Attenuation of vacuum ultraviolet light in liquid argon

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Abstract. The transmission of liquid argon has been measured, wavelength resolved, for a wavelength interval from 118 to 250 nm. The wavelength dependent attenuation length is shown to be present for pure argon. It is shown that no universal wavelength independent attenuation length can be assigned to liquid argon for its own fluorescent light due to the interplay between the wavelength dependent emission and absorption. A decreasing transmission is observed below 130 nm in both chemically cleaned and distilled liquid argon and assigned to absorption by the analogue of the first argon excimer continuum. For not perfectly cleaned argon a strong influence of impurities on the transmission is observed. Two strong absorption bands at 126.5 and 141.0 nm with approximately 2 and 4 nm width, respectively, are assigned to traces of xenon in argon. A broad absorption region below 180 nm is found for unpurified argon and tentatively attributed to the presence of water in the argon sample.

PACS. 29.40.Mc Scintillation detectors – 33.20.Ni Vacuum ultraviolet spectra – 61.25.Bi Liquid noble gases

1 Introduction

Liquid rare gases have been proven to be excellent media for particle detection with fluorescence detectors due to their high fluorescence efficiency. Liquid argon and xenon, in particular, are used for rare event search in astroparticle physics and benefit from the outstanding scintillation properties of liquid rare gases. Several already existing or future experiments use argon as detector medium, both in scintillation counters and time projection chambers, covering nearly all present fields of astroparticle physics like direct search for dark matter particles (MiniCLEAN [1], DEAP-3600 [2], ArDM [3], WARP [4], DarkSide [5], and DARWIN [6]), neutrino physics (ICARUS [7] and GLACIER [8]), or the search for neutrinoless double-beta decay (GERDA [9] uses a liquid argon veto).

Since some of the detector systems mentioned above use large quantities of liquid argon the scintillation light may have to traverse long distances from the position where it is produced to the detector. It is therefore important to study the attenuation of vacuum ultraviolet (VUV) light in liquid argon. However, an intensive literature search returned no publication showing a wavelength resolved attenuation length measurement for pure liquid argon. Ishida et al. [10] measured, among other noble gases and admixtures, the wavelength integrated attenuation length of pure liquid argon for its own scintillation light. The measuring principle was based on recording the amount of scintillation light (obtained by irradiating liquid argon with a heavy ion beam) at seven different distances between 137 mm and 737 mm from the light detector. For that purpose, liquid argon was contained in a steel cell with seven entrance windows for the heavy ion beam. Light detection was performed at the end of the steel cell via a Pyrex® window coated with sodium salicylate, which shifted the VUV scintillation light into visible light to enable detection with a regular photomultiplier. A wavelength integrated attenuation length of (66±3) cm in pure liquid argon was obtained in this experiment.

Recently we have measured the emission spectrum of liquid argon in the vacuum ultraviolet spectral range. The main result of this study [11] will be shown again here (section 3.1). It is obvious that the wavelength integrated measuring principle cannot disentangle the influence of possible impurities like water vapor, oxygen, or other noble gases, which have a huge effect on the transmission. It has also to be noted that impurities, depending on their concentrations, affect both the emission spectrum as well as the transmission spectrum. Any wavelength integrated measuring principle can therefore only measure a combined effect of emission and transmission, and consequently the obtained "attenuation length" strongly depends on impurities. We will show that an attenuation length (for wavelength-integrated light) in the sense of an exponential decay of intensity and a reduction of light to 1/2 cannot be applied due to the wavelength dependence of the absorption. We will also show that traces of other rare gases, xenon in particular in the case of liquid argon, can contribute significantly to light absorption. Since Ishida et al. [10] used the same experimental setup to measure all noble gases and admixtures, the probability of a noble gas contamination is high in these experiments but cannot be identified due to the wavelength-integrated data taking method. Wavelength resolved reference data for the
absorption of xenon-doped liquid argon are available from a measurement by Raz and Jortner in 1970 [13] and will be used to estimate the xenon impurity concentration in our experiments.

