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Negative Ion Time Projection Chamber operation with SF_6 at nearly atmospheric pressure

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Abstract: We present the measurement of negative ion drift velocities and mobilities for innovative particle tracking detectors using gas mixtures based on SF_6. This gas has recently received attention in the context of directional Dark Matter searches, thanks to its high Fluorine content, reduced diffusion and multiple species of charge carriers, which allow for full detector fiducialization. Our measurements, performed with a 5 cm drift distance Negative Ion Time Projection Chamber, show the possibility of negative ion operation in pure SF_6 between 75 and 150 Torr with triple thin GEM amplification, confirming the attractive potentialities of this gas. Above all, our results with the mixture He:CF_4:SF_6 360:240:10 Torr demonstrate for the first time the feasibility of SF_6^- negative ion drift and gas gain in He at nearly atmospheric pressure, opening very interesting prospects for the next generation of directional Dark Matter detectors.

Keywords: Dark Matter detectors (WIMPs, axions, etc.); Gaseous imaging and tracking detectors; Micropattern gaseous detectors (MSGC, GEM, THGEM, RETHGEM, MHSP, MICROPIC, MICROMEGAS, InGrid, etc); Time projection chambers

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2018 JINST 13 P04022

Contents

1 **Introduction** 1

2 **Motivations** 2

3 **The NITPC detector** 3

4 **The Beam Test Facility and the Experimental Setup** 4

5 **Drift velocity and mobility measurements** 6

6 **Conclusions** 10

2018 JINST 13 P04022

1 Introduction

The study of the nature of Dark Matter (DM) and of the properties of neutrinos share the same challenging requirements on the simultaneous optimisation of the active volume, energy resolution and background rejection capabilities of the detector. Powerful tools in event discrimination are the topological and directional signature of the event. Gaseous Time Projection Chambers (TPCs), thanks to their inherently 3D tracking reconstruction, dE/dx measurement, particle identification capabilities and versatility in terms of target gas can potentially provide very useful observables for such rare event searches.

A modification of the conventional TPC approach involves the addition to the gas mixture of a highly electronegative dopant, creating a Negative Ion TPC (NITPC) [1]. In this configuration, the primary electrons generated by charged particles during gas ionisation are captured at very short distances ($\leq$ 100 µm) by the electronegative molecules, creating negative ions. Thanks to the TPC electric field, these anions drift to the amplification plane at the anode. Here the extra electron is stripped from the negative ions and gives rise to an electron avalanche. Since anions are much more massive than electrons and can therefore exchange energy with the gas molecules much more efficiently, their diffusion is strongly reduced in their drift towards the anode. Thanks to this, longitudinal diffusion of $\sim$ 1 mm/m can be reached without need of magnetic fields (to be compared to $\sim$ 20 mm/m with standard electron drift) [1].

This idea was first proposed by Martoff in 2000 [1] and was then successfully demonstrated in the directional DM search field with CS_2-based gas mixtures by DRIFT [2, 3]. DRIFT gas choice of CS_2:CF_4:O_2 at 30:10:1 Torr is motivated by:

- low target density, obtained using low total gas pressure, to lengthen low energy nuclear recoil tracks;
- CS_2 to obtain negative ion drift;

- CF_4 to provide a spin-odd target for Spin-Dependent (SD) WIMP interactions (Fluorine);
- O_2 to enhance the amplitude of the secondary peaks in the time signals. Recently, in fact, DRIFT observed in its gas the presence of multiple charge carriers, whose time signal intensity can be amplified by the addition of a small quantity of O_2. These carriers are faster than the main one and are identified as negative ions with a mass smaller than CS_2^- [4]. Given the different mobilities of anions of different masses, the difference between their time of arrival effectively provides a measurement of the absolute position of the event along the drift direction. In most direct DM search experiments, a critical background resides in Radon Progeny Recoils (RPRs) coming from detector surfaces and mimicking WIMP interactions. The possibility to measure the event position also in the drift direction allows to effectively reject these events (a technique usually referred to as fiducialization), and allowed DRIFT to reach a background-free region of interest [5].

