Heavy Superheated Droplet Detectors as a Probe of Spin-independent WIMP Dark Matter Existence

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At present, application of Superheated Droplet Detectors (SDDs) in WIMP dark matter searches has been limited to the spin-dependent sector, owing to the general use of fluorinated refrigerants which have high spin sensitivity. Given their recent demonstration of a significant constraint capability with relatively small exposures and the relative economy of the technique, we consider the potential impact of heavy versions of such devices on the spin-independent sector. Limits obtainable from a CF$_3$I-loaded SDD are estimated on the basis of the radiopurity levels and backgrounds already achieved by the SIMPLE and PICASSO experiments. With 34 kgd exposure, equivalent to the current CDMS, such a device may already probe to below $10^{-6}$ pb in the spin-independent cross section.

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I. INTRODUCTION

The direct search for evidence of weakly interacting massive particle (WIMP) dark matter continues to be among the forefront efforts of experimental physics. Such searches are traditionally classified as to whether for spin-independent or spin-dependent WIMP channels, of which the first has generally attracted the most attention. The current status of search efforts is defined by a number of projects, including DAMA/NaI-NAIAD, CDMS, ZEPLIN, ZEPLIN, and EDELWEISS. Because of their high spin sensitivity, the spin-independent sector also provides significant constraints on the spin-dependent phase space. In fact, this sector is also largely constrained by the results of DAMA/NaI-NAIAD and CDMS.

Among the other experiments in the spin-dependent sector are two using superheated droplet detectors (SDDs): SIMPLE (C$_2$ClF$_2$-loaded) and PICASSO (C$_3$F$_{10}$-loaded), which have recently demonstrated an ability to achieve competitive results with significantly reduced measurement exposures. This is partially because of their high fluorine content, but also the result of their intrinsic insensitivity to the majority of common backgrounds which complicate other types of direct search experiments. The impact is clear if one considers that the current results were obtained with detectors of 42 and 19.4 g active mass, respectively, vis-a-vis the recent Kamioka report of essentially equivalent results with a 28 kgd exposure of a 300 g CaF$_2$ scintillator.

Given this performance, together with the relative inexpensiveness of the technique, the question naturally arises as to whether or not SDDs might have a similar impact on spin-independent measurements. Since the cross section scales with the squares of both the mass number and the WIMP-nucleus reduced mass, exploring the spin-independent channel of WIMP interactions suggests a detector composition with nuclei of a significantly higher mass number. Although several readily available “heavy” refrigerants exist (eg. CF$_3$Br, CF$_3$I, XeF$_6$, ...), the problem of density-matching the suspension gels in order to achieve a homogeneous dispersion of the refrigerant without introducing additional radio-contaminants, together with the belief that the current impact in the spin-dependent sector derives almost exclusively from the fluorine content, has discouraged their development. For this reason, some recent attention has focused on the development of a gel-free bubble chamber approach. While avoiding the problems of density-matching, this technique requires a significant extension of the metastability lifetime of the refrigerant, which is severely degraded by surface nucleations on the container walls. Recently, the Chicago group has succeeded in achieving lifetimes of up to several hours.

We discuss in Sec. II the SDD in general and the feasibility of fabricating a CF$_3$I device. The expected background contributions to the device operation are analyzed using the current results from the SIMPLE and PICASSO experiments. Sec. III provides projections of the results to be expected from a CF$_3$I-based search which show that its implementation has the potential to make a significant contribution to the search activity. Conclusions are drawn in Sec. IV.

II. A “HEAVY” SDD

A. Fabrication Considerations

A SDD is a dispersion (emulsion) of small droplets of superheated liquid freon fixed in a hydrogenated gel, each droplet of which functions as a mini-bubble chamber. Current device constructions rely on density matching of the gel ($\rho \sim 1.3$ g/cm$^3$) with the refrigerant in order to produce a homogeneous distribution of droplets in the gel during its setting in the fabrication process. In the

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case of “heavy” refrigerants, $\rho \sim 2 \text{ g/cm}^3$; the common practise of adding heavy salts such as CsCl to the gel in order to raise its density is discouraged for dark matter search applications by its introduction of radioactive contaminations.

