Index of refraction, Rayleigh scattering length, and Sellmeier coefficients in solid and liquid argon and xenon

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Abstract

Like all the noble elements, argon and xenon are scintillators, i.e. they produce light when exposed to radiation. Large liquid argon detectors have become widely used in low background experiments, including dark matter and neutrino research. However, the index of refraction of liquid argon at the scintillation wavelength has not been measured and current Rayleigh scattering length calculations disagree with measurements. Furthermore, the Rayleigh scattering length and index of refraction of solid argon and solid xenon at their scintillation wavelengths have not been previously measured or calculated. We introduce a new calculation using previously measured data in liquid and solid argon and xenon to extrapolate the optical properties at the scintillation wavelengths using the Sellmeier dispersion relationship. As a point of validation, we compare our extrapolated index of refraction for liquid xenon against the measured value and find agreement within the uncertainties. This method results in a Rayleigh scattering length for liquid argon at the triple point of $55 \pm 5$ cm, a Rayleigh scattering length of $40 \pm 4$ cm for solid argon at the triple point, and a Rayleigh scattering length of $14 \pm 1$ cm for solid xenon at the triple point.

Keywords: Rayleigh scattering, index of refraction, liquid argon, liquid xenon, solid argon, solid xenon

1. Introduction

Noble element detectors are widely used in many particle and astro-particle physics experiments. Detectors like this are useful for low background, low mass particle detection such as dark matter and neutrino research. For example, the DEAP/CLEAN collaborations use large liquid argon detectors in their dark matter searches [1], MicroBooNE will use a 170 ton liquid argon time projection chamber for...
neutrino research [2], and the successful LUX [3] dark matter experiment uses liquid xenon as a detection media.

Noble elements are useful active detector media due to their scintillation properties, emitting vacuum ultraviolet (VUV) photons when excited by radiation (128 \textit{nm} for argon, and 174 \textit{nm} for xenon [4]). Many liquid noble detector use some form of optical readout to take advantage of this property, but event energy and vertex reconstruction can both be effected by the details of photon propagation. In the case of large liquid argon detectors the location of an event within a detector is used to ascertain its scientific significance [1]. Event location is determined through optical modelling [1, 2] that takes into account the Rayleigh scattering length and index of refraction at the scintillation wavelength. While these values have been measured in liquid xenon at the triple point [5, 6], currently the index of refraction of the scintillation wavelength in liquid and solid argon and solid xenon have not been measured. In the case of liquid argon at the scintillation wavelength, there is a discrepancy between a calculated Rayleigh scattering length of 90 \textit{cm} [4] and a measured Rayleigh scattering length of 66±3 \textit{cm} [5]. The values obtained by these two groups differ by 27% which motivated a need to further clarify the precise Rayleigh scattering length at the scintillation wavelength of liquid argon.

2. Rayleigh Scattering

Rayleigh scattering was first proposed by Lord Rayleigh in his 1899 paper [7]. Rayleigh scattering is the process of light elastically scattering off of particles smaller than the wavelength of light in a medium so the wavelength of the light does not change. The length of travel for a photon through a medium before Rayleigh scattering is strongly dependent on the wavelength of the light as well as the optical properties of the material. This can be seen in the Rayleigh equations that follow. Knowing the wavelength dependent Rayleigh scattering length of an individual material is useful in reconstructing the path that light would travel through it.

The original Rayleigh scattering length formulation was expressed as [7]

\[ l = \frac{1}{\sigma N}, \]  \hspace{1cm} (1)

where \( l \) is the Rayleigh scattering length, \( N \) is the number density and \( \sigma \) is the Rayleigh cross section. This cross section can be written as,
\[ \sigma = \frac{24\pi^3}{N^2\lambda^4} \left[ \frac{n^2 - 1}{n^2 + 2} \right]^2, \]

where \( n \) is the index of refraction, and \( \lambda \) is the wavelength of light. It was later found through experiment that this equation was not sufficient to describe the cross section of anything other than a spherical gas molecule [8] [9]. Einstein updated the equation in 1910 to account for the thermodynamics of liquids and solids [8] which resulted in