Unfortunately, CS_2 has a combination of low flash point (−30°C), high vapor pressure (400 Torr) and low explosive mixture limit in air (1.3%), which requires great care in handling the material, especially with the addition of O_2 in an underground site. For these reasons, recently SF_6 has been proposed as an alternative and safer capture agent able to maintain all the interesting features of negative ion drift, while at the same time easing the handling and possibly extending the sensitivity in the WIMP search [6, 7]. In [6], ^{55}Fe energy spectra, together with gas gain, diffusion and mobility measurements obtained with a single 400 µm thick GEM are presented for pure SF_6 between 20 and 100 Torr. These results established for the first time the employment of this gas for negative ion drift operation, and verified the expected thermal diffusion of SF_5^- and SF_6^- at low E/N reduced drift fields. Above all, they demonstrated the possibility of fiducialization along the drift direction through the use of minority carriers (SF_5^-), with performances consistent with [4], laying the groundwork for the future use of SF_6.

2 Motivations

Our work sets into the framework of expanding the study and the possibilities of exploitation of SF_6 negative ion drift, as discussed in section 1. For our measurements, we choose to test a triple GEMs amplification with pure SF_6 between 75 and 150 Torr and SF_6 added to Ar:CO_2 and He:CF_4 mixtures. This was motivated by:

- the interest in the international community for the development of a DM directional experiment from 1–10 m^3 towards the ton-scale in the framework of the CYGNUS project [7], based on a NITPC approach with SF_6. The scenarios considered in this context are pure SF_6 at 20 or 200 Torr and He:SF_6 at 60:20 Torr or 600:200 Torr, to assess the proposed experiment sensitivity to different physics cases in terms of WIMP mass versus Spin Independent (SI) or SD coupling. With its high Fluorine content, the choice of SF_6 has a clear advantage over CS_2:CF_4 mixtures for both SI and SD searches. The low He atomic number can extend the experiment sensitivity down to few GeV WIMP masses, and its low density may allow to reach nearly atmospheric pressure operation while still maintaining tracks from low energy nuclear recoils long enough to be measured.

2018 JINST 13 P04022

- the promising scintillating features of He:CF_4 mixtures for TPC application with GEM amplification and optical readouts [8, 9]. Together with a charge signal, in fact, the avalanche mechanism in the amplification region creates an accompanying scintillation light, that can be detected by optical devices and used for track reconstruction. Three-dimensional tracking has been recently shown with a 7 Liter active volume prototype with 20 cm drift distance, triple thin GEM amplification and light readout through a CMOS camera and a PMT [10], obtaining a $\sim$ 100 µm tracking resolution on minimum ionising particles. The advantage of this technique is that, with the proper large aperture and suitable focal length lens, large areas can be imaged with high tracking precision at reduced costs and number of channels.

- the possibility of employing a TPC with He and ^{3}He target gases for the detection of fast (through nuclear recoil) and thermal (through thermal capture on ^{3}He) neutrons [11]. Such a detector would be very interesting, for example, to precisely measure spectrum and direction of the neutron fluxes from natural radioactivity present in the underground experimental halls.

- the possibility to compare with and extend to lower E/N values the measurements presented in [6], since this information is particularly important for large volume and long drift distance detectors.

3 The NITPC detector

The NITPC detector is composed by a 5 cm long field cage with an inner diameter of 7.4 cm, enclosed on one end by the cathode (a thin copper layer deposited on a PCB) and on the other by the GEMPix device [12, 13]. The 0.5 cm thick polycarbonate field rings, with 1 cm pitch, were manufactured with a 3D printer and 1 mm diameter silver wires were encased in to provide uniformity to the drift field. Resistors of 1 GΩ connecting the wires provide the partition of the HV through the field cage, that is provided by an external CAEN N1470 power supply. The GEMPix is obtained coupling three standard CERN GEMs with a $3 \times 3\,\mathrm{cm}^2$ active area to a Quad-Timepix ASIC (4 Timepix chips). A picture and a schematic of the NITPC is shown in figure 1.

