid within the shock enclosure decrease and the expansion process slows [17]. As the temperature and pressure continue decreasing to their critical values, a vapor-liquid interface is formed which may generate a proto-bubble of submicron critical radius r_c = 2σ(T)/Δρp of the vapor state at which the pressure difference (∆p = p_v – p_l) – vapor, l-liquid) overcomes its surface tension σ. If this is not achieved, growth is impeded by interfacial viscous forces and conduction heat loss, and the proto-bubble collapses; otherwise, the droplet evaporation generates an expanding gas bubble, accompanied by a pressure wave. The time scale for proto-bubble creation is sub-nanosecond; complete droplet evaporation occurs over milliseconds.

Only 2–6% of the total energy release appears acoustically. The acoustic energy release is given by [22]:

\[ E = A^2 \tau = -\frac{4\rho_i r_b^6}{3c \tau^4}, \]  

from which the acoustic power is

\[ K = A^2 = \frac{4\rho_i r_b^6}{c \tau^4}, \]

where \( \rho_i \) is the liquid density, \( c \) is the speed of sound in the liquid, \( r_b = r_b(\tau) \) is the bubble radius, \( \tau \) is the expansion time of the bubble, and \( A \) is the signal amplitude.

Proto-bubble formation occurs if the particle energy deposition \( E \) satisfies [12]:

\[ E \geq E_c = 4\pi c^2 \left( \sigma - T \frac{\partial \sigma}{\partial T} \right) + \frac{4\pi r^3}{3} \frac{\partial \varnothing}{\partial T}, \]

and

\[ \frac{dE}{dx} \geq E_c L_c \equiv LET_c, \]

where \( T \) is the SED operating temperature, \( \rho_v(T) \) is the vapor density, and \( h_v(T) \) is the heat of vaporization. The \( E_c/L_c \) is the critical deposited energy density required for proto-bubble nucleation, with \( L_c = \Lambda r_c \) the effective ionic energy deposition length, and \( \Lambda \) an empirical liquid-dependent parameter. Both \( E_c \) and \( r_c \) when displayed as a function of the reduced supersaturation \( s = (T - T_b)/(T_c - T_b) \) with \( T_c, T_b \) the critical and boiling temperature of the liquid at a given pressure [21] respectively, are seen to provide “universal” curves for different liquids (Fig. 1(a)). Similarly, the response of the SEDs to a given irradiation type lie on “universal” curves for different liquids (Fig. 1(a)).
curves when displayed in $s$ (Fig. 1(b)); sensitivity to $\gamma$’s and electrons is seen for $s \geq 0.51$, increasing with $s$.

Unlike $E_c$ and $r_c$, $\Lambda$ however appears to exhibit no universal behavior when evaluated in terms of reduced superheat [23]. Whereas $r_c$ and $E_c$ are well-defined thermodynamically, $\Lambda$ is without concrete theoretical basis and must be determined experimentally. It is constant for a liquid at a given $T$, $p$ [24]. It generally ranges from 2 to 12.96 [21], with reported values increasing up to 20 and larger for neutron-induced recoil thresholds above 1 MeV [24–28]. Studies with heavy ions indicate that $\Lambda$ may also depend on the ion mass number, decreasing with number increase [29].

3. New irradiation results and discussion

All measurements were performed using 150 ml versions of the standard ($< r > = 30 \pm 7.5 \mu m$) SIMPLE science (900 ml) detector. Because of uncertainties in the recompression capability to restore the original droplet size distribution, several pairs of near-identical SEDs were fabricated, one of which was neutron-then-$\alpha$ irradiated, and the second with the sequence reversed. The following pairs of SEDs were considered:

(i) two C2ClF5 SEDs (1.3% and 1.4%wt) fabricated to provide a droplet size distribution peaked at $\sim 10 \pm 1 \mu m$.

(ii) two C2F6S SEDs (1.3% and 1.8%wt) similarly fabricated, with a reduced liquid fractionating time to yield a droplet size distribution peaked at $\sim 23 \pm 3 \mu m$.

(iii) a third pair of SEDs (1.3% and 2.1%wt) made with a stiffer gel by adding twice as much gelatin. The liquid fractionating time was adjusted to provide a droplet size distribution peaked at $\sim 10 \pm 1 \mu m$.

Two additional SEDs were also fabricated:

(iv) a 150 ml SED with the stiffer gel (as above) and 2.3 g (1.3%wt) of C2F6S fractionated to give a larger distribution peaked at $\sim 23 \pm 3 \mu m$.

(v) a 150 ml SED with the standard gel and 3 g (1.7%wt) of C2ClF5 fractionated to give a droplet size distribution peaked at $\sim 5.5 \pm 3 \mu m$.

From polarimetry measurements, the rate of helix formation $\chi = 0.50$ [30]: since the helix concentration is $c_{helix}=\chi c_{gel}$ [31,32] and the standard gel $c_{gel}=8 \times 10^{-3} \text{ g cm}^{-3}$, $c_{helix}=16 \times 10^{-3} \text{ g cm}^{-3}$ for the "stiff" SED. The elastic modulus is given by $G'=325 \text{ Pa leading to a Young's modulus of } Y_{standard}=3 G'=975 \text{ Pa}$ for the standard gel and $Y_{stiff}=6 \text{ kPa}$ [30]. In the case of PICASSO, with a concentration of acrylamide $\sim 6\%$ and Bis-acrylamide $=0.16\%$, $Y_{PICASSO}=5 \text{ kPa}$ according to [33], and the "stiff" gel reproduced that of PICASSO.

