

# Development of Bubble Chambers With Enhanced Stability and Sensitivity to Low-Energy Nuclear Recoils

W.J.Bolte<sup>a</sup>, J.I.Collar<sup>a,\*</sup>, M.Crisler<sup>b</sup>, J.Hall<sup>a</sup>, D.Holmgren<sup>b</sup>, D.Nakazawa<sup>a</sup>, B.Odom<sup>a</sup>, K.O'Sullivan<sup>a</sup>, R.Plunkett<sup>b</sup>, E.Ramberg<sup>b</sup>, A.Raskin<sup>a</sup>, A.Sonnenschein<sup>a</sup> and J.D.Vieira<sup>a</sup>

<sup>a</sup>*Enrico Fermi Institute and Kavli Institute for Cosmological Physics, University of Chicago, IL, USA*

<sup>b</sup>*Fermi National Accelerator Laboratory, Batavia, IL, USA*

The viability of using a Bubble Chamber for rare event searches and in particular for the detection of dark matter particle candidates is considered. Techniques leading to the deactivation of inhomogeneous nucleation centers and subsequent enhanced stability in such a detector are described. Results from prototype trials indicate that sensitivity to low-energy nuclear recoils like those expected from Weakly Interacting Massive Particles can be obtained in conditions of near total insensitivity to minimum ionizing backgrounds. An understanding of the response of superheated heavy refrigerants to these recoils is demonstrated within the context of existing theoretical models. We comment on the prospects for the detection of supersymmetric dark matter particles with a large  $CF_3I$  chamber.

PACS number(s): 95.35.+d, 29.40.-n, 05.70.Fh, 68.55.Ac

\* Corresponding author. E-mail: collar@uchicago.edu

The positive identification of sporadic signals from among comparatively frequent backgrounds is common to any experiment at the forefront of particle physics. The challenge faced by direct searches for cold dark matter particles [1] is in this respect extraordinary: signal rates as small as one low-energy nuclear recoil (few keV) per ton of detector mass per year are predicted for the nuclear scattering of supersymmetric Weakly Interacting Massive Particle (WIMP) candidates, if they comprise the bulk of dark matter halos able to explain galactic evolution and dynamics [2]. A number of detector techniques have been developed during the last two decades for this purpose [1]. Simplicity of design, optimal target materials, rapid scaling to the ton regime and an excellent background rejection are the defining qualities of the next-generation detectors that should soon explore the vast range of WIMP masses and couplings still allowed.

The use of moderately superheated liquids has been proposed as a possible fast route towards this goal [3,4]. A concentrated energy deposition from certain particles can lead in these to the rupture of metastability and the formation of visible bubbles. Two experiments, SIMPLE [5] and PICASSO [6] exploit this approach, benefiting from an intrinsic insensitivity to most backgrounds, discussed below. Both experiments implement the method using superheated droplet detectors [7] (SDDs, a.k.a. bubble detectors), where small drops ( $r \sim 10 \mu m$ ) of the active liquid are dispersed in an insoluble gel or viscoelastic medium. In a SDD the gel provides a smooth liquid-liquid interface that impedes the continuous triggering (inhomogeneous nucleations) on surface defects, gaskets, motes, etc. that is observed even in the cleanest bubble chambers. As a result, the lifetime of the superheated state is considerably extended, to the point that a WIMP search can be performed.