The GEM [14] is a Micro Pattern Gas Detector composed by a 50 µm thick Kapton insulating foil, electroplated with copper on both sides, where conical holes with 70 µm external diameter are etched with a chemical process. If a voltage is applied between the two sides of the GEM foil, a very high electric field is generated in the holes. When an electron traverses the hole, avalanche multiplication takes place giving typically 20 secondary electrons for each primary electron (with the exact value depending on gas density, gas mixture and applied electric field). The triple GEM configuration provides not only higher gas gain but also higher stability, with respect to single or double GEMs [15]. In the GEMPix, the GEM foils are held rigid by gluing them to a frame and then superimposing them on top of the pixels chip. The system is powered by the HVGEM [16], a dedicated high voltage supply developed by Laboratori Nazionali di Frascati, which works as an active HV divider. The HVGEM can power seven floating independent channels (for the 3 GEMs, plus the two transfer fields, the induction field and the drift field), each isolated up to 5 kV to ground and with a high sensitivity ($\sim$ 1 nA) current meter.

The Timepix [17] is part of the second generation of the Medipix chip family, fabricated in the IBM 250 nm CMOS process, with a 256×256 matrix of $55 \times 55\,\mu\mathrm{m}^2$ square pixels. The

2018 JINST 13 P04022

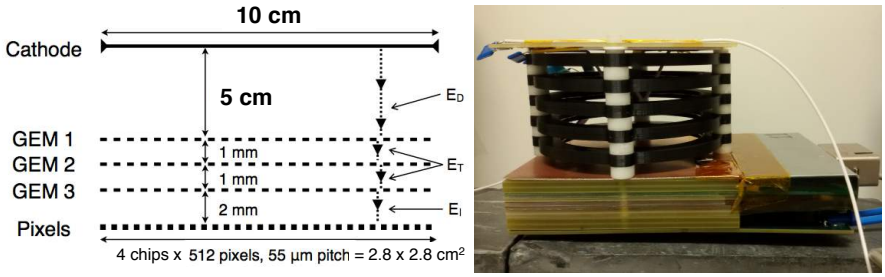

Figure 1. NITPC detector: on the left a schematic showing the distances between the cathode, the GEMs and pixels planes, and on the right a picture showing the full detector, with the field cage polycarbonate field rings in black and the PCBs composing the GEMpix below.

ionisation produced in the gas and amplified in the triple GEM system above is directly collected by the Timepix, that works as a charge collecting anode.[1] Each pixel in the matrix can individually measure either the number of hits above a threshold (Counting or Medipix mode), or the charge deposited via Time Over Threshold method (TOT), or the time of arrival (TOA). A distinctive feature of the Timepix, not available in other chips used in this field (i.e. ATLAS chips in D3 [18]), is the possibility to extend the acquisition time window to fractions of a second, thanks to the adjustable clock sampling frequency and the clock counter dynamic range of 11810 counts. This is very important when measuring time of arrival of negative ions, where typical sampling frequencies of 40–50 MHz as [18] are too fast for anions, that are 10^3–10^4 times slower than electrons.

The Timepix is controlled and readout by the FITPix interface [19], based on a FPGA integrated circuit that can handle both hardware and software triggers. The Fitpix is fully powered from an USB port, making it easy to operate from a common computer. The Pixelman software[2] allows to manage the Fitpix and to select Timepix mode of operation, acquisition time window, sampling frequency, trigger, pixels general settings and to perform chip thresholds equalisation.

4 The Beam Test Facility and the Experimental Setup

The Beam Test Facility (BTF) is a beam transfer line of the double ring electron-positron collider of the DAΦNE accelerator complex at Laboratori Nazionali di Frascati [20]. In the BTF, electrons of energy between 25 and 750 MeV with a repetition rate of 50 Hz and a 10 ns pulse can be produced, with a multiplicity between 1-10^8 particles per bunch [21]. In our setup, we choose to optimise the intensity of the beam rather than the dimension, given the short time available for the measurements. We used a 450 MeV electron beam with a multiplicity of about 500 particle per bunch. This resulted

[1] Typical Timepix pixels possess silicon semiconductor sensors bump-bonded on top, where the silicon is needed to produce the charge at the passage of an ionising particle. In our detector approach no silicon is therefore required.