\[ l^{-1} = \frac{16\pi^3}{6\lambda^4} \left[ kT\rho^2\kappa_T \left( \frac{\partial n^2}{\partial \rho} \right)_T^2 + \frac{kT^2}{\rho c_v} \left( \frac{\partial n^2}{\partial T} \right)_\rho^2 \right], \]

where \( k \) is the Boltzmann’s constant, \( T \) is the temperature, \( \rho \) is the liquid density, \( \kappa_T \) is the isothermal compressibility and \( c_v \) is the heat capacity. The second term is small enough to be negligible [9], so the equation becomes

\[ l^{-1} = \frac{16\pi^3}{6\lambda^4} \left[ kT\rho^2\kappa_T \left( \frac{\partial n^2}{\partial \rho} \right)_T^2 \right]. \]

The following step is used to estimate \( \left( \frac{\partial n^2}{\partial \rho} \right)_T \) [4]. The wavelength dependence of the index of refraction can be expressed with the Lorentz-Lorenz equation [10] [8],

\[ \frac{n^2 - 1}{n^2 + 2} = \rho \left( a_0 + \sum_i \frac{a_i}{\lambda^2 - \lambda_i^2} \right). \]

In this equation, \( \rho_m \) is the density in moles per unit volume, \( \lambda_i \) is the wavelength corresponding to the \( i^{th} \) resonance, \( a_i \) is the constant related to the strength of the resonance, and \( a_0 \) is an experimentally determined constant. This expression of the expansion is used for constant temperature and density. This expression is used to get \( \left( \frac{\partial n^2}{\partial \rho} \right)_T \) in terms of \( n^2 \) and equation 4 becomes

\[ l^{-1} = \frac{16\pi^3}{6\lambda^4} \left[ kT\rho^2\kappa_T \left( \frac{\partial n^2}{\partial \rho} \right)_T^2 \right]. \]

For this equation to be valid it is important to use the index of refraction at the specific conditions of temperature, density and wavelength [8]. There are also material dependent correction factors than can be added to equation 6 that do not apply in the case of nobles [9]. This expression for Rayleigh scattering length will be used in the extrapolations discussed later in this paper.
3. Measurements

With the exception of liquid xenon at the triple point [6], the index of refraction of the scintillation wavelengths of solid and liquid argon and solid xenon. There have been a few measurements of the various optical properties of solid and liquid argon and solid xenon.

- Sinnock and Smith [11] measured the index of refraction as a function wavelength at different temperatures in solid and liquid argon and xenon. These measurements were made between the wavelengths of 350 nm and 650 nm with an error of ±0.5%.

- Bideu-Mehu et al. [12] measured the index of refraction of room temperature argon and xenon gas between the wavelengths of 140 nm and 174 nm and used these values to find the Sellmeier coefficients for the gas based Sellmeier equation.

- Ishida et al. [5] measured the attenuation length of liquid xenon and argon at the scintillation wavelengths. They found values of 66 ± 3 cm for argon at 87 K and 29 ± 2 cm for xenon at 196 K.

- Solonov et al. [6] measured the index of refraction and attenuation length of liquid xenon.

Seidel et al. calculated the Rayleigh scattering length for liquid argon and xenon. Seidel’s calculated values were 90 cm for argon and 30 cm for xenon (the authors did not include the error on their calculation). Their extrapolations disagree with the measured attenuation lengths of Ishida et al. [5] for liquid argon. The calculated Rayleigh scattering length was in error with the measured values for xenon, however a different method of calculation was used for xenon. In the case of argon, Seidel et al. [4] used data from from Bideu-Mehu et al. [12] to extrapolate the dielectric constant at the scintillation wavelengths. Bideu-Mehu measured the index of refraction of argon and xenon at 275 K between the wavelength of 250 nm and 140 nm [12]. These values were fit using the simplified Sellmeier equation for gasses for the relationship between the index of refraction and wavelength. The simplified Sellmeier equation for gasses is,

\[ n - 1 = \frac{N e^2}{8\pi^2\epsilon_0 mc^2} \sum \frac{f_i}{\lambda_i^{-2} - \lambda^{-2}} \]  