Here we follow the concept that the optical properties of liquid rare gases are similar to those of dense gases. From the latter some information is available to predict the attenuation. Detailed studies of the issue have been performed in the context of VUV excimer lasers of pure rare gases [15]. Attenuation on the long wavelength side of the resonance lines of dense krypton and xenon gas has been described in ref. [15] in that context. Both absorption due to optical transitions starting from the van der Waals minimum of the lower potential curve in the region of the so-called first excimer continuum and scattering due to increasing fluctuations of the index of refraction when approaching the resonance lines from longer wavelengths have been identified in that reference as the reason for light attenuation extending far out into the red wings of the resonance lines. Rayleigh scattering in rare gas liquids has also more recently been discussed in ref. [10] resulting in a scattering length of 90 cm. Here we assume that the processes discussed in these publications are also relevant for the attenuation in liquid argon for wavelengths close to the resonance lines of argon at 104.82 nm 106.67 nm [17], and longer wavelengths.

After a description of experimental details the VUV transmission of the best quality liquid argon achieved in this study will be presented. Then, it will be shown that traces of xenon in liquid argon lead to significant absorption bands, in part exactly at the peak of the emission band of liquid argon, and the data will be discussed using the results from ref. [14]. Attenuation by impurities is a well known effect in the vacuum ultraviolet spectral region, and rare gas impurities are particularly critical because they are not removed by regular chemical gas purification techniques. It will be shown that these chemical purification steps are mandatory to remove regular impurities like oxygen and water vapour.

2 Experimental details

2.1 Experimental setup

The concept of the experiment was to use a very basic optical setup to avoid complications in the interpretation of the results due to changes in the geometry when the gas cell is cooled down (fig. 1). Vacuum ultraviolet light was produced by a deuterium lamp with a MgF2 window (Cathodeon Model V03). It was attached to an outer vacuum cell of 100 mm diameter (CF 100 cross piece). An inner cell made from copper was installed in the center of this vacuum cell. This inner cell had two optical MgF2 windows of 5 mm thickness and 25 mm diameter and could be filled with liquid argon. The distance between the inner surfaces of the MgF2 windows was 58 mm, which is the relevant distance for the transmission measurements. The light emitted from the deuterium lamp was collimated by a stack of 3 apertures with 4 mm diameter and sent through the center of the inner cell. Another set of apertures was mounted behind the inner cell. A f=30 cm vacuum spectrometer (McPherson model 218) was mounted at the outer cell opposite to the deuterium lamp. It was equipped with a VUV image intensifier (micro channel plate, MCP) with MgF2 entrance window, S20 multi-alkali cathode, and a diode array. The spectrometer could measure a spectrum of about 60 nm width in one exposure, and the center wavelength could be set manually.

The cryogenic setup and gas handling were analogous to those described in refs. [13][15]. The setup including a schematic drawing of the gas handling system is shown in fig. 2. The inner cell was held in place and cooled by a copper rod attached to a dewar filled with liquid nitrogen. Temperature was measured using PT100 resistors and could be controlled by sending an electrical current through a 47 Ohm, 20 W resistor, which was soldered onto the copper rod. Gas filling and circulation were performed via a heat exchanger. The outer gas system consisted of a closed cycle of a rare gas purifier (SAES model MonoToRR® Phase II, PS4-MT3) and a metal bellows pump to circulate the gas through the system. The gas could be filled into the system from a gas bottle with a pressure reducer. The cryogenic cell was added as a bypass to the gas cycle so that the gas in that cell could be continuously refreshed by condensation and evaporation. A stainless steel storage volume was attached to the outer gas system, and could be cooled to liquid nitrogen tem-

Fig. 1. Transmission measurements in liquid argon were performed by sending light from a deuterium lamp (A), which was attached to an evacuated CF-100 crosspiece (outer cell, G), through a 58 mm long inner cell made from copper (B) containing liquid argon. Apertures (D), both in front and behind the inner cell, were used to avoid stray light. The inner cell (B) was attached to a liquid nitrogen dewar for cooling (not shown) and was sealed with MgF2 windows (E) on the optical axis. Gas in- and outlet were provided via the connections (F). The evacuated inner cell kept at the same temperature was used for recording reference spectra. A f=30 cm VUV spectrometer was placed at the light output (C) and used to record the spectra.

