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Special Article - Tools for Experiment and Theory

Spectroscopic study of light scattering in linear alkylbenzene for liquid scintillator neutrino detectors

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Abstract We have set up a light scattering spectrometer to study the depolarization of light scattering in linear alkylbenzene. The scattering spectra show that the depolarized part of light scattering is due to Rayleigh scattering. The additional depolarized Rayleigh scattering can make the effective transparency of linear alkylbenzene much better than expected. Therefore, sufficient scintillation photons can transmit through large liquid scintillator detector, such as that of the JUNO experiment. Our study is crucial to achieving an unprecedented energy resolution of $3 \% / \sqrt{E (MeV)}$ required for the JUNO experiment to determine the neutrino mass hierarchy. The spectroscopic method can also be used to examine the depolarization of other organic solvents used in neutrino experiments.

1 Introduction

Organic scintillators have played a key role in the 60-yearlong research history of neutrinos [1]. They were chosen as the targets in the liquid scintillator detectors where antielectron neutrinos were first discovered by Cowan and Reines [2], in the KamLAND experiment where the large mixing angle (LMA) solution of the solar neutrino problem was confirmed [3], in the Daya Bay experiment where the neutrino oscillation mixing angle θ_{13} was precisely measured [4], and in the Borexino experiment where the spectrum of protonproton neutrinos was recently observed [5]. Because of the unexpectedly large value of θ_{13} , the neutrino mass hierarchy and the leptonic *CP*-violating phase can be measured in the next generation of neutrino oscillation experiments [6].

Recently, the Jiangmen Underground Neutrino Observatory (JUNO) experiment has been approved, aiming to measure the neutrino mass hierarchy using an organic liquid scintillator detector [7]. There will be 20 kilotons of liquid scintillator filling the huge spherical central detector of JUNO, whose diameter will be about 35 m. A ternary hydrogenous organic liquid is chosen as the detector target in JUNO, with the solvent being linear alkylbenzene (LAB), whose formula is $C_6H_5C_nH_{2n+1}$ (n = 10–13), and the primary and secondary solutes being 2,5-diphenyloxazole (PPO) and 1,4bis[2-methylstyryl]benzene (bis-MSB), respectively. Reactor anti-neutrinos can be detected via the inverse beta-decay reaction $\bar{\nu}_e + p \rightarrow e^+ + n$. Each positron will rapidly annihilate with an electron and produce a pair of γ -rays. Each neutron will be finally captured by a proton and release a γ -ray. The γ -rays will product scintillation in the LAB. The solutes, PPO and bis-MSB, will transfer the wavelengths of scintillation to longer wavelength to avoid self-absorption by the LAB [8]. In the discussion of light transmission in the liquid scintillator, the reference wavelength is usually chosen as 430 nm, around which the intensity of the scintillation spectrum is at a maximum.

The JUNO experiment will determine the neutrino mass hierarchy by measuring a very precise and high-statistics reactor anti-neutrino energy spectrum [9]. The method demands an unprecedented energy resolution of $3\%/\sqrt{E(\text{MeV})}$, i.e., about 1200 photon electrons per MeV [7]. The large scale of the central detector will make scintillation photons transmit several tens of meters before they finally arrive at the photomultipliers and then transfer into photon electrons. The photons attenuate exponentially when they transmit in a liquid. The detection challenge requires that the solvent of the liquid scintillator, LAB, should be highly



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transparent to scintillation photons [6]. After purification, LAB can have a large attenuation length which is about 20 m at 430 nm [10]. Even if the scintillation photons vanish exponentially after transmitting 20 m, the energy resolution can still hardly reach the requirement of the JUNO experiment. Attenuation contains two sub-processes, scattering and absorption [11]. Since the liquid scintillator should be dustfree, the scattering process of LAB should be Rayleigh scattering. Because Rayleigh scattering only changes the directions of photons, the scattered photons still have a chance to be detected in a 4π detector. It has been found that Rayleigh scattering can increase the effective transparency of scintillation in large liquid scintillator detectors [12]. Simulations have shown that if the Rayleigh scattering length of LAB at 430 nm is 30 m, the energy resolution of the central detector of JUNO can satisfy the requirement for measurement of the neutrino mass hierarchy [13].