The droplet size distributions were measured in randomly-selected slices of each gel matrix, taken from randomly-selected sites in each SED volume, by an optical microscope (Olympus Model Bx 60 M). The results in each slice were similar. The resulting distributions were fit both with a Gaussian ($\text{mean} < R >, \sigma$) and a Lorentzian (peak $< R >, \Gamma$), and seen to exhibit the Lorentzian profile which included a non-negligible larger radii tail.

All SEDs but (v) were injected with 0.94 Bq of a U3O8 solution; the (v) SED was doped with 0.37 Bq of U3O8 during fabrication. Neutron irradiations were performed either on the $\gamma$-shielded PRR thermal column (Case (i) above) or with a 1 mCi AmBe source located at 1.5 m from the SEDs to provide 2–3 recoil events per min (Cases (ii)-(v)). Each SED was placed inside a temperature-controlled, circulating water bath; the bath temperature was temperature- monitored with a 0.1 °C uncertainty. Detector responses were recorded for up to 24 h at operating temperature and pressure (hereafter OTP) of 9 °C and 2 bar, except in the case of the (iv) SED with OTP of 27 °C and 1 bar, and the (v) SED with OTP of 1 bar over a temperature range of 5–13 °C.

All results were obtained using standard SIMPLE instrumentation, comprising a single electret microphone with 0–16 kHz sensitivity, without amplification or filtering. Analysis was also standard, using the natural logarithm of the squared amplitude of the principal harmonic of the power density spectrum ($\ln(K)$) for each event with frequency 450–750 Hz and decay constant of 5–40 ms [34].

The measured power distribution of the irradiation sequences are shown in Figs. 2–5, with the "(a)" displaying the neutron-then-$\alpha$ irradiations, and the "(b)" the $\alpha$-then-neutron irradiations; each response distribution was normalized to unity:

Case (i). the results shown in Fig. 2(a), comprising 794 recoil and 532 $\alpha$ events, indicate the existence of the gap between the recoil and $\alpha$ power distributions. The $\alpha$ distribution is asymmetric tailing to higher powers. In Fig. 2(b), with 672 recoil and 631 $\alpha$ events, the recoil distribution is virtually identical to that of Fig. 2(a). In comparison with Fig. 2(a), the $\alpha$ distribution is significantly broadened in its lower power structure, with its peak shifted downwards to $\ln(K)=9.5$. The two response distributions are almost separated.

The apparent shift of the neutron distribution in Fig. 2(a) from that of previous measurements [4] with a 900 ml science detector.
is a result of the smaller SED size, and pressure wave attenuation. The mean of the sound distance in the larger device was \( \sim 5 \text{ cm} \), vs. the 4 cm of the devices herein, yielding an amplitude increase of \( \sim 20\% \) to peak at \( \ln(K) = 7.5 \).

Case (ii). In contrast to Case (i), the neutron-then-\( \alpha \) result in Fig. 3 (a), comprising 727 and 607 events, indicates an increased overlap of the two power distributions. The recoil distribution is shifted to higher power, while the peak of the \( \alpha \) distribution remains

![Fig. 3. Recoil- and \( \alpha \)-induced event power distributions as in Fig. 2: big droplet distribution.](image)

![Fig. 4. Recoil- and \( \alpha \)-induced event power distributions: stiffer gel SEDs.](image)

![Fig. 5. (a) Spectra of recoil- and \( \alpha \)-induced event power distributions: \( \text{C}_4\text{F}_{10} \) SED at 27 °C/1 bar, with stiffer gel and \( <r > \sim 23 \mu \text{m} \); (b) \( \text{C}_2\text{ClF}_5 \) SED at 9 °C/1 bar, \( <r > \sim 5.5 \mu \text{m} \).](image)
virtually unaltered with a slight decrease in its low power tail to overlap the high power tail of the recoil response. The histogram of the reversed irradiation sequence (770 recoil and 598 α events), is shown in Fig. 3(b); the α response peak is similar to Fig. 2(b) but its low power tail reaches to ln(K) ~ 8. The recoil distribution, no longer symmetric, exhibits a high power tail spanning the α distribution, with its peak now shifted downward to ln(K) = 7.8 as in Fig. 2(b). The recoil response is slightly higher than Figs. 2(b) or 3(b); the α response, slightly lower than either.

Case (iii). Fig. 4(a) comprises 480 recoil and 570 α events. Both responses are shifted upwards in power relative to Fig. 2(a), with the exception of the lower limit of the α-response. The α-distribution is seemingly narrower, and accompanied by a secondary high power component. The recoil distribution is of higher power than either Figs. 2(a) or 3(b), as also the α-distribution with respect to Figs. 2(b) or 3(b); it appears somewhat broadened, but in fact remains contained between ln(K) = 7–10. Both distributions overlap, and include high power tails. In Fig. 4(b), comprising 281 neutron- and 453 α-induced events, the α peak has returned to that of Fig. 3(a), with the principal distribution again seemingly narrower and accompanied by a significant low power tail. The recoil distribution is broader than in Fig. 2(b) as a result of an enhanced high power tail.