Fig. 2. Schematic drawing of the gas handling system is shown here. The cryogenic setup and gas handling were analogous to those described in refs. [13][15]. The setup including the cryogenic cell and the gas handling system is shown in the figure. The inner cell was held in place and cooled by a copper rod attached to a dewar filled with liquid nitrogen. Temperature was measured using PT100 resistors and could be controlled by sending an electrical current through a 47 Ohm, 20 W resistor, which was soldered onto the copper rod. Gas filling and circulation were performed via a heat exchanger. The outer gas system consisted of a closed cycle of a rare gas purifier (SAES model MonoToRR® Phase II, PS4-MT3) and a metal bellows pump to circulate the gas through the system. The gas could be filled into the system from a gas bottle with a pressure reducer. The cryogenic cell was added as a bypass to the gas cycle so that the gas in that cell could be continuously refreshed by condensation and evaporation. A stainless steel storage volume was attached to the outer gas system, and could be cooled to liquid nitrogen tem-
mission spectra when the inner cell was filled with liquid
to improve the statistics. The concept was to record trans-
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which the higher diffraction-order signals from the grat-
length cutoff of the detection system up to 250 nm above
was generated by minimizing the squared deviations of the
vided center wavelengths of 90, 120, 150, 180, and 210 nm
was used for the experiments described below. The indi-
er cell at the same temperature, changes in the transmis-
evacuated. Since both spectra were recorded with the in-
reference spectra recorded with the inner cell cooled and
onds and the time differences between the measurements
accuracy of time measurement was of the order of sec-
data based on the findings shown in fig. 3 and fig. 4. The
carefully documented for corrections in the analysis of the
liquid. The time at which each spectrum was recorded
tra had been measured with the inner cell filled with the
inner cell (see fig. 1), which could be either evacuated or filled
with liquid argon, and detected by a spectrometer. The cell was
operated as a bypass (valve D) of a closed gas system in which
the argon gas was circulated with a metal bellow pump (C)
through a rare gas purifier (B). A heat exchanger (E) was in-
stalled between the warm and cold parts of the gas system. A
small storage volume (A) could be used to distill the gas prior
to filling it into the system. A turbo molecular pump (F) was
used to evacuate the system prior to filling.

2.2 Experimental method

The basic concept was to divide the spectra obtained with
the inner cell filled with liquid argon by the corresponding
reference spectra recorded with the inner cell cooled and
evacuated. Since both spectra were recorded with the in-
ner cell at the same temperature, changes in the transmis-
sion due to geometrical effects (expansion and contraction of the
rod holding the cell etc.) could be excluded. A fixed
set of center wavelengths of 90, 120, 150, 180, and 210 nm
was used for the experiments described below. The indi-
vidual spectra overlapped by 30 nm. An overall spectrum
was generated by minimizing the squared deviations of the
optical signal in the overlapping regions [19]. This allowed
us to measure absorption spectra from the short wave-
length cutoff of the detection system up to 250 nm above
which the higher diffraction-order signals from the grat-
ing monochromator would have required edge filters to be
used. The operation and readout of the detector was com-
puter controlled and allowed to select the exposure time.
Exposures could be repeated automatically several times
to improve the statistics. The concept was to record trans-
mission spectra when the inner cell was filled with liquid
argon for every setting of the central wavelength and cor-
responding reference spectra with a cooled and evacuated
inner cell. A necessary correction is to take the change in the
index of refraction at the inner surfaces of the MgF$_2$
windows into account between the case of an empty cell
and a cell filled with liquid argon (Fresnel formula). A test
to correct our raw data with this Fresnel formula using the
index of refraction of MgF$_2$ [20] and liquid argon (scaled from the
gas phase) [21,22] did not change the overall spectral shape, in particular, the decreasing trans-
mision towards shorter wavelengths (described in section
3.1) remained. Therefore, the "Fresnel correction" was not
applied.

