

## Mobility of thorium ions in liquid xenon

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### Abstract

We present a measurement of the <sup>226</sup>Th ion mobility in LXe at 163.0 K and 0.9 bar. The result obtained,  $0.240 \pm 0.011$  (stat)  $\pm 0.011$  (syst) cm<sup>2</sup>/(kV-s), is compared with a popular model of ion transport.

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### 1. Introduction

Free ions in the liquefied noble gases at low concentrations represent a theoretically straightforward condensed matter system. This is largely because of the liquid's atomic nature, implying relatively few microscopic internal degrees of freedom [1]. Despite this fact, the experimental study of these systems has long been confounded by the difficulty in reproducibly generating and transporting ions of known identity while controlling for the effects of impurity ions. Accordingly, there have been relatively few transport studies for ions in the liquefied noble gases other than LHe reported in the literature to date. Furthermore,

studies that deal only with molecular ions [1–5], are more common than those that deal with monatomic species [6,7]. There is therefore a clear need for additional data on monatomic ion transport particularly in the heavy noble gases. In the present work we consider the system of monatomic Th ions in LXe.

This research also has important applications to the physics of ionizing particle detectors, especially in the area of rare event searches that use a liquefied noble gas as the detection medium [8–10]. For these systems, the transport properties of Th and U ions are of concern because these elements in trace amounts represent a large source of background radioactivity. It is therefore desirable to understand how electric fields might be used to draw these ions out of the detection medium [10]. For LXe-based detectors, Cs and Ba ions are also interesting because their respective separation from the detection medium and subsequent identification can greatly enhance the

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sensitivity to solar neutrinos and to double-beta decay [8,9]. The motivation for the present study is work involving Ba ion retrieval from LXe as part of R&D for a large double-beta decay experiment [11].

In this paper, we report on the measurement of the mobility of  $^{226}\text{Th}$  ions in liquid Xe at 163.0 K and 0.9 bar pressure. We also compare our results and those of other workers with the theoretical calculations of Hilt et al. [2].

## 2. Experiment

Our system, schematically shown in Fig. 1, consists of a vacuum-insulated cell which we fill with about  $10\text{ cm}^3$  of LXe. The cell is cooled by a cold finger that forms a thermal link with a liquid nitrogen reservoir. The temperature is maintained at  $163.0 \pm 0.1\text{ K}$  by a resistive heater. The cell, built almost entirely out of UHV-compatible materials, is baked at 400 K for several days, after which it is pumped to a  $7.0 \times 10^{-8}$  mbar vacuum by means of two turbomolecular pumps. When filling the cell, we pass research-grade Xe

