

# Xenon Sampling System for LUX

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## Abstract

We describe a xenon purity analysis system we have developed used by the LUX dark matter experiment based on a mass spectrometry technique. The device is fully automated, simple, compact and is integrated into the LUX circulation system allowing for hourly, in-situ sampling from several ports. The sensitivity of the spectrometer is enhanced by several orders of magnitude by the presence of a liquid nitrogen cold trap, and many impurity species of interest can be detected at the level of one part-per-billion or better. In the case of Kr, a troublesome internal background for xenon based dark matter experiments, the sensitivity is sub one part-per-trillion. We have used the technique to screen the LUX xenon before, during, and after the first underground science run, and these measurements have proven useful. This is the second application of the cold trap mass spectrometry technique to an operating physics experiment, first employed for EXO-200.

### *Keywords:*

Noble gas, xenon, purification, SAES purifier, cold trap, mass spectrometry, mass spectroscopy, Dark Matter

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## 1. Introduction

The LUX collaboration has conducted the first dark matter search at the Sanford Underground research facility in South Dakota becoming the leading spin-independent WIMP cross section limit [ref]. The LUX detector is a dual phase xenon TPC in which energy deposition and the 3D coordinate of an event can be reconstructed from the primary scintillation (S1) and the subsequent secondary scintillation (S2) collected on two arrays congaing 61 PMTs each. To ensure that radioactive backgrounds in the detector materials do not obscure the WIMP signal, a comprehensive materials screening program was employed during the construction of the experiment[ BG paper]. This program measured and certified the radiopurity of all passive detector materials, including the PMTs, TPC instrumentation, the cabling, the xenon vessel, and the shielding materials. In this article we describe a complementary program to study the purity of the xenon source itself. This campaign has been carried out with in-situ measurements during the surface run in 2012 and the underground science run in 2013. These measurements have allowed us to verify that the xenon stockpiles after gas chromatography [ref] are suitable for their intended purposes in LUX, to monitor the performance of the xenon gas purifiers, and to independently monitor the radioactive backgrounds from  $^{85}\text{Kr}$  and  $^{39}\text{Ar}$  beta decay daily.[ref]

The measurements described in this article are based on a cold trap mass spectrometry technique which was developed to study  $\text{O}_2$ ,  $\text{N}_2$ , and  $\text{CH}_4$  impurities in xenon [CT, Purifier, tritium paper]. The method has been extended for the purposes of krypton detection as well [6] and was first implemented for xenon screening use in EXO-200. [EXO paper]

Mass spectrometry has several attractive features as an analysis technique. It can detect both electronegative and non-electronegative impurity species, including  $\text{O}_2$  and some of the problematic noble gases which contain radioactive isotopes. It allows each impurity species to be identified and counted individually unlike mass spec techniques that can only measure one species at a time[ref]. It also allows the purity of the xenon to be screened, and possibly corrected, prior to detector operations, which is similar to how all other EXO-200 and LUX detector materials are treated. Further, the technique is simple and cost effective only requiring readily available laboratory equipment. The analysis system takes up minimal space and can be plumbed in directly to any circulation panel allowing for for in-situ, hourly purity results at the sub ppt level for multiple impurity species simultaneously, making the method attractive over other more complex techniques which only detect krypton, off site and on a significantly larger time scale [Aprile, and the Germans].

We have found many useful applications for having an in situ automated xenon sampling device which will be described in detail. Mainly for LUX, we could ensure the Kr content of the xenon each time the xenon was moved or there was a potential for air contamination. Xenon100 suffered a year long setback due to a minor leak in the reticulation pump which took days to show up in the liquid TPC via data analysis, something that would have been caught by the daily gas sampling program developed LUX. Just one liter of air breaching the plumbing is enough to exceed the radioactive background goals for the LUX experiment. It is important to note that minimal xenon is lost during

the sampling which consumes only milligrams of xenon per sample, the bulk of the sampled xenon is simply recovered after analysis.