Case (iv). The results of the neutron-then-α irradiations (364 recoil and 231 α-events) are shown in Fig. 5(a). The two power distributions are clearly non-coincident, the results being similar to those of both Figs. 3(a) and 4(b). There is no significant overlap of the primary response distributions, the small overlap arising from the high power recoil response tail which extends well into the α distribution.

Case (v). The α-then-recoil response at 9 °C and 1 bar (909 α and 874 recoil events) is shown in Fig. 5(b). Both power distributions are asymmetric as in Fig. 2(a), and of lower power relative to both the α- and recoil responses of Figs. 2–4; the α response is of higher power than the recoil event, unlike the results of Ref. [11].

Table 1 contains a summary of the SED characteristics and observed power distribution parameters. In all cases, the results manifest a clear separation between recoil and α-induced event peaks. With the exception of Case (i), there is generally a partial overlap of the low power α and high power recoil distributions. The recoil spectra of the “(b)” Figs. include the simultaneously occurring α-decay during the neutron irradiations which contribute to the higher power tails. In Case (ii), all α distributions generally appear symmetric, as also the recoil response of Fig. 3(a) whose peak power has also risen slightly relative to Case (i). In Case (iii), the α distributions appear narrower, whereas the recoil distributions appear broader. The results of Case (iii) manifest structure in the higher power region of the α-event distributions.

Previous studies [35] with different droplet size distributions, gel stiffness and mixed irradiations have shown that the observed signal amplitudes of a larger droplet size distribution are uniformly larger than a smaller-sized as might be expected from Eq. (2). Larger droplet size expansions are also accompanied by an increased probability of sympathetic bubble nucleations which can combine to yield increased signal amplitudes. With stiffer gels, the signal amplitudes are also larger than with the “standard” gel [33]. One might think that the decreased gel elasticity would restrict the range of bubble expansion relative to the less stiff, resulting in narrower power distribution. This is observed for the α-irradiation, but not for the recoil event distribution. In Case (iv), with an increased operating temperature of 27 °C, the gel becomes more liquid and the signal amplitudes increase since there is less resistance to the bubble expansion.

The liquids themselves (as well as of Ref. [11]) are not significantly different in their characteristic properties, as seen in Table 2. Apart from the obvious differences in instrumentation, the experiments also differ in their measurement observable. The total energy release in a bubble nucleation is the superheat of the droplet; for C4F10 at 1 bar and 27 °C, Etotal ~ 2.1 × 10−8 J, well in excess of a 10 MeV energy deposition (1.6 × 10−12 J). Although only a fraction of the energy appears in the acoustic signal, the same-sized droplet releases the same energy whether triggered by α or recoil interactions. For comparison purposes, we convert Fig. 5(a) from ln(K) to AP = log(∫0 t A dt) via ∫0 A(t)2 dt = ∫0 A02 e−λt dt ~ A02(λ/2), with j = [rec, α], the λ’s taken from the signal analysis, and normalizing the AP of the α recoil mean to 1. The result, in Fig. 6(a), yields an overlap of the two event distributions, in reasonable agreement with Fig. 10 of Ref. [7] at 27.5 °C, as also the salient features of each distribution with the exception of the inverted tail structure of the α distributions. The same analysis applied to the data of Fig. 2(a) yields Fig. 6(b) which preserves both the shapes of the event distributions and the peak separations but not the gap, suggesting that the α distribution derives from larger droplets of larger r than the recoil response.

Table 1

<table>
<thead>
<tr>
<th>Case</th>
<th>Fig.</th>
<th>Description</th>
<th>Recoil distribution (K_{\text{min}}) – ln (K_{\text{peak}}) (K_{\text{max}})</th>
<th>α distribution (K_{\text{min}}) – ln (K_{\text{peak}}) (K_{\text{max}})</th>
<th>% overlap</th>
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<tr>
<td>(i)</td>
<td>2(a)</td>
<td>&lt; r &gt; ~ 10 μm; std gel</td>
<td>6.5 – 7.5 – 9.2</td>
<td>9.7 – 10 – 12</td>
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<tr>
<td>(ii)</td>
<td>3(a)</td>
<td>&lt; r &gt; ~ 23 μm; std gel</td>
<td>6.5 – 7.5 – 9.2</td>
<td>8.8 – 9.5 – 12</td>
<td>1.56</td>
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<tr>
<td>(iii)</td>
<td>3(b)</td>
<td>&lt; r &gt; ~ 23 μm; std gel</td>
<td>6.9 – 8.3 – 10</td>
<td>9 – 10 – 12</td>
<td>0.82</td>
</tr>
<tr>
<td>(iv)</td>
<td>4(a)</td>
<td>&lt; r &gt; ~ 10 μm; stiff gel</td>
<td>7.9 – 9.0 – 12</td>
<td>8 – 9.7 – 12</td>
<td>12.5</td>
</tr>
<tr>
<td>(v)</td>
<td>4(b)</td>
<td>&lt; r &gt; ~ 10 μm; stiff gel</td>
<td>7.8 – 10.2 + 1</td>
<td>10 – 11.2 – 12.5 + 14</td>
<td>6.07</td>
</tr>
<tr>
<td>(vi)</td>
<td>5(a)</td>
<td>&lt; r &gt; ~ 5.5 μm; std gel</td>
<td>6.8 – 7.0 + 1</td>
<td>9 – 10.4 – 13</td>
<td>211</td>
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<tr>
<td>(vii)</td>
<td>5(b)</td>
<td>&lt; r &gt; ~ 5.5 μm; std gel</td>
<td>7.4 – 7.8 – 11</td>
<td>2 – 2.2 – 7.5</td>
<td>18.9</td>
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Table 2