The goal of the present study is to assess the feasi-

bility of employing bulk quantities of superheated liquid instead, i.e., to use a conventional bubble chamber, an alternative for WIMP searches first put forward by Hahn [8]. Large, stable bubble chambers have been previously proposed for other rare-event searches (e.g., nucleon decay, superheavy elements [9]) but no dedicated attempt to extend the superheated times was made. The rapid uncontrollable foaming of a conventional chamber following its decompression was bypassed in accelerator experiments by a precise timing of the pulsed beam injection to coincide with the few ms of usable radiation-sensitive superheated time in each pressure cycle. The motivation to explore this apparently more problematic approach arises from the difficulty to manufacture SDDs out of the most interesting available industrial refrigerants, e.g.,  $CF_3I$  and  $CF_3Br$ . These liquids constitute ideal supersymmetric WIMP targets [10] due to the presence of both fluorine (optimal for spin-dependent neutralino couplings [11]) and a heavy nucleus (maximally sensitive to coherent spin-independent couplings) [12,13]. Their density is nevertheless severely mismatched with respect to that of a water-based SDD gel matrix, leading to inhomogeneous, unstable emulsions during the fabrication process. Saturation of the matrix with inorganic salts can help alleviate this issue, but leads to exacting requirements on the alpha-emitter radiopurity of the gel [6], exacerbated by the observed tendency of complex actinide salts to migrate to the droplet-gel boundary [12,14], where their ability to create an undesirable alpha-recoil background is the greatest. A first attempt to measure the attainable stability of bulk superheated liquid was made within the context of the SIMPLE experiment, using a rudimentary plastic chamber where the active fluid was fully encapsulated by a thick sheath of viscoelastic liquid [15] to avoid evaporation and nucleations on chamber walls. The chamber held 30 g of R-115 ( $C_2ClF_5$ ) superheated for up

to 12 hours at an underground depth of 1,500 m.w.e. [16], with no other precautions against neutron or radon backgrounds. This behavior revealed the possibility to control the sources of instability in a bubble chamber and prompted the further experimentation described here.

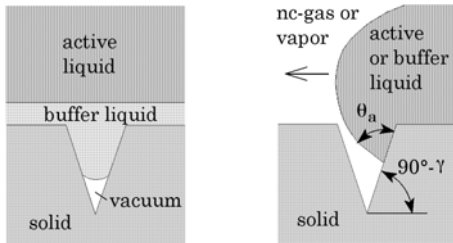


FIG. 1. *Left:* Use of a buffer liquid to isolate microscopic surface cavities able to act as inhomogeneous nucleation centers in a bubble chamber [21]. Mass transfer into the cavity can still lead to boiling, but deactivation is possible in the absence of noncondensable (nc) gas (see text). *Right:* Direct pouring of a liquid during chamber filling can lead to vapor entrapment in cavities when the advancing contact angle  $\theta_a$  is larger than the groove angle  $2\gamma$  [19]. Filling by slow vapor condensation after evacuation instead leads to efficient wetting of cavities, including those reentrant.

The mechanism leading to the nucleation of the gaseous phase and possible ensuing phase transition in the bulk of a superheated liquid (homogeneous nucleation) is described by a classical theory [17] where the probability of spontaneously generating a protobubble of radius larger than  $r_c$  is computed as a function of pressure, temperature and thermophysical properties of the liquid. If this critical radius is reached or surpassed, the vapor nucleus grows unchecked and metastability is lost. Protobubbles smaller than  $r_c$  collapse back onto themselves, producing no phase transition. In the case of a radiation-induced nucleation, the local heating (“hot spike” [18]) from a particle’s energy deposition can be responsible for the formation of a critically-sized nucleus, only if this energy is concentrated over a small enough region, comparable to  $r_c$ . This leads to the mentioned insensitivity to most backgrounds by imposing a threshold in stopping power, amply surpassed by nuclear recoils but not by minimum-ionizing particles (MIPs) [3,4]. For moderate degrees of superheat like those necessary to sensitize the liquid to low-energy nuclear recoils,  $r_c$  is typically  $\sim$ few tens of nm, and the rate of homogeneous nucleation is entirely negligible as a source of instability ( $\ll 10^{-20}$  bubbles/kg/day [17]).

