Non-Proportionality in the Scintillation Response and the Energy Resolution Obtainable with Scintillation Crystals

P. Dorenbos, J. T. M. de Haas, and C. W. E. van Eijk

Abstract—**A review and new data are presented on the absolute photon yield emitted by "classical" ($NaI(Tl^+)$, $CsI(Tl^+)$, $CsI(Na^+)$, $CaF_2(Eu^{2+})$, $Bi_4Ge_3O_{12}$, and $CdWO_4$) and "modern" (BaF_2, $Gd_2SiO_5(Ce^{3+})$, $YAlO_3(Ce^{3+})$, $Lu_2SiO_5(Ce^{3+})$, $Lu_3Al_5O_{12}(Sc^{3+})$, and $K_2LaCl_5(Ce^{3+})$) scintillation crystals after absorption of X-rays and γ-rays of energies ranging from 5 keV to 1 MeV. Factors influencing the energy resolution with which high energy photons can be detected with scintillator-photomultiplier combinations are reviewed. Attention is especially focused on the effects of nonproportionality in the scintillation response on the energy resolution.**

I. Introduction

DURING the 20 years after the discovery of the scintillator $NaI(Tl^+)$ by Hofstadter in 1948 [1] significant effort was put into the research and development of other scintillators for the detection of ionizing radiation, in particular high energy photons (X-rays and γ-rays). Besides $NaI(Tl^+)$, "classical" scintillators like $CdWO_4$ in 1948 [2], $CsI(Tl^+)$ in 1950 [3], $CsI(Na^+)$ in 1965 [4], $CaF_2(Eu^{2+})$ in 1966 [5], were discovered in this period. In 1973 Weber and Monchamp [6] reported on the luminescence properties of $Bi_4Ge_3O_{12}$ (BGO). This material is of particular interest because of its high density of 7.13 g/cm^3 which provides a good stopping power for high energy gamma rays [6], [7]. With the exception of the work on BGO, the activity in scintillator research was quite low during the seventies. A renewed interest in scintillators was initiated by the demand for high density and fast scintillating crystals for applications in medical diagnostics, high energy physics and high count rate experiments. This research, starting around 1980, is still actively being pursued and has resulted in several new scintillators e.g., the fast scintillation component of BaF_2 in 1982 [8], [9], $Gd_2SiO_5(Ce^{3+})$ in 1983 [10], $YAlO_3(Ce^{3+})$ in 1980 [11], [12], $Lu_2SiO_5(Ce^{3+})$ in 1990 [13]–[16], and recently (1994) $Lu_3Al_5O_{12}(Sc^{3+})$ [17], [18], and $K_2LaCl_5(Ce^{3+})$ [19].

In scintillator research one is particularly interested in the scintillation emission spectrum, the scintillation decay time, and the absolute scintillation light yield. The latter is usually expressed in terms of the number of photons created in the crystal per unit of absorbed ionizing energy (photons/MeV). It is desirable for the scintillation yield to be independent of energy. Deviations, which are known as nonproportionality (or nonlinearity) in the scintillation efficiency (or response), may result in difficulties in the determination of the energy of the detected radiation. It was already noted in 1967 by Aitken, *et al.* [20] that the nonproportionality of the scintillation response of $CsI(Tl^+)$ near energies of 4.5 to 5.9 keV makes this material completely worthless as a proportional spectrometer for energy analysis in this region. Another aspect of nonproportionality is its degradation of the energy resolution obtainable with scintillators.

The phenomenon of nonproportional response and its relation with energy resolution has been studied quite extensively for the classical scintillators, especially for $NaI(Tl^+)$. However, for the "modern" scintillators, this aspect gained very little to no attention at all during the last 15 years. The recent literature mainly reports on studies at energies ranging from the 59.5 keV gamma emission of ^{241}Am to the 1.33 MeV gamma emission of ^{60}Co. Studies at energies lower than 59.5 keV at which nonproportionality is most prominent are quite scarce.

In this work, we will start with a short review about the theory of the energy resolution obtainable with scintillators optically coupled to a photomultiplier tube (PMT). Next, a review of the existing data on the nonproportionality of $NaI(Tl^+)$, $CsI(Tl^+)$, $CsI(Na^+)$, $CaF_2(Eu^{2+})$, $Bi_4Ge_3O_{12}$ is followed by new data on BaF_2, $CdWO_4$, $Gd_2SiO_5(Ce^{3+})$, $YAlO_3(Ce^{3+})$, $Lu_2SiO_5(Ce^{3+})$, $Lu_3Al_5O_{12}(Sc^{3+})$, and $K_2LaCl_5(Ce^{3+})$. A compilation is given of the absolute photon yield, the photoelectron yield in PMT's with bialkali photocathodes, and the energy resolution at 662 keV obtainable with the above mentioned scintillators. Finally, we will discuss the factors determining the experimentally observed values of the energy resolution.

II. Theory on Energy Resolution

In discussing the factors determining the energy resolution obtainable with scintillators optically coupled to photomultiplier tubes we follow the review on this subject presented by Birks in his book [21]. Additional information can be found in the work by Breitenberger [22].

The average total charge $\overline{Q_0}$ contained in the anode pulse delivered by a photomultiplier tube equals

$$\overline{Q_0} = \overline{N}\overline{p}\overline{M} \tag{1}$$

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where $\overline{N}$ is the mean number of photons created in the scintillator upon absorption of a gamma quantum. $\overline{p}$ is the average transfer efficiency and represents the probability that a photon from the scintillator results in the arrival of a photoelectron at the first dynode which subsequently undergoes the full multiplication in the PMT. $\overline{M}$ is the mean electron multiplication factor of the PMT.