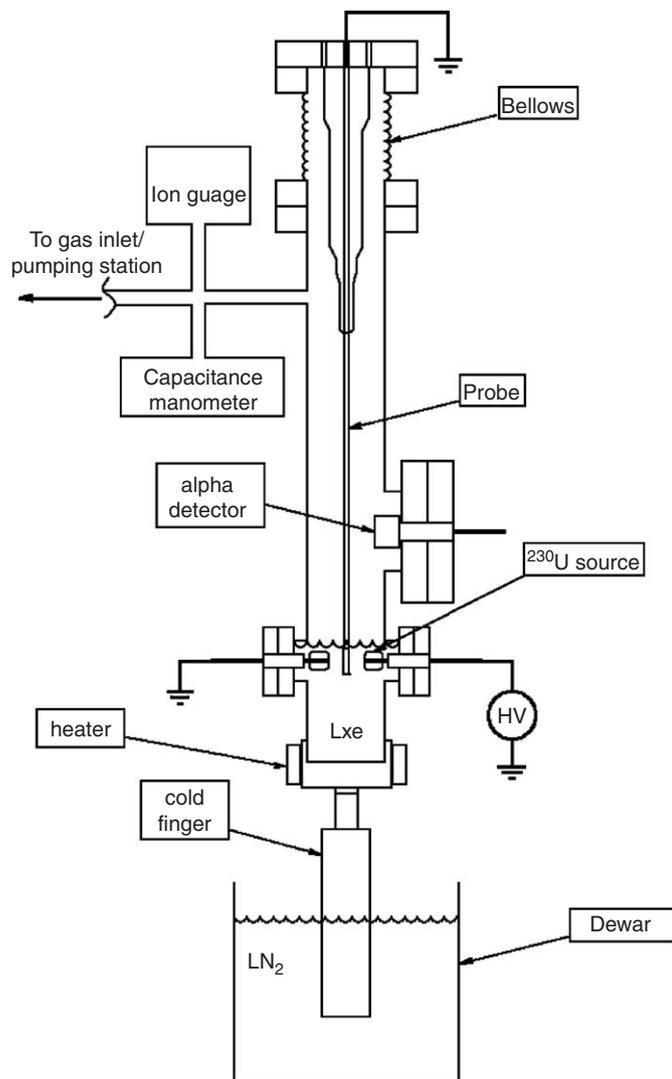


Fig. 1. Schematic of the probe cell.

(which, as supplied, is quoted as 99.999% pure) through a hot Zr getter [12], which is rated to remove reactive impurities to the  $< 1$  ppb level. These precautions are taken to minimize any effect of impurities that might influence ion transport characteristics.

Th ions are produced by a  $^{230}\text{U}$  source mounted inside the cell. The  $^{230}\text{U}$  was obtained as a decay product of  $^{230}\text{Pa}$ , in turn produced in the reaction  $^{232}\text{Th} + p \rightarrow ^{230}\text{Pa} + 3n$  on natural thorium at a cyclotron [13]. After irradiation, the  $^{230}\text{Pa}$  was left to  $\beta$ -decay for a few of its 17.4 day half lives, allowing some build-up of  $^{230}\text{U}$ , which was then chemically separated and electroplated onto a platinum disk. Fig. 2 shows the decay scheme for the final  $^{230}\text{U}$  source.

$^{226}\text{Th}$  ions are formed as a result of the 20.8 day  $\alpha$  decay of  $^{230}\text{U}$ . The average recoil energy of  $\sim 100\text{ keV}$  extracts  $^{226}\text{Th}$  atoms from the surface in a highly ionized state. As the  $^{226}\text{Th}$  ion enters the liquid it exchanges charge with the surrounding Xe atoms and reaches equilibrium in some final charge state. Although the energetics of the charge exchange in principle favor a final state containing  $\text{Xe}^+$  and  $\text{Th}^+$  [6], a large number of the emerging Th ions can also recombine with free ionization electrons that arise from the original  $\alpha$  decay [6]. This effect presents a major problem as it greatly suppresses the ion yield. We collect ions on a stainless steel probe which we lower into the liquid. To collect ions we place the probe at ground potential while applying a positive bias to the source electrode. The tip of the probe is fashioned into a  $5\text{ mm} \times 16\text{ mm}$ , 2 mm thick rectangular stainless steel plate, ensuring a spatially uniform electric field in the vicinity of the source. The uniformity of the field is verified by constructing a computer model of the electrostatics with the MAXWELL software package [14].

The ions that land on the probe are detected by retracting the probe vertically to a counting station,