An alternative approach, at least in principle, is to match in viscosity rather than density. An estimate of the minimum viscosity ($\eta$) required to trap the droplets during the fabrication process is obtained by equating the viscous and Archimede’s forces:

$$\eta = \frac{2r^2gt(\rho_b - \rho_0)}{9D}$$  \hspace{1cm} (1)

where $r$ is the average droplet radius, $D$ is the height of the gel, $t$ is the time for a droplet to fall a distance $D$, and $\rho_0(\rho_b)$ is the CF$_3$I (gel) density. For $t = 1$ hour (the time required for the setting of the gel during cooling), $\rho_b(\rho_0) = 2 \times 10^3 \text{ kg/m}^3(1.3 \times 10^3 \text{ kg/m}^3)$, $r = 35 \times 10^{-6}\text{ m}$ and $D = 5 \times 10^{-2}\text{ m}$, this yields 0.13 kg/m/s. We have recently succeeded to produce a gel matrix, using the standard SIMPLE ingredients with the addition of agarose to modify the viscosity rather than density match, as well as shift upwards the sol-gel transition temperature \cite{11}, with a measured $\eta = 0.17 \text{ kg/m/s}$. This has permitted production of a prototype CF$_3$I-based SDD with 1-3 times the concentration of the SIMPLE devices. The Chicago group has similarly succeeded in developing a SDD prototype of CF$_3$I with a polyacrylamide-based gel \cite{12}.

The process results in a homogeneous distribution of micron-sized CF$_3$I droplets, and a device insensitive to $\gamma$’s and $\beta$’s at lower temperatures while sensitive to reactor neutron irradiations via the induced recoils of C, F and I. Since all direct search experiments rely on the detection of WIMP-induced nuclear recoil events, the neutron response provides an understanding of the device response to WIMPS, and of an essential background component.

In the current fabrication protocol however, and unlike the current $\text{C}_2\text{ClF}_3$ fabrications \cite{13}, about 50% of the refrigerant dissolves into the gel due to its high solubility in the high hydrogen bond content matrix, consistent with the solubility of CF$_3$I in water (16% of the gel) and glycerin (78% of the gel). Abrupt bubble nucleation of large droplets in the suspension leads to an unchecked growth of small fractures in the gel via absorption of the dissolved refrigerant by the bubbles, and a relatively rapid degrading of the detector performance. There is also a significant presence of clathrates hydrates at low temperature, implying that the device cannot be stored at temperature below 0°C because clathrates hydrates break down locally the metastability of the droplets. Although the current SIMPLE background level would suggest moderately long lifetimes in the underground site, various techniques to include the use of gelifying agents not requiring water as a solvent or the use of others techniques to inhibit the diffusion of the dissolved gas, are being explored.

B. Background Considerations

The sensitivity of a CF$_3$I-based SDD dark matter measurement is defined by the device response. Following the thermal spike model of Seitz \cite{13}, there are two thresholds for bubble nucleation: (i) the deposited energy must be larger than the work of formation of a critical embryo ($E_c$), and (ii) $E_c$ must be deposited within a distance of the order of a critical radius.

Fig. 1 shows how the two thresholds combine into the mass number ($A$)-dependent threshold recoil energy $E_{thr}^A$. The bubble nucleation efficiency of an ion of mass number $A$ recoiling with energy $E$ is given by the superheat factor $S_A(E) = 1 - \frac{E_c}{E}$. $E_c$ is set to 8 keV (37°C for all the recoiling ions (C, F and I) as in the case of the most recent SIMPLE measurement \cite{7}. $E_{thr}$ can be set as low as 6.5 keV before onset of the gel melting at 40°C; the stopping power is $\geq 100 \text{ keV/µm}$ for temperatures up to $\sim 5$°C above the gel melting point. In fact, whenever all the recoiling ions stop within a pressure- and temperature-dependent critical distance, $E_{thr}$ and $S_A(E)$ do not depend on $A$ \cite{15}. $E_{thr}^A$ (Fig. 1) was calculated as a function of the operating temperature and pressure by using the method employed by SIMPLE for $\text{R-12}$- and R-115-loaded SDDs \cite{15,16,17}, with thermodynamic parameters taken from Ref. \cite{18} and recoiling ion stopping powers precalculated with SRIM 2003 \cite{19}. As evident, the calculations foresee the same $E_{thr}^A$ and $S_A$ for F, C, I at temperatures above $\sim 29°C$ (2 atm).