It was found that a more serious experimental problem
appears due to condensation of residual gas components in the
outer vacuum onto the surfaces of the MgF$_2$ windows of the
inner cell, a process which will be called "fogging" in the text below. A series of transmission spectra of the
cooled and evacuated inner cell recorded at various times
after cooling the inner cell down is shown in fig. 3. An analysis of these data showed that the transmission was
reduced exponentially with time at each wavelength po-

tion. This is documented in fig. 4 and allowed a proper
correction of each spectrum according to the time at which it
had been recorded after the start of the cooldown. The
measurement of the time dependent fogging of the MgF$_2$
windows (fig. 3) was performed 21 hours after the mea-
surement of pure liquid argon. This was approximately
the time difference which was necessary to heat up the
cell to room temperature again, without thermally dam-
aging the experimental setup. Therefore, changes in the
composition of the residual gas components in the outer
insulation vacuum between the measurement of pure liq-
uid argon and the measurement of the fogging of the MgF$_2$
windows can be excluded.

Since it was found that the liquid argon could be re-
moved from the inner cell more quickly than condensed
into it, the reference spectra were recorded after the spec-
tra had been measured with the inner cell filled with the
liquid. The time at which each spectrum was recorded
with respect to the start of cooling the inner cell was
carefully documented for corrections in the analysis of the
data based on the findings shown in fig. 3 and fig. 4. The
accuracy of time measurement was of the order of sec-
onds and the time differences between the measurements
of the cell filled with liquid argon and the corresponding
reference measurements were between one and two hours
depending on the intensity of the D$_2$ lamp in the different
spectral regions and consequently the required exposure
times. The overall accuracy was of the order of minutes
given by the exposure time of the data recording system.
The statistical errors of the exponential fits of the trans-
mision at each wavelength (fig. 4) were considered and
incorporated in the total error analysis.

Besides the short-term degradation of the spectra shown
in figs. 3 and 4 a long term modification (weeks) of the
emission from the deuterium lamp was found. Exposure
to the residual gas in the outer cell obviously led to a re-
duction of intensity in the short wavelength part of the

![Fig. 2. Schematic drawing of the experimental setup. Broad-
band VUV light was sent from a deuterium lamp through the
inner cell (see fig. 1), which could be either evacuated or filled
with liquid argon, and detected by a spectrometer. The cell was
operated as a bypass (valve D) of a closed gas system in which
the argon gas was circulated with a metal bellow pump (C)
through a rare gas purifier (B). A heat exchanger (E) was in-
stalled between the warm and cold parts of the gas system. A
small storage volume (A) could be used to distill the gas prior
to filling it into the system. A turbo molecular pump (F) was
used to evacuate the system prior to filling.](image-url)
Fig. 3. The transmission of the empty and cooled inner cell is reduced with time by condensation of substances from the insulating vacuum of the outer cell onto the MgF$_2$ windows. A series of transmission spectra with respect to the time of filling the dewar with liquid nitrogen is shown. Spectra in the time interval from 2 to 3 hours were used to correct the data recorded for the actual transmission measurements of liquid argon. Spectra obtained with argon were typically measured 2 hours and reference spectra typically 3 hours after the start. Detailed time keeping in combination with interpolation (see fig. 4) between the data shown above was performed to use the appropriate corrections to the data before calculating the transmission of liquid argon itself.

Fig. 4. Time dependence of the transmission due to "fogging" for a set of different wavelengths, together with exponential fits to the data.

VUV emission due to carbon hydroxides in the residual gas, which were cracked by the VUV light and coated the MgF$_2$ window of the lamp. This effect is known in VUV optics and could be reversed by letting the lamp operate in an oxygen containing atmosphere for several hours.