Light scattering in several organic solvents, including LAB, has been studied experimentally [14]. It has been found that light scattering in LAB is depolarized at 90°. It is well known that the Rayleigh scattering of light by small isotropic dielectric spheres at 90° is completely polarized [15]. The Rayleigh scattering length of LAB will be about 40 m at 430 nm if only the polarized part of light scattering is considered. The depolarized part of light scattering in LAB has been treated as a result of absorption/reemission [14]. Because 430 nm is far from the absorption band of LAB [8], the weak absorption including the absorption/reemission of LAB at 430 nm is due to the optical purities [16,17]. Recently, it has been argued that the depolarized part of light scattering in LAB is also Rayleigh scattering because of molecular anisotropy [18]. Then the depolarization of light scattering in LAB is an intrinsic property of LAB. A significant difference between the depolarized Rayleigh scattering and absorption/reemission is that the wavelengths of the deflected photons will remain in the former process and change in the latter [19]. Whatever the depolarized part of light scattering is, the total scattering length of the LAB in the previous experiment [14] will shorten to about 30 m at 430 nm because the quantum efficient of photomultipliers is typically flat around 430 nm. However, optical purities in LAB of different productions or purifications are various in types and amounts. The LAB in the JUNO experiment may not have the same absorption/reemission as the LAB in the previous experiment. Therefore, the total scattering length of LAB in the JUNO experiment will be guaranteed to be less than 30 m at 430 nm [18] only if the depolarized part of light scattering is Rayleigh scattering.

To distinguish whether the depolarized part of light scattering in LAB is Rayleigh scattering or absorption/reemission is crucial for the JUNO experiment to achieving the unprecedented energy resolution of $3 \%/\sqrt{E (\text{MeV})}$ to measure the neutrino mass hierarchy. In this paper, we studied spectroscopically the depolarization of light scattering in LAB. The experimental setup used for the scattering spectrum measurements is described in Sect. 2. Results and discussions of the scattering spectra and the depolarization of LAB are presented in Sect. 3. A summary is given in Sect. 4.

2 Experimental setup

A schematic diagram of the light scattering spectrometer is shown in Fig. 1. The light source was a commercial Spectra-Physics BeamLok 2060 argon ion continuous-wave laser. The beam from the laser had a divergence of 0.45 mrad with a beam diameter of 1.7 mm. The light beam supplied by the laser was vertically polarized with the extinction ratio better then 100:1.

The beam first passed through a circular continuously variable reflective neutral density filter which can adjust light intensity continuously. The beam was then collimated by two reflection mirrors and two apertures. Between the mirrors and apertures, a beam splitter was used to split the beam into two parts. One part of the beam remained on the original path and the other, perpendicular to the original path, was received by a power meter to monitor the stability of the laser beam. Since the optical components would induce spurious depolarization, a Glan-Laser calcite polarizing prism was put behind the splitter to improve the extinction ratio to be better than 5000:1. After passing through two apertures the laser beam enter a cell whose light path was 50 mm. The cell was of strain-free fused quartz of very high optical quality. The cell was mounted on an optical bench whose height could be adjusted for final alignment of the cell with respect to the incident beam. This alignment was not changed during the experiments. The cell, which could be emptied or filled with LAB, was held firmly in position during measurements by a removable spring yoke. The attenuation length of the batch of LAB sample was about 20 m [10].

Following the cell, two apertures, with holes of diameter 1 mm normal to the laser beam, served to limit the field of view of the spectroscope and prevent stray light from enter-



Fig. 1 A schematic diagram of the light scattering spectrometer

ing the spectroscope. The first aperture was 65 mm behind the cell. The second aperture was 790 mm after the first aperture. A Glan–Thompson calcite polarizer was mounted on a motorized precision rotation mount (Thorlabs PRM1/MZ8) 115 mm behind the second aperture. The scattered photons entered the slit of a Shamrock SR-500i grating monochromator which was 53 mm behind the calcite polarizer. The light was finally collected by an Andor Newton EMCCD DU971P CCD detector. The temperature of the CCD detector was cooled to -75 °C to decrease noise. The experiment was held in a clean room with room temperature controlled at 21 ± 1 °C. The scattered photons were collected horizontally and vertically separately by the Glan-Thompson calcite polarizer. The horizontal and vertical scattering spectra were given by the spectroscope with the CCD detector.

