The isotope dependence of the vapor pressure has been studied for a number of noble elements, but little credible data exist for xenon. Indeed, early measurements gave results much smaller than theoretical expectations, and in the case of one study, a measurement based on distillation, the value obtained was zero dependence. Recently, an urgent need to understand this dependence has arisen from the interest in enriching $^{136}$Xe for use in the search for neutrinoless double beta decay. Other interest has centered on the use of the triple point (TP) of xenon as a temperature standard. The isotopic dependence of TP properties has been reviewed by Tew.

The search for neutrinoless double beta decay is one of the most pressing problems in particle physics today. If observed, it (i) would allow the absolute mass of the neutrinos to be established, (ii) would identify that neutrinos are Majorana particles, (iii) would help to understand the origin of neutrino mass, and (iv) might help to explain the dominance of matter over antimatter in the Universe. $^{136}$Xe is a very attractive target for such studies because it can be made largely free of radioactive contamination, it can be made into detectors of high sensitivity, and it offers excellent self-shielding against external backgrounds. Several groups are constructing or proposing xenon-based detectors for such studies. The dominant cost of such proposals comes from the high cost of xenon and the need for isotopic enrichment to limit the total xenon volume used. Current proposals are aimed at a sensitivity to probe the lifetimes predicted if neutrinos have an inverted hierarchy. Three separate groups have recently shown results that favor the normal hierarchy. It is important then that concepts for detectors that can provide the sensitivity required to reach the normal hierarchy be developed. For $^{136}$Xe, the required lifetime sensitivity is about $10^{30}$ years, implying a need for an exposure of order 1 kt·yr. Such a detector might employ 50 tonnes of this nuclide with a 20 year exposure to reach the required sensitivity, which will be prohibitively expensive unless more economical ways of enriching the xenon can be found. At present, the only technique available is centrifuge separation and the number of providers is very limited. Distillation is a possible alternate method for large-scale isotopic enrichment, but the absence of credible data on the isotopic variation of the vapor pressures in xenon precludes any meaningful feasibility study of this alternative.

The measurement of the vapor pressure differences for various substances has usually been carried out either by studying the separation of the isotopes under distillation or by...
directly measuring the vapor pressure differences of the isotopes. A comprehensive review of the early experimental data is given by Jancso and Van Hook,\(^6\) while a review of the current available data and the theoretical treatment is given by Tew.\(^6\) The direct measure of the vapor pressure is normally regarded as the most reliable method, and one usually quantifies the vapor-pressure isotope effect (VPIE) by the ratio \(\ln \left( \frac{P_i}{P_h} \right)\), where \(P_i\) and \(P_h\) are the vapor pressures of a light isotope \((l)\) and a heavy \((h)\) isotope. When the vapor pressure differences are as small as expected for xenon, the measurement requires extremely high purity control and temperature control and is understood that isotopic effects on phase equilibria can be better carried out by analyzing the fractionation factor \(\alpha_{l-h} = (n_l/n_h)/(c_l/c_h)\), where \(n\) and \(c\) denote the mole fraction of the isotope \(i = l, h\) in the vapor and condensed phases, respectively. The fractionation factor is nearly equivalent to the reduced partition function ratio for the condensed phase \((f_l)\) and ideal vapor phases \((f_h)\), and thus the following relations hold for noble elements:\(^6\)

\[
\text{VPIE} := \ln \left( \frac{P_i}{P_h} \right) \approx \ln \left( \frac{f_l}{f_h} \right) \approx \ln (\alpha_{l-h})
\]

(1)

The most recent results for argon, krypton, and xenon are summarized in Tables 1 and 2 for measurements and expectations near their TP and boiling point (BP) temperatures, respectively.

