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Present status of liquid rare gas scintillation detectors and their new application to gamma-ray calorimeters

Tadayoshi Doke[a,*], Kimiaki Masuda[b]

[a] *Kikuicho-Branch, Advanced Research Center for Science and Engineering, Waseda University, Kikuicho-17, Shinjuku-ku, Tokyo 162-8555, Japan*

[b] *Solar-Terrestrial Environment Laboratory, Nagoya University, Nagoya 464-8601, Japan*

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Abstract

Scintillation from liquid rare gases has a fast decay component with a high photon yield which is comparable to that of NaI(Tl), although the wavelength is in the vacuum ultraviolet. On the basis of these properties, many investigators have tried to realize liquid rare gas scintillation detector. In particular, liquid Xe is expected to be an excellent detector medium for γ-rays because of its fast response, its large atomic number, and high density. However, it is very difficult to achieve sufficient light collection as scintillator, because the light emission is in the vacuum ultraviolet. Recently, some groups have reported liquid Xe/Kr homogeneous scintillation calorimeters for high-energy γ-rays using reflectors method, but their energy resolution was not as high as expected. In this situation it seems that the realization of liquid rare gas detectors may not be easy. In this paper, we review the physical properties of scintillation from liquid rare gas, the scintillation yields, the decay time constants and attenuation lengths. We summarize the results obtained with liquid rare gas scintillation detector. The present status of development of liquid Kr/Xe homogeneous scintillation calorimeters by using reflected light is described. A new device which observes only direct light is proposed as high-energy γ-ray detector or homogeneous electromagnetic shower calorimeters for γ-rays. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Liquid rare gas scintillation detector; γ-ray calorimeter; Scintillation yield; Decay constant; Attenuation lengths

1. Introduction

Since the successful application of liquid Ar sampling calorimeters in physics experiments, which were initiated by Knies and Neuffer [1], Engler et al. [2], and independently by Willis and Radeka [3] in 1974, great progress has been made in the field of liquid rare gas detectors. In particular, since 1977, many new devices based on rare gas liquids have been proposed in the fields of elementary particles and astrophysics experiments. These devices can be roughly categorized into two types as follows:

(1) Homogeneous liquid rare gas calorimeters.

(2) Liquid Ar or liquid Xe time projection chambers (TPC).

* Corresponding author. Tel.: + 81 3 3203 4363; fax: + 81 3 3203 3231; e-mail: tdoke@mn.waseda.ac.jp

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The homogeneous calorimeters in category (1) may be subdivided into the following two modes; (i) ionization mode, including photoionization, and (ii) scintillation mode. For both modes, an excellent energy resolution is expected [4]. The idea of category (2) was originally proposed by Rubbia for liquid Ar detectors [5], and now this technique is being successfully applied to liquid xenon detectors [6–8].

Although liquid rare gas TPC techniques are still under development, homogeneous calorimeters operated in ionization mode have already been developed and successfully applied to some elementary particle experiments [9–17]. The homogeneous calorimeter operated in scintillation mode is not yet applied in scientific studies, although many physicists working in the field of elementary particle experiment are interested in this type of detectors. The first proposal of a liquid Xe homogenous scintillation calorimeter was presented for an experiment of the SSC project by Chen [18]. The prototype models have been tested by some groups, but the expected energy resolution has not been achieved so far as described in Section 4. This review will focus on the present status of developments of liquid rare gas scintillation detectors including homogeneous electromagnetic shower calorimeters.

In this paper, first, the major properties of scintillation from liquid rare gases are summarized, and then, the progress of investigations on γ-ray spectrometers by using liquid rare gases is reviewed. The current status of development of liquid rare gas homogeneous scintillation calorimeters is discussed and, finally, a new scintillation γ-ray detection device is proposed.

2. Physical properties of scintillation from liquid rare gases

The essential knowledge for using liquid rare gas as scintillation detector medium is: (1) their wave length, (2) photon emission yields, (3) scintillation decay time constants, and (4) attenuation length for emitted photons in the liquid. Here we consider only liquid Ar, Kr and Xe because they are the main detector media used. The wave lengths of photons emitted from these liquids are in the vacuum ultra-violet (VUV), 128 nm for liquid Ar, 147 nm for liquid Kr, and 174 nm for liquid Xe. The measurement of the VUV-light is not so easy. Accordingly, this becomes a disadvantage for the application of these detector media. In this section, the current investigations of the parameters other than wave length are described.

2.1. Scintillation photon emission yields

In the mid-1980s, the Waseda University–LBL collaboration group investigated the correlation between scintillation light and ionization produced in liquid Ar using relativistic ions from the Bevalac [19–21]. With the data obtained by this collaboration as well as data obtained previously, they summarized the LET dependence of scintillation yield in liquid Ar [22]. Fig. 1 shows the LET dependences of the scintillation yield in liquid Ar as well as that in liquid Xe [23]. The flat-top responses given by theoretical limits of scintillation yields are shown. The decrease with LET in the low LET region is due to "escape electrons" and that in the high LET region is due to the so-called "quenching" of scintillation light [22]. The figure also shows the response of NaI(Tl) which is similar to liquid Ar [25]. Based on these results, Doke et al. estimated the absolute photon yield, which can be

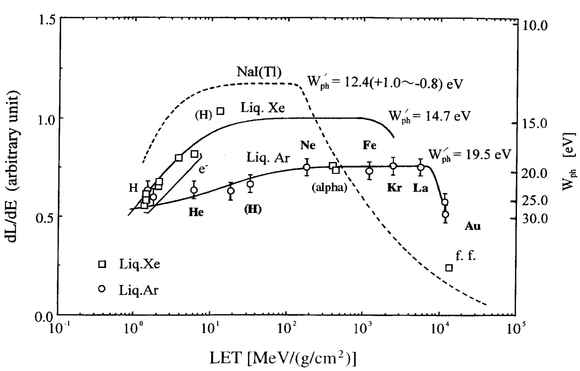

Fig. 1. LET dependence of scintillation yields in liquid Ar, liquid Xe and NaI(Tl). H, He, Ne, Fe, Kr, La and Au represent relativistic ions in the experiments, e^-, (H), (α) and (f.f.) represent 1 MeV electrons, 20 and 40 MeV protons, α-particles and fission fragments, respectively. The curve of NaI(Tl) was cited from the paper of Murray and Meyer [24].

expressed by a reciprocal of the average energy required for production of one photon by ionizing radiation, $1/W'_{ph}$, at the flat-top of the LET dependency of scintillation yield of liquid Ar or Xe [22,23,25]. Using these results, they also estimated W_{ph}-values in liquid Ar and Xe for relativistic electrons and α-particles [4,23,25]. Recently, Doke and Masuda evaluated W_{ph} -values in liquid Ar and Xe for relativistic electrons, α-particles, and relativistic heavy particles using the relations between ionization and scintillation measured at the same time and estimated the maximum possible errors [26]. The results mentioned above are listed in Table 1 together with other values experimentally obtained. In this table, W'_{ph} corresponds to the "intrinsic" W_{ph}-value when neither quenching nor "escape electrons" occur. The W'_{ph} value is given by the Eq. (1),

$$W'_{ph} = W/(1 + N_{ex}/N_i), \tag{1}$$

where W is the average energy required for production of an electron–ion pair, and N_{ex} and N_i are, respectively, the number of excited atoms and the number of ion pairs produced by the ionizing radiation. These "intrinsic" W_{ph}-values are applicable only to relativistic heavy ions from Ne to La. The values in parenthesis are those experimentally obtained by other groups [27–34]. The errors shown for our values in the table are not the maximum possible errors as estimated in Ref. [26], but reasonable estimates. For the other experimental values, the errors are given by the authors. The W_{ph}-values in NaI(Tl) for relativistic electrons recently measured by Miyajima et al. are shown in the table as Refs. [35,36]. For particles other than relativistic heavy ions mentioned above, the W_{ph}-value is affected by "escape electrons" and/or quenching processes. Accordingly, $W_{ph} = 14.2$ eV (or 12.5 and 12.3 eV) obtained for 100 keV electron beam in liquid Xe by Seguinot et al. [32,33] should not be compared with $W'_{ph} = 14.7$ eV estimated by the Waseda and LBL group, but with $W_{ph} = 24$ eV estimated for 1 MeV electrons which takes the effect of "escape electrons" into consideration [22,26]. If the value of 14.2 eV does not include any experimental error, it means that perfect recombination immediately occurs along the particle track as in the case of relativistic heavy ions. From the intensity and energy of the electron beam used in the experiment, it can be excluded that complete recombination immediately occurs under zero electric field. Seguinot et al. also observed the W-value of 9.76 eV in liquid Xe using the same method [32,33]. This value is also very small compared to the usual value (15.6 eV [37]) and is comparable to the band-gap energy (about 9.2 eV [38,39]) in liquid Xe. Theoretically, the ratio of the W-value and the gap energy should be larger than 1.5 because the W-value includes the energy losses due to sub-excitation electrons and also the collisions between electrons and the crystal lattice [37]. The ratio of 9.7 eV/15.6 eV is almost equal to that of 14.2 eV/24 eV. If the estimation of the energy deposition in the detection part of the experiment of Seguinot et al. is smaller than the actual one, it is easily explained why both these small values were