![FIG. 1: Variation of $E_{thr}^A$ with temperature for each of the CF$_3$I components. The vertical line indicates the temperature below which $\alpha$’s are below the stopping power threshold of the detector.](image-url)

The reason why the threshold recoil energies $E_{thr}^A$ for direct detection of I, F and C ions in Fig. 1 do not coincide for all temperatures is that while in the range $E_{thr}^A < E_c$, below $\sim 24°C$ a F ion above $E_c$ and below $E_{thr}^A$ has not enough stopping power to trigger a nucleation. More generally, a particle above $E_c$ but below the
stopping power threshold cannot directly produce a bubble nucleation, and can only be detected indirectly, with lower efficiency, through a secondary recoiling ion. Since inelastic scattering will in general involve the absorption of part of the available energy by either the recoiling nucleus or the scattered particle, this extends to all types of scattering.

The energy $E_{\text{total}}$ (kinetic+mass) needed by an incident particle of mass $m$ in order to produce a non-relativistic recoiling ion of kinetic energy $E_R$ and mass number $A$ is given by

$$E_{\text{total}} = E_R + \sqrt{E_R^2 + 2AE_R - 2\sqrt{E_{\text{total}} - m^2} \sqrt{2AE_R \cos \theta}}$$

(2)

where $m$ is the incident particle mass, $\theta$ the recoil angle, and the mass of both nucleons has been approximated to 1 GeV/c². Since $\cos \theta \leq 1$, it is straightforward to show that $E_{\text{total}}$ is at least that corresponding to the recoiling nucleus linear momentum $\sqrt{2AE_R}$:

$$E_{\text{total}} \geq E_R + \sqrt{E_R^2 + 4m^2 + 2AE_R} \geq \sqrt{2AE_R}$$

(3)

Applying Eq. (3) to $^{12}$C, the lightest isotope in CF₃I, it is clear that no light radiation below $E_{\text{total}} = 5.4$ MeV can produce via elastic scattering non-relativistic recoiling ions of energy higher than 5 keV. For the chosen threshold of 8 keV, the minimum value of $E_{\text{total}}$ becomes 6.9 MeV. Therefore, under pressure and temperature operating conditions such that $E_{\text{thr}}^{\text{A}}$ is 8 keV, low stopping power light radiations below $E_{\text{total}} = 6.9$ MeV cannot produce a bubble nucleation either directly or via a recoiling ion.

Both the $E_c$ and $dE/dx$ thresholds can be tuned via the operating temperature and pressure conditions to render the SDD insensitive to energetic gamma-rays, X-rays, electrons and other radiations depositing less than ~200 keV $\mu$m⁻¹; the SDD is essentially sensitivity-limited to neutrons and $\alpha$-particles. This insensitivity is not trivial: the SDD at 37°C and 2 bar is effectively “blind” to $\gamma$ backgrounds below 6.9 MeV. Given the ~10⁷ evt/kgd environmental $\gamma$ rate observed in an unshielded 1 kg Ge detector [20], this blindness to $\gamma$’s is equivalent to an intrinsic rejection factor several orders of magnitude larger than the bolometer experiments with particle discrimination [21].

The external background component is primarily muons and environmental neutrons. At 1500 mwe, the ambient muon flux is ~10⁻⁶ muons/cm²s. The response of SDDS, of both low and high concentrations, to cosmic-ray muons is well-studied [21, 22]: the SDD muon response is similar to that of $\gamma$’s, with the threshold sensitivity to these backgrounds occurring for $s = [(T - T_b)/(T_c - T_b)] \geq 0.5$, where $T_c$, $T_b$ are the critical and boiling temperatures of the refrigerant. SDD operation at 37°C and 2 bar ($s \sim 0.3$) is sufficiently below threshold for this contribution to be neglected [7], and the predominant external background is neutron.

The response of low concentration SDDS to various neutron fields has been studied extensively [14, 23, 24]; the high concentration SDD response to neutrons has been investigated using sources of Am/Be, ²⁵²Cf [17, 21] and monochromatic low energy neutron beams [15, 21], and is in good agreement with thermodynamic calculations. Since the detectors are further insensitive to neutron energies below threshold, the neutron contribution can be reduced or eliminated by external moderation. The background is also addressed by the gel and the water bath used to maintain the devices at operating temperature, which are themselves neutron moderators.