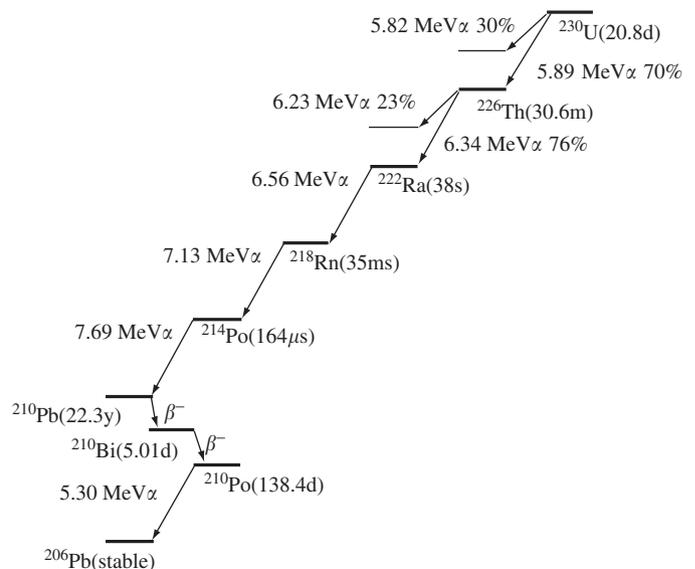


Fig. 2. Decay scheme for  $^{230}\text{U}$ . Decays with branching ratios less than 5% are not shown.

located above the liquid surface. It consists of a 50 mm<sup>2</sup> active area ion-implanted Si detector [15] that is read out using standard pre- and post-amplification electronics. The ion yield is determined by counting the  $\alpha$  particles emitted in the 30.6 min <sup>226</sup>Th decay, along with those produced by the subsequent  $\alpha$  decays of <sup>222</sup>Ra, <sup>218</sup>Rn, and <sup>214</sup>Po (see Fig. 2).

It should be pointed out that in addition to the <sup>226</sup>Th ions, the <sup>230</sup>U source also produces ions arising from the other  $\alpha$  decays in the chain. These can also drift to the probe and be detected at the counting station. However, because of their considerably shorter lifetimes, they present a negligible background to the <sup>226</sup>Th signal provided some short time elapses between the probe extraction and the counting. In addition, the low angular acceptance of our  $\alpha$  particle detection system ( $\sim 5\%$ ) ensures that the probability of detecting more than one  $\alpha$  particle from the 4- $\alpha$  decay sequence of a given Th atom is negligibly small.

We measure the ion mobility in liquid Xe by determining the ion transit time from source to probe at a given probe distance and electric field. This is achieved by biasing the source with an alternating potential produced by a computer driving a high voltage amplifier [16]. The system is set to produce a square wave switching between  $V_{\text{drift}}$  and  $V_{\text{sup}}$  with respect to the probe, where  $V_{\text{drift}}$  can be varied from 1.0 to 4.0 kV, and  $V_{\text{sup}}$  is fixed at  $-4.0$  kV.  $V_{\text{drift}}$  is set for a time  $t_{\text{drift}} \approx 1$  s, very long compared to the  $\sim 20$   $\mu$ s switching time of the high voltage system. During each full cycle, the voltage is reversed for a time  $t_{\text{sup}}$ . When this occurs, the ions that do not reach the probe during  $t_{\text{drift}}$  are drawn back toward the source. This occurs when  $t_{\text{drift}}$  is too short for the ions to complete the journey to the probe. We maximize our total ion integration time by having  $t_{\text{sup}} \leq t_{\text{drift}}$  and  $|V_{\text{drift}}| < |V_{\text{sup}}|$  [6]. During each data taking run,  $V_{\text{drift}}$  is set to a particular value and the field is switched for 1800 s. The constant duration of collection runs ensures that the amount of ion loss due to radioactive decay during collection is uniform. We then measure the ion yield at the counting station over a period of 2 h to allow sufficient time for the collected <sup>226</sup>Th ions to decay before the next run. Data are taken at four different electric fields, using four source-probe voltages and two source-probe separation distances.

We determine the background by performing 2-h counts between data runs in which no ions are collected. In these runs the source-probe voltage remains reverse-biased. The observed rate during the background runs is averaged over all runs, giving the mean background rate and an associated statistical error. The resulting value of  $3.16 \pm 0.73$  counts per 2-h run is subtracted from the number of counts observed at each  $t_{\text{drift}}$ . When ions are collected, the typical rate is  $\sim 30$  counts per run. Sources of background include cosmic rays and neutral Th atoms that evaporate from the surface of the liquid and diffuse through the gas.