Table 1
Measurements and estimates of the average energy needed to produce a scintillation photon, W_{ph}(eV) in liquid Xe

Liquid rare gas	Relativistic electrons	α-particles	Relativistic heavy particles (W'_{ph} (eV))
Ar	25.1 ± 2.5	27.5 ± 2.8	19.5 ± 2.0
Xe	23.7 ± 2.4	19.6 ± 2.0	14.7 ± 1.5
	(< 35)[a]	(16.3 ± 0.3)[b]	
	(67 ± 22)[c]	(39.2)[d]	
	(29.6 ± 1.8)[e]		
	(14.2)[f], $(12.5, 12.3)$[g]		
	(42 ± 0.6)[h]		
NaI(Tl)	$(17.2 \pm 0.4, 16.5 \pm 0.4)$[i]		

[a]Ref. [27]; [b]Ref. [28]; [c]Ref. [29]; [d]Ref. [30]; [e]Ref. [31]; [f]Ref. [32]; [g]Ref. [33]; [h]Ref. [34]; [i]Refs. [35,36].

obtained in their experiment. In the design of electron collectors, generally we must strive to suppress the secondary electron emission from the collector. In the electron collector of their equipment, however, such a device could not be seen.

Recently, Ospanov and Obodovski measured the energy dependence of scintillation yields for soft X-rays and low-energy γ-rays. They obtained the variation of the specific photon emission yield (photon intensity per unit deposited energy) versus photon energy [40]. Although their result is not shown in Fig. 1, it is similar to that of NaI(Tl) [41] and is consistent with the results obtained by Barabanov et al. [42] which are shown in the low LET part of the curve of liquid Xe in Fig. 1.

In any case, the W_{ph}-values in liquid Ar and liquid Xe are almost comparable to that in NaI(Tl) crystal [35,36]. So, the number of photons emitted from liquid rare gases by high-energy electrons or γ-rays in the GeV region are sufficient to achieve excellent energy resolution. This consideration was proven by the Waseda–MIT–Columbia group using high-energy ^{27}Al ions. Namely, they achieved an excellent energy resolution, better than 0.5% (rms) for an energy deposition of 2.5 GeV [43]. In this case, however, the origin of scintillation light is highly localized. So, there is no non-uniformity problem in light collection. This situation is entirely different from the broad origin of scintillation in electromagnetic shower calorimeter.

2.2. Decay time constants

The scintillation light from pure liquid Ar, Kr and Xe has two decay components for α-particles or fission fragments due to de-excitation of singlet and triplet states of excited dimers ($R_2^* \rightarrow 2R + h\nu$). The decay shapes for electrons, α-particles and fission fragments in liquid Ar and Xe are shown in Fig. 2a, a′, and b [44] and that for electrons in liquid Kr in Fig. 2c [45]. However, it seems that the scintillation from liquid Xe for relativistic electrons has only one decay component (see Fig. 2b). This is due to the slow recombination between electrons and ions produced by relativistic electrons, since this component disappears if some electric field is applied. Fig. 3 shows the decay shape of scintillation light from liquid Xe under an electric field of 4 kV/cm, which consists of the usual two decay components [45]. In Table 2a and Table 2b, the decay time constants of scintillation light from liquid rare gases excited by 1 MeV electrons and α-particles are listed.

As seen from Fig. 2b, in liquid Xe, there is a clear difference in pulse shapes of scintillation excited by 1 MeV electrons and α-particles. Such a difference in pulse shapes by electrons and heavy particles might be used for particle identification or for improvement of the e/π ratio in hadron calorimeters [46].

Liquid Ar or liquid Kr doped with Xe is important as a scintillation detector medium. In these liquids, Xe_2^* is formed through the following energy transfer process;

$$\mathrm{Ar^* \text{ or } Kr^* + Xe + Xe \rightarrow Xe^* + Xe + Ar \text{ or } Kr}$$

$$\mathrm{\rightarrow Xe_2^* + Ar \text{ or } Kr,}$$

where the photon emitted has the same wavelength as that from pure liquid Xe. The formation of Xe_2^* is proven by spectroscopic measurements. Fig. 4 shows the variation of the emission spectra for the concentration of doped Xe in liquid and solid Ar [49]. Namely, the origin of the light emitted from Xe-doped liquid Ar or Kr is Xe_2^*. The variation as shown in Fig. 4 is energetically possible in Xe-doped liquid Kr, too. Recently, Russian and Italian groups observed the expected decay shapes of scintillation in liquid Kr + Xe (1.7%) [50] and in liquid Ar + Xe (0.01% $\sim$ a few %) [51]. Thus, the scintillation from Xe-doped liquid Ar or liquid Kr has a fast decay shape without any slow component as seen in pure liquid Ar or Kr and, in addition, the possibility of the contamination by electronegative molecules is very small compared with the case of N_2 or CO_2 doping. Thus, they consider that Xe-doped liquid Ar or Kr is an excellent detector medium for scintillating calorimeters [52].

2.3. Attenuation length

Until recently an attenuation length of scintillation light longer than 1 m could not be achieved in liquid rare gases [53]. In 1992, Waseda and IETP groups found independently the attenuation length of scintillation light longer than 1 m in liquid Xe