Nucleations can nevertheless also occur on microcavities, scratches or imperfections naturally present even in the smoothest surfaces (e.g., glass) or in motes, partially or totally wetted by the liquid. The (inhomogeneous) nucleation rate on these cavities is only minimally increased with respect to the extremely small (homogeneous) bulk rate if the fluid has a zero contact angle with the surface, i.e., if the cavity is well wetted [17,19]. The actual

source of the inhomogeneous nucleations that limit a bubble chamber’s stability is instead any entrapped gas in these cavities [20], which can act as a vaporization initiator, allowing mass transfer from the fluid to the unwetted cavity volume [19,21]. Once nucleation is initiated in such a cavity, the superheat required to sustain boiling on it drops to a much lower value than what is required for homogeneous nucleation [19,21], i.e., destabilization occurs. It is however important to distinguish between cavities filled by superheated fluid vapor and those filled by noncondensable gas or a binary. In the first case, cooling or pressurization can lead to nucleation site deactivation by recondensing the trapped vapor. In the second, and in particular for reentrant cavities, deactivation can be arduous, albeit continued boiling may lead to an eventual depletion of the gaseous volume [19].

Once the nature of the problem is understood, precautions can be taken that lead to an enhanced bubble chamber stability: *i)* only smooth glass or quartz surfaces are allowed to be wetted by the superheated liquid, thereby reducing the number of available cavities. *ii)* A layer of a low-density buffer liquid can be allowed to form a “lid” above the (immiscible) active liquid [22], with all rough metallic parts (bellows, diaphragms, gaskets) coming in contact with the buffer only. *iii)* This same buffer liquid can be used to create a layer that fills cavities, previously evacuated to remove noncondensable gases (Fig. 1, left) [21]. Cavity filling can be improved by transferring the buffer (a step prior to the addition of the denser active liquid) by slow condensation of its vapor into the chamber rather than pouring. This ensures maximum wetting of even reentrant cavities [19,25] (Fig. 1, right). In the particular case of  $CF_3I$ , the shape of the meniscus in the interface between these and the buffer “lid” reveals a highly preferential wetting of glass and quartz by the buffer when water is used, a positive indication of the effectiveness of the buffer. To some extent, these methods reproduce the advantages of the smooth liquid-liquid interfaces in SDDs. *iv)* Exhaustive cleaning of glass surfaces [23] in clean-room conditions and filtering of all gases and fluids lead to a reduction in the number of large motes present (cavities smaller than  $O(r_c)$  cannot in principle act as nucleation centers). Some known cleaning techniques also have the desirable effect of improving surface wetting by the buffer [24]. *v)* After application of these techniques in the chambers and operating conditions described below, a periodic long recompression ( $\sim 200$  s) is observed to effectively deactivate the few boiling centers that can still sporadically appear due to vapor diffusion through the buffer layer, or cavity exposure to vapor during radiation-induced boiling.

Small bubble chamber prototypes up to 50 c.c. in active volume can be built for moderately-superheated refrigerants using commercially available pressure-resistant quartz vials [26]. Pressure cycling is achieved with a three-way valve or its equivalent and temperature con-

trol by means of a double-bath [27]. Fast triggering ( $<10$  ms) of bubble photography and recompression can be performed by use of a piezoelectric microphone to detect the acoustic emission that accompanies nucleations [12] or by monitoring the pressure increase caused by bubble growth. These simple devices have been used to study chamber stability and response to radiation sources.

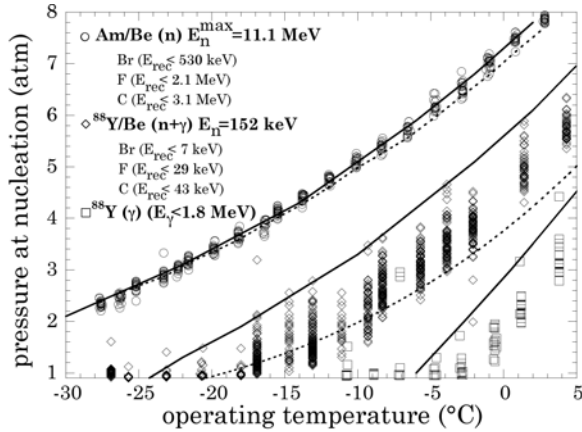


FIG. 2. Response of a  $CF_3Br$  chamber to radiation sources and comparison with theoretical models. Lines indicate the pressure below which full sensitivity to the source is expected according to different theoretical models (the experimental points represent the appearance of the first bubble upon decompression, see text). Insensitivity to gamma interactions in conditions that nevertheless afford good sensitivity to low-energy nuclear recoils has been demonstrated (see text).