For the purpose of characterizing the system a test in Xe gas was also done. The cell was filled with 0.2 bar of room-

temperature Xe gas and the probe was alternately brought close to the <sup>230</sup>U source and raised to the counting station, where ions were respectively collected and counted. This was repeated many times over several hours to build up counting statistics. The electric field between source and probe for ion collection was roughly  $\sim 200$  V/cm for this test. Shown in Fig. 3 are the  $\alpha$  spectrum and time histogram for the collected Th and its decay daughters. For comparison, the <sup>230</sup>U spectrum, as measured in vacuum at LLNL [17], is also shown. In performing this test it was found that the ion yield the yield in Xe gas remained constant when the source-probe electric field was varied, in contrast to the situation in LXe, where increasing the electric field was accompanied by a slight increase in the ion yield. Presumably, the mechanism for the behavior in LXe is that the ions recombine with free ionization electrons, thereby neutralizing and suppressing the ion yield. This effect is offset by increasing the source-probe electric field, causing free electrons to be swept away before they can recombine. By contrast, in gaseous Xe the ionization density is much smaller so these effects are largely absent. For the range of electric fields used in these experiments the ion yield in LXe was found to be on average  $\sim 200$  times smaller than the corresponding yield in gaseous Xe. From this we conclude that, under the present conditions, ion recombination with free ionization electrons is roughly  $\sim 200$  times more efficient in LXe than it is in gaseous Xe.

### 3. Results

Shown in Fig. 4 are plots of the ion yield vs.  $t_{\text{drift}}$ . For each data point, the ion yield is determined by dividing the number of detected  $\alpha$  particle counts by the number of cycles, giving the ion yield per cycle. The average background in these units is  $\leq 0.002$ , and the statistical error bounds on each data point were computed using the Feldman–Cousins prescription [19]. We find that the ion yield is consistent with zero (after background subtraction) when  $t_{\text{drift}}$  is short, while at longer times, the ion yield per cycle increases linearly with  $t_{\text{drift}}$ . The abscissa of the join point between these two behaviors is the ion transit time. We fit the ion yield data to the following two-parameter piecewise linear function:

$$Y = 0 \quad t_{\text{drift}} < t_{\text{tr}}$$

$$Y = m(t_{\text{drift}} - t_{\text{tr}}) \quad t_{\text{drift}} \geq t_{\text{tr}}$$

where  $Y$  is the ion yield per cycle,  $m$  is the slope (ion yield per cycle per unit  $t_{\text{drift}}$ ), and  $t_{\text{tr}}$  is the ion transit time. The value of  $t_{\text{tr}}$  yielded by the fit is used to calculate the ion drift velocity. To perform the fits, and to calculate the statistical error on  $t_{\text{tr}}$  in each case, we use an algorithm due to D.J. Hudson [20] that is designed specifically to handle the difficulties that arise when performing a piecewise fit. One peculiarity of such a fit is that the error bounds are almost