[2] http://aladdin.utef.cvut.cz/ofat/others/Pixelman/Pixelman.html.

2018 JINST 13 P04022

in a beam-spot dimension of about 1 mm in Y (the drift direction in the detector) and 2 mm in X , with about 2 mrad angular divergence, as measured by the BTF beam monitoring. In order to operate the NITPC below atmospheric pressure, the detector was positioned inside a steel vacuum vessel, equipped with a 50 µm thick Mylar window to allow the electron beam to enter the active volume with negligible intensity loss and reduced multiple scattering. After sealing, for each measurement the vessel was pumped down to less than 0.1 Torr with a dry scroll vacuum pump and then filled with the chosen gas mixture to the operating pressure. We monitored the overall pressure inside the vessel during data taking with a capacitance manometer and repeated the above procedure any time the total pressure increased above 2% with respect to its original value. Since our vessel is made of steel instead of acrylic as in [6], we do not expect a significant amount of water or other contaminants to come from this part of the apparatus. On the other hand, we have some evidence of a not perfect sealing of the very thin Mylar windows. For this reason, we believe our contamination to be mainly air. In section 5 we will discuss the possible effects of such gas impurity on our measurements. The vessel was positioned on top of a table, that can be moved with 1 mm steps with 100 µm uncertainty in X and Y directions, at 110 cm distance from the beam exit point from the vacuum pipe. This allowed to precisely position the electron beam at different drift distances from the GEMPix readout and hence perform the mobility and drift velocities measurements here presented. A picture of the experimental setup showing the end of the beam line and the vacuum vessel is shown in figure 2.

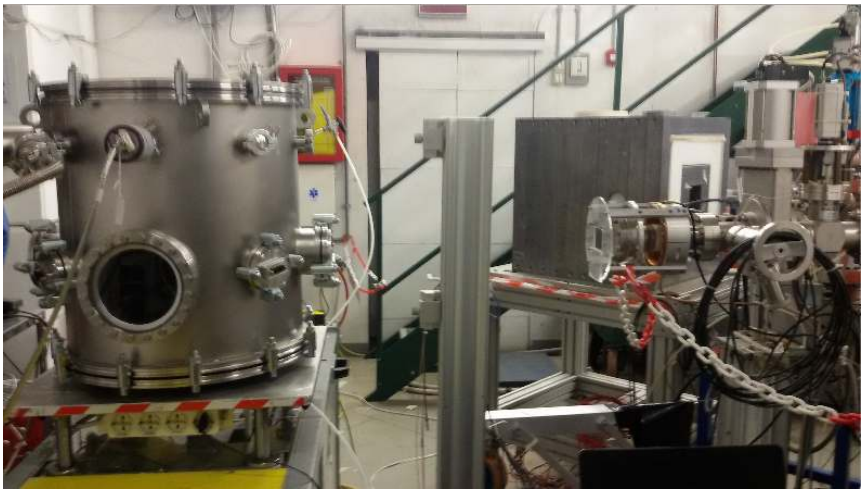

Figure 2. Experimental setup at the BTF, with the electron beam going from right to left into the vacuum vessel (cylindrical stainless steel container on the left).

2018 JINST 13 P04022

5 Drift velocity and mobility measurements

Several measurements in various conditions have shown that electron capture by the electronegative SF_6 molecule occurs very rapidly, with a mean free path of about a micron at the pressures and drift fields considered in our measurements [6, 22]. The negative SF_6 ions are produced by attachment of electrons to the SF_6 molecule, originating a metastable excited ion SF_6^{-*} (eq. (5.1)) that can decay mainly through auto-detachment (eq. (5.2)), collisional stabilization (eq. (5.3)) or auto-dissociation (eq. (5.4)) [22]:

$$SF_6 + e^- \rightarrow SF_6^{-*} \quad \text{(attachment, metastable)} \tag{5.1}$$