## 2. Purity Requirements

There are two aspects of xenon purity requirements that can be screened for using the gas sampling technique. First, the xenon stockpiles should be relatively free of electronegative impurities for successful TPC operations. For example, the  $O_2$  concentration should be less than 1 ppb in order to collect ionization over the 20 cm drift length of the detector[8]. Other electronegative impurity species may have larger or smaller attachment coefficients. For example, the attachment coefficient of  $N_2$ , measured in liquid Ar, is about a factor of 1000 smaller than that of  $O_2$  [9]. Second, Krypton and argon impurities are problematic because they include the beta emitters  $^{85}Kr$  and  $^{39}Ar$ . After gas chromatography, LUX requires a Kr and Ar concentration of  $<5 \cdot 10^{-12}$  g/g and  $<1 \cdot 10^{-6}$  g/g respectively. With Q values of 687 keV and 565 keV respectively, these beta decays occur in the fiducial volume defeating the power of xenon self-shielding and contribute to the WIMP search background. Clearly the krypton goal is much more stringent than the argon goal, so krypton contamination is the more serious concern. Note that the mass spectrometry technique does not detect  $^{85}Kr$  and  $^{39}Ar$  directly, but instead detects the stable and abundant isotopes  $^{84}Kr$ ,  $^{86}Kr$ ,  $^{82}Kr$  and  $^{40}Ar$ , using the standard isotopic abundances ( $\sim 2 \cdot 10^{-11}$   $^{85}Kr/natKr$  [10] and  $\sim 8 \cdot 10^{-16}$   $^{39}Ar/natAr$  [11]). The concentrations of the radioactive components can then be inferred from the isotopic abundances, when these are known. Radon is another radioactive noble gas which is a serious concern. However, radon cannot be detected by our method, but a limit can be placed on external radon diffusion by knowing the activity in the surrounding air and measuring Kr and Ar accumulation in the xenon overtime.

## 3. Methodology

The xenon gas analysis technique used for LUX is similar to the method described in Refs [5] [6] [EXO]. We use a residual gas analyzer (SRS RGA-200) mass spectrometer aided by a liquid nitrogen coldtrap to measure the impurity content of xenon gas, the diagram representing the basic setup is shown in figure 1. The RGA requires a high vacuum to operate ( $10^{-5}$  torr or less), so only modest amounts of xenon gas can be admitted into the device at one time. This is accomplished with a precise vacuum leak valve. The RGA measures the partial pressures of the relevant atomic masses, and these partial pressures are proportional to both the concentration of the various gas species and to their flow rate through the leak valve. By opening the leak valve further and further, we can increase the flow rate to an arbitrarily high value, resulting in higher and higher RGA partial pressures for all species. In principle this allows very small concentrations of impurities to be detected above background levels. In practice, however, the use of very high flow rates will cause the RGA to saturate due to the high partial pressure of the bulk xenon gas. Once the pressure of the xenon rises above  $10^{-5}$  torr, the

RGA will be unable to operate, and this limits the impurity sensitivity of the RGA to about one part-per-million. Since we desire to detect impurities at the part-per-billion level or better, we must prevent RGA saturation by removing the bulk xenon from the sampled gas. This is accomplished by placing a liquid nitrogen cold trap between the leak valve and the RGA. In the cold trap, the xenon partial pressure is held fixed by the liquid nitrogen bath at the vapor pressure of xenon ice at 77 K ( $1.8 \times 10^3$  torr), independent of the leak valve setting and the flow rate. With these conditions in the coldtrap impurity species of interest such as  $N_2$ ,  $O_2$ , He, Ar, Kr,  $CH_4$  remain gaseous and pass through unabated while the bulk xenon is captured [ref]. Depending on flow, the xenon leaving the coldtrap has up to six orders of magnitude enhancement in the impurity to xenon ratio and is passed to the RGA for measurement. The xenon partial pressure is further reduced below  $10^{-5}$  torr at the RGA by including vacuum plumbing with impedance after the cold trap. With the xenon pressure now held fixed, we can increase the flow rate, and therefore the partial pressures of many impurity species, by several orders of magnitude. In laboratory bench tests this method has achieved sensitivity to 0.12 ppb of  $O_2$ , 0.3 ppt of Kr (g/g) and 5.0 ppt of  $CH_4$ . Impurities which do not pass through the cold trap at liquid nitrogen temperature, such as  $H_2O$  or heavy hydrocarbons, cannot be detected in this way, but in some cases they may be detectable as an excess background level in the the cold trap plumbing after it returns to room temperature.