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<th>Liquid</th>
<th>Tc (K)</th>
<th>pc (kPa)</th>
<th>Tb (K)</th>
<th>ν-OTP (g cm−3)</th>
<th>Ec-OTP (keV)</th>
<th>R0-OTP (μm)</th>
<th>s-OTP (μm)</th>
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<td>C2ClF2</td>
<td>385.0</td>
<td>4136</td>
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<td>9.7</td>
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<tr>
<td>CCl2F5</td>
<td>353.1</td>
<td>3158</td>
<td>250.8</td>
<td>2.53</td>
<td>7.8</td>
<td>0.0341</td>
<td>0.31</td>
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<tr>
<td>C4F10</td>
<td>386.3</td>
<td>2389</td>
<td>271.4</td>
<td>1.49</td>
<td>42.1</td>
<td>0.0749</td>
<td>0.25</td>
</tr>
</tbody>
</table>

a 2 bar.
b 1 bar.
4. Analysis

Since larger droplet size distributions and gel stiffness are common to both irradiations, there is no obvious differential to provide the difference between the two response distributions. We examine the incident radiation interaction and nature of the particle energy loss in the liquids.

4.1. Neutron-induced nuclear recoil events

Elastic neutron scattering on target nuclei provides nuclear recoils with the maximum recoil energy \( \text{ER}_A \) of an ion A given by

\[
\text{ER}_A = \omega_A \frac{\text{En}}{1 + \lambda^2}, \qquad \omega_A = 0.28, \quad \text{for C}, \quad \omega_A = 0.19, \quad \text{for F}, \quad \omega_A = 0.11.
\]

For the AmBe source, the incident neutron energy \( \text{En} \) ranges to 10 MeV, yielding \( \text{ER}_C \approx 2.8 \text{MeV}, \text{ER}_F \approx 2 \text{MeV} \) and \( \text{ER}_\text{Cl} \approx 1.1 \text{MeV} \). In the reactor irradiations (\( \text{En} \approx 300 \text{keV} \)), recoils with maximum \( \text{ER}_C \approx 84 \text{keV}, \text{ER}_F \approx 57 \text{keV}, \text{and } \text{ER}_\text{Cl} \approx 33 \text{keV} \) are obtained; at OTP, \( \text{ER}_C =\text{ER}_F = 8 \text{keV} \) and \( \text{ER}_\text{Cl} = 120 \text{keV} \), so that carbon ions are unobserved.

Inelastic scattering also generates nuclear recoils for neutron energies above the reaction threshold (120 keV for F, 1.3 MeV for Cl and 4.8 MeV for C). However, the contribution of these reactions is very reduced, as the neutron moderation by the SED hydrogenous gel matrix down scatters the incident neutrons to energies below the reaction threshold [39]. Among transmutation reactions with positive Q-value, \( ^{35}\text{Cl}(n,p)^{35}\text{S} \) and \( ^{35}\text{Cl}(n,\alpha)^{32}\text{P} \) are in principle of interest because the emerging ions have a minimum energy of 17 keV \( ^{35}\text{S} \) and 104 keV \( ^{32}\text{P} \) that can provoke a bubble nucleation. These reactions however have cross sections smaller than those of elastic scattering in Cl by \( \approx 1-7 \) orders of magnitude, and their contribution to the detector signal is generally small (with exceptions in thermal neutron fields). There are also high LET Auger cascades from environmental \( \gamma \) interactions with the Cl, with a threshold of \( \approx 16 \) °C at 2 bar, which prescribes its operation at lower temperatures.