3 Results and discussion

Firstly, the output wavelength of the argon ion laser was set to 488 nm and the cell was empty. The slit width was set to 0.2 mm and then the spectroscopic resolution would be about 1 nm. In order to make the laser operate stably, the output power was set to the optimized one which was approximately 300 mW at 488 nm. The output beam power was so high that for safety a reflective neutral filter was used to reduce the input laser intensity to the organic sample. The filter reflected about 99.6% laser light. Although a light shield was constructed from the cell to the CCD detector, there were still peaks around 488 nm in the spectra caused by stray light. The vertical and horizontal scattering spectra for the empty cell with an integration time of 5 s are shown in Fig. 2. The maximum intensities of both spectra were near to each other because the stray light had been reflected so many times that it would be almost unpolarized. The flat intensities of scattering spectra away from 488 nm were caused by the CCD detector noise.

The cell was then filled with LAB without any movement. The vertical and horizontal scattering spectra for LAB with an integration time of 5 s are shown in Fig. 3. Because of light scattering from LAB, the intensities of peaks were larger than those for the empty cell. All the wavelengths of peaks in the scattering spectra in Figs. 2 and 3 were exactly the same. Thus it can be concluded that the scattered photons from LAB are caused by Rayleigh scattering.

In the light of the above spectroscopic study, the peak background in the spectra caused by stray light has to be suppressed for quantitative analysis of the depolarization of Rayleigh scattering in LAB. The output wavelength of the argon ion laser was then set to 457.9 nm so that the output power could be lowered to approximately 50 mW. Stray light in the lab was successfully suppressed. However, very few scattered photons could be collected by the spec-



Fig. 2 a The vertical scattering spectrum and **b** the horizontal scattering spectrum of the empty cell for 488 nm incident laser beam. The slit width was 0.2 mm and the integration time of the CCD was 5 s



Fig. 3 a The vertical scattering spectrum and b the horizontal scattering spectrum of LAB for 488 nm incident laser beam. The slit width was 0.2 mm and the integration time of the CCD was 5 s

troscope because of the 0.2 mm narrow slit. The slit width was then widened to 1 mm and the integration time of the CCD increased to 20 s. The widened slit lowered the spectroscopic resolution to about 5 nm. The fluorescence spectra of a mixture of LAB and bis-MSB have shown that the spectroscopic properties of the Rayleigh scattering of LAB and the absorption/reemission of bis-MSB are totally different [19]. Thus if there was absorption/reemission, it can still be distinguished from Rayleigh scattering in the scattering spectra. The vertical and horizontal scattering spectra of the empty cell are shown in Fig. 4. The horizontal scattering spectrum was almost flat, while there was only a very small peak around 457.9 nm in the vertical scattering spectrum which might be caused by light scattering from air or cell.

The vertical and horizontal scattering spectra for LAB are shown in Fig. 5. There are obvious peaks around 457.9 nm in the vertical and horizontal scattering spectra. The depolarization of Rayleigh scattering in LAB can be characterized by



Fig. 4 a The vertical scattering spectrum and **b** the horizontal scattering spectrum of the empty cell for 457.9 nm incident laser beam. The slit width was 1 mm and the integration time of the CCD was 20 s



Fig. 5 a The vertical scattering spectrum and **b** the horizontal scattering spectrum of LAB for 457.9 nm incident laser beam. The slit width was 1 mm and the integration time of the CCD was 20 s

the depolarization ratio [20]. The directly measured quantity from the scattering spectra is

$$\rho_v = \frac{H_v}{V_v},\tag{1}$$

where H_v and V_v are the horizontal and vertical intensities of the scattered photons from a vertically polarized incident beam. The depolarization ratio ρ_u of an unpolarized incident beam is defined as

$$\rho_u = \frac{H_v + H_h}{V_h + V_v},\tag{2}$$

where H_h and V_h are the horizontal and vertical intensities of the scattered photons from a horizontal polarized incident beam. When the scattering angle is 90°, $H_v = H_h = V_h$ and then the depolarization ratio ρ_u can be obtain by

 Table 1
 Depolarization ratio of LAB

$ \rho_v $ (457.9 nm) Present work	$ \rho_u $ (457.9 nm) Present work	$ \rho_u (430 \text{ nm}) $ Ref. [18]	$ \rho_u (405 \text{ nm}) $ Ref. [22]
0.18 ± 0.01	0.30 ± 0.02	0.31 ± 0.04	0.31 ± 0.02

$$\rho_u = \frac{2\rho_v}{1+\rho_v},\tag{3}$$

which is the specific result for Krishnan's relations at the 90° scattering angle [21].