$$SF_6^{-*} \rightarrow SF_6 + e^- \quad \text{(auto-detachment)} \tag{5.2}$$

$$SF_6^{-*} + SF_6 \rightarrow SF_6^- + SF_6 \quad \text{(collisional stabilization)} \tag{5.3}$$

$$SF_6^{-*} \rightarrow SF_5^- + F \quad \text{(auto-dissociation)} \tag{5.4}$$

with the relative abundances of the decay channels depending on the electron energy, gas pressure, temperature, and drift field. Other negative ion species such as F^- or SF_4^- can be produced, but with much lower probabilities, due to the much lower cross section and higher electron energy required. Therefore, in our working conditions we expect only two anion species to be observable, SF_6^- and SF_5^-.

The possibility to have two or more species of charge carriers can provide a measurement of the event position along the drift direction in gas-based TPCs, allowing to reject RPRs background in DM and other rare event searches, as discussed in section 1. The measurements in [6] show that SF_5^- mobility is only 6.9% higher than SF_6^- at a E/N of about 39 Td and the relative SF_5^-/SF_6^- peak amplitude ratio is between 1.5–3% in the same regime. Since we performed our measurements in the range from 3 to 30 Td and our drift distance is limited to 5 cm, unfortunately we do not expect to be able to distinguish the minority carrier species in our data. This is consistent with what was observed in [6], where SF_5^- and SF_6^- peaks could not be resolved for drift distances shorter than 10 cm. For these reasons, we can assume the detector discussed in section 3 to be sensitive only to the main SF_6^- time signal.

To determine the negative ion drift velocity, we measured the Timepix TOA of the collected charges at five different drift distances (1, 2, 3, 4 and 5 cm) from the GEMPix, with the data acquisition triggered externally through the Fitpix interface thanks to a signal provided by the BTF system monitor. For each gas mixture and pressure, we repeated the same measurement at the drift fields of 250 V/cm, 530 V/cm, 640 V/cm, 750 V/cm and 860 V/cm.

Before any measurement, we adjusted the pixel thresholds as most homogeneously as possible, with an automatic threshold equalisation procedure provided by the Pixelman software. Since this method can be applied only one chip at a time, non-uniformities are still possible over the full area of our readout, that is composed by 4 different chips (Quad-Timepix). For these reasons, in our analysis we determine the drift times and drift velocities independently for each chip and then average the results.

For each chip, we perform a gaussian fit to the peak of the TOA distribution for each of the five drift distances in the range of ±1 FWHM around the maximum and we take the mean and sigma of the fit as the central value and error of the drift times of the SF_6^- anion. As discussed in section 4,