Fig. 7(a) displays track-averaged Bragg curves calculated using SRIM [40] at OTP for fluorine ions of various initial \( \text{ER}_F \) in \( \text{C}_2\text{ClF}_5 \); the 0-depth entry equals the stopping power at \( \text{ER}_F \). The inset, displaying the 5–100 keV recoils, indicates the \( \text{C}_2\text{ClF}_5 \) LET\(_C\) to be exceeded over penetration depths of \( \approx 0.4 \) mm, significantly below the smallest droplet sizes and \( \approx 10 \times \text{L}_C \); the lighter carbon ions penetrate larger depths due to their higher recoil energies and smaller stopping power. The higher \( \text{En} \) of AmBe-generated recoils range up to 2.8 MeV with LET > LET\(_C\); calculated from the data of Table 2 and \( \Lambda_{\text{C2ClF5}} = 1.40 \) [41]. For \( \text{C}_4\text{F}_{10} \), the situation is somewhat different as seen in Fig. 7(b), which shows several track-averaged recoil F ion Bragg curves in \( \text{C}_4\text{F}_{10} \) at 27 °C, together with the LET\(_C\).
derived from Fig. 6 of Ref. [7] and respective $E_i(T)$, $r_c(T)$. The profiles are generally similar to Fig. 7(a), inset, but range to $\sim 0.07$ $\mu$m, with LET > LETc over distances of $\leq 0.04$ $\mu$m $\sim r_c$.

For comparison, Fig. 8 display the Bragg curves for carbon and chlorine recoil ions, calculated with the recoil energies corresponding to the same incident neutron energies of Fig. 7(a).

The neutron mean free path with $E_n = 10^{-8}-10^{-1}$ MeV is $\sim 0.2$–0.3 cm, far larger than the largest droplet diameters: subsequent elastic scatterings occur in the gel or other droplets. For the low energy recoils, the track-averaged path lengths for which LET > LETc are of order $L_c$, and $O(1)$ proto-bubble formation is anticipated; increased proto-bubble production is expected for the higher energies.

4.2. Alpha decay events

For U$_3$O$_8$, the main $\alpha$ decay energies are $E_{\alpha} = 4.2$ and 4.7 MeV [42]. Although an $\alpha$ decay is itself a “recoiling” $^4$He ion, its larger kinetic energy produces a significantly different track-averaged Bragg curve relative to a target ion recoil (Fig. 9). As indicated, $\alpha$'s originating on a droplet surface would generally achieve LET > LETc in C$_2$ClF$_5$ over several microns in the liquid following several tens of microns penetration with LET < LETc. Droplets with diameters less than a minimum penetration distance $p_c$ cannot support bubble nucleation since the $\alpha$ transits the droplet without achieving LETc.

4.3. Alpha-recoil event separation

From the experiments of Section 2, two aspects seem evident:

(1) the same droplet size distribution produces two different acoustic responses, and
(2) the acoustic response profiles of each are different. The first is addressed from the above: it is clear that $\alpha$'s (as well as high energy nuclear recoils) in general have a higher proto-bubble generation capacity. Each proto-bubble formation within a droplet serves as an evaporation center, and the droplet evaporation time is accordingly reduced to $\tau_0/n_0$, where $n_0$ is the number of proto-bubbles created by radiation type $j$, and $\tau_0$ is the droplet evaporation time for $n_0 = 1$.

Since the bubble and droplet have the same mass, $r_0 = (\rho/\rho_v)^{1/3}r_d$, and Eq. (2) can be re-expressed in terms of $r_d$ as

$$K = A_0 n_0^{1/4},$$

with $A_0^2 = 4\pi\rho (\rho/\rho_v)^2 e^{-1}\tau_0^{-4}$. With $n_0 = 1$, the recoil signal distribution should then mirror the droplet size distribution. This is observed experimentally, as seen in Fig. 10 [5]. Moreover, for the same droplet size, the $n_0^2$ serves as an amplification factor for separating the two particle-induced acoustic power responses if the $\alpha$ proto-bubble formation is sufficiently $> 1$.

Assuming for the sake of argument that $n_0 = 2$ with the measured $r_{\text{max}} = 22$ $\mu$m, $r_{\text{min}} = 11$ $\mu$m from Fig. 9, $A_0^2 = 5.456 \times 10^{-6}$ from Eq. (6a) and $n_0 = 6.4$ from Eq. (6b), consistent with the results of Diemand et al. [43]. A similar analysis of the partial separation of the $\alpha$ and recoil peaks in Fig. 5(a) with $r_{\text{max}} = 40$ $\mu$m and $2\tau_{\text{min}} = 38$ $\mu$m for the C$_2$F$_5$ with $n_0 = 2$ gives $A_0^2 = 3.361 \times 10^{-7}$ (5.378 $\times 10^{-6}$) and $n_0 = 4.8$ (2.4) in agreement with Ref. [7]. The increased proto-bubble formation in $\alpha$-droplet interactions amplifies the acoustic signal amplitudes above those of the recoil.

Fig. 8. (a) Bragg curves of recoil carbon ions of various initial energies in C$_2$ClF$_5$ together with the LETc; (b) same for chlorine in C$_2$ClF$_5$.

Fig. 9. SRIM-computed Bragg curves for 4.0 MeV $\alpha$'s in C$_2$ClF$_5$ and C$_4$F$_{10}$ at their OTPs. The respective LETc, obtained from Refs. [5,6] are also indicated. The $p_<$, $p_>$ define the track-averaged penetration depths for each, between which the $dE/dx \geq$ LETc.

The situation appears significantly different for C$_4$F$_{10}$, where the maximum of the Bragg peak is virtually coincident with the LETc. The energies of the $^{238}$U and $^{234}$U daughters are 86 keV ($^{230}$Th) and 72 keV ($^{234}$Th) [42]. These have penetration depths of $\sim 0.10$ $\mu$m, $0.08$ $\mu$m of which exhibits LET > LETc; in contrast to the $\alpha$, the daughters behavior more like the target recoils of Figs. 7 and 8 and can mimic a recoil event.