Thus, in the case of the SDD, the background issue is almost entirely determined by the radiopurity of the device construction. The SDD consists of two components: the refrigerant, and the gel matrix. The refrigerant determines the response of the device, and is singly distilled during detector fabrication. The metastability limit of a superheated liquid is described by homogeneous nucleation theory [25], which gives a limit of stability of the liquid phase at approximately 90% of the critical temperature for organic liquids at atmospheric pressure, and an estimate of the spontaneous nucleation rate of

$$R_s = N_p \sqrt{\frac{2\tau}{\pi M}} e^{-\frac{10^{1800}}{3kT(\tau - P_v)/\rho_t}}$$

(4)

where $N_p = \frac{NA\mu_l}{M_{\text{mol}}}$ = 6.15 × 10²¹ cm⁻³ with $N_A$ Avogadro’s number, $M_{\text{mol}}$ the molar mass and $\rho_t$ the liquid density; $\tau$ is the surface tension, $m$ is the molecular mass, $k$ is Boltzman’s constant, and $P_l(P_v)$ is the liquid (vapor) pressure. At 40°C, $R_s$ ~ 10⁻¹⁸⁰⁰ nucleations/kgd, and decreases by three order of magnitude per degree with decreasing temperature: at 37°C this contribution is entirely negligible, as verified to within experimental uncertainties in Ref. [22].

The gel is the key factor in considerations of the device backgrounds. The current SIMPLE gel ingredients used in the CF₃I prototype are purified using pre-eluted ion-exchanging resins specifically suited to actinide removal; the freon is single distilled; the water, double distilled. The presence of U/Th contaminations in the gel, measured at ≤ 0.1 ppb via low-level $\alpha$ spectroscopy, yields an overall $\alpha$-background level of < 0.5 evts/kg freon/d. The $\alpha$ response of SDDS has been studied extensively [17, 21]. The SRIM-simulated dE/dx for $\alpha$’s in CF₃I has a Bragg peak at 700 keV and ~193 keV/µm, which sets the temperature threshold for direct $\alpha$ detection to ~37°C (Fig. 1). Below this temperature, $\alpha$’s can only be detected through $\alpha$-induced nuclear recoils. Radon contamination is low because of the 2 atm overpressure, water immersion, and short Rn diffusion lengths of the SDD construction materials (glass, metal); the measured Rn contamination of the glass is at a level similar to that of the gel.
III. POTENTIAL IMPACTS

In order to assess the potential of a “heavy” SDD to contribute to the dark matter search, we assume from the above discussion the background of a potential CF$_3$I-based search to be at the level reported by the SIMPLE dark matter search. We further assume, for comparison purposes, a 34 kgd exposure equivalent to that of CDMS-II (easily achievable, e.g., by operating for 34 days a detector array with only 1 kg active mass). The expected number of background events is then 34. Assuming 34 events are “observed”, the highest total expected event number compatible at 90% C.L. with the “observation” is 42.8, implying an upper limit to the total rate of 1.26 evts/kgd.

Currently, the discrimination capability of the SDD experiment is limited to the rejection of coincident device signals in the detector mosaic, which addresses only penetrating neutrons. The bubble nucleation process is however a four-stage process, the last two of which can generate an acoustic pulse and the last of which generally provides the recorded signal. The formation of a high temperature, high pressure zone (stage 2) is followed by its rapid expansion ($10^{-9}$ s) to a size at which the pressure inside the bubble almost equals the external pressure; if the bubble diameter is above a critical length, the fourth stage then sets in. The bubble expansion in the third stage is solely attributable to the transformed energy of the incident particle, whereas the fourth stage is due to the energy stored in the liquid. The full signal should therefore consist of both a fast and slow pulse, the fast component of which depends on the nature of the incident radiation. To what extent this is resolvable remains in question, and the feasibility of measuring this stage as a discrimination technique using ultrasound technology is being explored.