Calibrations using neutron sources having a well-defined maximum energy (11.1 MeV for  $^{241}Am/Be$ ) or monochromatic neutron emission (152 KeV for  $^{88}Y/Be$ ) have allowed measurement of the response of the liquids to nuclear recoils down to 4 keV in the case of  $CF_3I$  and to establish agreement with theoretical models of this response. Data points in Fig. 2 represent the appearance of the first bubble nucleation upon decompression in the presence of each source (i.e., as the energy threshold for radiation-induced nucleation is reduced), each point corresponding to a compression/decompression cycle. For sufficiently-high source intensities and/or slow decompression rates this bubble is the result of a recoil with an energy close to the well-defined maximum that these sources can produce. These maximum recoil energies are indicated by labels in the figure, for each recoiling species. Solid lines represent the theoretical expectations (Seitz model [18,28]) for the onset of sensitivity to maximum-energy recoils, i.e., should trace the top boundary of the data points. Their dispersion towards lower pressures is expected from a progressive onset of sensitivity, which is not well described by a step-function [6] as naively assumed in the Seitz model. A review of the theoretical background leading to these predictions can be found in [12]. A good agreement with the data is observed by best-fitting the single free parameter in this model. The

best value obtained ( $a \sim 4$  in the notation of [12]) is compatible with previous [28] and most recent [29] studies. Since the predicted onset of response to the source for each recoiling species is not exactly the same (differing by a small fraction of an atm), the lines represent the first species expected to react to the source (Br and F, closely matched, for  $CF_3Br$ ). A calibration is planned where tagging of gamma rays emitted in inelastic 2.4 MeV neutron scattering will allow to separate the contributions from each species. Dotted lines correspond to the predictions of a modern phenomenological “reduced superheat” theory, the forte of which is its simplicity. It generates remarkably accurate predictions for lighter refrigerants used in SDDs [30], but may need further refinement for  $CF_3Br$  and  $CF_3I$  [31]. The effect of Moliere electron straggling in the formalism of Archard [32] was included in the calculation of  $^{88}Y$  (gamma) response.

The photonuclear  $^{88}Y/Be$  source employed emitted a mixed field of  $\sim 10^8$  high-energy gammas and just  $3.5 \times 10^3$  monochromatic neutrons per second: Fig. 2 illustrates the much higher degree of superheat (lower pressure at a given temperature) necessary to become sensitive to the gamma component once the Be sheath, the actual neutron emitter, is removed from the source. This allows for a dramatic demonstration of insensitivity to photoelectrons in operating conditions that nevertheless would ensure an optimal response to WIMP interactions. For instance, from the figure, at  $-10^\circ C$  and 1 atm no response to MIPs is observed, while sensitivity to WIMP-induced recoils more energetic than the maximum recoil energies produced by  $^{88}Y/Be$  seems guaranteed. A recently procured  $^{124}Sb/Be$  source ( $E_n = 24$  keV) will be used to produce recoil energies as low as  $\sim 1$  keV in  $CF_3I$ , an unprecedented test of a WIMP detector.

Prototypes containing a few tens of c.c. of active liquid remain superheated for periods of several minutes on the average in a shallow-depth laboratory (6 m.w.e.). The reduced ambient neutron flux in this site was characterized using a  $^3He$  detector surrounded by several configurations of neutron moderator and absorber (Bonner spheres) calibrated using known neutron sources, and deconvolved following an approach similar to [33]. Taking the measured fast neutron spectrum as input to a MCNP-POLIMI simulation [34] of the energy depositions in the chamber, the observed spontaneous nucleation rate is found to be in agreement with the expected neutron-induced recoils at this depth (Fig. 3). For superheated times  $t_{SH}$  longer than a few seconds, no observable excess of nucleations on walls can be inferred from bubble photography using two orthogonal cameras, which allows for 3-D reconstruction of nucleation sites with  $\sim 1$  mm precision. For shorter  $t_{SH}$  a small excess of wall events, evident in the figure, is observed. Sporadic boiling sites can be deactivated as previously described. The duty (live) time was  $\sim 65\%$  during these runs. The insensitivity (rejection factor) to MIPs at  $-10^\circ C$  and 1 atm is