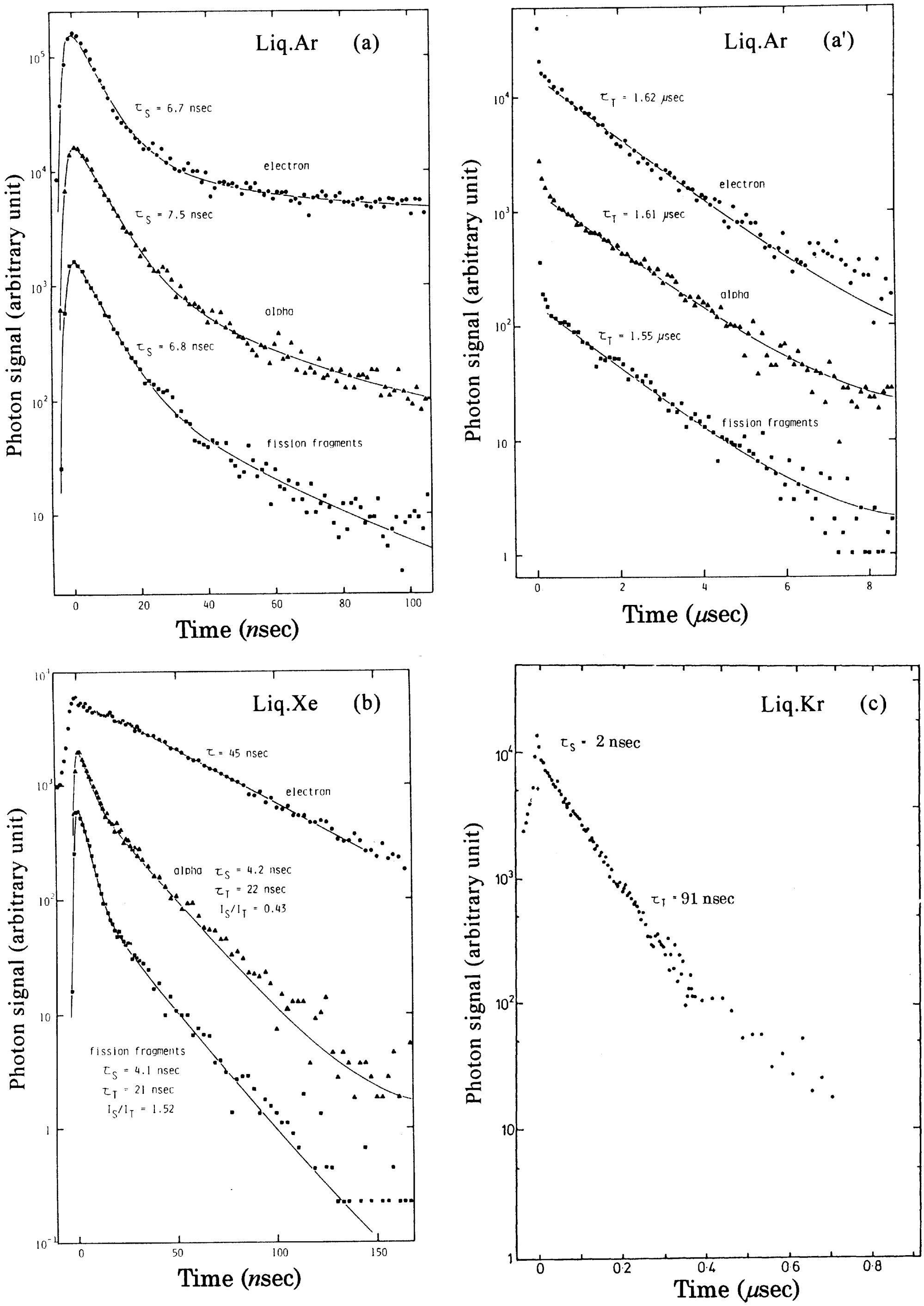

Fig. 2. (a,a′) Decay curves obtained for the scintillation light from liquid Ar excited by electrons, by α-particles and fission fragments for the short time range (a) and for the long time range (a′). A slightly slow rise for electron excitation in Fig. 2a may be due to recombination. (b) Decay curves obtained for luminescence from liquid Xe excited by electrons, α-particles and fission fragments. The slow decay in the electron excitation is due to electron–ion recombination. (c) Decay curves obtained for the scintillation light from liquid Kr excited by electrons.

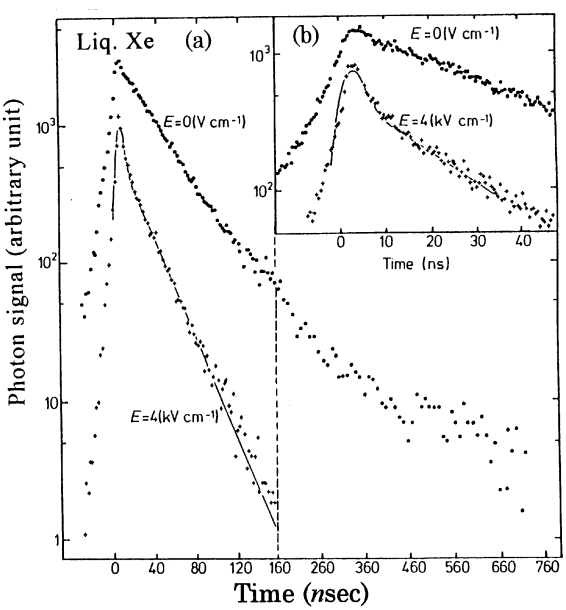

Fig. 3. Decay curves obtained for the scintillation light from liquid Xe with and without an electric field. The long time range (a) and the short time range (b) are indicated. Note the change of time scale at 160 ns in (a).

and Kr [54,55]. In both measurements, a flat black reflector surface (aluminum plate) was used to define the effective solid angle of scintillation light for the photomultiplier. Most recently, however, Ishida et al. found that such a black reflector (Al_2O_3) slightly reflects ultraviolet light and as a result an apparent attenuation length longer than 1 m was observed in liquid Xe. To make the reflectivity negligibly small, they tested black reflectors with irregular surfaces as shown in Fig. 5 [56]. Using such reflectors, they measured again the attenuation length of scintillation in liquid Xe and obtained an attenuation length of about 30 cm which is almost the same as the other data recently reported [29,53]. In addition, they found the attenuation length longer than 1 m in liquid Ar + Xe (3%) and liquid Kr + Xe (3%) using the same reflector, although it is about 60 cm in pure liquid Ar and about 80 cm in pure liquid Kr [56]. These results are shown in Table 3. The reason why an attenuation length in Xe-doped liquid Ar or Kr is longer than that in pure liquid rare gas is not clear, but we think that this tendency may be explained by assuming that the attenuation length of scintillation light is reduced by Rayleigh scattering [56]. If so, the short attenuation length in liquid Xe is not problematic for light collection.

In any case, it is certain that Xe-doped liquid Ar or Xe-doped liquid Kr has an attenuation length longer than 1 m. These mixtures should be very useful for scintillation calorimetry because these liquids have no slow recombination component as found in pure liquid Ar.

3. Usual scintillation detectors

The use of the scintillation light from liquid rare gases was considered to show a fast response compared with that in ionization mode. At first, the idea of scintillation Xe detector appeared to detect γ-rays in medical application and physics experiment [27,42], but their energy resolutions were worse than that of NaI(Tl) crystals in the MeV region. Such poor energy resolution comes mainly from the low light collection efficiency for ultraviolet photons and its non-uniformity. In spite of such a situation, recently an Italian group, led by Belli, began to use liquid Xe scintillation detector for detection of signals due to recoil nuclei followed by the elastic scattering of massive “dark matter” particles [57,58]. The ICARUS group is trying to use “proportional scintillation light” which is multiplied by electrons produced by recoil nucleus due to massive “dark matter” particles, under high electric field for discrimination of background γ-rays [59,60]. “Proportional scintillation” is included in the “ionization mode”, because its light intensity is proportional to the number of primary electrons and its essential process is “ionization”. Thus, this review does not include “proportional scintillation”.

If liquid Xe scintillation is used for emission of a large number of photons such as in electromagnetic showers due to high-energy γ-rays or electrons, the low light collection efficiency mentioned above does not become an obstacle for achieving good energy resolution. In such electromagnetic shower calorimeters, however, uniformity in light collection is required to achieve good energy resolution.

Table 2
(a) Decay times for fast (τ_S) and the slow (τ_T) components of scintillation light from liquid Ar, Kr and Xe excited by 1 MeV electrons. τ_R, the recombination time, and the intensity ratios I_S/I_T of fast component to the slow components are also shown. All decay times are in ns
(b) Decay times for fast (τ_S) and the slow (τ_T) component of scintillation light from liquid Ar, and Xe excited by α-particles. The intensity ratios I_S/I_T of the fast component to the slow component are also shown. All decay times are in ns

(a)	Liquid Ar	Liquid Kr	Liquid Xe
τ_S	6.3 ± 0.2^a ns	2.0 ± 0.2^a ns	
	(5.0 ± 0.2 ns	(2.1 ± 0.3 ns	(2.2 ± 0.3 ns
	for $E = 6$ kV/cm)^b	for $E = 4$ kV/cm)^b	for $E = 4$ kV/cm)^b
	6 ± 2^b		
τ_T	1020 ± 60^a, 1590 ± 100^b ns	91 ± 2^b ns	34 ± 2^b ns
	(860 ± 30 ns	(80 ± 3 ns	(27 ± 1 ns
	for $E = 6$ kV/cm)^b	for $E = 4$ kV/cm)^b	for $E = 4$ kV/cm)^b
τ_R	< 1 ns		45^a ns
I_S/I_T	0.083^b	0.01^b	0.05
	(0.045 for $E = 6$ kV/cm)^b 0.3^a	(0.02 for $E = 4$ kV/cm)^b	for $E = 4$ kV/cm)^b

(b)	Liquid Ar		Liquid Xe
τ_S	7.7 ± 1.0^a ns		4.3 ± 0.6^a ns
	$\sim 5^b$ ns		3^c ns
τ_T	1660 ± 100^a ns		22 ± 1.5^a ns
	1200 ± 100^b ns		22^a ns
I_S/I_T	1.3^a		0.45 ± 0.07^a
			1.5^a ns

[a]Ref. [44]; [b]Ref. [45].
[a]Ref. [44]; [b]Ref. [47]; [c]Ref. [48].

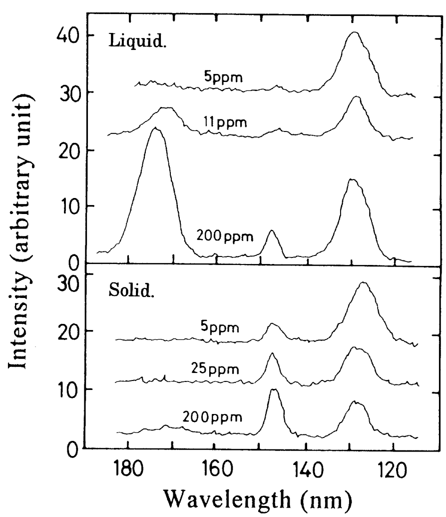

Fig. 4. The luminescence spectra of liquid and solid argon doped with various concentration of xenon.

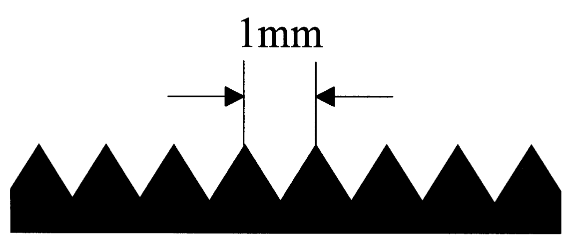

Fig. 5. The black reflector with irregular surface.

