Isotopic Dependence of Vapour Pressure in Xenon

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Introduction and Motivation

The search for neutrinoless double beta (0νββ) decay is one of the most pressing problems in particle physics today. If observed, it would:

i. identify that neutrinos are their own antiparticles, *i.e.* Majorana particles;
ii. shed light on the absolute mass scale of the neutrinos;
iii. help explain the dominance of matter over antimatter in the Universe.

^{136}Xe is an attractive nuclide to search for 0νββ. To probe the normal ordering of neutrino masses, ~1 meV, ~50 t of this isotope are required. In general, economic or background issues dictate isotopic enrichment.

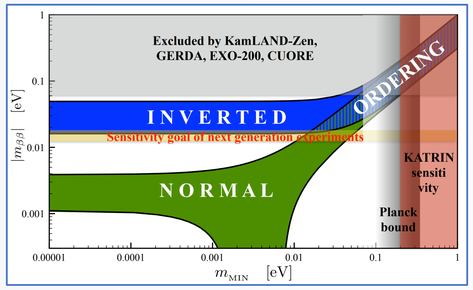


Figure 1. Effective Majorana mass vs. the smallest neutrino mass [1].

We are exploring the use of distillation for xenon enrichment. This poster reports the first credible measurement of the separability of xenon isotopes by distillation and hence the first measure of the xenon vapour pressure isotope effect (VPIE).

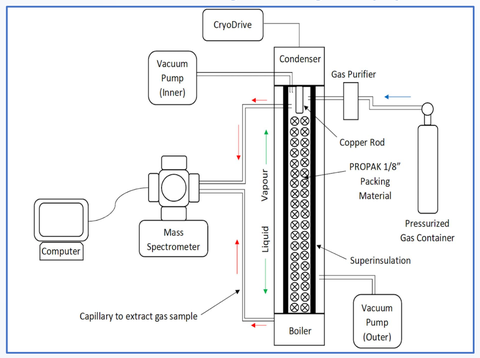
Experimental Methods

A 1.8m high cryogenic still has been built at Carleton University:

- 25 mm inner diameter vacuum insulated reflux column filled with PROPAK packing material [2];
- a condenser is placed at the top of the column and a re-boiler placed at the bottom;
- the still is operated at 100% reflux.

Figure 2. Experimental setup showing gas entering the top of the still.

All measurements were made at a pressure of 105 Pa. The vapour is sampled at the top and bottom of the packed bed using a large bore quadrupole mass spectrometer to determine the isotopic abundances. The ion detector in the spectrometer is operated in current mode to reduce the non-linearity in the response.



Data Analysis and Results

The spectrometer was sequenced to sample the top and then the bottom repeatedly, acquiring each mass sample over a period of approximately 20 minutes. Asymptotic values are reached after about 15 hours, and after this the continued sampling serves to improve the precision of the measurement.

Typical mass spectra for the three elements are shown in Figure 3. Each of the isotopes is cleanly separated. Shapes of the peaks are not Gaussian so to determine the relative peak intensities, the strongest clean line is used as a template and the spectrum is then fitted as a sum of the peaks plus a constant background. Peak intensities are then normalized to the total intensity. This method of taking ratios of ratios eliminates most of the systematic errors.

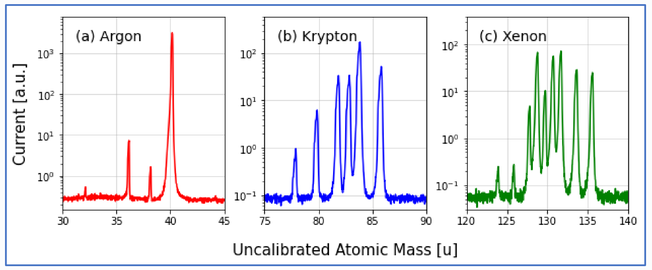


Figure 3. Mass spectra for argon, krypton and xenon.

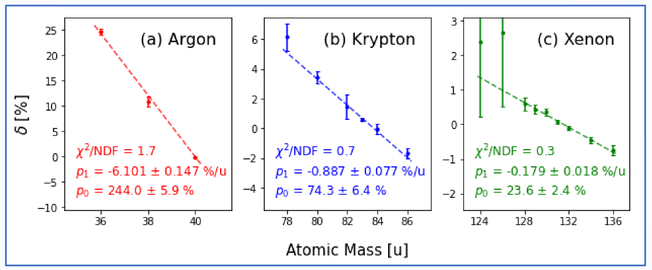


Figure 4. Fractional difference between the top and bottom of the column for each isotope.

In order to produce a measure of the VPIE one needs to calibrate the still. This is done by running first using argon and then krypton, for which measurements of the VPIE have been published [3], and then taking data for xenon.

As a first approximation one can simply assume the height of a theoretical plate (HETP) is the same for each element [4]. Then the VPIE's should be proportional to the slopes of the curves in Figure 4. This approximation gives very good agreement with previous data when comparing argon and krypton.

Discussion and Conclusion

The minimum number of stages (*N*) required to achieve the measured fractional differences can be calculated using the Fenske equation [5] and the expected VPIE for argon [3,6,7], then validated with krypton - Figure 5 (a):

- N = 42.5 ± 2.5 stages
- HETP = 4.23 ± 0.25 cm

(vs. manufacturer's HETP of 3 cm for hydrocarbons at higher temperatures)

Due to the method of this measurement, a comparison of our results can be made not only for existing values of the vapour pressure differences between ^{36}Ar/^{40}Ar, ^{80}Kr/^{84}Kr and ^{130}Xe/^{136}Xe - Figure 5 (a), but also against the expectation values for all stable isotopes - Figure 5 (b).

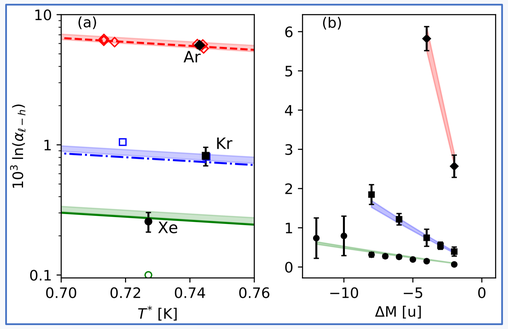


Figure 5. VPIE between fluids' isotopes as a function of (a) dimensionless temperature parameter and (b) ΔM, difference between the heaviest isotope and lighter masses. The VPIE is $\ln \alpha_{l-h} \simeq \ln (p_l/p_h)$, where p_h and p_l are the vapour pressures of the heavy and light isotopes. The lines represent values from [7] for argon (dashed), krypton (dot-dashed) and xenon (solid), while the filled regions are the corresponding expectations from [6]. Open data points are those available in literature [8] 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.

These results will allow a study of the feasibility of distillation as a technique for isotopic enrichment of xenon and resolves a long-standing anomaly in the xenon VPIE data. It demonstrates for the first time the linear dependence of the VPIE across a range of isotopes and can be relevant to the use of the triple point of xenon as a temperature standard.

References

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