$\gtrsim 10^9$  from the absence of any observable reaction to the  $\sim 10^6$  gamma interactions per second induced by the  $^{88}\text{Y}$  source within the active volume (Fig. 3). As discussed above, good sensitivity to WIMP-recoils is nevertheless expected in these conditions. This *intrinsic* rejection factor can be compared with the best ( $\sim 10^4$ ) achieved using complex cryogenic WIMP detectors [1]. It should permit construction of much larger chambers in the ton or multi-ton regime essentially without any concern for MIPs, including from elevated concentrations of  $^{14}\text{C}$ .

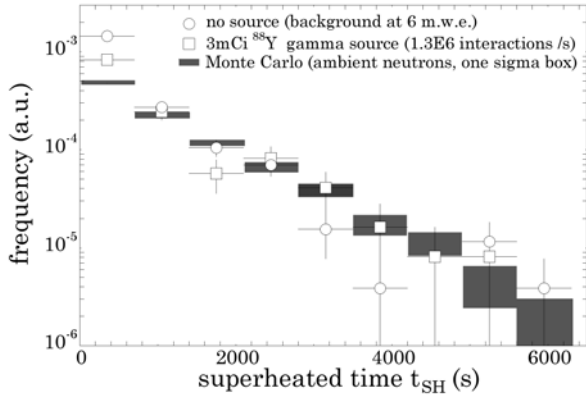


FIG. 3. Distribution of duration of the superheated state  $t_{SH}$  in a 12 ml  $\text{CF}_3\text{Br}$  bubble chamber operated at 6 m.w.e.,  $-10^\circ\text{C}$  and atmospheric pressure. When fitted by a form  $\propto e^{-t_{SH}/\tau}$  and after rejection of inhomogeneous nucleations during decompression, all cases depicted yield  $\tau \sim 1220$  s.

The encouraging outcome from these tests has led to the construction of a steel recompression chamber housing 2 kg of  $\text{CF}_3\text{I}$  in an inner quartz vessel [35]. A bellows mechanism compensates the pressure inside and outside of this vessel, which is both sealed against Rn penetration and low in its emanations, measures against alpha-recoil backgrounds. Provisions to ensure the long-term stability of this fire-extinguishing compound at the envisioned running temperature ( $40^\circ\text{C}$ ) are in place [36]. The behavior of this chamber at 6 m.w.e. ( $\langle t_{SH} \rangle \sim 60$  s) remains in agreement with the simulated ambient neutron contribution. Ongoing tests of  $\text{CF}_3\text{I}$  neutron and gamma response yield similar results to those discussed. To further assess the prospects of this new approach to WIMP detection, this chamber will be operated during 2005 within a neutron shield at the 300 m.w.e. depth of the MINOS near gallery on FNAL grounds (COUPP, the Chicagoland Observatory for Underground Particle Physics). The WIMP sensitivity that can in principle be achieved with COUPP is already competitive with the best present searches [35]. Besides those mentioned, there are unique advantages (neutron rejection ability, rapid target replacement, room temperature operation and low cost [35]) that distinguish ton-sized bubble chambers from their competitors in this exciting endeavor of dark matter detection: a successful COUPP would lead us to attempt their construction.

We are indebted to F.d'Errico, J.Ely, D.Jordan, E.Padovani, D.Quéré, and in particular to R.Hildebrand for being a constant source of inspiration. Work supported by the Kavli Institute for Cosmological Physics (NSF grant PHYS-0114422) and NSF CAREER award 0239812.