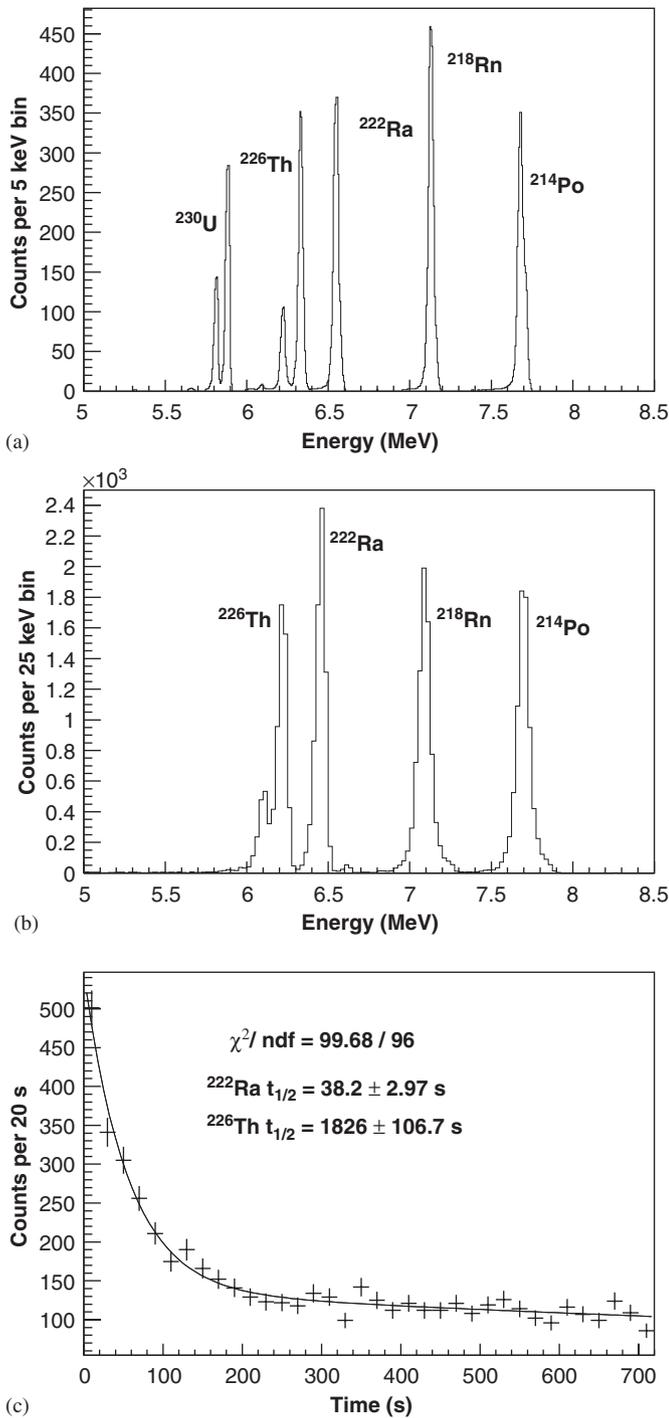


Fig. 3. (a)  $\alpha$  spectrum of the  $^{230}\text{U}$  source, taken under vacuum [17]. The  $^{230}\text{U}$  peaks are apparent at 5.82 and 5.89 MeV, while  $^{226}\text{Th}$  is identified by the peaks at 6.23 and 6.34 MeV.  $^{222}\text{Ra}$ ,  $^{218}\text{Rn}$ , and  $^{214}\text{Po}$  correspond to the peaks at 6.56, 7.13, and 7.69 MeV, respectively. (b)  $\alpha$ -decay spectrum for ions that are collected on the probe in 0.2 bar of ambient Xe gas. Here, the peaks are broadened and shifted to lower energy as a result of losses in the Xe gas. (c)  $\alpha$ -decay time histogram for the sum over all counts in the alpha spectrum shown in (b). The best fit is the superposition of two exponentials with half-lives of  $1826 \pm 106.7$  s and  $38.2 \pm 2.97$  s. These match the  $^{226}\text{Th}$  and  $^{222}\text{Ra}$  half-lives, whose published values are 1830 and 38 s, respectively [18].

always asymmetric [20]. The results of the fits are summarized in Table 1.