### Table 1. Experimental and Theoretical Values Reported for VPIE of Ar and Kr Closest to Their TP Temperature\(^a\)

| Fluid | Method | \(\Delta m\) | \(T\) (K) | \(10^3\) VPIE | Reference |
|-------|--------|--------------|----------|----------------|-----------|
| Ar    | DP     | 83.806       | 63.0 (6) | 17             |           |
|       | D      | 84.20        | 6.15 (24) | 18             |           |
|       | DP     | 83.81        | 6.50 (1)  | 19             |           |
|       | Theor. | 83.806       | 6.45 (32) | 3              |           |
|       | Theor. | 83.806       | 6.40      | 4              |           |
| Kr    | DP     | 115.778      | 1.05 (2) | 20             |           |
|       | Theor. | 115.775      | 0.88 (4)  | 3              |           |
|       | Theor. | 115.775      | 0.814     | 4              |           |

*For the experimental results, the technique (DP, differential pressure; or D, distillation) is indicated.*

### Table 2. Experimental and Theoretical Values Reported for VPIE of Ar, Kr, and Xe Closest to Their BP Temperature\(^a\)

| Fluid | Method | \(\Delta m\) | \(T\) (K) | \(10^3\) VPIE | \(<\) AMU | Reference |
|-------|--------|--------------|----------|----------------|---------|-----------|
| Ar    | DP     | 87.360       | 5.73     | 1.4325         | 17      |           |
|       | D      | 87.45        | 5.50 (8) | 1.38 (2)       | 18      |           |
|       | DP     | 87.22        | 5.940 (2) | 1.485 (1) | 19      |           |
|       | Theor. | 87.302       | 5.82 (31) | 1.46 (8) | 3       |           |
|       | Theor. | 87.302       | 5.78      | 1.445          | 4       |           |
| Kr    | DP     | 119.97       | 0.81     | 0.202          | 20      |           |
|       | Theor. | 119.93       | 0.80 (4) | 0.20 (1)      | 3       |           |
|       | Theor. | 119.93       | 0.75      | 0.188          | 4       |           |
| Xe    | D      | 130–136      | 165.05   | 0.1            | 2       |           |
|       | DP     | 132–136      | <0.025   | <0.006         | 1       |           |
|       | Theor. | 130–136      | 0.292 (15) | 0.073 (4) | 3       |           |
|       | Theor. | 132–136      | 0.185    | 0.046          | 4       |           |

*For the experimental results, the technique (DP, differential pressure; or D, distillation) is indicated.*
motivated in large measure by the fact that this yields directly the quantity needed for the design of a practical production facility, but also because it avoids the difficulties of a differential pressure study noted above. The cryogenic distillation column is calibrated using argon and krypton, for which there are measured values for the VPIE. All measurements were made at a pressure of 101.28 kPa, with variations below 0.5%, and the same molar vapor flow rate was used in each case. The VPIE for xenon can be inferred from these data, and our results are in agreement with the theoretical values, in contrast with the older measurements.

THE MEASUREMENT

An outline of the still is displayed in Figure 1. The apparatus consisted of a cryogenic still with a 1.8 m tall reflux column of 25 mm inner diameter. This inner column is wrapped in a multilayer insulation and placed in a vacuum jacket. The column was filled with a random packing, PROPAK 0.16” (by Cannon Industries) made of electropolished stainless steel. A cold head at the top was cooled by an Oxford Cryogenics refrigeration system operated at a fixed cooling rate. A heater in the re-boiler was controlled to maintain a fixed pressure in the vapor phase. The pressure was monitored using an MKS Baratron with an accuracy of 0.12%. Typically, the reading was stable to within 0.1% through the runs. Heat was supplied to the re-boiler in a pair of side arms to ensure mixing of the liquid volume. The total volume of liquid in the re-boiler, measured using a differential pressure sensor connected between the top and the bottom of the boiler, was about 300 cm$^3$. The vapor flow in the column was determined from the known latent heat of vaporization and the measured electrical power supplied to the boiler. External heat leaks were 2% of the applied heater power as determined by measuring the evaporation rate in the absence of applied power. The flow was typically 0.25 g/s for krypton and similar molar flows for the other gases. These low flows are about 4% of the flooding flow for this column. The gas velocity in the column is about 5 cm/s, while the pressure drop across the column was about 110 Pa or about 0.1% of the total pressure. Under these conditions, variations in the temperature or VPIE along the column can be neglected.

A mass spectrometer was used to measure the relative abundances of isotopes at either the top of the column or the bottom. Sampling was done through capillary tubes with an inner diameter of 0.5 mm, and a procedure described below for purging the capillaries prior to each measurement was followed. The mass spectrometer consisted of an EXTREL quadrupole with a 19 mm bore and a mass resolution of 1:2000. Ions were detected using a channel multiplier. A current sensitive preamplifier was used to minimize non-linearity due to pileup seen when using pulse counting electronics.