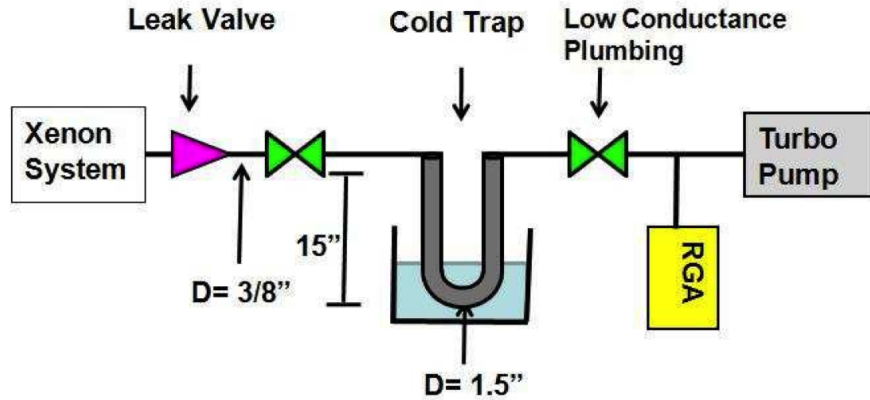


Figure 1: Basic setup of the purity analysis technique. A coldtrap is used to remove the bulk xenon, enhancing the impurity to xenon ratio of several species of interest by up to six orders of magnitude before passing the sample to a RGA for analysis.

For the LUX experiment we have integrated the xenon gas analysis system into the xenon circulation system and automated the sampling procedure using pneumatic valve via slow control, the plumbing schematic is shown in Figure 1. Xenon gas is collected from the various locations of interest along the circulation path including the input/output of the xenon gas purifier, the output of the

TPC, directly from the bottle farm and the xenon storage vessel. The gas sampling is done directly from the port of interest with the lines permanently plumbed in, reducing potential for air leaks and other systematics associated with having to draw samples into bottles and shipping them elsewhere for analysis [ref]. In the case where we sample a high pressure source such as a gas cylinder or SRV (storage recovery vessel), the sampling port is located at the output of a pressure regulator. The pressure of the xenon gas sampled is set either by the regulator (for a high pressure source) or by the pressure of the LUX xenon gas system which does not exceed 3 atm absolute. In either case the sample pressure is typically between 1 and 1.5 atmospheres in the 0.80 liter sample volume (about 5 to 9 grams of xenon). A diagram showing sampling ports plumbed into the LUX experiment and a schematic of the analysis system is shown in figure 2.