For the analysis of the data described below it was important to study the performance of the detector carefully. An important issue was to find the zero level in the spectra, which has to be subtracted from the raw data before the ratio between transmission spectrum and reference spectrum is calculated. First, background spectra were recorded by closing a valve in front of the spectrometer and subtracted from the subsequent measurements. Fig. 5 exhibits an example of the raw data measured in the most critical, short wavelength region (120 nm central wavelength setting at the monochromator). The thin (red) line shows the data obtained in a measurement of the inner cell evacuated and the thick (black) line shows data recorded in a measurement with the inner cell filled with liquid argon. Relevant are two background levels in which scattered light is taken into account. The first pedestal (I in fig. 5) which appears left and right in the spectra is due to the fact that the diode array is somewhat larger than the diameter of the MCP and sees no signal. After a transition phase (II in fig. 5), the second step is observed in the region (III in fig. 5) where the MCP and the diode array are both active. This level is used as a background level based on data which were recorded with wavelength settings for which no light can be expected: In our analysis we used wavelengths, which were shorter than the cutoff wavelength of the MgF$_2$ windows ($\lambda < 113$ nm [23]).

A critical examination of the emission spectrum based on the emission spectrum of the lamp and the background issues of the detector showed that transmission measurements down to 118 nm can be performed with the setup described here. The light output from the deuterium lamp below 118 nm is too low to provide reliable data (see fig. 5). Transmission spectra shown below will therefore always start at this wavelength.

3 Results and discussion

3.1 Pure liquid argon

The best result, obtained so far, for the wavelength dependence of the transmission of VUV light through 58 nm liquid argon is shown in fig. 6 in comparison with the emission spectrum observed in ref. [13]. All purification steps described in section 3.2 below (usage of a gas purifier and distillation of liquid argon) were applied to record these data. Since the raw data showed a transmission slightly above 100% for long wavelengths, we compensated this so far unresolved systematic effect by scaling the 210 nm center wavelength transmission measurements to 100%. The transmission is close to 100% above 130 nm. It decreases towards shorter wavelengths and reaches 70% at 118 nm, the shortest wavelength of our measurements. It is believed that this decrease in transmission is due to argon itself and could be attributed to absorption by transitions corresponding to the analogue of the first argon excimer continuum. However, a residual unknown impurity, which in the presence of argon atoms causes this decrease, cannot be completely excluded. For an empirical description we fitted an exponential function (eq. [1]) with two parameters $a$ and $b$ to the data from $\lambda = 118$ nm to 140 nm. The parameter $b$ is a hint to the wavelength position of
strong resonant absorption by the analogue of the first argon excimer continuum.

\[ \frac{I(d, \lambda)}{I_0(\lambda)} = T(\lambda) = 1 - e^{-a(\lambda-b)} \]  

(1)

\[ I(d, \lambda) \text{ Light intensity after traversing a layer of thickness } d \]

For our case, \( d=5.8 \text{ cm} \), we obtain the following values for \( a \) and \( b \) together with their 1\sigma errors:

\[ a = (0.234 \pm 0.014) \text{ nm}^{-1} \]  

(2)

\[ b = (113.02 \pm 0.65) \text{ nm} \]  

(3)

The transmission data and the fit (red line) are shown in fig. 5 (upper panel). The dashed (red) lines correspond to the two extreme combinations of the 1\sigma errors of the fit parameters \( a \) and \( b \). The experimental method as described in section 2.2 leads to transmission values slightly above unity around \( \lambda=145 \text{ nm} \). This is caused by a systematic error, which cannot be resolved until a measurement of the wavelength dependent refractive index of liquid argon is available to provide a correct reflection value at the surface between liquid argon and the MgF\(_2\) windows (Fresnel correction). Consequently, the presented transmission curve should rather be seen as an upper limit for the real transmission. However, the decrease towards the short wavelength end of the presented transmission spectrum (see fig. 6) is visible in all measurements and cannot be attributed to a systematic error. This decrease is probably even underestimated, due to the optimistic transmission scaling described above. The empirical description for the wavelength dependence of the transmission (eq. 1) is now used to calculate the wavelength dependent attenuation lengths \( \lambda_{\text{att}}(\lambda) \). The attenuation of light, traversing a medium can be described by an exponential decay\(^3\) at each wavelength \( \lambda\):

\[ I(d, \lambda) = I_0(\lambda) \cdot e^{-\frac{d}{\lambda_{\text{att}}(\lambda)}} \]  