The mass calibration of the spectrometer was done by comparing the observed peaks with those expected for the gas introduced. There are small non-linearities, and these are included but precision mass measurements are not required. Of more interest are the intensity non-linearities with either mass or intensity. These are cancelled by the method of ratios used in the analysis explained below.

For an individual measurement, the detected current was sampled while the quadrupole scanned the selected mass range. Each scan took approximately 1 s, and data from a thousand scans were averaged to produce the mass spectrum. Data were taken in four run periods as listed in Table 3. While in 2018 the sampling between the top and the bottom capillaries were alternated in consecutive measurements, in

| run | fluid | date       |
|-----|-------|------------|
| 1   | Ar    | Oct. 5–12, 2018 |
| 2   | Kr    | Nov. 6–7, 2018  |
| 3   | Kr    | Apr. 17–19, 2019 |
| 4   | Xe    | Apr. 23–26, 2019 |
Five repetitions with the same end of column was performed before switch reading to the other end. Typical spectra from one such measurement for each of the three gases are shown in Figure 2. It can be seen that the isotopic peaks are well resolved and even the least abundant isotopes are clearly seen. The mass range sampled for each isotope was as shown in this figure. However, broad mass range spectra were also recorded for each gas to test for impurities. For the argon data, we see small peaks at mass 32 ($\text{O}_2$) and 44 ($\text{CO}_2$). No background peaks were identified at higher masses for any of the gases used. Thus, the possibility that any of the measurements are contaminated by molecules is small.

In order to extract the VPIE for xenon from the distillation separation data, one needs to calibrate the still and understand the mass transfer properties. It was decided to do this by taking initial data with argon and krypton for which the VPIE is known, and to determine from these results, a method to extrapolate to xenon. The initial data suggested that the best method was to assume a constant value for the height of the equivalent theoretical plate (HETP). The HETP is the height of packing required to achieve a theoretical stage, in which the liquid and vapor phases establish an equilibrium with each other. The HETP is frequently used to characterize the mass-transfer capability of packings.26 This decision was made before the xenon data were taken, and thus the resulting analysis can be considered blind.

![Mass spectra for (a) argon, (b) krypton, and (c) xenon.](data:image/png;base64,iVBORw0KGgoAAAANSUhEUgAAAAEAAAABCAYAAAAfS9krAAAACTpaaaaAAACCAIAAADApAAAABJRU5ErkJggg)

Figure 2. Mass spectra for (a) argon, (b) krypton, and (c) xenon.

![Average fractional difference between the top and bottom of the column, $\delta_i$, for each isotope of (a) argon, (b) krypton, and (c) xenon.](data:image/png;base64,iVBORw0KGgoAAAANSUhEUgAAAAEAAAABCAYAAAAfS9krAAAACTpaaaaAAACCAIAAADApAAAABJRU5ErkJggg)

Figure 3. Average fractional difference between the top and bottom of the column, $\delta_i$, for each isotope of (a) argon, (b) krypton, and (c) xenon. The data can be described by a linear dependence on the isotopic mass $m_i$, and results from the fit $\delta_i = p_1 m_i + p_0$ are presented for each fluid.

### DATA ANALYSIS AND RESULTS

In each measurement, a baseline for the current is estimated by averaging the last data points of the mass spectra, ranging approximately two atomic masses. This baseline is subtracted from all data points, and the associated standard deviation is assigned as a systematic error to all points. The mass range considered in the analysis is reduced to approximately one unit past the expected lightest and heaviest isotope of the target element, removing trace elements such as $\text{O}_2$ and $\text{CO}_2$ from the argon data. The resulting spectrum is then normalized to unit area.