If the experiment were further able to discriminate each of the 34 “observed” events as background, then it could be claimed that no WIMP has been observed, and the 90% C.L. upper limit to the WIMP rate becomes simply $42.8/34 = 0.668$ evts/kgd, obtained by setting to 10% the probability of observing no WIMP. This would also be the limit in the unlikely case of only 26 events, since at 90% C.L. at least 26 background events should be observed. We also show this limit, indicated as “no evts”, with the actual result to be obtained lying somewhere between the two contours.

A. Spin-independent Sector

1. Isospin-independent

Based on the above estimates of the upper limit on the WIMP rate, and following the procedures of Ref. [27], the projected spin-independent limits of Fig. 2 are displayed with those of some leading experiments [4, 5, 28, 29, 30]. These projections assume a standard spherical isothermal halo with a local density of 0.3 GeV/$c^2$, a halo velocity of 230 km/s, average Earth velocity of 244 km/s, and a galactic escape velocity of 600 km/s. A Helm form factor $F(qr_n) = 3(2q/r_n)^2 e^{-(qr_n)^2/2}$ with nuclear radius $r_n = \sqrt{\frac{2}{3}\pi^2y^2 - 5x^2}$ ($x = 1$ fm, $y = 0.52$ fm, $z[fm] = 1.23A^{\frac{1}{3}} - 0.6$) is assumed [27].

As seen in Fig. 2 with an exposure equivalent to that of CDMS, an experiment based on CF$_3$I-loaded SDDs would exclude a significant part of the 3σ C.L. DAMA region even without background discrimination. This is particularly true for the unexcluded region below $\sim 20$ GeV, which is better probed by the CF$_3$I than CDMS/Ge owing to the light nuclei presence. Since the halo velocity distribution has a cutoff at the galactic escape velocity $v_{esc}$, a nucleus of mass number $A$ in a detector of threshold $E_{thr}$ can only reveal WIMPs such that

$$E_{thr}^A \leq \frac{2M_w A m_p}{(M_w + A m_p)^2}(v_{esc} + v_E)^2,$$

where the small difference in neutron and proton mass ($m_p$), has been neglected; $v_E$ is the Earth velocity with respect to the Galaxy, so that $v_{esc} + v_E$ is the maximum...
WIMP speed with respect to the laboratory. Eq. 5 implies a threshold sensitivity in \( M_W \) given by

\[
M_{W_{\text{min}}} = \frac{A m_p}{\sqrt{2 A m_p E_{\text{thr}} (v_{\text{esc}} + v_E) - 1}}. \tag{6}
\]

As \( M_W \) approaches \( M_{W_{\text{min}}} \), the fraction of the incident WIMP current detectable through a nucleus of mass number \( A \) vanishes, and, if \( A \) is the lightest isotope, the exclusion plot has a vertical asymptote for \( M_W = M_{W_{\text{min}}} \). With the above parameters, a CF\(_3\)I detector with \( E_{\text{thr}} = 8 \text{ keV} \) can probe down to \( \sim 3 \text{ GeV}/c^2 \) with \(^{12}\)C and \( \sim 3.6 \) with \(^{19}\)F, while a germanium detector with \( E_{\text{thr}} = 7 \text{ keV} \) is sensitive down to \( \sim 5.9 \text{ GeV}/c^2 \) through \(^{70}\)Ge, and a silicon detector with same threshold to \( \sim 3.9 \text{ GeV}/c^2 \) through \(^{28}\)Si.

2. Isospin-dependent

Fig. 2 is obtained under either the assumption of isospin-independence or the approximation that \( Z \approx N \). For heavy nuclei, the latter breaks down, giving the experiments with a heavy component a sensitivity to a possible isospin-dependence of the WIMP interaction. Without a priori assuming isospin-independence, the cross section \( \sigma_A \) for the scattering of a spin-independent WIMP on a nucleus of mass number \( A \) is given by

\[
\sigma_A = \frac{4}{\pi} G_F^2 \mu_A^2 (g_p Z + g_n N)^2, \tag{7}
\]

where \( G_F \) is the Fermi constant, \( \mu_A \) the WIMP-nucleus reduced mass and \( g_{p,n} \) the WIMP-proton and WIMP-neutron spin-independent coupling strengths (i.e. the coupling coefficients, in units of \( \sqrt{2 G_F} \)).