2018 JINST 13 P04022

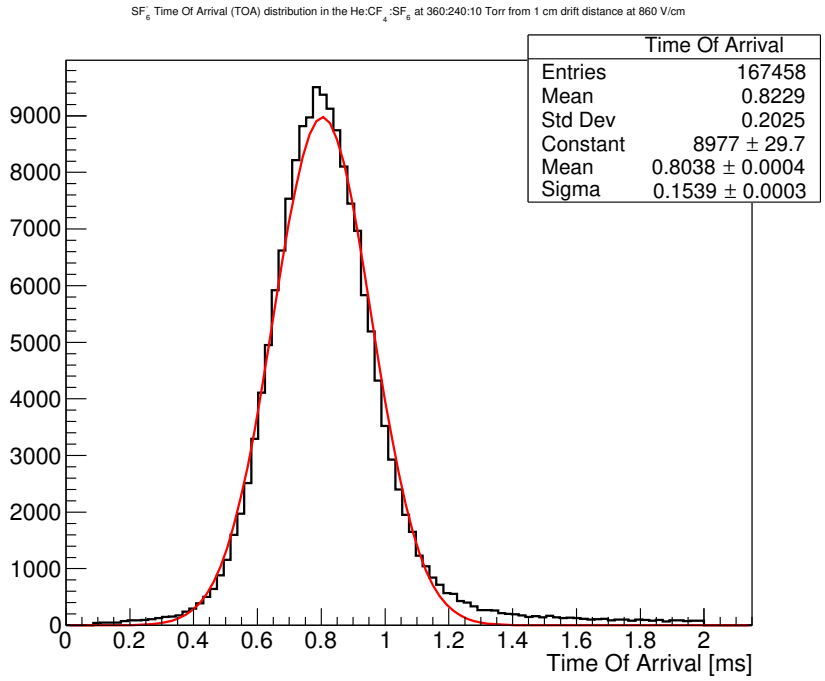

Figure 3. Example of the gaussian fit to the time of arrival distribution of the ionization cloud for the He:CF_4:SF_6 360:240:10 Torr mixture at 1 cm drift distance with a 860 V/cm drift field.

in order to produce ionisation in the gas of our detector, we used a 450 MeV electron beam with about 1–2 mm beam-spot and 2 mrad angular divergence at the beam exit point from the vacuum pipe. The air and other materials between the beam exit point and the gas inside the NITPC (a beam monitor, a 50 µm Mylar window and the gas inside the vacuum vessel) contribute for an additional total 1.9 mm smearing of the beam dimensions by multiple scattering. For these reasons we are not sensitive to the diffusion over the short 5 cm NITPC drift length. Beam dimensions and diffusion, however, affect only the time distribution width and not its central value, allowing us to measure the negative ion drift times properly.

An example of the gaussian fit to the TOA distribution of the ionisation cloud for the He:CF_4:SF_6 360:240:10 Torr mixture at 1 cm drift distance (summed over all chips) with a 860 V/cm drift field is shown in figure 3. The broad non-peak component at lower times with respect to the main peak observed by [6] and attributed to water vapor contamination, does not appear in our data. Since we believe our contamination to be mainly from air (as discussed in section 4) and since we do not observe any strange feature in our time signal, we do not have any strong reason to believe that this is affecting our measurement beyond the uncertainties we are assigning to it.

To extract the drift velocity and the error associated, we perform a linear fit of the drift times versus drift distances. We cross-checked our data analysis method with the conventional electron carriers gas mixture Ar:CO_2:CF_4 with a 45:15:40 ratio at atmospheric pressure, measuring a drift velocity of 2.06 ± 0.02 cm/μs in a 500 V/cm drift field, in agreement with already published results [23]. The linear fit to the SF_6^- drift velocities at 860 V/cm of drift field obtained in each Timepix chip for the He:CF_4:SF_6 360:240:10 Torr mixture is shown in figure 4.

2018 JINST 13 P04022

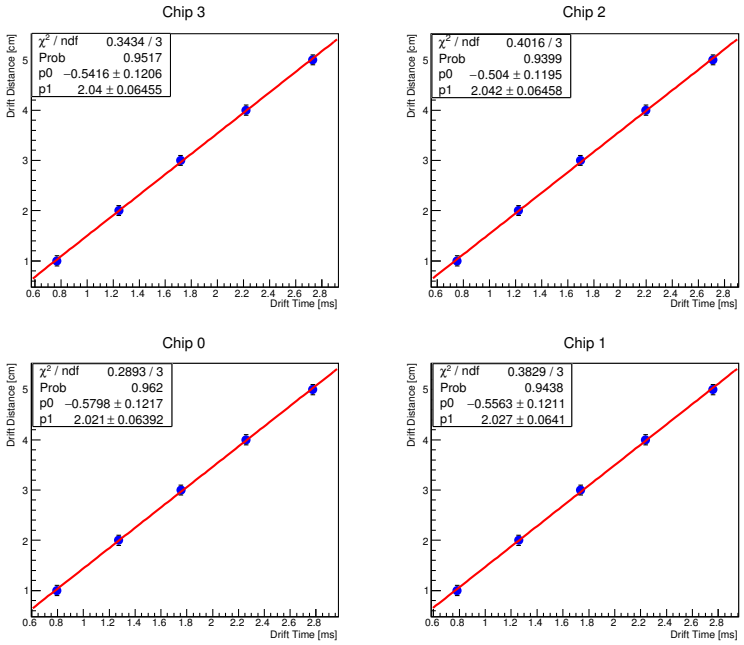

Figure 4. Chip by chip fit of the TOA versus drift distance, in order to extract the drift velocity of SF_6^- in $He:CF_4:SF_6$ 360:240:10 Torr (p_1 fit parameter).