Assuming for the sake of argument that $n_0 = 2$ with the measured $r_{\text{max}} = 22$ $\mu$m, $r_{\text{min}} = 11$ $\mu$m from Fig. 9, $A_0^2 = 5.456 \times 10^{-6}$ from Eq. (6a) and $n_0 = 6.4$ from Eq. (6b), consistent with the results of Diemand et al. [43]. A similar analysis of the partial separation of the $\alpha$ and recoil peaks in Fig. 5(a) with $r_{\text{max}} = 40$ $\mu$m and $2\tau_{\text{min}} = 38$ $\mu$m for the C$_2$F$_5$ with $n_0 = 2$ gives $A_0^2 = 3.361 \times 10^{-7}$ (5.378 $\times 10^{-6}$) and $n_0 = 4.8$ (2.4) in agreement with Ref. [7]. The increased proto-bubble formation in $\alpha$-droplet interactions amplifies the acoustic signal amplitudes above those of the recoil.
4.4. A simple response model

The question of differing acoustic distribution profiles is addressed via a “surface emission” model, based on Pan et al. [19] which can be used to estimate the anticipated response of SEDs to actinide doping, such as 241Am or U238O8. In this case, the atoms have an electrochemical affinity for both C2Cl2 and C2F10 molecules, as also do the complex ions with which they are normally associated [44–46]. In consequence, they should migrate towards the droplet surfaces to preferentially populate the larger droplet surfaces; at the least, larger droplets should have a larger number of α-emitters, hence higher decay probability. A surface origin for the emulsion by acting as a surfactant [46].

Assuming surface emission, only αs entering the shaded region of the droplet shown in Fig. 11(a) produce proto-bubbles, where \( \rho_\alpha, \rho_\geq \) demarcate the penetration depths of the Bragg curves over which the \( \rho > \rho_\geq \) (see Fig. 9). A bubble nucleation probability, automatically reduced to \(< 50\% \rangle \) by the α isotropic emission, is obtained by subtracting the volume intersection \( V_\alpha \) of the droplet sphere with a sphere of radius \( \rho_\alpha \) centered at the droplet radius, and normalizing to the droplet volume: \( P_{bn}(r) = \frac{1}{2} \left( V_\alpha - V_\rho \right)/V_d \), where

\[
V_c = \frac{4\pi}{3} \left( 8r_d - 3r_c^3 \right).
\] (7)

Similarly, an \( \alpha \) proto-bubble generation probability \( P_{ph} \), corresponding to the region of the Bragg curve between \( \rho_\alpha \) and \( \rho \) (i.e. with \( \rho > \rho_\geq \)) is obtained as \( P_{ph}(r) = \frac{1}{2} \left( V_\rho - V_\alpha \right)/V_d \). For the 4.2 MeV \( \alpha \)'s of the Section 3 experiments, droplets with \( 2r < \rho_\alpha = 22 \mu m \) do not support proto-bubble formation; those with \( 2r > \rho_\alpha = 25 \mu m \) do not contribute further proto-bubbles since the \( \alpha \) LET is below \( \rho_\geq \). Shown in Fig. 11(b), \( P_{bn} \) rises from zero at 11 \( \mu m \) to asymptotically approach 0.5 with increase in \( r_d \), for droplets with \( r > 12.5 \mu m \), no further proto-bubble formation occurs and \( P_{ph} \) decays. Given our interest in the signal production as a measure of the SED response, we neglect \( P_{ph} \) in the discussion hereafter.

The neutron-generated recoil signal is biased by the inherent geometric cross section of droplet size. As noted previously [5], the larger the droplet, the more likely it is a scattering, and the recoil window is initially populated by high amplitude events. This is seen in Fig. 12 taken from the data of Fig. 2(a): the first recoil events include amplitudes reaching to near 100 mV, although these rapidly drop to cluster above the mean of \( A = 43 \) mV. As the droplet depopulation progresses, the mean of the recoil event distribution first shifts downward, then rises to the mean of the Fig. 2(a) distribution, accompanied by the disappearance of events with \( A > 60 \) mV.

Measurements of pristine, α-doped detectors in contrast show no time-ordered droplet size-related geometric interaction probability, as seen in Fig. 12(b) taken from the data of Fig. 2(b). The lowest power \( \alpha \)-event occurs only after 125 previous events, with the bulk of event \( ln(A^2) \) \( \geq 9.7 \). The event spectrum however suggests that only the upper half of the droplet distribution is involved. Note that the (a) spectrum slope from event 20–150 is about that of the (b) spectrum minimum, giving possible evidence of an expected sound attenuation with increase in bubble populations and/or larger droplet depletion. Note however that initial neutron irradiation tends to remove the larger droplets first, so that the α doping is relegated to the smaller — which is the greater interest with respect to particle response discrimination since it likely relates to the gap between the two distributions observed in Fig. 2(a).