Fig. 3 shows the resulting spin-independent CF\(_3\)I exclusion projection for a 34 kgd exposure at \( M_W = 50 \text{ GeV}/c^2 \) following Ref. 31, together with the current results of some leading searches. In this representation, as clear from Eq. (7), the exclusion plots for a given \( M_W \) are generally ellipses. Even at this exposure level, a CF\(_3\)I SDD experiment could already contribute to the overall exclusion.

B. Spin-dependent Sector

Although intended for the spin-independent search effort, the CF\(_3\)I device remains sensitive in the spin-dependent sector, essentially through \(^{19}\)F and \(^{127}\)I. The spin-dependent exclusions for \( M_W = 50 \text{ GeV}/c^2 \) are shown in Fig. 4. As evident, with only 34 kgd the projected experiment would supersede the 16389 kgd NAIAD 2, or possibly yield a positive signal at this \( M_W \). The 50 GeV/c\(^2\) is chosen since it lies near the minimum of the various contours of Fig. 2 for larger or smaller \( M_W \), all results are generally less restrictive, and vary differentially.

The details of the isotopic composition are given in Table I. For completeness, \(^{13}\)C is included: its spins were evaluated in the odd group approximation 34.

Since \(^{127}\)I recoils can have high linear momentum, the
TABLE I: CF$_3$I spin parameters.

| Isotope | Z  | J* | $\langle S_p \rangle$ | $\langle S_n \rangle$ | abundance | Ref. |
|---------|----|----|----------------------|----------------------|-----------|-----|
| $^{19}$F | 9  | 1/2 | 0.441               | -0.109               | 100%      | [32, 35] |
| $^{19}$F | 9  | 1/2 | 0.075               | -0.109               | 100%      | [33]   |
| $^{127}$I | 53 | 5/2+ | 0.309               | 0.075                | 100%      | [36]   |
| $^{12}$C | 6  | 0+  | 0                   | 0                    | 98.9%     |       |
| $^{13}$C $^a$ | 6  | 1/2 | 0                   | -0.184               | 1.1%      |       |

$^a$Calculated in the odd group approximation using Ref. [37]

zero momentum transfer limit does not apply to this isotope, and the non-zero momentum transfer method of Refs. [36, 38, 39] must be applied in order to evaluate cross section limits from rate limits. Following a common choice, the results of Ref. [36] for a Bonn A potential have been used.

The two projections for CF$_3$I differ in the choice of the $^{19}$F shell model: the dotted ellipse is based on spin matrix elements (and structure functions) taken from Ref. [33], while calculation of the dot-dash ellipse employs the form factor of Ref. [27], which is independent of $a_{p,n}$. Here, the difference in orientation of the two projected ellipses is explained by the 92% lower ($S_n$) estimate of Ref. [33] with respect to the result of Ref. [32]. This leads to evaluate a smaller $^{19}$F neutron sensitivity, which tends to stretch the ellipse in the $a_n$ direction, and make it more horizontal. Incidentally, this shows that the spin-dependent response is fluorine-dominated, as expected from the 3 x larger amount of fluorine with respect to iodine.

IV. SUMMARY

At least two groups have succeeded in confronting the density-matching difficulties of “heavy” SDD fabrication by viscosity modification, and demonstrated the feasibility of producing high concentration prototype devices for R&D.

Given the fabrication feasibility, the pursuit of such experiments depends on the results to be obtained with the device implementation, which are seen to contribute competitively in both spin-independent and -dependent searches. Because of the tunable double thermodynamic thresholds of the device, a sensitivity approaching that of the present bolometric CDMS-II experiment could be economically achieved with a CF$_3$I-loading and similar exposure. This competitiveness is largely based on the device insensitivity to a majority of the backgrounds in the more traditional search devices; to remain competitive will require techniques of discriminating the remaining contribution. Since the recoil threshold can be tuned to as low as 6.5 keV, the results in particular would address the low mass region of the spin-independent parameter space still insufficiently explored by such searches. The same measurement would simultaneously contribute to the search in the spin-dependent sector, either ruling out the DAMA/NaI result completely or – more interestingly – obtaining a positive signal.

Given that the devices are robust, low maintenance and modular, and generally inexpensive in both construction and operation, large mass experiments can be easily envisioned. A small-scale measurement would provide information on the actual sensitivities of such an experiment, as well as new limits in the spin-independent sector.

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