The present status of development of homogeneous scintillation shower calorimeters will be described in the next section.

3.1. For α-particles

In the initial development phase of liquid rare gas detectors, α-particles have been used for the

Table 3
Comparison of attenuation length between pure liquids and mixtures

Liquid	Argon	Krypton	Xenon	Argon–xenon (3%) mixture	Krypton–xenon (3%) mixture
Peak wavelength of emmitted photons (nm)	128	147	174	174	174
Measured attenuation length (cm)	66 ± 3	82 ± 4	29 ± 2	170 ± 23 (1st exp.) 118 ± 10 (2nd exp.)	136 ± 11

investigation of liquid rare gas scintillation detectors, because of easiness of use. However, the data obtained before 1980s may not be reliable because the purity of gas used in the experiments was not verified. Because the difficulty in quantitative data comparison among the reports was seen even after 1980, we carefully choose some reliable data obtained by the following groups, Waseda University [61], Columbia University [62] and Technische Hochschule Darmstadt [63]. Fig. 6 shows a schematic drawing of the instrument used by Funayama et al. (Waseda group) [61]. The α (^{210}Po, or ^{212}Bi and ^{212}Po)-source was set 3–4 mm apart from the Pyrex glass window. The window was coated by sodium salicylate as the wavelength shifter and the scintillation light produced by an α-particle is converted to visible light to pass through the window and a light guide (acrylic cylinder). Fig. 7a and Fig. 7b shows the energy spectra of α-particles from ^{210}Po, ^{212}Bi and ^{212}Po sources in liquid Xe, respectively. In these cases, we used only the energy resolutions (8.6% FWHM, 8.1% FWHM) obtained for α-particles from ^{210}Po and ^{212}Bi for comparison, because the energy spectrum of α-particles from ^{212}Po is distorted by electrons emitted by the beta decay of the parent nucleus. Fig. 8 shows the liquid Xe gridded chamber with a photomultiplier which was used by Aprile et al. [62]. In this case, the ultraviolet photons emitted in liquid Xe are directly observed by an UV-sensitive photomultiplier through a CaF_2 window. The geometric condition of the chamber is not shown in the figure but the authors showed all

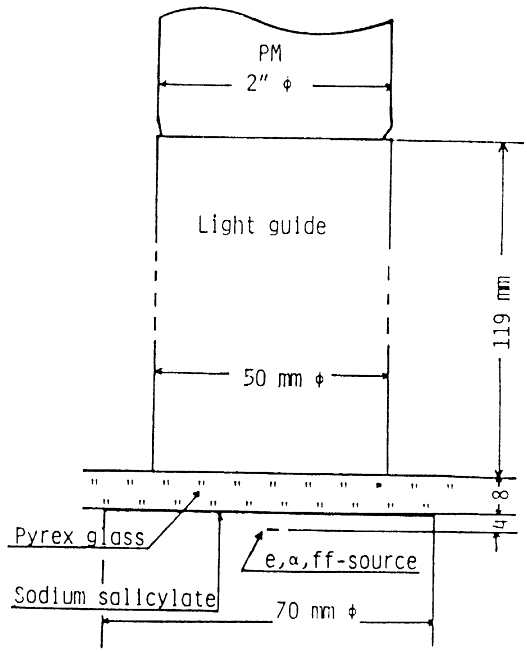

Fig. 6. Geometric arrangement of the apparatus by Waseda group for measurement of the scintillation light from liquid Ar and Xe excited by α-particles.

parameters to estimate the number of emitted photons in their paper. Fig. 9 shows the energy spectrum for α-particles from ^{241}Am measured under zero electric field. The energy resolution is 13% FWHM. Baum et al. directly observed scintillation from liquid Xe by an UV-sensitive photomultiplier through two quartz windows which are separated

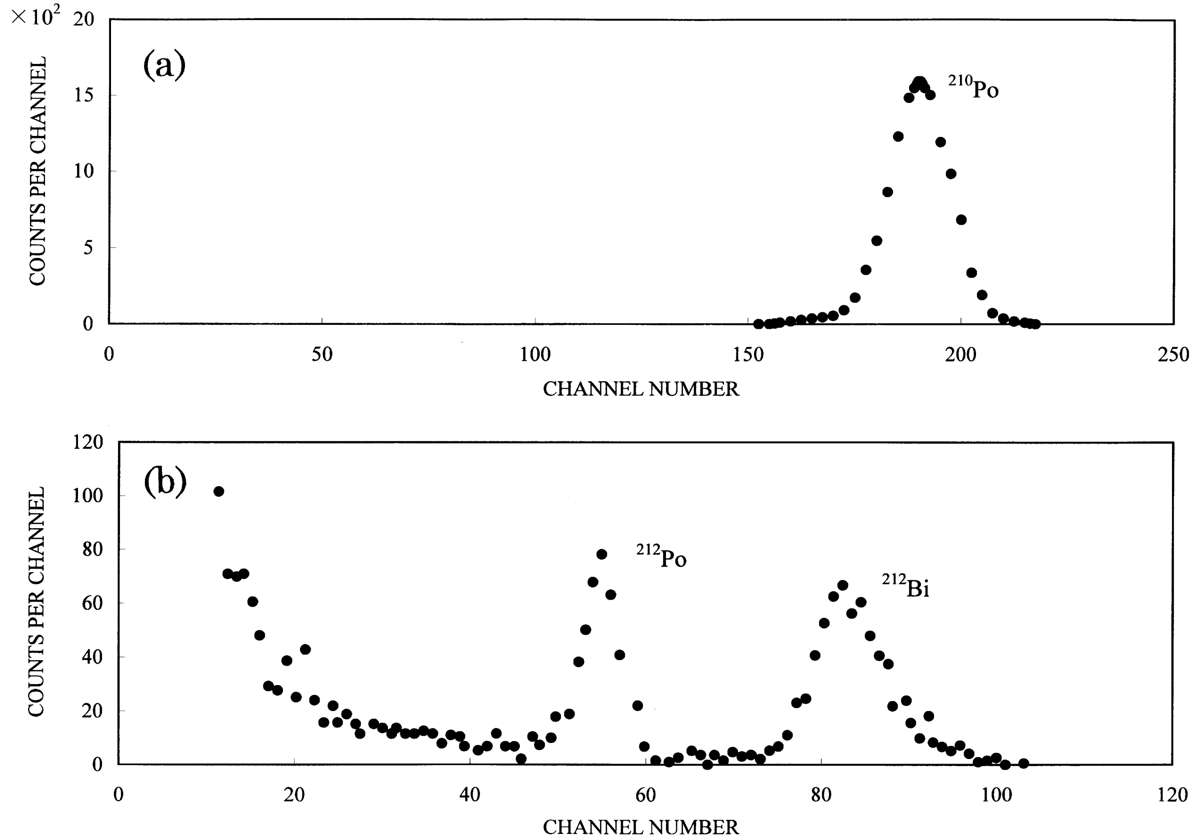

Fig. 7. Energy spectra of α-particles from ^{210}Po (a) and that of α-particles from ^{212}Bi and ^{212}Po (b) measured by the liquid Xe scintillation detector shown in Fig. 6.

by a 10 mm vacuum gap for thermal insulation [63]. Their result was worse than that by Funayama et al. but was comparable to that by Aprile et al. Recently, Ishida[1] [64] measured the variation of the energy spectrum against the distance between the α-source and the wavelength shifter coated Pyrex glass using the same instrument as used for measuring the attenuation coefficient of scintillation photons in liquid xenon [54]. In this case, sodium salicylate was used as a wavelength shifter as in the case of Funayama et al. [61].

[1] Ishida -1 and -2 in Fig. 10 were measured by their instrument for measuring the attenuation length in liquid rare gases shown in Ref [54].

Fig. 10 shows the relation between the energy resolution and the estimated number of photo-electrons. In this figure, the number of photo-electrons was estimated assuming that $W_{ph} = 19.5$ eV for α-particles and that the conversion factor of the wavelength shifter is nearly 100%, and the photons are isotropically emitted from the wavelength shifter. For the data of Baum et al., we assumed that the quantum efficiency of the PMT was 0.1 and the transparency of the quartz windows was 0.7. For other parameters, we used those given by the authors. Almost all data are on a line which is inversely proportional to the square root of the number of photoelectrons. From the figure, we can see that the best resolutions obtained for α-particles are near the theoretically expected line.

