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Fabrication and response of high concentration SIMPLE superheated droplet detectors with different liquids

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ABSTRACT

The combined measurement of dark matter interactions with different superheated liquids has recently been suggested as a cross-correlation technique in identifying WIMP candidates. We describe the fabrication of high concentration superheated droplet detectors based on the light nuclei liquids C_3F_8, C_4F_8, C_4F_{10} and CCl_2F_2, and investigation of their irradiation response with respect to C_2ClF_5. The results are discussed in terms of the basic physics of superheated liquid response to particle interactions, as well as the necessary detector qualifications for application in dark matter search investigations. The possibility of heavier nuclei SDDs is explored using the light nuclei results as a basis, with CF_3I provided as an example.

1. Introduction

The direct search for weakly interacting massive particle (WIMP) dark matter is generally based on one of five techniques: scintillators, semiconductors, cryogenic bolometers, noble liquids and superheated liquids. The last, in contrast to the others, relies on the stimulated transition of a metastable liquid to its gas phase by particle interaction: because the transition criteria are thermodynamic, the devices can be operated at temperatures and/or pressures at which they are generally sensitive to only fast neutrons, α's and other high linear energy transfer (LET) irradiations.

Only three WIMP search efforts employ the superheated liquid technique: PICASSO [1], COUPP [2] and SIMPLE [3], using C_4F_{10}, CF_3I and C_2ClF_5 respectively. Of the three, COUPP is based on bubble chamber technology: only PICASSO and SIMPLE employ superheated droplet detectors (SDDs). Because of their fluorine content and fluorine's high proton spin sensitivity, as well as their otherwise light nuclei content relative to Ge, I, Xe, W and others, they have generally contributed most to the search for spin-dependent WIMP-proton interactions. COUPP, with CF_3I, has also made a significant impact in the spin-independent sector.

A SDD consists of a uniform dispersion of micrometric-sized superheated liquid droplets homogeneously suspended in a hydrogenated, viscoelastic gel matrix. The phase transition generates a millimetric-sized gas bubble which can be recorded by either optical, acoustic or chemical means; both SDD experiments employ acoustic, while COUPP employs both acoustic and optical (the liquid is essentially transparent, whereas the gel matrix of the SDDs is at best translucent).

The significant difference between the two approaches is that SDDs are continuously sensitive for extended periods since the overall liquid droplet population is maintained in steady-state superheated conditions despite bubble nucleation of some droplets, whereas in the bubble chamber the bulk liquid is only sensitized between nucleation events, each of which precipitates the transition of the liquid volume hence requires recompression to re-establish the metastable state and leads to measurement deadtime. The advantage of the chamber approach is an ability to instrument large active target masses. SDDs have generally been confined to low concentration (<1 wt%: liquid-to-colloid mass ratio) devices, for use in neutron [4–11], and heavy ion [12] detector applications, with impact in heavy ion and cosmic ray physics, exotic particle detection and imaging in cancer therapy [13,14]. For rare event applications such as a WIMP search, however, higher concentration detectors are required: the PICASSO devices

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are ~1 wt% concentrations. SIMPLE detectors in contrast are generally of 1–2 wt%; concentrations above 2 wt%, in which the droplets are sufficiently close in proximity, tend to self-destruct as a result of massive sympathetic bubble nucleation and induced fractures.

Recently, variation of the target liquids with different sensitivities to the possible scalar and axial vector components of a WIMP interaction has been suggested as a technique in identifying WIMP candidates [15], specifically in the case of COUPP in combined measurements using CF_3I and C_4F_{10}. This measurement variation while maintaining equivalent sensitivities in the case of SDDs is not trivial, since device fabrication and operation depends on the individual thermodynamic characteristics of each liquid.

SIMPLE SDD fabrications generally proceed on the basis of density-matching the liquid with a 1.3 g/cm^3 food-based gel with low U/Th contamination: a significant difference in gel and liquid densities (as occurs with heavier nuclei liquids) results in inhomogeneous distributions of differential droplet sizes within the detector. Although this has been addressed by SIMPLE via viscosity-matching the gel [16,17], this approach is constrained by the SIMPLE gel melting at 35 °C, limiting the temperature range of the device and hence restricting the liquids employed. The traditional addition of heavy salts such as CsCl to raise the gel density, as originally used by PICASSO with its polyacrylamide-based gels [18], is discouraged since this generally adds radioactive contaminants which must be later removed chemically with the highest efficiency possible.