(7)

where \( n \) is the index of refraction, \( N \) is the number density of atoms or molecules, \( e \) and \( m \) are the charge and mass of the electron, \( \epsilon_0 \) is the vacuum permittivity, \( \lambda \) is the wavelength, \( \lambda_i \) is a resonance wavelength.
and $f_i$ is the Sellmeier coefficient found experimentally that corresponds to the resonance wavelength [13]. Equation 7 is derived from equation 5 using assumptions specifically applicable to gasses. Seidel et al. used the coefficients found by Bideu-Mehu to extrapolate the dielectric constants at the scintillation wavelengths. Seidel et al. neglected any temperature dependence and made an adjustment for the density change from gas to liquid. The reasoning for neglecting the temperature was based on the research done by Achtermann et al. [14], a study which examined the effect of temperature on argon gas between the temperatures of 298 K and 323 K. In the case of xenon, Seidel et al. used a previous liquid xenon index of refraction and Rayleigh scattering length measurements made in the ultraviolet [15] and extrapolated accordingly. Therefore, in the case of their xenon extrapolation, both temperature and state were properly accounted for, unlike their argon extrapolation. Seidel et al. then calculated the Rayleigh scattering length with equation 6 using their extrapolated indices of refraction.

4. Updated Analysis

In the extrapolations done by Seidel, the assumption was made that the index of refraction of argon would not change as a function of pressure, state or temperature or that the change would be nominal. This is contrary to other work demonstrating that the index of refraction does indeed depend on these parameters [16, 17, 13]. To avoid this problem, we sought out various measurements of the refractive index of liquid argon all done at the same temperature. Sinnock and Smith measured the refractive index of argon and xenon at different temperatures in the liquid and solid regions of these elements [11]. These measurements were done at different wavelengths. We used the index of refraction measurements in this experiment to extrapolate down to the scintillation wavelength. In the case of the liquid state, it was to verify or dispute previous measurements or extrapolations, in the case of the solid state, it was to provide the first numbers for index of refraction and Rayleigh scattering for this state. To do this we used the following Sellmeier dispersion relation for liquids and solids at constant temperature and density [13, 17],

$$n^2 = a_0 + \sum_i \frac{a_i\lambda^2}{\lambda^2 - \lambda_i^2}.$$  

In this case $a_0$ is a Sellmeier coefficient that accounts for the effect of UV resonances not included in the sum and $a_i$ are the Sellmeier coefficients that correspond with the $i^{th}$ resonances. The Sellmeier dispersion equation was derived from the Lorentz-Lorenz equation and the components have been experimentally determined. The data from Sinnock and Smith was fit using the first UV and IR resonance wavelength.
for each element. The wavelengths of their data fall between the surrounding resonance peaks listed in table 1 which will have the most influence on the fit. The fit equation used is

\[
n^2 = a_0 + \frac{a_{UV} \lambda^2}{\lambda^2 - \lambda_{UV}^2} + \frac{a_{IR} \lambda^2}{\lambda^2 - \lambda_{IR}^2},
\]

where \(\lambda_{UV}\) corresponds to the closest or first UV resonance and \(\lambda_{IR}\) corresponds to the closest or first IR resonance.

The data used for the fit was taken by Sinnock and Smith. The error reported in their original work was \(\pm 0.5\%\) [16] on the index of refraction, though the authors state that this was a conservative value [11].

| Element | UV Resonance \(\lambda\) (nm) | IR Resonance \(\lambda\) (nm) |
|---------|-------------------------------|-------------------------------|
| Argon   | 106.6                         | 908.3                         |
| Xenon   | 146.9                         | 827.0                         |

Table 1: Resonance wavelengths of argon and xenon. Argon UV resonance value obtained from [18], xenon UV resonance obtained from [12] and argon and xenon IR resonances sourced from [19].