A custom algorithm was implemented to identify all mass peaks and visually inspected for each spectrum. It associates data points to atomic masses, hence providing the peak shape measured for each mass. In the data analysis, to account for overlaps between adjacent peak tails and rises, as seen in the region from 82 u to 84 u in Figure 2b, the peak shape associated to the largest isotopic mass is used as a probability density function (PDF). This PDF is assumed the same for all masses in the same spectrum and summed to form a model for the full spectrum ($S$):

$$S(m) = \sum_{i \in I} \tilde{f}_i \cdot \text{PDF}(m - m_i)$$

where $m_i$ is the uncalibrated atomic mass of isotope $i$, $\tilde{f}_i$ is the fractional contribution to the spectrum from the same isotope,
and $f$ is the set of stable isotopes of the target element. A $\chi^2$-fit is performed to minimize the difference between data and $S(m)$, where $f_i$ and $m_i$ are the floating parameters, with initial values set to those expected from the natural gas composition. The fitted fractions are finally corrected by the sum of all fractions, $f_i = \bar{f}_i / (\sum \bar{f}_i)$.

The difference between the top ($f_i_{\text{top}}$) and bottom ($f_i_{\text{bot}}$) measured isotopic fractions was observed to evolve during approximately 15 h until reaching a period of stable running. Only measurements performed with a stable system are considered in this analysis. The fractional difference ($\delta_i$) between consecutive top-bottom averages is calculated by

$$\delta_i = \frac{f_i_{\text{top}} - f_i_{\text{bot}}}{(f_i_{\text{top}} + f_i_{\text{bot}})/2} \times 100$$

(4)

Imperfections in the measurements arising from the analysis procedure, such as the choice of the PDF shape, as well as from the system conditions during data taking, such as the quadrupole tuning, are accounted for as systematic errors. Figure 3 shows $\delta_i$ as a function of the atomic mass for (a) argon, (b) krypton, and (c) xenon, where it is observed that the data points can be described by a linear relationship.

In order to extract the VPIE for xenon from the distillation separation data, one needs to calibrate the still and understand the mass transfer properties. In principle, one can do this using computer codes developed for the simulation of distillation systems. However, such codes as ChemSep$^{27}$ rely on models for the mass transfer that are developed to fit the world data on distillation. Such datasets are strongly weighted by distillation of petrochemicals and alcohols at high temperatures and different fits give rather different extrapolations for noble fluids at cryogenic systems. Instead, for this report, the still was effectively calibrated using argon. This calibration was then tested using krypton, for which VPIE data are also available, and then used to interpret the xenon data.

In order to extract the physically interesting quantities such as the VPIE from the data, it is necessary to account in some way for the still properties. Following Wagner et al.$^{26}$ for conditions of 100% reflux, the height of a vapor transfer unit is the same as the HETP, and when the liquid holdup is a small fraction of the void volume in the packing, the dominant term is $\propto \sqrt{u_V/D_{Vf}}$, where $u_V$ is the vapor velocity through the packing and $D_{Vf}$ is the vapor phase self-diffusion coefficient. This ratio is expected to be very similar for the noble gases, which can be checked for argon and krypton using the available data.

Under conditions of 100% reflux, the minimum number of stages ($N$) required to achieve the measured fractional differences in Figure 3 can be calculated using the Fenske equation$^{28}$

$$N = \frac{\ln (\delta_{i-\text{top}})}{\ln (\alpha_{i-\text{bot}})}$$

(5)

where $\delta_{i-\text{top}} = \frac{(1 + \delta_i / 200) / (1 - \delta_i / 200)}{(1 + \delta_i / 200) / (1 - \delta_i / 200)}$. For argon, we use eq 2 to calculate $10^3 \ln (\alpha_{36 - 40}) = 5.82 \pm 0.31$ at BP (see Table 2) along with the still data $\ln (\delta_{36 - 40}) = 0.248 \pm 0.06$ and obtain $N = 42.5 \pm 2.5$ stages, which is equivalent to HETP = 4.23 ± 0.25 cm. This compares with the manufacturer’s value of about 3 cm for hydrocarbons at much higher temperatures. If we assume the same number of stages or HETP for krypton, we obtain a VPIE of $10^3 \ln (\alpha_{36 - 40}) = 0.82 \pm 0.13$, in good agreement with values in Table 2, also shown in Figure 4a.

Figure 4. VPIE between fluids isotopes as a function of (a) $T$K and (b) $\Delta M = f - h$. The lines represent values from numerical simulations$^3$ for argon (dashed), krypton (dot-dashed), and xenon (solid), while the filled regions are the corresponding expectations from eq 2$^3$ accounting for uncertainties in the parameters. Open data points are those presented in Tables 1 and 2 for argon (diamonds), krypton (squares), and xenon (circle) representing the average for the same temperature, while solid points with error bars are measurements from this work.