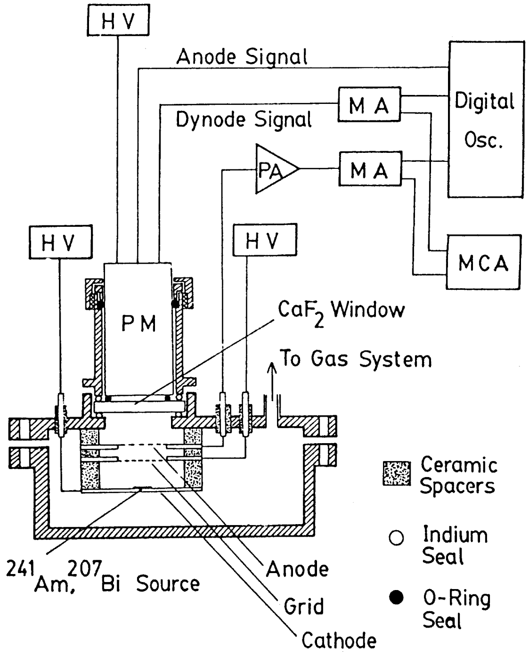

Fig. 8. A gridded ionization chamber with a UV-sensitive photomultiplier used for measurement of α-particles from ^{214}Am.

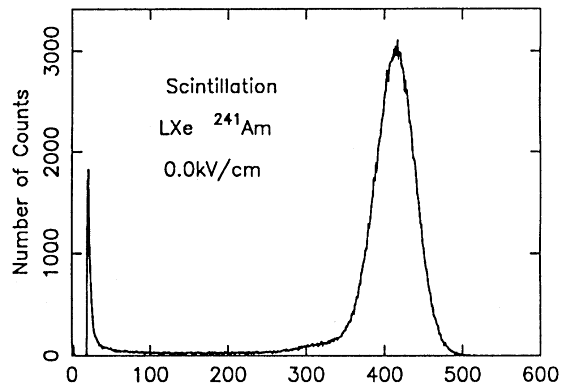

Fig. 9. The energy spectrum of α-particles from ^{214}Am directly observed by a UV-sensitive photomultiplier is shown in Fig. 8.

3.2. For γ-rays

The experiments for γ-ray detectors were made only with liquid Xe as detection medium. Since the first trial of medical application of liquid Xe scintillation detector was reported by Lavoie [27], many tests have been made but very few results show good energy resolution with clear experimental conditions. Here, the results obtained by Barabanov et al. [42] for γ-rays and by Ospanov and Obodovski [40] for X-rays are discussed for evaluation of the energy resolution.

At first, Barabanov et al. used a cylindrical quartz flask with a 5 mm thick Teflon reflector placed inside of the flask and with a single photomultiplier for γ-ray observation. The dimensions of the space filled with liquid xenon are 27 mm diameter and 18 mm length. They could observe the pulse height spectrum for 59 keV γ-rays from ^{241}Am and found that its peak largely depends on the distance between the source and the photomultiplier. To improve this dependency, they put two photomultipliers on opposite sides of the detector. Fig. 11 shows the improved liquid Xe scintillation spectrometer. They used a quartz cylinder filled with liquid Xe and a 5 mm thick Teflon reflector. Both ends of the cylinder were sealed with photomultipliers with quartz windows. The dimensions of the cylindrical volume filled with liquid Xe were 27 mm diameter and 12–30 mm length. For γ-ray measurements, the following sources were used: ^{241}Am (26, 59 keV), ^{57}Co (122 keV), ^{137}Cs (662 keV) and ^{22}Na (511, 1275 keV). As a result, they found the following facts:

1. Without the Teflon reflector a total absorption peak from ^{137}Cs is not distinguishable from the background of the Compton-scatter distribution.
2. Energy resolution for 122 keV γ-rays is 12% for the detector of smallest Xe thickness (12 mm) which is of the same order of magnitude as NaI(Tl) crystals of the same size. Results for lower energies (26, 59 keV) showed that the energy resolution is approximately proportional to the inverse of the square root of the γ-ray energy in the energy range lower than 100 keV.
3. Energy resolution for 662 keV γ-rays is the same as that for 122 keV or slightly worse.
4. Scintillation yield, the number of photons produced per unit energy of γ-ray, varies by 30%

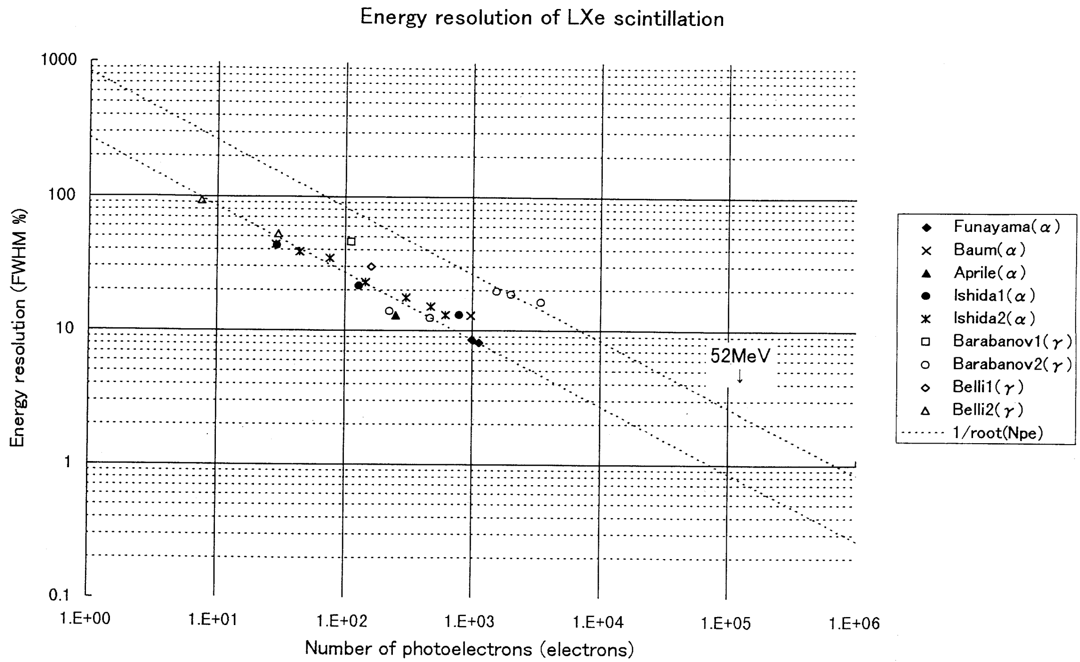

Fig. 10. The relation between energy resolutions (FWHM) and estimated number of photoelectrons obtained for α-particles and γ-rays.

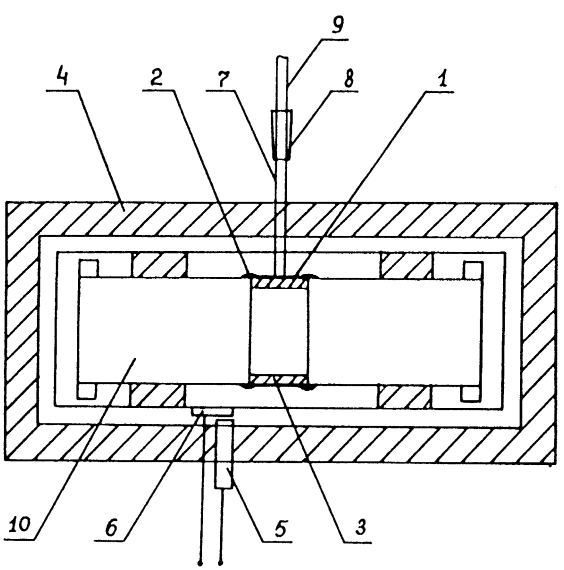

Fig. 11. Schematic diagram of a liquid xenon scintillation spectrometer used by Barabanov et al.: (1) quartz container, (2) araldite, (3) Teflon reflector, (4) foam plastic shell, (5) liquid nitrogen sprayer, (6) thermoresistance, (7) quartz pipe, (8) vacuum grease, (9) metal pipe, and (10) quartz photomultiplier.

in the energy range 50–2000 keV as shown in Fig. 12, which is similar to that of NaI(Tl) crystals.
5. As a reflector, apart from Teflon, aluminum covered by a thin film of LiF and different wavelength shifters were used but the energy resolution became worse in all these cases.

The discrepancy between 2 and 3 is explained by the authors as follows:

1. The additional spread of the total absorption peak for the high-energy γ-rays can arise as a result of fluctuation in energy distribution of Compton-scattering electrons and non-linearity of light output as a function of energy as seen in Fig. 12.
2. The poor reflection of Teflon for UV emission of liquid Xe may cause an additional spread of the total absorption peak because of the non-uniformity of the light collection. Such a spread would be larger for γ-rays with energies in the range higher than 0.5 MeV since they are absorbed over the whole liquid Xe volume.