(4)

\[ \lambda_{\text{att}}(\lambda) = \frac{d}{\ln \left( \frac{I_0(\lambda)}{I_0(\lambda)} \right)} \]  

(5)

Equation 5 is used to calculate the wavelength dependent attenuation lengths \( \lambda_{\text{att}}(\lambda) \) of the light for distance \( d \). Using eq. 6 the wavelength dependent attenuation length was calculated. The result is shown in fig. 7. Note that no unique attenuation length can be given for liquid argon in the VUV spectral range. However, combining a source spectrum with the transmission data, the wavelength integrated intensity versus distance traversed by the light in liquid argon can be calculated. This is demonstrated using the important example when fluorescence light of argon itself (fig. 6 lower panel) is attenuated and thereby modified in its spectral shape during its path through liquid argon. The modification of the spectral shape with distance is calculated using eq. 4 with the obtained attenuation lengths \( \lambda_{\text{att}}(\lambda) \) from eq. 6 and is depicted in fig. 8. It shows the “reddening” of VUV light propagating in liquid argon just like sunlight observed around sunset. Wave-length integrated data for the light flux \( F(x) \) versus distance is calculated. The result is shown in fig. 9 along the path of light using eqs. 7 and 8 thereby taking the modification of the spectrum into account:

\[ F(x) = \frac{\int_{118 \text{ nm}}^{140 \text{ nm}} I(x, \lambda) \, d\lambda}{\int_{118 \text{ nm}}^{140 \text{ nm}} I_0(\lambda) \, d\lambda} \]  

(7)

\[ = \int_{118 \text{ nm}}^{140 \text{ nm}} \frac{I_0(\lambda) \cdot e^{-\frac{x}{\lambda_{\text{att}}(\lambda)}} \, d\lambda}{\int_{118 \text{ nm}}^{140 \text{ nm}} I_0(\lambda) \, d\lambda} \]  

(8)

\(^3\) Equation 4 neglects any potential fluorescence-light emission. This is expected to be justified since the emission of the deuterium lamp is weak below about 130 nm and the distance between the inner cell and the detector is large (\( \sim 45 \text{ cm} \)).
Fig. 6. The transmission of well purified and distilled liquid argon (upper panel) is shown in comparison with the emission of liquid argon (lower panel) as taken from ref. [13]. The red line (upper panel) shows the exponential fit according to eq. 1. The dashed red lines correspond to the two extreme combinations of the 1σ errors of the fit parameters a and b (see text).

Fig. 7. Wavelength-dependent attenuation length calculated from the fit in fig. 6. The shaded area represents the error margins corresponding to the dashed lines in fig. 6. Above 140 nm the calculated attenuation length diverges due to the transmission approaching unity.

The results are shown in fig. 8. As mentioned above no unique attenuation length exists in liquid argon. The flux decrease with distance depends on the combination of the spectrum at the starting point and its modification by the wavelength dependent attenuation, and has to be calculated for each wavelength independently. Wavelength integrated values have to be obtained by integrating the spectrum afterwards and comparing this integral with the integral of the spectrum at the starting point. The flux reduction is not exponential as the comparison with an exponential function in fig. 8 shows. The nominal 1/e value with respect to the initial flux of the argon fluorescence light is 1.63 m.

3.2 Effect of xenon impurities

Although the 58 mm length of the absorption cell was too short for measuring reliable transmission values for wavelengths longer than about 130 nm it had the right length for studying the effect of impurities and gas handling. The
The transmission of chemically purified but not distilled liquid argon (upper panel) is shown in comparison with the fluorescence spectrum of liquid argon (lower panel [13]). The two pronounced absorption dips at 126.5 and 141.0 nm are related to xenon impurities, as described in detail in ref. [14]. Note that the xenon absorption feature at 126.5 nm is in almost exact coincidence with the peak of the argon emission spectrum.