4.1. Fit Verification

Solovov et al. [6] measured the index of refraction of the scintillation wavelength of liquid xenon at the triple point. We fit the liquid triple point data from [11] with equation 9 and used this fit to extrapolate to the scintillation wavelength. The extrapolation error was calculated using the covariance matrix, as described in [20]. In figure 4 we include the data point from Solovov et al. demonstrating that our extrapolation is consistent with the measurement, to within the given uncertainties. This result verifies that fitting the data from [11] with equation 9 is effective in extrapolating the index of refraction at the scintillation wavelengths. The difference between the Solovov point and the extrapolated value is less than 2.5\%. 
Figure 1: Sinnock data [11] with fit and Solovov [6] point. The line is the extrapolation and points corresponds to data points. A dashed line is placed at 174 nm, the scintillation wavelength of Xenon.

5. Results

5.1. Sellmeier Coefficients

The first step in analyzing the data from [11] was to fit with equation 9 and find the Sellmeier coefficients. These will be useful in determining the index of refraction at any wavelength between the UV and IR resonances. The results of this fit can be found in tables 2 - 3. For the case of liquid xenon at the triple point, the Solovov point [6] was included in the fit to improve the accuracy of the coefficients at this temperature. In fact, as one can see from table [3] including this point decreases the uncertainties in two of the Sellmeier coefficients by an order of magnitude compared to the other temperatures.
| T (K) | $a_0$    | $a_{UV}$  | $a_{IR}$  |
|------|---------|-----------|-----------|
| Solid |         |           |           |
| 20   | 1.4±0.1 | 0.30±0.09 | 0.0011±0.007 |
| 30   | 1.4±0.1 | 0.30±0.09 | 0.00047±0.007 |
| 40   | 1.3±0.1 | 0.29±0.09 | 0.00071±0.007 |
| 50   | 1.4±0.1 | 0.27±0.09 | 0.0019±0.007 |
| 60   | 1.3±0.1 | 0.29±0.09 | 0.0011±0.007 |
| 70   | 1.3±0.1 | 0.21±0.09 | 0.0011±0.007 |
| 80   | 1.3±0.1 | 0.29±0.09 | 0.0011±0.007 |
| 83.81| 1.3±0.1 | 0.29±0.09 | 0.00087±0.007 |
| Liquid|         |           |           |
| 83.81| 1.24±0.09 | 0.27±0.09 | 0.00047±0.007 |
| 86   | 1.24±0.09 | 0.27±0.09 | 0.00085±0.007 |
| 88   | 1.23±0.09 | 0.27±0.09 | 0.00085±0.007 |
| 90   | 1.26±0.09 | 0.23±0.09 | 0.0023±0.007 |

Table 2: Argon Sellmeier coefficients. 83.81 K is the argon triple point.
| T (K) | $a_0$     | $a_{UV}$  | $a_{IR}$  |
|-------|----------|----------|----------|
| Solid |          |          |          |
| 80    | 1.6±0.3  | 0.6±0.2  | 0.001±0.03 |
| 90    | 1.6±0.3  | 0.6±0.2  | 0.0009±0.03 |
| 100   | 1.6±0.3  | 0.6±0.2  | 0.0009±0.03 |
| 110   | 1.6±0.3  | 0.6±0.2  | 0.001±0.03 |
| 120   | 1.6±0.3  | 0.6±0.2  | 0.001±0.03 |
| 130   | 1.5±0.2  | 0.6±0.2  | 0.001±0.03 |
| 140   | 1.5±0.2  | 0.6±0.2  | 0.0009±0.03 |
| 150   | 1.5±0.2  | 0.6±0.2  | 0.0008±0.03 |
| 162.35| 1.4±0.2  | 0.6±0.2  | 0.0008±0.03 |
| Liquid|          |          |          |
| 162.35*| 1.5±0.02 | 0.38±0.01| 0.009±0.01 |
| 166   | 1.4±0.2  | 0.4±0.2  | 0.001±0.02 |
| 170   | 1.4±0.2  | 0.4±0.2  | 0.002±0.02 |
| 174   | 1.4±0.2  | 0.4±0.2  | 0.002±0.02 |
| 178   | 1.4±0.2  | 0.4±0.2  | 0.002±0.02 |

Table 3: Xenon Sellmeier Coefficients. *This Fit includes the point from [6]. 162.35 K is the xenon triple point.