The top panel of figure 5 shows the SF_6^- drift velocities as a function of the drift field, measured in the gas mixtures and pressures reported in table 1. In each case, the total HV applied to the triple GEMs corresponded to charge gain of the order 10^3 and it varies because different gas mixtures with different total pressures require different electric fields to generate the same gas amplification. All the drift velocities shown in figure 5 are in the range of cm/ms, demonstrating that we are effectively measuring the drift of negative ions and not electrons.

For ions of mass m drifting in a gas with molecules of mass M, the drift velocity can be written as [24]:

$$v_d = \left(\frac{1}{m} + \frac{1}{M}\right)^{1/2} \left(\frac{1}{3kT}\right)^{1/2} \frac{eE}{N\sigma}, \tag{5.5}$$

where E the drift field, N the gas density and σ is the ion-gas molecule total cross-section. All the measurements in top panel of figure 5 are consistent with this expectation, showing a linear dependence on the drift field E and reduced velocities for higher pressures. The mobility $\mu = v_d/E$, is obtained from the drift velocity values of top figure 5 and the results are shown in table 1.

In order to compare our results at different gas densities, we define the reduced mobility as:

$$\mu_0 = \frac{v_d}{E}\frac{N}{N_0}, \tag{5.6}$$

where $N_0 = 2.687 \times 10^{19}\,\mathrm{cm}^{-3}$ is the gas density at STP (0°C and 760 Torr). These are shown in the bottom panel of figure 5 as a function of the reduced field E/N in Townsend units (1 Td = $10^{-17}\,\mathrm{V\,cm}^2$), with the conservative uncertainty of 2% assigned to the drift field and pressure (see also section 4).