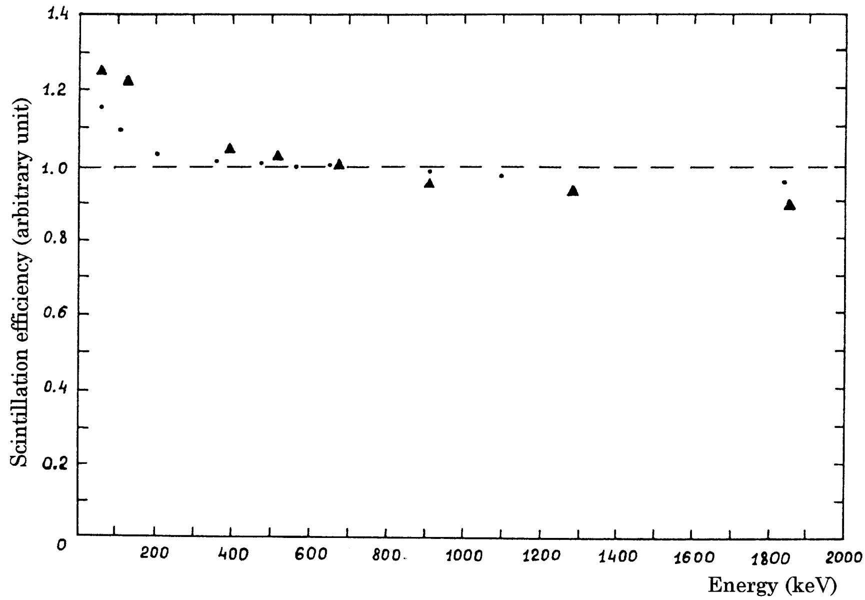

Fig. 12. Scintillation efficiency versus energy for liquid xenon (triangles) and NaI(Tl) (dots: results from Ref. [48]).

Although the authors described that the energy resolution is not improved in the high-energy region, the energy spectrum of the γ-rays from ^{22}Na, in the literature, shows the improvement of energy resolutions for 511, and 1275 keV γ-rays as seen from Fig. 13. Namely, the energy resolution is 19.7% for 511 keV γ-rays and 16.2% for 1275 keV γ-rays. To estimate the energy resolution of 1275 keV γ-rays, we used the half-width on the higher-energy side of the peak. Thus obtained data are plotted in Fig. 10 which include the data with γ-rays by Belli et al. [31,65] and α-particles. Here, we assume a quantum efficiency of 10% for their photomultipliers with quartz windows and the effective solid angle for γ-rays detected at the center of the sensitive volume. The data for γ-rays with energies higher than 0.5 MeV are not on the line for α-particles but on the other line giving worse energy resolution, as expected from the non-uniformity of light collection.

The energy dependence of the specific scintillation yield for X-rays in the energy range lower than 59.5 keV was obtained by Ospanov and Obodovski [40]. Their result supports the increase in scintillation yield in the low-energy region in Fig. 12 which includes the data for NaI(Tl) for comparison. The LET dependence of liquid Xe is clearly larger than that of NaI(Tl). This may be the reason why the energy resolution becomes worse with increase in energies of γ-rays in the high-energy region.

Chepel et al. are trying to use a liquid Xe detector to PET [66,67] and van Sonsbeek et al. are developing new liquid or solid Xe γ-ray detectors [32,64], but they could not yet show any good result as a γ-ray spectrometer.

4. Homogenous scintillation calorimeter

4.1. Reflection-type calorimeter

In 1988, M. Chen et al. proposed construction of a liquid Xe scintillating electromagnetic shower calorimeter with excellent energy resolution in the order of 10^{-3}(rms) for SSC- and LHC-experiments [18]. Fig. 14 shows a cross-sectional view of the proposed liquid Xe scintillating calorimeter. This calorimeter consists of two parts of a barrel and an endcap, the latter is of the sampling type. To keep uniformity in light collection, the unit cell is

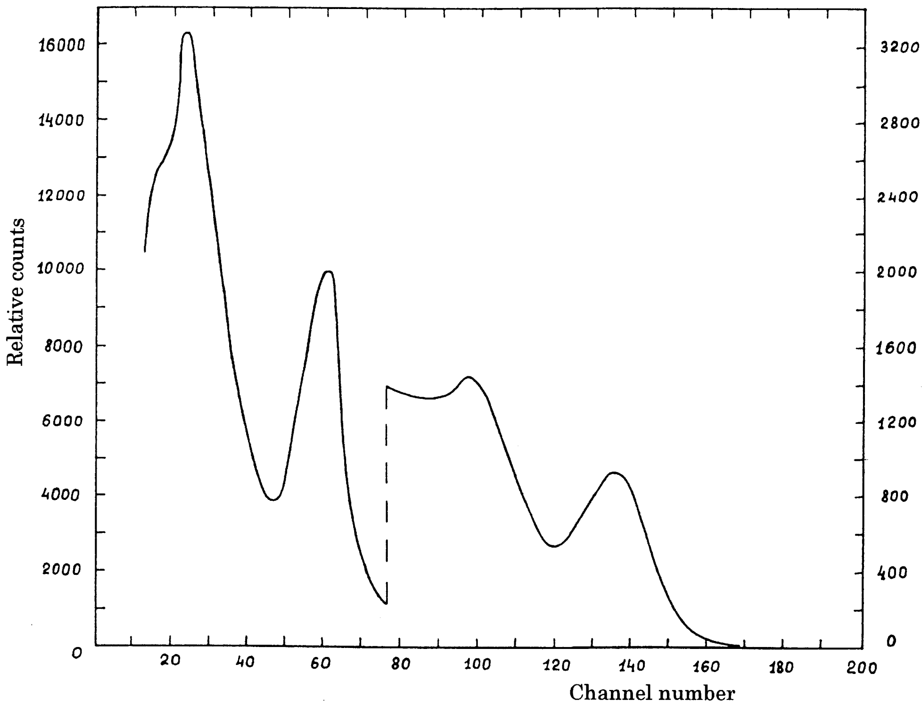

Fig. 13. Pulse-height spectrum for liquid xenon spectrometer exposed to 511 and 1275 keV γ-rays with 20 mm xenon thickness.

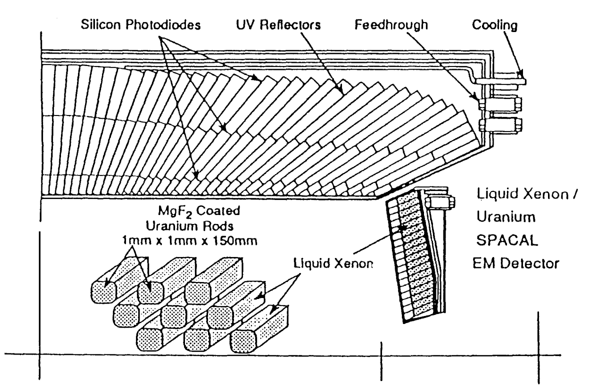

Fig. 14. A side view of one quadrant of a liquid Xe electromagnetic calorimeter proposed for SSC/LHC with silicon photodiodes, UV reflector, warm and cold feedthrough and cooling ports. The calorimeter is formed of a long barrel and two tapered endcaps.

surrounded by four aluminum surfaces coated by MgF_2 layers thicker than 4 wavelengths for photons emitted from liquid Xe. The reflectors are used to achieve total internal reflection. A typical unit cell uses three layers of UV-sensitive silicon photodiodes as shown in the figure. The uniformity in sensitivity of silicon photodiodes is excellent compared to photomultipliers. A major concern for keeping good energy resolution is to achieve uniform light collection in the unit cell. Besides the investigations on reflectivity of wall surfaces, as described in the former section, several silicon photodiodes have been developed by the MIT or ITEP groups [70,71] and the Waseda group [43,72].

Although M. Chen's proposal was not adopted by the SSC, the prototype of the calorimeter whose light collection system consists of 7×7 unit cells [52,70] as shown in Fig. 15 was constructed by the MIT–ITEP group and its reflectivity of wall surfaces in an unit cell was tested as follows.