5.2. Index of Refraction and Rayleigh Scattering Lengths

The Sellemeier coefficients obtained by fitting the data from Sinnock and Smith [11] were then used to extrapolate the index of refraction and Rayleigh scattering length at the scintillation wavelengths. The values can be found in tables 4-5 and the extrapolations are graphically depicted in figures 3-5.
Figure 2: The index of refraction for argon extrapolation. A dashed line is placed at 128 nm, the scintillation wavelength of Argon.
Figure 3: The Rayleigh scattering length of argon extrapolation. A dashed line is placed at 128 nm, the scintillation wavelength of Argon.
| $T$ (K) | $n$     | $l$ (cm) |
|---------|---------|----------|
| Solid   |         |          |
| 20      | 1.52±0.07 | 149±4    |
| 30      | 1.53±0.07 | 97±10    |
| 40      | 1.52±0.074| 75±8     |
| 50      | 1.51±0.07 | 67±7     |
| 60      | 1.51±0.07 | 53±5     |
| 70      | 1.51±0.07 | 47±5     |
| 80      | 1.50±0.07 | 42±4     |
| 83.81   | 1.50±0.07 | 40±4     |
| Liquid  |         |          |
| 83.81   | 1.46±0.07 | 55±5     |
| 86      | 1.45±0.07 | 54±5     |
| 88      | 1.45±0.07 | 54±5     |
| 90      | 1.43±0.07 | 62±6     |

Table 4: Solid and liquid argon index of refraction and Rayleigh scattering length extrapolations at the scintillation wavelength. 83.81 K is the argon triple point.
Figure 4: Index of refraction of solid xenon extrapolation. A dashed line is placed at 174 nm, the scintillation wavelength of xenon.
Figure 5: The Rayleigh scattering length extrapolation. A dashed line is placed at 174 nm, the scintillation wavelength of xenon.

| $T$ (K) | $n$       | $l$ (cm) |
|---------|-----------|-----------|
| Solid   |           |           |
| 80      | 1.90±0.03 | 25±2      |
| 90      | 1.90±0.04 | 23±2      |
| 100     | 1.89±0.04 | 21±1      |
| 110     | 1.89±0.04 | 19±1      |
| 120     | 1.89±0.04 | 18±1      |
| 130     | 1.88±0.03 | 17±1      |
| 140     | 1.88±0.03 | 16±1      |
| 150     | 1.87±0.03 | 15±1      |
| 162.35  | 1.87±0.03 | 14±1      |

Table 5: Solid and liquid xenon index of refraction and Rayleigh scattering length extrapolations at the scintillation wavelength. 162.35 K is the xenon triple point.
Table 6: This a summary of the results of our extrapolations made by fitting the Sinnock data at the triple point with the Sellmeier equation. Both the liquid and solid triple point values are included. These values are compared with previous calculations and measurements. The previous index of refraction and Rayleigh scattering length calculation come from [4]; error bars were not included in the original work. The previous argon scattering length measurements come from [5] and the previous xenon index of refraction measurement is from [6].

6. Conclusion

Our updated analysis used the data from Sinnock et al. [11] extrapolate the wavelength dependent index of refraction through argon at constant temperature and state. Using this index of refraction, we calculated a Rayleigh scattering length in liquid argon at 88 K of 54±5 cm. This is closer to the value measured by Ishida et al. [5] than the extrapolated value by Seidel et al. [4], implying that using a Rayleigh scattering length of 90 cm, in optical reconstruction would result in misconstructing detector events. We tested the accuracy of our extrapolation method against a measured point in xenon and we were able to predict the value within experimental error.

The data taken by Sinnock et al. [11] also gave us the opportunity to produce values for the index of the refraction and Rayleigh scattering lengths of solid argon and xenon at different temperatures at the scintillation wavelengths. These values may be useful in future detectors or experiments that take advantage of the scintillation properties of these elements in a solid state. All of the results for the index of refraction and Rayleigh scattering length are collected in table 6.

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