Thus, the question of liquid variation in SDDs naturally raises the questions of whether or not such "other" SDDs can in fact be fabricated, much less operated, and with what sensitivity. We here describe our fabrications and testing of small volume (150 ml), high concentration (1–2 wt%) SDD prototypes with C_3F_8, C_4F_8, C_4F_{10}, CCl_2F_2 and CF_3I including for completeness a "standard" C_2ClF_5 device of the SIMPLE dark matter search effort [3]. Section 2 provides an overview of the device fabrication, and describes the experimental testing of the products. The response of superheated liquids to irradiations in general, and liquid characteristics necessary to dark matter searches is discussed in Section 3, and applied to the fabricated SDD test results, with the salient aspects of particle discrimination as observed by SIMPLE identified in Section 4. Section 5 discusses the considerations necessary to the fabrication and implementation of heavier nuclei SDDs, to include the introduction of a figure of merit based on the light nuclei results by which an initial screening of possibilities can be made in the absence of a complete thermophysical description of the liquids: The fabrication and analysis of a CF_3I is described as an example. Conclusions are formed in Section 6.

2. Light nuclei detectors

For light liquids, SDD construction generally consists of two parts: the gel, and the liquid droplet suspension. The variation of the liquid densities with temperature is shown in Fig. 1, and can be divided into three basic density groups:

(i) C_2ClF_5, C_3F_8,
(ii) CCl_2F_2,
(iii) C_4F_{10}, C_4F_8.

For those in groups (i) and (ii) with $\rho \sim 1.3$ g/cm^3, small variations in the current C_2ClF_5 recipes are indicated; for the more dense liquids of group (iii), viscosity matching is necessary using an additive as discussed in detail in [16,17].

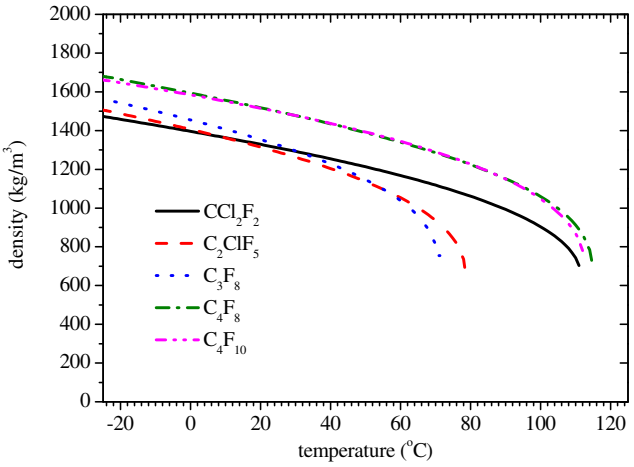

Fig. 1. Variation of liquid densities with temperature [19].

2.1. Gel fabrications

The basic SDD ingredients have been described previously [20]. In the density-matched, "standard" case of C_2ClF_5, the gel composition is 1.71 wt% gelatin, 4.18 wt% polyvinylpyrrolidone (PVP), 15.48 wt% bi-distilled water and 78.16 wt% glycerin. The gelatin is selected on the basis of its organic origins to minimize the U/Th impurity content; the glycerin serves to enhance the viscosity and strength of the gel, and wet the container surfaces. The presence of the PVP (i) assists in fracture control by viscosity enhancement which decreases diffusion, (ii) improves the SDD homogeneity and reduces the droplet sizes via its surfactant behavior, (iii) decreases the liquid solubility [21], (iv) inhibits clathrate hydrate formation, and (v) reduces the migration of α-emitters to droplet boundaries as a result of actinide complex ion polarity [22].

The basic process, minus several proprietary aspects, has been described in [20]. The ingredients are first formed: powdered gelatin (Sigma Aldrich G-1890 Type A), bi-distilled water and pre-eluted ion exchange resins for actinide removal are combined and left for 12–15 h at 45 °C with slow agitation to homogenize the solution. Separately, PVP (Sigma Aldrich PVP-40T) and exchange resins are added to bi-distilled water, and stirred at ~65 °C for 12–15 h. Resins and glycerin (Riedel-de-Haën No. 33224) are combined separately, and left in medium stirring at ~50 °C for 12–15 h.

The PVP solution is then slowly added to the gel solution ("concentrated gel"), and slowly agitated at 55–60 °C for 2 h. The resins in all are next removed separately by filtering (Whatman 6725–5002A). The glycerin and concentrated gel are then combined at ~60 °C, outgassed at ~70 °C, and foam aspirated to eliminate trapped air bubbles. The solution is left at 48 °C for 14 h with slow agitation to prevent bubble formation.

For the viscosity-matched protocol required for the C_4F_8 and C_4F_{10}, the gel composition is essentially the same as in the density-matched recipe, with a small agarose (Sigma Aldrich A0576) addition effected by combining it with glycerin at 90 °C, then adding it to the concentrated gel mix prior its filtration.

Following resin purification, the gel yields measured U/Th contamination levels of < 8.7 mBq/kg ^{238}U, < 4.9 mBq/kg ^{235}U and < 6.9 mBq/kg ^{234}U.

2.2. Droplet suspension fabrications

The specific protocol for fabrication of a liquid droplet suspension depends on the thermodynamic properties of the liquid. The