To achieve an excellent energy resolution, the light collection efficiency of the photodetector should not depend on the position where the scintillation is produced. Also, the inner surface of the wall should have a high reflectivity for ultraviolet

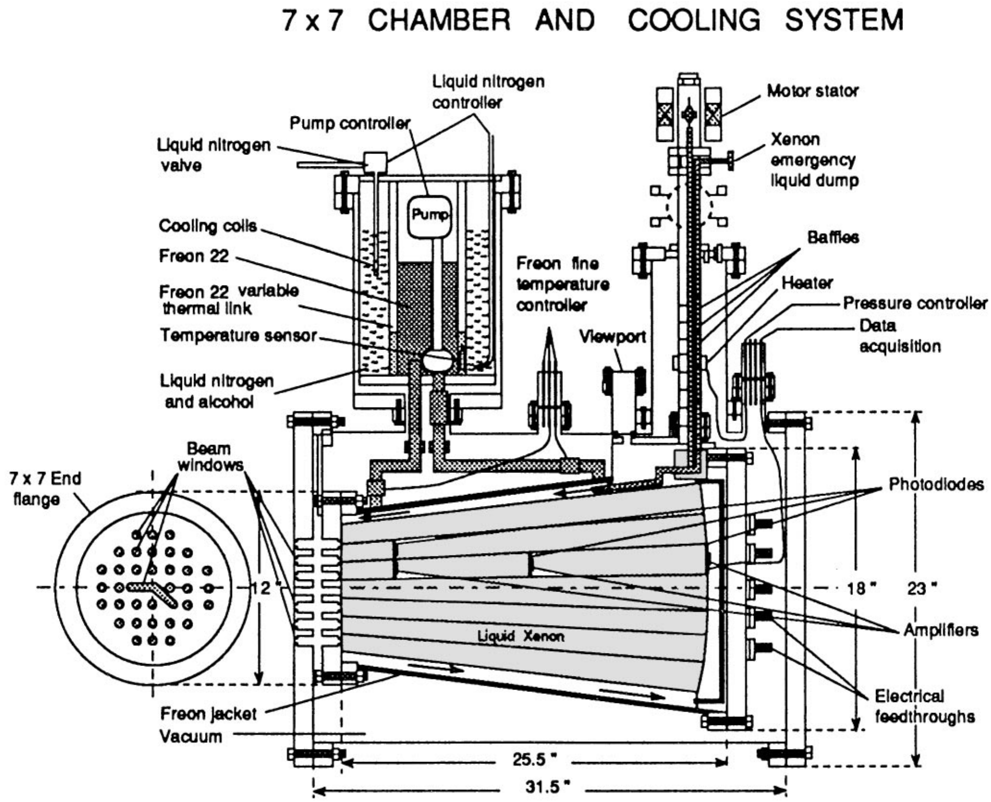

Fig. 15. Full length prototype liquid Xe calorimeter, consisting of 7 × 7 unit cells, constructed by the MIT–ITEP group.

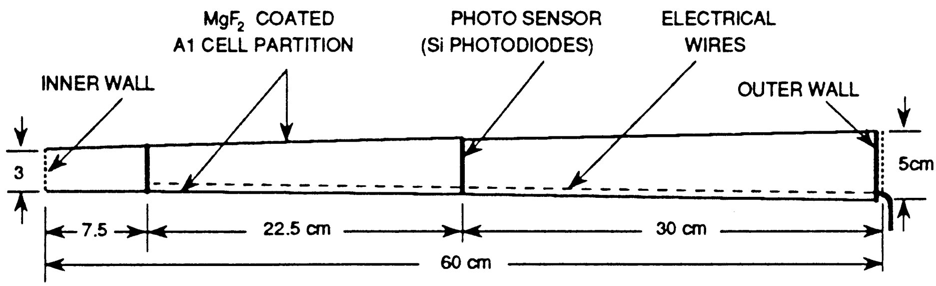

Fig. 16. A cross-sectional view of the unit cell of liquid Xe scintillation calorimeter. In this cell, three Si photodiodes are used as photon sensors.

photons to efficiently collect the scintillation light by the photodetectors. If we need additional position sensitivity in the scintillating calorimeter, the liquid Xe space in the calorimeter should be divided into many small cells. Fig. 16 shows a cross-sectional view of the unit cell. The MIT–ITEP group has tested many kinds of reflectors; Kapton, Al, glass plates and Cu clad printed circuits (290 μm thick) coated by aluminum and MgF_2 which is used as a protector against oxidation of the aluminum surface. Thus, they obtained reflectivities in the range 85–90% [70]. Since the refractive index of liquid Xe (1.56 [73]) for the wavelength of liquid Xe scintillation is larger than that of MgF_2 (1.45), most of the scintillation light in liquid Xe causes total reflection against aluminum mirrors coated with MgF_2 layers 4 times thicker than the wave lengths [70]. In addition, they tested the uniformity of light collection in the test chamber, using a photodiode as a photo-sensor and an aluminum surface coated by MgF_2 as a reflector. As seen in Fig. 17, the result shows a very flat response [70]. In this experiment, a lattice collimater was put in front of the photodiode to remove photons with large incident angle. From the figure, we cannot estimate how many photons are discriminated to make such a flat response because only the relative light yield is given as the vertical scale. We guess that a large amount of scintillation light was discriminated by the collimater and, as a result, the energy resolution deteriorates in the low-energy region. This type of calorimeter has not yet been tested for high-energy electrons or γ-rays.

Recently, however, the ITEP-Dubna and MIT collaboration group has constructed a liquid Xe/Kr scintillating calorimeter of the same type as mentioned above but the reflection walls were coated by wavelength shifter for converting the VUV light to visible light [74,75]. They used photomultiplers sensitive to visible light and aluminized Mylar reflectors partly covered with strips of wavelength shifter (p-terphenyl) as seen in Fig. 18. Fig. 19 shows the variation of light collection efficiency versus distance from the window surface of the photomulitiplier [76]. The ordinate of this figure shows the number of photoelectrons per MeV. If the quantum efficiency of the PMT is 10%, the number of effective photons is 100/MeV from the figure. This is only 0.2% of the photons produced as estimated from the W_{ph}-value. Also, Fig. 20 shows the variation of the light collection efficiency for transverse coordinate [75]. The energy resolution for liquid Xe was 7.5%(rms) for 348 MeV electrons [74] which corresponds to $\sigma/E \sim 5\%/(E\,(\mathrm{GeV}))^{1/2}$. This value is 2.5–3 times worse than that obtained by the Monte Carlo simulation. By fitting the energy resolution obtained to Monte Carlo simulation results, they estimated the attenuation length of the scintillation photons in the liquid Xe to be 3–5 cm [74].

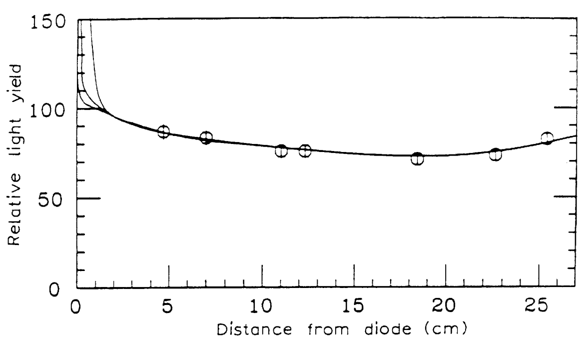

Fig. 17. The measured light collection acceptance, using cosmic muons, versus the distance to the diode, in a 27 cm long mirror system. Three Monte Carlo curves, corresponding to different light collimators from top to bottom (8 × 8 grid and 12 mm deep; 32 × 32 grid and 3 mm deep, and 128 × 128 grid and 1 mm deep), showing the non-uniform region near the diode decreases as the number of grids increases.

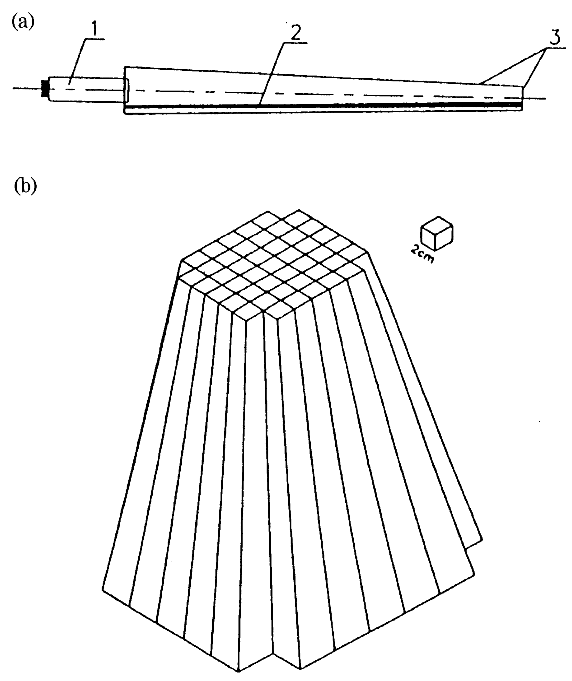

Fig. 18. Single calorimeter cell (a): (1) PMT, (2) fragment of p-terphenyle strip, (3) Mylar pyromidal reflector; and reflector structure in the calorimeter (b).