This supports the procedure proposed to measure the VPIE for xenon and in this case yields $10^3 \ln (\alpha_{36 - 40}) = 0.26 \pm 0.04$. As depicted in Figure 4a, this value is consistent with theoretical predictions but in disagreement with previous measurements.

Figure 4b presents a comparison between our measurements and values from eq 2 for all stable isotopes as a function of $\Delta M = I - h$, using the heaviest mass as the reference in all cases. The value for $^{36}\text{Ar}^{40}\text{Ar}$ is used for calibration, but the measurement for $^{38}\text{Ar}^{40}\text{Ar}$ is reported here for the first time. We observe agreement within errors between data and prediction for all isotopes of krypton and xenon. The data is summarized in Table 4.

## Discusson and Conclusions

A possible background to any measurement relying on mass spectroscopy could come from molecules having the same mass as one of the isotopes under measurement. In the present experiment, several measures were taken to guard against this. First, research grade gases (99.999% purity) were used and the gases passed through a getter expected to remove non-noble impurities to below ppb levels. Second, for each gas, a scan was carried out over a wide mass range and no lines were seen above the noise level, except in the case of the argon data where we see traces of $\text{O}_2$ at mass 32 and $\text{CO}_2$ at mass 44. Finally, if there were a contaminant contributing, it would likely have a vapor pressure quite different from that of the noble fluid and thus be completely separated by the column. This would give rise to strong departures from the linear relation seen in the separation data.

The VPIE for xenon is extremely small, and measurements are difficult. In this experiment, the use of a tall still, a modern mass spectrometer, and a technique that used a double ratio analysis technique to control systematic errors allowed the effect to be detected. In order to improve the precision, a taller still, eight times higher than in the present experiment, will be used. This device, which is under construction, should produce...
Table 4. All the Measurements from This Work for the Three Fluids in Terms of Atomic Mass (A), Fractional Difference $\delta_A$, and VPIE

| fluid | A | $\delta_A$ (%) | $p_1$ (%)/u | $\Delta m$ (u) | $10^3$ VPIE |
|-------|---|----------------|-------------|---------------|-------------|
| Ar    | 36| 24.57 (0.61)   | -4          | 5.82 (0.31)   |
|       | 38| 10.8 (1.0)     | -2          | 2.57 (0.28)   |
|       | 40| -0.063 (0.003) | 0           | 0             |
| Kr    | 80| 3.5 (0.4)      | -4          | 0.82 (0.13)   |
|       | 82| 1.5 (0.8)      | -2          | 0.35 (0.21)   |
|       | 83| 0.60 (0.10)    | -0.89 (0.08)| 0.15 (0.08)   |
|       | 84| -0.04 (0.33)   | 0           | 0             |
|       | 86| -1.71 (0.35)   | 2           | -0.39 (0.12)  |
| Xe    | 124| 2.4 (2.2)     | -12         | 0.7 (0.5)     |
|       | 126| 2.6 (2.1)     | -10         | 0.8 (0.5)     |
|       | 128| 0.59 (0.19)   | -8          | 0.31 (0.06)   |
|       | 129| 0.43 (0.13)   | -7          | 0.27 (0.06)   |
|       | 130| 0.36 (0.10)   | -0.179 (0.018)| -6   | 0.26 (0.04) |
|       | 131| 0.08 (0.06)   | -5           | 0.19 (0.04)   |
|       | 132| -0.10 (0.07)  | -4           | 0.15 (0.04)   |
|       | 134| -0.46 (0.08)  | -2           | 0.07 (0.04)   |
|       | 136| -0.74 (0.14)  | 0            | 0             |

Effects eight times larger but with the same absolute measurement accuracy. The design and construction of a production still would be a very complex engineering undertaking as the still would probably be of order of 1 km in height. Such a device might be supported inside a mining shaft. A precision measure of the VPIE would reduce the uncertainties in such an undertaking.

This work has demonstrated that isotopes of xenon can be separated by distillation and has resolved a long-standing anomaly in the xenon VPIE data. The measurement present here agrees well with the theoretical predictions in previous studies. The data also demonstrate, for the first time, the linear dependence of the VPIE across a range of isotopes, and this is consistent with the theoretical predictions.

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**Notes**

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