Fig. 5 shows a plot of the computed ion drift velocity as a function of electric field. The data are consistent with the linear relation  $v = \mu E$ , where  $v$  is the ion drift speed,  $\mu$  is the mobility, and  $E$  is the electric field. From a fit to the data in Fig. 5 we find an ion mobility  $\mu = 0.240 \pm 0.011$  cm<sup>2</sup>/(kV-s). This fit is obtained by forcing the straight line to go through the origin and it yields  $\chi^2/\text{ndf} = 2.02/3$ . A fit not constrained to go through the origin results in  $\mu = 0.268 \pm 0.031$  cm<sup>2</sup>/(kV-s), a  $y$ -intercept of  $-0.095 \pm 0.088$  cm/s, and  $\chi^2/\text{ndf} = 1.14/3$ . This is fully consistent with the original result.

The mobility and its statistical error are determined largely by the data taken at the lowest electric field. This point is more precise for two reasons. First, the ion transit time at this electric field is the longest of the four datasets, meaning that the fractional error on the velocity is the smallest. Secondly, in this dataset the ion transit time happens to be within  $\sim 0.01$  s of the nearest  $t_{\text{drift}}$  at which we collected data, and this results in a precise measurement. While, in principle, better accuracy could be obtained by repeating the measurements and always taking a point near this optimal time, the 20.8 day half life of the  $^{230}\text{U}$  source makes it impractical to repeat the entire experiment. It is also true that the data from which the first point is determined was recorded earlier in the lifetime of the  $^{230}\text{U}$  source, resulting in higher statistics. Systematic uncertainties on the mobility derive from small mechanical vibrations of the probe when the electric field is switched, and from the accuracy of the voltage gain in the amplifier. These contributions are estimated to give a combined error of  $\pm 0.011$  cm<sup>2</sup>/(kV-s). Table 1 shows the absolute errors on the voltages, drift distances and electric fields. The systematic error on the ion mobility was computed by shifting the electric field and drift distance values by their corresponding errors and re-doing the fit.

#### 4. Discussion

To help us understand our result, we have considered a transport model due to Hilt et al. [2] (henceforth referred to as HSK). In this model the microscopic properties of the ion and its detailed coupling to the liquid atoms are neglected, and the ionic mobility is determined solely by the equation of state of the liquid in question. The ion is modeled as a point charge whose electric field exerts a pressure on the liquid of sufficient magnitude for a microscopic ice layer to form around the ion. The ion and its attached cluster of Xe atoms then behave as a positively charged ball of ice whose radius is determined by the distance at which the electric field is no longer of a magnitude sufficient to compress the liquid into the solid state [2]. In this theory, no distinction is made between the mobilities of ions of different identities, and the drag suffered by the ion and its attached cluster is computed from the bulk viscosity of the liquid.