Given the above, the original droplet size distribution is modified by a geometric cross section factor \( e_g = \left( \frac{\rho}{\rho_{max}} \right)^2 \) to \( N(r) = P_{bn} e_g N(r) \), as shown schematically in Fig. 13 for a normalized Gaussian distribution of \( \langle r_d \rangle = 15 \pm 5 \mu m \), with \( P_{bn} \) computed for
$E_\alpha = 4.2\text{ MeV}$. The gray contour represents the initial gaussian distribution; the black, its geometric-correction. The red curve is $P_{bn}$, and the purple is the un-normalized convolution result. As evident, for diameters below the $p_\text{c}$ cut, the droplets are insensitive to the $\alpha$'s: the irradiation samples only the upper half of the SED droplet sizes. Higher energy $\alpha$ detection requires larger droplet sizes or distributions.

Fig. 14(a) displays the anticipated “acoustic power response” of 4.2 MeV $\alpha$'s incident on SEDs with geometrically-weighted Gaussian size distributions of the several $<r_d>$ of this study, now cast as a function of $\ln(r_d^6)$: the amplification factor $n_b^6$ is neglected in order to examine the effect of $P_{bn}$ on the resulting response, and the result un-normalized to indicate its relative strength. For droplet size distributions with $<r_d> \leq p_\text{c}$, the probability cut samples only the large radius tail of the distribution, yielding an asymmetric response distribution which becomes increasingly symmetric as $p_\text{c}$ increases. As $p_\text{c}$ increases, the sensitivity of the small droplet distributions vanishes.

Note that the two smallest of the five considered $<r>$ distributions do not appear in Fig. 14(a), since the $P_{bn} \sim 10^{-8} - 10^{-4}$.

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Note that the two smallest of the five considered $<r>$ distributions do not appear in Fig. 14(a), since the $P_{bn} \sim 10^{-8} - 10^{-4}$.

Fig. 14. (a) convolution of the bubble nucleation efficiency with Gaussian droplet size distributions of $<r_d> = 5 \pm 3, 10 \pm 3, 15 \pm 5, 20 \pm 7$ and $25 \pm 8 \mu m$ and $E_\alpha = 4.2\text{ MeV}$.

(b) same as (a) but with the simulated response now normalized.
Given that the SED may however contain upwards of $2 \times 10^8$ droplets in its distribution, events will occur which manifest themselves in a normalized presentation. Fig. 14(b) displays the results now normalized (as generally presented in calibrations). As evident, the smallest responses are highly asymmetric since only the large $r$ tail of the distribution is being sampled; as $o_r$ increases above $p_o$, the $\alpha$ response becomes increasingly symmetric.

Since the decay daughters also share the droplet surface origin, the reverse considerations apply with $p_o = 0.1 \, \mu m$: the contribution is a priori reduced by 50%, but essentially 100% otherwise. The contribution from $\alpha$-emitters outside the droplets is estimated at $\leq 1.5\%$ [48, 49].

The results of the model for the Section 3 experiments are seen in Fig. 15 with daughter contributions neglected. Fig. 15 (a) corresponds to Case (i): the recoil distribution is obtained by rescaling the abscissa as $A_2 = \xi_2 r_5 n_2^2$ with $n_2 = 1$ and adjusting the abscissa by $\xi_2 = -7.5$ to overlap the recoil distribution of Fig. 2(a). The $\alpha$ distribution is similarly obtained, with $n_\alpha = 0$ for $r_d \leq 11 \, \mu m$ and $n_\alpha = 1.6/\mu m$ for $r > 11 \, \mu m$; the same $\xi_2$ shift is employed. The observed gap between the two and asymmetry in the $\alpha$ distribution of Fig. 2(a) is indicated.

Fig. 15(b) corresponds to Case (ii), with the $<r_d> = 22 \pm 2.5 \, \mu m$ droplet size distribution, $p_o = 19 \, \mu m$ obtained from Fig. 9 and $n_\alpha = 1.6$ regardless of size above $r_d = 11 \, \mu m$. The event distributions are overlapped, as seen experimentally.

Fig. 15(c) corresponds to Case (iv), with the $<r_d> = 22 \pm 2.5 \, \mu m$ size distribution, $p_o = 19 \, \mu m$ obtained from the C$_4$F$_{10}$ Bragg curve of Fig. 9 and $n_\alpha = 1.8$ regardless of size above $<r_d> = 9.5 \, \mu m$. Although the simulated $\alpha$ event distribution exhibits the same structural symmetry as Fig. 15(b), the response overlap is decreased; the lower $p_o$ is apparently compensated by the $n_\alpha$ of C$_4$F$_{10}$.

Fig. 15(d) corresponds to Case (v), with the experimentally-measured $<r_d> = 5.5 \, \mu m$ droplet size distribution approximated by a Lorentz distribution tailing to $\sim 16 \, \mu m$, $p_o$ from Fig. 9 and $n_\alpha = 1.6/\mu m$ for $r_d > 11 \, \mu m$. Both simulated event distributions occur at a significantly reduced power and exhibit the structural asymmetry of the experimental results; the recoil power distribution however remains below that of the $\alpha$, in contrast to Ref. [11].