2018 JINST 13 P04022

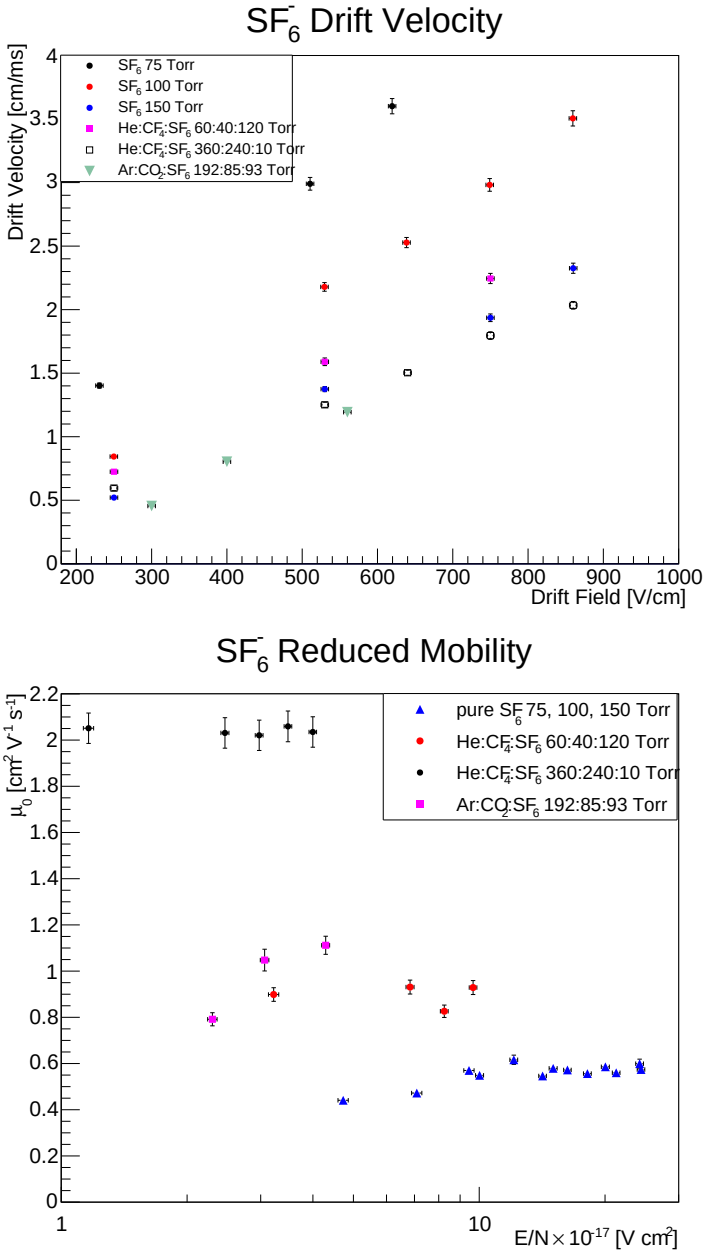

Figure 5. SF_6^- drift velocities (top) and reduced mobilities (bottom) in pure SF_6 (at 75 Torr, 100 Torr and 150 Torr), in Ar:CO_2:SF_6 at 192:85:93 Torr and He:CF_4:SF_6 at 60:40:120 Torr and 360:240:10 Torr.

For each gas mixture, the measured reduced mobility is nearly constant, as expected in the low E/N regime. Our results on SF_6^- mobility in pure SF_6 are consistent with [6] and [22] between 10 and 30 Td of E/N and extend the measurements to the region below 10 Td. The measured SF_6^- mobility in He:CF_4:SF_6 360:240:10 Torr is nearly 4 times larger than in pure SF_6 and twice the value in He:CF_4:SF_6 at 60:40:120 Torr and Ar:CO_2:SF_6 at 192:85:93 Torr. Given eq. 5.6 and 5.5, this seems to indicate that the total cross-section for SF_6^- interactions in the He:CF_4 gas mixture is smaller than in Ar:CO_2 and in pure SF_6 in the E/N range we explored.

2018 JINST 13 P04022

Table 1. SF_6^- mobility for the different gas mixtures and pressures studied in this work.

Gas	Partial Pressure [Torr]	Triple GEMs Voltage [V]	SF_6^- Mobility [$cm^2\ V^{-1}\ s^{-1}$]
SF_6	75 ± 2	1140 ± 5	5.8 ± 0.2
SF_6	100 ± 2	1240 ± 5	4.3 ± 0.1
SF_6	150 ± 3	1440 ± 5	3.00 ± 0.08
$He:CF_4:SF_6$	60 : 40 : 120 ± 4	1460 ± 5	2.9 ± 0.1
$He:CF_4:SF_6$	360 : 240 : 10 ± 10	1640 ± 5	2.36 ± 0.06
$Ar:CO_2:SF_6$	192 : 85 : 93 ± 7	1480 ± 5	2.8 ± 0.2

6 Conclusions

In this paper we studied the drift velocities and mobilities of SF_6^- ions in various gas mixtures. This was motivated by the recent attention given by the directional DM search community to the use of electronegative gases with high Fluorine content, namely SF_6, in order to reduced diffusion and allow for full detector fiducialization [6, 7].

After having reproduced the measurements of SF_6^- mobilities in pure SF_6 available in the literature [6, 22], we obtained the first evidence of SF_6^- drift in a He mixture at nearly atmospheric pressure, namely He:CF4:SF6 at 360:240:10 Torr. Moreover, although we did not extract precise gain information, we demonstrated for the first time the possibility to obtain gas amplification from SF_6^- with triple thin GEMs, both at low and nearly atmospheric pressure.

These results are of great interest in the context of the CYGNUS project, for the development of a ton-scale directional DM detector [7], with $He:SF_6$, possibly at 1 bar. Thanks to the attractive scintillating features of $He:CF_4$ mixtures, this result is considerably relevant for TPC detectors based on optical readouts [8–10], opening the prospect of negative ion operation with SF_6 also for these devices. We are currently working on a more precise study of thin GEMs gain capabilities in a negative ion operation mode, in order to better understand the avalanche mechanism and maximise gain and energy resolutions. This will be the subject of an upcoming publication.

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2018 JINST 13 P04022