Note that the xenon absorption feature at 126.5 nm is in almost exact coincidence with the peak of the argon emission spectrum. The transmission spectra shown in figs. [6] and [10] could only be observed after a series of about 20 measuring campaigns with stepwise improved gas handling. Here we follow this way backwards to emphasize the most critical issues by describing them first. Already in the emission spectra of ref. [13] it was found that it is difficult to remove other rare gases from argon, xenon in particular. Why krypton never played a role in our experiments cannot be answered by the authors, so far. In general, xenon (or may be in some cases krypton) may be contained in some gas bottles, or it may be released from components in the gas system if it had been used with xenon or krypton prior to argon measurements. Poly-tetra-fluor-ethylene (PTFE, Teflon) is one of the materials, which is critical because it is often used for gaskets, e.g. in the connector of the pressure reducer to the gas bottle. Measurements exposing a 1 mm thick Teflon sheet of 16 g weight for 1 h to 2 bar xenon led to an increase of 25 mg in weight, which was released again with a time constant of 2 to 3 days when it was again exposed to ambient air. This behaviour is also in agreement with the observation that another rare gas pressure reducer, used on a xenon bottle more than a year ago, still led to xenon impurities of more than 50 ppm even after several purgings.

The problem is that xenon (or krypton if it were contained in a gas system) cannot be removed by regular rare gas purifiers because they make use of chemical reactions and can therefore not act on rare gases. The situation is even more critical in experiments with a cryogenic cell because xenon (or krypton) impurities will condense in the experimental cell with time from all parts of the gas system at higher temperatures. Absorption features which could be attributed to xenon were indeed observed in the experiments described here. In fig. [10] (upper panel) the transmission of a liquid argon sample is shown for which the conventional chemical purification had been applied but the gas was not distilled before being used for the measurement. The fluorescence of liquid argon is again shown for comparison (lower panel). This demonstrates that there is, unfortunately, an almost exact coincidence between one of the xenon absorption features at 126.5 nm and the peak of the argon emission spectrum. For the assignment of the absorption dips a comparison of the absorption spectrum with the emission spectrum of our unpurified argon gas is shown in fig. [11]. A low-energy electron beam excitation method described in ref. [24] and references therein was used to record the emission spectrum in the gas phase. The resonance lines of xenon at 146.96 and 129.56 nm [17] are clearly visible (see fig. [11] lower panel). According to ref. [14] the pronounced absorption dip at 141.0 nm can be attributed to a perturbed atomic transition of xenon ($^1S_0 \rightarrow ^3P_1$, 146.96 nm). The second absorption dip at 126.5 nm, however, cannot be assigned to a perturbed atomic transition but rather to a trapped exciton (Wannier-Mott type) impurity state [14]. The widths of the absorption bands are 2.0 and 3.8 nm, respectively. Xenon is therefore a very critical impurity, which can as a pure rare gas be easily underestimated in practical applications.

The concentration of xenon in the argon gas used in the present experiments could be estimated from emission measurements performed in the gas phase (fig. [11] lower panel) to be smaller than 3 ppm. This is obtained from a comparison with data shown in ref. [25]. The concentration of xenon could also be estimated from a comparison with measurements by Raz and Jortner [13]. There, the optical density of liquid argon doped with 3 ppm of xenon was
measured in a wavelength range from 120 nm to 160 nm, and for an optical path length of 1 cm. The transmission minimum at 141.0 nm is also clearly visible. A recalculation of their data to an optical path length of 5.8 cm and a subsequent comparison of the areas of the transmission minima at 141.0 nm lead to a concentration of approximately 140 ppb xenon for the undistilled argon sample used in our experiment. The effect of the fractional distillation can also be estimated. A comparison of the area of the small transmission minimum in pure liquid argon at 141.0 nm (fig. 6 upper panel), which is still visible but reduced compared to the undistilled liquid argon (e.g. fig. 10 upper panel) leads to a concentration of approximately 7 ppb. Consequently, the xenon contained in the argon was reduced by a factor of approximately 20 due to fractional distillation.