The analysis system consists of a 0.5 l bottle used as a buffer volume for sampled xenon and 0.3 l of additional plumbing to the sampling ports, a capacitive manometer, three vacuum leak valve set at various leak rates, a U- shaped liquid nitrogen cold trap, a low-conductance plumbing element, an SRS RGA-200, two cold cathode vacuum gauge, two turbo pumps, two scroll pumps, six pneumatic VCR valves and instrumentation necessary for automation via a slow control interface. The capacitance manometer (MKS Baratron model 627B) measures the pressure in the sample buffer volume (0.8 l total) before and during the impurity measurement. The leak valves (part number of the ones with the dial?) admit the sampled gas to the cold trap at much reduced pressure and allows for control of the flow rate into the trap a three preset values. The cold trap consists of a U-shaped segment of 3.8 cm OD stainless steel tube with a height of 38 cm designed to be immersed in a liquid nitrogen bath. The low conductance element is a 10 cm segment of 0.6 cm and 0.95 cm OD plumbing with one right angle bends. A port leading to the SRV is also plumbed to the analysis system to allow for 99.9% of the 5 to 9 grams of sampled xenon to be recovered after the conclusion of the impurity measurement, only several milligrams of xenon are consumed for a measurements. The cold cathode gauge allows the total pressure at the RGA to be monitored and recorded. We have found it necessary to limit this pressure to less than  $1.0 \times 10^{-5}$  torr to prevent RGA saturation effects. The analysis system includes a port to allow for a sample bottle filled with calibration xenon to be attached. We prepared the calibration xenon gas with known concentrations of O<sub>2</sub>, N<sub>2</sub>, He, Ar, Kr, and CH<sub>4</sub>, by mixing known quantities of these impurities with a known amount of xenon. Except where otherwise noted, all of our impurity measurements are quantified by comparing to such calibration xenon. Once a xenon gas sample has been collected we analyze the sample for impurities as follows. The RGA is prepared to measure and record the partial pressures as a function of time of the relevant species, as shown in Figure 3. We typically monitor, in atomic mass units, 28 (N<sub>2</sub>), 32 (O<sub>2</sub>), 18 (H<sub>2</sub>O), 132 (Xe), 2 (H<sub>2</sub>), 4 (He), 15 (CH<sub>4</sub>), 40 (Ar), and 82, 84, 86 (Kr). We cool the cold trap with liquid nitrogen while it is still being pumped to ultra-high vacuum with the turbo pump, and then the xenon gas is admitted into the cold trap in three steps. First, the leak valve at the lowest flow setting is opened which admits xenon into the cold trap at a very small flow rate, below  $10^{-4}$  standard liters per minute (SLPM). Xenon ice forms in the cold trap and the analysis system is purged of residual trace impurities by