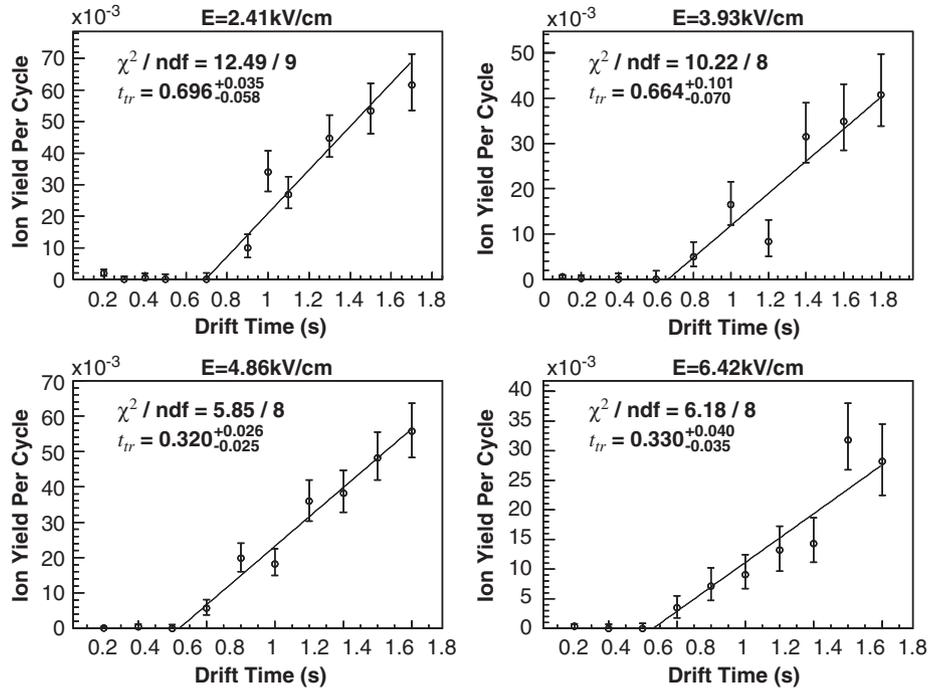


Fig. 4. Ion yield  $Y$  as a function of drift time  $t_{\text{drift}}$  for the four electric fields 2.41, 3.93, 4.86, and 6.42 kV/cm. Error bars shown are statistical only. We take the transit time in each case to be the knee point in the fit.

Table 1

Electric fields ( $E$ ), voltages ( $V_{\text{drift}}$ ), and drift distances ( $d$ ) used in this experiment, showing their associated systematic errors

$d (\times 10 \text{ cm})$	$V_{\text{drift}} (\times 10 \text{ kV})$	$E (\text{ kV/cm})$	$t_{tr} (\times 10 \text{ s})$	$v (\times 10 \text{ cm/s})$
$3.87 \pm 0.08$	$9.32 \pm 0.14$	$2.41 \pm 0.06$	$6.96^{+0.35}_{-0.58}$	$5.56^{+0.27}_{-0.46}$
$5.87 \pm 0.08$	$23.10 \pm 0.24$	$3.93 \pm 0.07$	$6.64^{+1.01}_{-0.70}$	$8.84^{+1.13}_{-0.93}$
$3.87 \pm 0.08$	$18.88 \pm 0.10$	$4.86 \pm 0.11$	$3.20^{+0.26}_{-0.25}$	$12.1^{+0.98}_{-0.94}$
$5.87 \pm 0.08$	$37.71 \pm 0.32$	$6.42 \pm 0.11$	$3.30^{+0.40}_{-0.35}$	$17.8^{+2.15}_{-1.89}$

The measured transit times ( $t_{tr}$ ) and velocities ( $v$ ) are the values returned by the fit. For these last two quantities, only statistical errors are given.

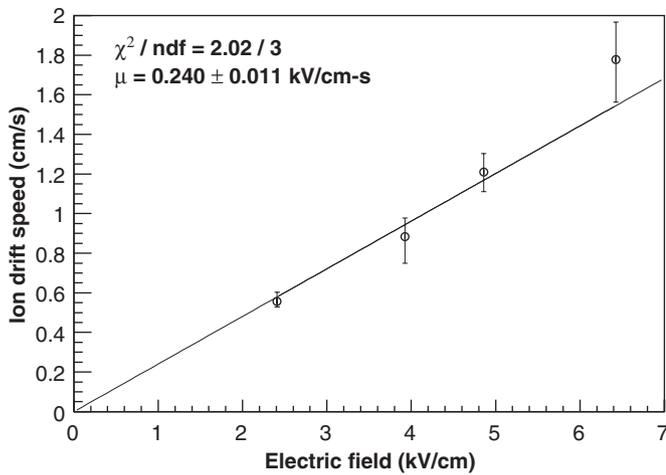


Fig. 5. Global fit to the ion drift velocity as a function of electric field for the entire data set.

Shown in Table 2 are some selected experimental mobilities for atomic ions in LXe. From these results it is clear that the HSK model does quite well in predicting the

overall scale of the observed mobilities. However, the significant difference between the  $^{208}\text{Tl}^+$  and  $^{226}\text{Th}^+$  numbers suggests that these species differ enough microscopically that the HSK model, when applied to them, begins to break down.