5. Summary

The acoustic response of SEDs to nuclear recoil and $\alpha$ events is seen to fundamentally depend on the relation between the
incident particle energy and distribution of droplet size in the detector. Low energy nuclear recoils of the target nuclei yield an O (1) proto-bubble formation, as also the α-decay daughters; α’s and high energy recoils, generally more. Each proto-bubble is formed on time scales less than for a complete bubble nucleation event, hence serves as an additional evaporation center for the bubble nucleation, providing an amplification factor to the acoustic signal power. In the case of actinide α-doping, the SED response can be estimated via a simple model based on the intersection of two spheres, one a droplet and the other of radius \( p_\alpha \) obtained from the intersection of the particle Bragg curve with the LET\(_c\) of the liquid at its OTP. The model is seen to address the profile variations via the droplet size distribution and incident α energies. As seen for Gaussian droplet size distributions with means \( < r > \) below \( p_\alpha \), the α-response is asymmetric in its lower amplitudes since the α’s sample only the larger droplets of the distribution. As the distribution mean increases above \( p_\alpha \), the response becomes increasingly asymmetric as more of the smaller droplets become involved. The apparent decrease in power of the α response in Case (ii) is a result of the increased distribution \( < r > \) above the \( p_\alpha \) of the \( \text{U}_3\text{O}_8 \). The model is usable with all droplet distributions and actinide α-emitters; applied to the results of Section 3, it reproduces well the experimental results. The model is not directly applicable to the observations of Ref. [11] since the α irradiation is initiated external to the SED and hence more properly relates to α surface emission from the SED containment walls. It does however provide some further insight into the results, which are attributed by the authors to the smaller \( n_\alpha \) formation resulting from partial α passage through the \( < r > \) distribution. As seen from the Bragg curves of Section 4, proto-bubbles are generated only when the \( \alpha \) de/dx \( \geq \) LET\(_c\); this range is reduced compared to the full particle range in the droplet liquid. The proto-bubbles moreover serve as triggers to the bubble nucleation: the total acoustic energy release of a droplet, which is the \( P \) variable of Ref. [11] (and not the signal power) is \( \sim r_\alpha^n \), and the signal amplitude differences depend only on the relative difference in the \( < r_1 > \), \( < r_2 > \) sizes. The \( n_\alpha = \text{range/L}_E \) of Ref. [11] is questionable, as also that \( L_M = E_f / \text{de/dx} \), since the denominator obtained from the stopping power curve reflects only the zero-thickness value of the respective Bragg curve. The more likely explanation of the apparent response inversion is that the \( 241\text{Am} \) α’s, degraded by their air passage, must transit \( \sim 11 \mu \text{m} \) of glycerol before their de/dx approaches the CCl\(_2\)F\(_5\) LET\(_c\); at OTP – droplets within this distance are effectively blind to α’s, and the α response must come entirely from the \( < r_1 > \) droplets within an approximately hemispherical shell of \( \sim 17 \mu \text{m} \) thickness at a 11 \( \mu \text{m} \) distance from the SED-air interface, in contrast to the neutron irradiations which sample the entire SED droplet population (in particular \( < r_2 > \) with larger geometrical cross section). The simple model neglects a number of presumably higher order effects including α-emission outside the droplet, ion straggling and nucleation efficiencies, and can therefore only be considered a first approximation. Also not covered by the present model is the case of non-actinide α-emitter doping in which the decay daughters may diffuse into the droplets. This is to be addressed in a forthcoming paper. Despite this, the above description is consistent with observations, and captures the essence of the involved response physics in SED devices. The sensitivity of a detector to the range of decay \( E_\alpha \) is clearly dependent on the distribution of droplet size in the device, requiring careful characterization measurements and establishment of fabrication protocols for reproducibility. The question of recoil - α acoustic power separation is dependent on a gap existence, hence whether the \( n_\alpha^4 \) amplification factor is sufficient to promote the lower power tail of the α response above the high power tail of the recoil response. The principle problem is the estimation of proto-bubble formation probability, which is far from clear. The \( P_{\alpha}(r) \), not discussed herein, describes only the droplet sensitivity to multiple proto-bubble formation and gives only the droplet volume capable of supporting proto-bubble formation, not the number per micron. As seen by the particle Bragg curves, the number of proto-bubbles generated by particle interactions depends on the de/dx of the particle in the target liquid, and its relation to the LET\(_c\) of the fluid at OTP. In the case of C\(_2\)Cl\(_5\), the LET\(_c\) is well-below the Bragg peak, providing α proto-bubble formation probabilities over 5–15 \( \mu \text{m} \) of penetration; for C\(_4\)F\(_10\), LET\(_c\) is at or just below the maximum of the Bragg peak. The simulation of Figs. (b) and (c) however support similar \( n_\alpha \)'s. The difficulty in defining the LET\(_c\) of a liquid derives from the presence of \( \Lambda \) in Eq. (3), and use of the stopping power curves in its determination since the de/dx from the latter at a given incident energy is only the 0-thickness penetration value of the Bragg curve (which for light ions is generally well-below the maximum de/dx). Significant further investigation of the involved molecular dynamics and proto-bubble formation mechanics is required.

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