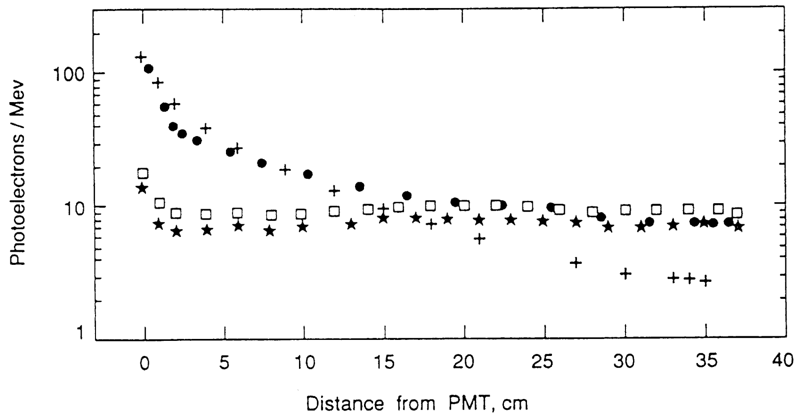

Fig. 19. Measured uniformity: peak positions of scintillation spectra as function of the distance between the beam and the photodetector, for liquid Xe (crosses) and liquid Kr (dots) with aluminized Mylar reflectors and wavelength shifter-coated PMT; for liquid Xe (squares) and liquid Kr (stars) with wavelength shifter-evaporated Al-Mylar reflectors and UV-blind PMT.

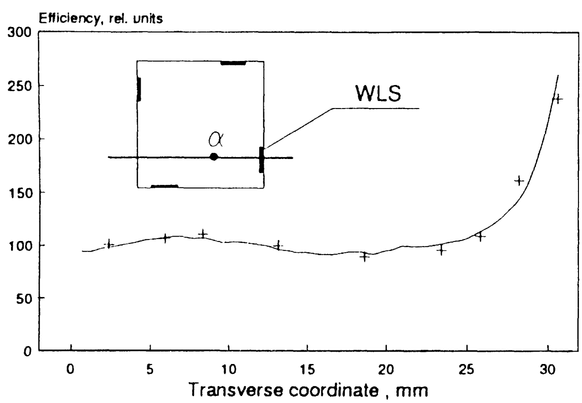

Fig. 20. Efficiency of light collection measured with the movable α-source. Crosses and curve are measured and Monte Carlo simulated, respectively, for the cell shown in Fig. 18.

On the other hand, Braem et al. observed scintillation light in liquid Xe in a rectangular cell with Al reflectors coated with MgF_2 of the dimensions $7.4 \times 7.4 \times 60\ cm^3$ for 5 GeV electrons using silicon photodiodes; they obtained the W_{ph}-value and the light attenuation length from comparison with a stimulated result [28]. The attenuation length is reasonable but the W_{ph}-value is too large compared with others as shown in Table 1.

Recently, the refractive index of liquid Xe for intrinsic scintillation light, which is important to determine the incident condition of scintillation light, was measured by Barkov et al. [73]. The value was 1.5655 ($\pm$ 0.0024 $\pm$ 0.0078) which is consistent with the extrapolated value from data at longer wavelengths [29,77].

Also, Ypsilantis's group at CERN, independently from the MIT–ITEP group, proposed the simultaneous use of both ionization and scintillation signals in an electromagnetic shower calorimeter [78]. Although some fundamental tests were performed [32], such a simultaneous processing both signals results in a more complicated structure, and this disadvantage is too great compared with the merits of using both signals.

4.2. *Direct scintillation calorimeter (new proposal)*

According to calculations by the MIT–IETP group, the best energy resolution (rms) can be expected to be $2\%/[E\ (\mathrm{GeV})]^{1/2}$ [75]. For γ-rays of energy lower than 1 GeV, however, this energy resolution is not always sufficient when compared with the requirements from experimental physicists. To achieve the energy resolution of 1–2% (FWHM) for $\sim$ 50 MeV γ-rays, we propose to use the direct scintillation from liquid Xe without reflection. To achieve this, the liquid Xe chamber should be surrounded by VUV-sensitive photomultipliers immersed in liquid Xe. The photons reflected by the wall surface may be detected, but a large fraction of emitted scintillation photons should be directly detected by the photomultipliers. Even if its fraction is 30% and the quantum efficiency of photomultiplier is 10%, the number of photoelectrons is estimated to be $50\times10^6/24\times0.3\times0.1=6.25\times10^4$. This is statistically sufficient to achieve 1% (FWHM) energy resolution. The extrapolation of the line for γ-rays or α-particles in Fig. 10 gives 2.5% or 1.4% (FWHM). Recently, Hamamatsu Photonics Co. developed new VUV-sensitive photomultipliers which can operate at liquid Xe temperatures. In usual photomultipliers, the so-called "bialkali" material is used as photocathode. However, they do not work at temperatures lower than $-100°C$ [79] except for Cs_3Sb. Recently, Araujo et al. found that a VUV-sensitive photomultiplier (R1668) with a photocathode of SbRbCs, supplied from Hammamatsu Photonics Co., works at temperature lower than $-100°C$ [80]. We asked the company to fabricate a new photomultiplier of 5 cm diameter and 3 cm height with the same photocathode material (SbRbCs) as R1668 and a gain of 10^6–10^7. The first product is under testing. If such small size photomultipliers can be used for the above-mentioned purpose as mentioned above an energy resolution of 1–2% can be expected. Fig. 21 shows such a calorimeter designed for 52 MeV γ-rays emitted from $\mu\rightarrow e\cdot\gamma$ decays [81,82]. Recently, Orito and Mori simulated the position of the interaction point of γ-rays from the difference in signal size of individual photomultipliers. The result showed that position accuracy of about 1 mm for the conversion point can be obtained [82]. This resolution is sufficient for $\mu\rightarrow e\cdot\gamma$ decay experiments. We are currently designing such a calorimeter needed for this experiment.

5. Summary

In this paper, the present status of investigations of the basic properties of scintillation from liquid rare gases and of liquid rare gas scintillation spectrometers for electrons, γ-rays, α-particles and heavy ions over a wide energy range is reviewed. In

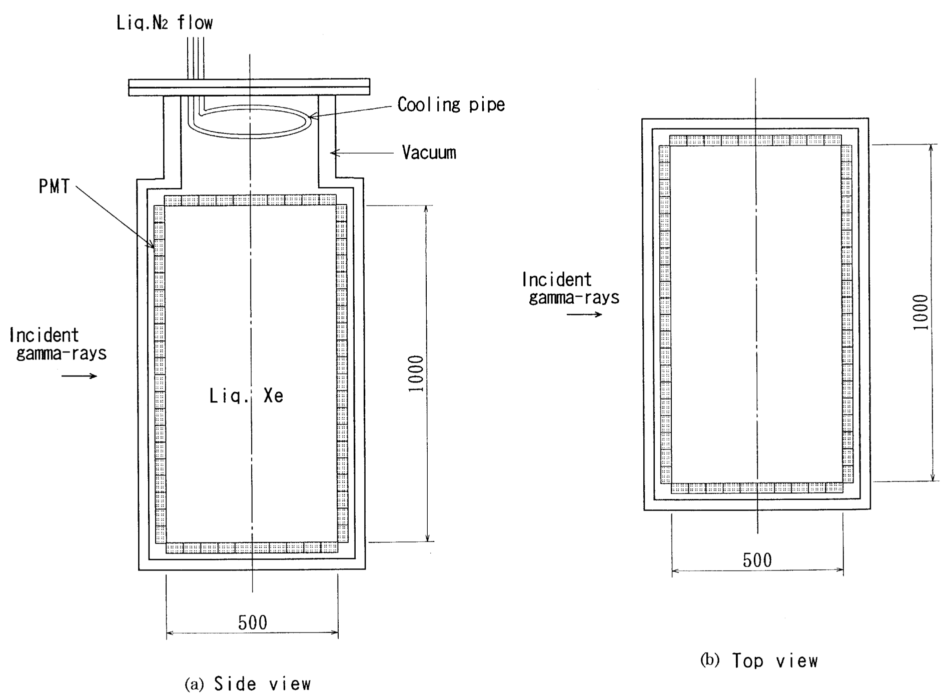

Fig. 21. The cross-sectional side view (a) and the top view (b) of a liquid Xe scintillation homogenous calorimeter designed for 52 MeV γ-rays from $\mu\rightarrow e\cdot\gamma$ decay.

addition, a new liquid Xe γ-ray spectrometer which observes direct scintillation is proposed for $\mu \rightarrow e \cdot \gamma$ decay experiments.

6. For Further Reading:

The following references are also of interest to the reader: [68] and [69].

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