The scattering intensities H_v and V_v can be obtained from the scattering spectra by subtracting the noise background. The results derived from Eqs. (1) and (2) are listed in Table 1. The scattering spectra of LAB were measured three times. The statistical uncertainty of the depolarization ratio is about 3%. The bidirectional repeatability of the polarization angle controlled by a precision rotation mount is $\pm 0.1^\circ$ and it causes an error of 1%. The stability of the laser beam was monitored by a power meter during the measurement and the instability of the laser beam causes an error of 1%. Therefore, the depolarization ratio of LAB is measured to be 0.30 ± 0.02 at 457.9 nm.

Recently the depolarization ratio of LAB at 405 nm has also been measured independently giving a result of 0.31 \pm 0.02 [22]. From Table 1, the depolarization ratios of 405, 430 and 457.9 nm for LAB are the same within 1 σ . It is well known that the depolarization ratio is a weak function of wavelength over the visible range [20,23]. Therefore, we can conclude that the depolarized part of light scattering in LAB is Rayleigh scattering even if we can not directly study the depolarization of light scattering of LAB at 430 nm, the main region of interest, due to the limitation of light source. Then the total light scattering length of Rayleigh scattering in LAB should be less than 30 m at 430 nm [18].

Experimentally, even gases can have very small depolarization ratios due to molecular anisotropy [24]. The light scattered by liquids is generally depolarized [25,26]. Polarized Rayleigh scattering has been used to approximately describe light scattering in water Cherenkov detectors [11,27] and rare-gas liquid detectors [28] because the depolarization ratio of water and rare gas liquids are small. However, the depolarization of Rayleigh scattering in LAB is too large to be ignored. The typical fluorescence time of absorption/reemission for liquid scintillators is several nano-seconds to which large detectors are sensitive [29,30]. To treat the depolarized part of Rayleigh scattering as absorption/reemission would increase the time response in simulations of large scintillator detectors and might affect the spatial resolutions and the detection efficiencies [31,32]. Aside from the JUNO experiment [9], LAB has been used in the Daya Bay experiment [33], RENO experiment [34] and the upcoming SNO+ experiment [35,36]. It would also be used in the planned LENA [37] and Hanohano observatories [38].

The previous scattering experiment has also shown that the light scatterings in dodecane, pseudocumene (PC) and phenylxylylethane (PXE) are also depolarized [14]. PC is currently used as the solvent in the Borexino experiment [32,39] and PXE has been investigated as a back-up solution for the Borexino experiment [40]. The solvent used in KamLAND experiment consists of 20 % PC and 80 % normal dodecane (ND) [12]. The diameters of the liquids scintillator detectors in Borexino and KamLAND are 8.5 and 13 m, respectively. If the depolarized parts are also Rayleigh scattering, the scattering lengths of PXE and PC will shorten to about 10 m and the scattering length of the mixture of PC and ND will be also shorter than expected. The spectroscopic method can be used for the Borexino and KamLAND experiments to investigate whether the depolarization of light scattering in PXE, PC and ND are due to Rayleigh scattering or absorption/reemission.

4 Summary

A light scattering spectrometer has been set up to investigate the depolarization of light scattering in LAB. It has been shown that the depolarized part of light scattering in LAB is due to Rayleigh scattering. The additional depolarized Rayleigh scattering can increase the effective transparency of LAB. Our study will help the JUNO experiment to achieve the unprecedented energy resolution of $3\%/\sqrt{E(MeV)}$ to determine the neutrino mass hierarchy measurement. Our result is also useful to other neutrino experiments which use LAB as the solvent for their liquid scintillators detectors. The spectroscopic method can be also used to test the depolarization of PXE, PC and ND for the Borexino and KamLAND experiments.

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