It should be pointed out that all of the calculations assume an ionic charge state of +1, which may not be correct. This is based on the argument that the energetics of charge exchange favor such a final state [6], but this assertion has not yet been convincingly verified experimentally. The HSK model predicts that a more highly charged ion should experience a larger drift force, but that it should also experience an increased drag force because it forms a larger ice layer around itself. These two effects nearly cancel, giving a net square-root dependence of the mobility on the charge state. Accordingly, we find that assuming an ionic charge other than +1 does not improve the compatibility between the theory and the data for any of the cases considered here.

Overall, it can be concluded that HSK provides a reasonable rough approximation of the ion mobility for the

Table 2  
Ion mobilities in LXe, comparing a model due to Hilt, Schmidt and Khrapak (HSK) [2] to the experimental data.

Ion	$T$ (K)	$P$ (bar)	Mobility ( $\text{cm}^2 \text{kV}^{-1} \text{s}^{-1}$ )	
			HSK	Expt.
$^{208}\text{Tl}^+$	163.0	0.9	0.31	$0.133 \pm 0.004^a$
$^{226}\text{Th}^+$	163.0	0.9	0.31	$0.240 \pm 0.011 \pm 0.011^b$
$\text{Ba}^+$	168.0	1.2	0.33	$0.211^{+0.020c}_{-0.012}$

<sup>a</sup>Ref. [6].

<sup>b</sup>This work.

<sup>c</sup>Ref. [7].

systems under consideration. However, it remains to be seen whether the model produces the correct temperature dependence in all cases. In addition, it is worth understanding the situations in which the ion identity becomes important because this would offer clues as to how HSK could be simply improved.

A theory put forward by Davis, Rice and Meyer (henceforth referred to as DRM) might offer some clues as to how this might be accomplished [1]. These authors have derived an expression for the positive ion mobility in which the internal structure of the ion and its interaction with the liquid are taken into account. In this model the drag force experienced by the ion is determined by the microscopic interaction potentials rather than the bulk properties of the liquid. It is conceivable, at least in principle, that as the ion–liquid interaction increases in strength, bound states between the ion and several liquid atoms can occur, particularly as the temperature of the liquid is lowered. Nevertheless, the treatment reported in Ref. [1] is specific to conditions in which this effect is not present, i.e., the ion–liquid interaction is too weak and the liquid temperature is too high. Indeed, the DRM model has only been compared with experimental data from systems involving molecular ions in liquids at temperatures well above the liquid triple point, namely  $\text{He}_2^+$  in LHe [4],  $\text{Ar}_2^+$  in LAr [1], and  $\text{Kr}_2^+$  in LKr [3]. While these systems are satisfactorily described by the model, it is clear that some modification will be necessary in order to treat the case of atomic ion mobilities close to the triple point. This is because the atomic ions under consideration have interaction strengths that are tens of times stronger [21,22] than those of the molecular ions previously considered.

To investigate these details experimentally, one could perform an accurate measurement of the temperature dependence of the ion mobility in several different ion–liquid systems. This would require multiple, well-understood and well-controlled sources of ions of known identity. A complementary approach would be to use the  $^{226}\text{Th}$  ion source in several different liquefied noble gases at various temperatures.

## 5. Conclusion

We have measured the  $^{226}\text{Th}$  ion mobility in LXe at 163.0K and standard pressure and have found it to have a value of  $0.240 \pm 0.011$  (stat)  $\pm 0.011$  (syst)  $\text{cm}^2/(\text{kV}\cdot\text{s})$ . The HSK model gives the correct order of magnitude when used to calculate ion mobilities, however it cannot account for the significant difference between our result and the one measured for  $^{208}\text{Tl}^+$  by other workers. A more detailed study of this effect may help to suggest how the HSK model can be modified to account for different ion identities. A possible route might be to generalize the DRM treatment to the case of atomic ions that form bound states with liquid atoms.

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