- 
- [1] R.J. Gaitskell, *An. Rev. Nucl. Part. Sci.* **54**, 315 (2004).
  - [2] G. Bertone *et al.*, *Phys. Rep.* **405**, 279 (2005).
  - [3] V. Zacek, *Nuovo Cimento* **A107**, 291 (1994).
  - [4] J.I. Collar, *Phys. Rev.* **D54**, R1247 (1996).
  - [5] J.I. Collar *et al.*, *Phys. Rev. Lett.* **85**, 3083 (2000).
  - [6] N. Boukhira *et al.*, *Astrop. Phys.* **14**, 227 (2000).
  - [7] R.E. Apfel, *Nucl. Instr. Meth.* **162**, 603 (1979).
  - [8] B. Hahn, *Nucl. Phys. B (Proc. Suppl.)* **36**, 459 (1994).
  - [9] G. Harigel *et al.*, *Nucl. Instr. Meth.* **216**, 355 (1983); K. Behringer *et al.*, *Phys. Rev.* **C9**, 48 (1974).
  - [10] J.I. Collar *et al.*, *Procs. 1st Workshop on the Identification of Dark Matter*, Sheffield (1996), World Scientific.
  - [11] J. Ellis and R.A. Flores, *Phys. Lett.* **B263**, 259 (1991).
  - [12] J.I. Collar *et al.*, *New J. Phys.* **2**, 14.1 (2000).
  - [13] F. Mayet *et al.*, *Phys. Lett.* **B538**, 257 (2002); V.A. Bednyakov *et al.*, *Phys. Rev.* **D63**, 095005 (2001).
  - [14] L.K. Pan *et al.*, *Nucl. Instrum. Meth.* **A420**, 345 (1999).
  - [15] Aquasonic, Parker laboratories, Fairfield, NJ.
  - [16] J. Puibasset, Ph.D. thesis (Université Paris VI, 2000).
  - [17] M. Blander and J.L. Katz, *AIChE J.* **21**, 833 (1975).
  - [18] F. Seitz, *Phys. Fluids* **1**, 1 (1958).
  - [19] V.P. Carey, "*Liquid-Vapor Phase-Change Phenomena*", (Hemisphere, Washington, 1992).
  - [20] M.G. Buiuid and M.V. Sussman, *Nature* **275**, 203 (1978).
  - [21] P. Reinke, *Exp. Heat Transfer* **10**, 133 (1997); P. Reinke, Ph.D. thesis (Swiss Federal Institute of Technology, 1996).
  - [22] M.A. Grolmes and H.K. Fauske, in *Procs. 5th Intl. Heat Transfer Conference*, Tokyo (1974).
  - [23] J. Bardina in "*Particles on Surfaces*", K.L. Mittal ed., (Plenum Press, NY, 1988), and references therein.
  - [24] M.D. Lelah *et al.*, *Ceram. Bull.* **58**, 1121 (1979).
  - [25] D. Quéré, private communication.
  - [26] Griffin-Worden vessel, Kontes Glass Co., Vineland NJ.
  - [27] L. Bond *et al.*, *Nucl. Phys. B (Proc. Suppl.)* **138**, 68 (2005).
  - [28] M. Harper, PhD thesis (U. of Maryland, 1991), *Nucl. Sci. Eng.* **114**, 118 (1993), *Nucl. Instr. Meth.* **A336**, 220 (1993); Ch. Peyrou in "*Bubble and Spark Chambers*", R.P. Shutt ed., (Academic Press, NY, 1967); S.C. Roy *et al.*, *Nucl. Instr. and Meth.* **A255**, 199 (1987).
  - [29] M. Das *et al.*, *Nucl. Instr. and Meth.* **A531**, 577 (2004).
  - [30] F. d'Errico, *Nucl. Instr. Meth.* **B184**, 229 (2001).
  - [31] F. d'Errico, private communication.
  - [32] H. Niedrig, *J. Appl. Phys.* **53**, R15 (1982).
  - [33] P. Belli *et al.*, *Nuovo Cimento* **A101**, 959 (1989).
  - [34] S.A. Pozzi *et al.*, *Nucl. Instr. Meth.* **A513**, 550 (2003).
  - [35] J. Bolte *et al.*, *Procs. 5th Workshop on the Identification of Dark Matter*, Edinburgh (2004), World Scientific.
  - [36] M.K. Donnelly *et al.*, NIST Technical Note 1452.