3.3 Absorption in unpurified liquid argon

A transmission spectrum of liquid argon as taken directly from the gas bottle (Ar 5.0, i.e. a nominal purity of 99.999%) is shown in fig. 12. Besides the two absorption bands at 141.0 and 126.5 nm, which have been attributed to a xenon impurity, a broad absorption is observed below 180 nm with a minimum transmission of about 45% at 145 nm. The absorption/emission structure around 121 nm may be due to hydrogen released from the water or an artifact related to the Lyman-α line of hydrogen emitted by the deuterium lamp. The broad absorption between about 125 and 180 nm is believed to be mainly due to water vapor contained in the argon gas, which has not been circulated through the rare gas purifier. The temperature dependent absorption of solid water in the VUV had been studied in refs. 26, 27.

It has to be noted that the transmission shown in fig. 12 was only reached when the system had already been used for a long time with many purification cycles with the rare gas purifier. In the beginning of the experiments the transmission reduced towards shorter wavelengths starting at 180 nm reaching zero at about 150 nm. This means that chemical gas purification is mandatory for fluorescence detectors using liquid argon as detector material.

Finally, it should be noted that the transmission measurement had in one test experiment been extended to the visible spectral range. The data are not easy to interpret because of the higher diffraction orders of the grating in the VUV monochromator. However, since all structures in the spectra appeared at wavelength positions which could be related to first order structures we can conclude that no significant absorption exists in liquid argon in the longer wavelength UV and visible spectral range.

4 Summary

It has been shown that the light propagation in liquid argon, which is frequently and in large quantities used for particle detection, is strongly wavelength dependent. However, a purification level can be reached where an effective attenuation length on the order of 1.6 m can be obtained for its own fluorescence emission. If much longer propagation lengths are possible will become clear when experiments with higher accuracy will be performed, using longer absorption cells and improved light sources.

We found a systematic reduction of transmission in liquid argon from about 140 nm towards shorter wavelengths and attribute this to absorption in argon by transitions which are called the "first continuum" in emission. Note that the atomic and molecular structure of the heavier rare gases neon through xenon are so similar that essentially all effects measured for one of the species can also be found in the other ones. Therefore, an absorption feature analogous to the one observed here must also be expected, e.g., in particle detectors using liquid xenon as the scintillating material.

Special care has always to be taken to avoid impurities. This is common knowledge as far as regular chemical rare gas purification is concerned. In the present paper it is shown that other rare gases, xenon in particular, lead to absorption bands, which in the case of xenon perfectly coincide with the emission band of argon. Therefore, it is important to test liquid argon used as scintillation material for the presence of this impurity.

5 Outlook

It may be worthwhile to explore the region near the wavelength cutoff of the present experiment (120 to 130 nm) with better statistics. A light source described in ref. 28 emitting strongly in this region could be used for that purpose. Experiments with synchrotron radiation could also be considered 29. Critical opalescence (in dense gases),
Rayleigh scattering (in general), and fluctuations of the index of refraction driven by temperature gradients etc. (in the liquid phase) will have to be considered in experiments with longer optical path lengths than that used in the present experiment.

Xenon and krypton added deliberately in well defined quantities could provide information about the absorption features of mixed Ar-Xe and Ar-Kr molecules and their absorption spectra. Absorption and emission experiments in the liquid and the gas phase could be performed for such a study. As an experimental improvement cryogenic baffles in front of the inner cell and cooled, e.g., to liquid helium temperature should be installed to avoid the degradation of the transmission of the inner cell’s optical windows by condensation of impurities from the insulating vacuum surrounding the inner cell.

Finally, we are aware of the fact that a setup in which monochromatic light would be used instead of the broadband emission of a deuterium lamp would be superior to the setup presented here because possible problems with fluorescence induced by the probe light could then be avoided. The light intensity at the exit of a monochromator, however, would have been too weak and the light would have had a divergence related to the aperture ratio of the monochromator, which would have required a much more complex optical setup. The issue of fluorescence will be tested in a separate experiment by trying to observe light through an additional window of the inner cell through which scattered or reemitted light can be observed at 90° with respect to the incident light.

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