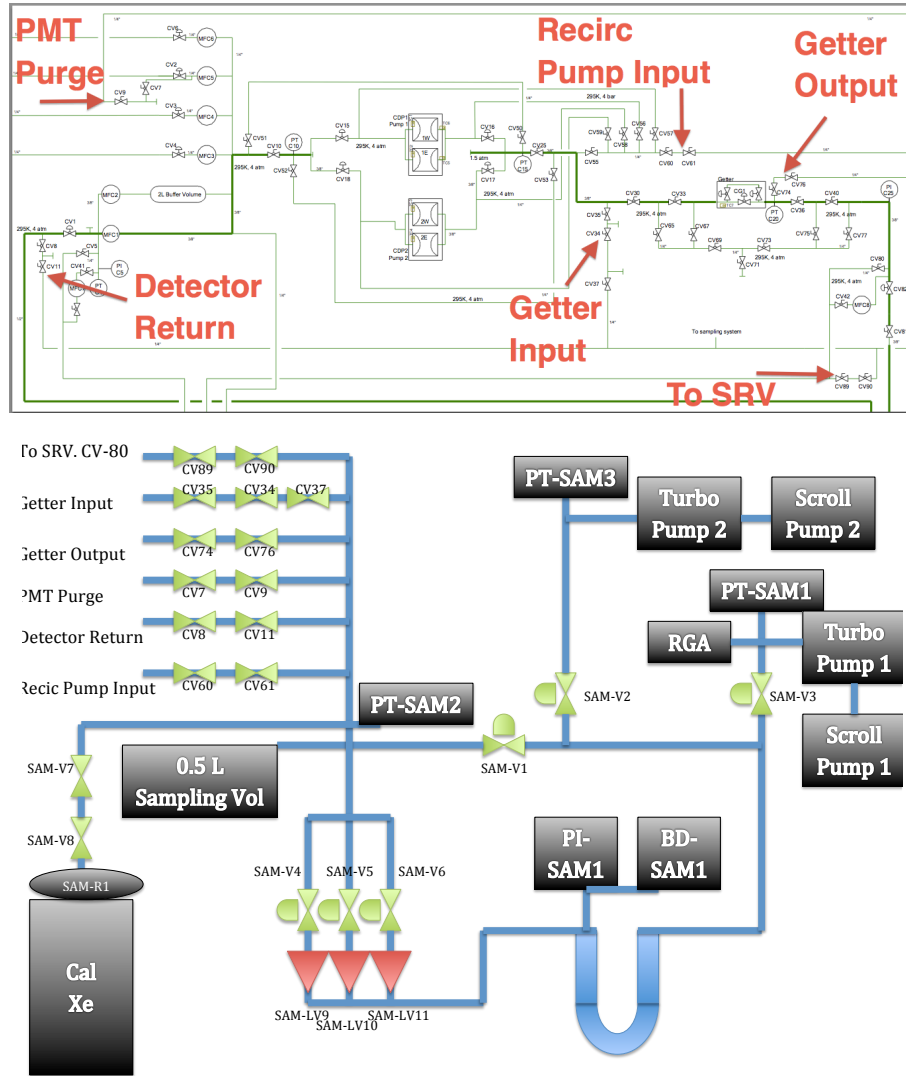


Figure 2: Top: Xenon sampling ports plumbed into the LUX circulation and handling system. Bottom: A schematic of the purity analysis system, the total sampling volume is 0.80 liters. Typical sample pressures are between 1 and 1.5 atmospheres.

the flowing gas. We wait ten minutes for the backgrounds to stabilize before proceeding. Second, we open the leak valve to a larger flow rate, usually corresponding to an flow rate of  $\sim 0.2$  SLPM. This coarse measurement is performed to ensure no  $N_2$  from an air leak or other impurity is present at amounts that could saturate the RGA signal, if the xenon is deemed pure then the system moves

on to the next higher flow stage for the  $1 \cdot 10^{-12}$  g/g Kr measurement. Third, the main leak valve is opened corresponding to flow rates of  $\sim 2$  SLPM gaining another factor of 10x in sensitivity from the coarse measurement. The higher flow rate used in this step allows impurities to be observed with high sensitivity, typically one part-per-billion to one part-per-trillion or better depending on the impurity species. The xenon gas in the sample bottle is slowly depleted as it flows into the cold trap. This causes the sample bottle pressure, and the flow rate through the leak valve, to decrease in time while the measurement takes place (see Figure 3). After a measurement period of three minutes, the leak valve is closed, the cold trap is allowed to warm to room temperature, and the xenon in the cold trap and the sample bottle is collected in the recovery bottle with liquid nitrogen. After the sample one final RGA mass spectrum of the cold trap at room temperature to look for impurity species such as  $\text{H}_2\text{O}$ , solvents, or heavy hydrocarbons which would be trapped in the cold trap at liquid nitrogen temperature. Note that to achieve the best sensitivity, the highest possible flow rate should be used for each measurement, limited only by RGA saturation effects.

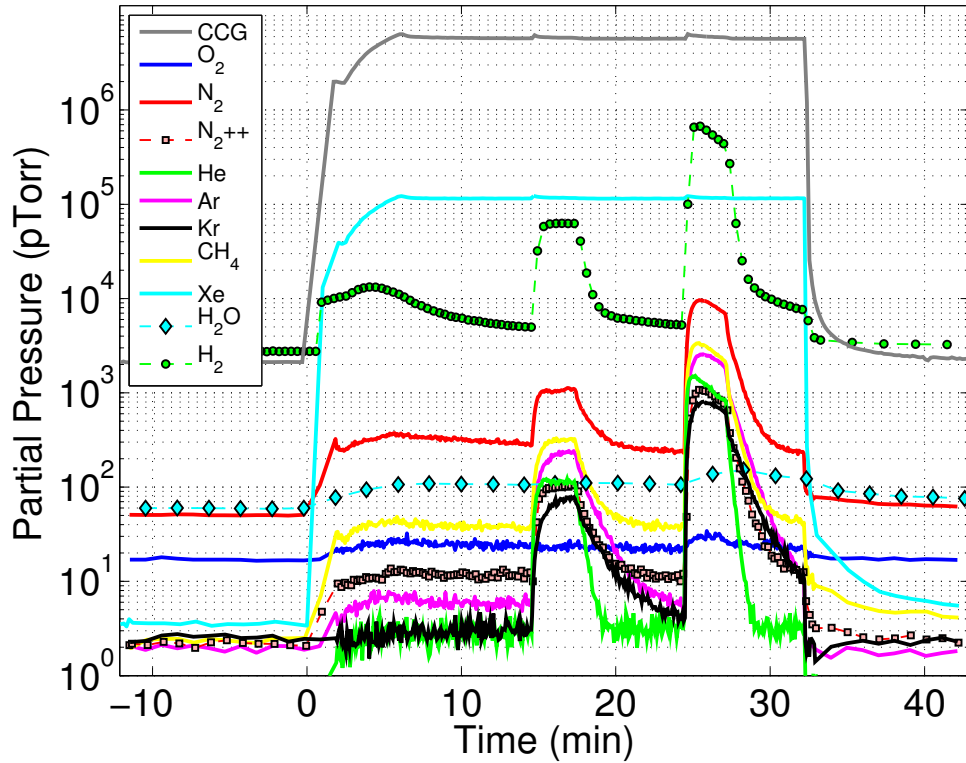


Figure 3: Typical RGA calibration measurement.

## 4. Purity Measurement

The measurements described in this article differ from that of Ref. [5] in that only a modest quantity of xenon gas is available for analysis in each sample, typically one standard liter, note that since xenon freezes in the coldtrap the sample is entirely recovered after the analysis. The sample pressure noticeably decreases during the measurement process, and so the flow rate through the leak valve between subsequent samples. We account for the changing flow rate as follows. We normalize the average integral of the RGA signal to the integral of the flow rate which have been found to be constant [refs, EXO ref] . Therefore the integrated RGA partial pressure of the impurity partial pressures, which are measured in the first three minutes of flow, also depend on the average flow rate as the number of impurities that pass by the RGA per unit time is proportional to the flow. As shown in Figure 4, we have measured this dependence for O<sub>2</sub>, Ar, and Kr, at our nominal flow setting, using xenon gas from our calibration cylinder. The data is well described by a simple exponential fit, and we use this fit to derive an expression for a sample bottle pressure correction factor (CF): (Talk about the gain correction from Xe PP, show plot) (Show formula for how the Figure of Merit is calculated. Normalized to Flow rate, XePP, and calibration signal.)

First, we correct for the flow rate dependence at our nominal leak valve setting by assuming the input-pressure correction. We take a systematic error of 10% on this correction. Secondly, we correct for a drift in the RGA gain by normalizing to the partial pressure of xenon which is the one true constant between all samples as it represents the xenon ice vapor pressure at 77 K in the coldtrap system.

To monitor for daily variations in the system response, including changes in the RGA gain, we periodically calibrate the analysis system using our cylinder of calibration xenon gas. Figure 5 shows the variations in the system response to various impurities for our calibration xenon over a period of six months. To account for these variations, we normalize the signal to the corresponding xenon vapor pressure, and we take the the deviation from the xenon vapor pressure in calibration measurements to be a systematic uncertainty. For most of our measurements, this is the dominating uncertainty in the impurity concentration measurement.