The energy resolution R, defined as the full width (ΔE) of the peak in the pulse height spectrum at half the maximum intensity (FWHM) divided by its energy, is related to the fractional (or relative) variance $v(Q_0)$ of Q_0 as

$$R = \left(\frac{\Delta E}{E}\right)_{FWHM} = 2.35\sqrt{v(Q_0)}. \tag{2}$$

Note, the fractional variance of a variable x is defined as $v(x) = var(x)/\overline{x}^2$, where $var(x)$ is the variance and $\overline{x}$ the mean value of x. $v(Q_0)$ can be written as [21]

$$v(Q_0) \approx \left[v(N) - \frac{1}{\overline{N}}\right] + v(p) + \frac{1+v(M)}{\overline{N}\overline{p}}. \tag{3}$$

In (3), $v(N)$ is the variance in the number of photons generated in the crystal which in case of Poisson statistics equals $\overline{N}^{-1}$. The bracketed term in (3) therefore represents the variance in the number of photons other than due to Poisson statistics. $v(p)$ is known as the transfer variance, and the last term in (3) is known as the photomultiplier variance. $v(M)$ is the variance in the photomultiplier gain and can be written as

$$v(M) = \sum_{i=1}^{n}\prod_{j=1}^{i}\frac{1}{\overline{\delta}_j} \tag{4}$$

with n the number of dynodes in the photomultiplier tube and $\overline{\delta}_j$ the mean multiplication factor at the jth dynode. The mean gain $\overline{M}$ is simply

$$\overline{M} = \prod_{j=1}^{n}\overline{\delta}_j. \tag{5}$$

In discussing the energy resolution, it is useful to introduce the terms intrinsic resolution R_i, transfer resolution R_p, and photomultiplier resolution R_M; see (6). The intrinsic resolution also known as intrinsic broadening or intrinsic linewidth is the equivalent of the bracketed term in (3). The transfer resolution and photomultiplier resolution correspond with the second and last term in (3), respectively

$$R^2 = R_i^2 + R_p^2 + R_M^2 = 5.55v(Q_0). \tag{6}$$

Below, we discuss the factors which determine R_i, R_p, and R_M. There are two main contributions to the intrinsic resolution

$$R_i^2 = R_{np}^2 + R_{inh}^2. \tag{7}$$

One is connected with the nonproportional response (R_{np}) of the scintillator to γ-quanta as a function of energy and will be discussed in Section V-A. The other (R_{inh}) is connected with inhomogeneity of the crystal causing local variations in the scintillation light output. A possible origin can be a concentration gradient or local fluctuations of luminescence centers in a scintillation crystal. The number of scintillation photons is then dependent on the point in the crystal at which the scintillation pulse is created.

There are many factors which determine the transfer efficiency p and connected with this, the transfer resolution R_p. For example: i) the wavelength λ of the photon and the quantum efficiency (QE) of the PMT at this wavelength, ii) the transmittance of the scintillator and the reflectivity of its reflecting covering, iii) the optical coupling to the window of the PMT, iv) angle of incidence of the photon on the photocathode, v) nonuniformity of the photocathode, vi) the photoelectron collection efficiency at the first dynode.

The combined effect of intrinsic and transfer resolution are often named scintillator resolution R_s defined as $R_s^2 = R_i^2 + R_p^2$. For an ideal scintillation detector, R_i, R_p, and consequently R_s will be zero. The resolution is then given by

$$R = R_M = 2.35\sqrt{\frac{1+v(M)}{\overline{N}\overline{p}}}. \tag{8}$$

With this equation, R_M can be calculated from experimentally determined photoelectron yields and values of $v(M)$.

III. Non-Proportional Response

A. Classical Scintillators (Review)

The first observation of the nonproportional response of NaI(Tl$^+$) was by Pringle and Standil [23], and one of the first studies was reported by Engelkemeir in 1956 [24]. Engelkemeir found that as the gamma ray energy decreases from about 1 MeV to 60 keV, the (relative) light yield gradually increases by about 15%, see also Fig. 1. Then the yield drops by about 5% near 40 keV and below 33 keV the yield appeared to increase again. It was suggested by Engelkemeir that the changes in yield near 40 keV are associated with the K-shell absorption edge at 33.2 keV of the iodine ions in the NaI host crystal. Later studies by Jones [25], Hink [26], and Brunner, *et al.* [27] revealed much more clearly the relationship with the K-edge; also changes in the light yield near the L absorption edges of iodine were observed [27].

It appears rather difficult to obtain reasonably accurate values for the scintillation response at low energy. Kaiser, *et al.* [28] observed that the response of NaI(Tl$^+$) at energies below 20 keV is sensitive to the surface treatment of the crystal, whereas at higher energies it is insensitive to the treatment. Hill and Collinson, however, observe that the surface treatment has a negligible influence on the response at low energy [29], [30]. They find that the response is sensitive to the strain in the crystal. Based on the small penetration depth of low energy X-rays in NaI(Tl$^+$), Meggitt [31] considered the effect of a surface layer of reduced scintillation efficiency to explain the decrease of the response at low energy.

CsI(Tl$^+$) also appears to show nonproportional response especially near the K-shell and L-shell absorption edges of cesium and iodine ions [20], [32], [33] (see Fig. 2). Figs. 3 and 4 show results for CsI(Na$^+$), and CaF_2(Eu^{2+}). Both of these figures and Figs. 1 and 2 were originally published by Aitken *et al.* [20]. Fig. 5 shows results for $Bi_4Ge_3O_{12}$ and CsI(Tl$^+$)

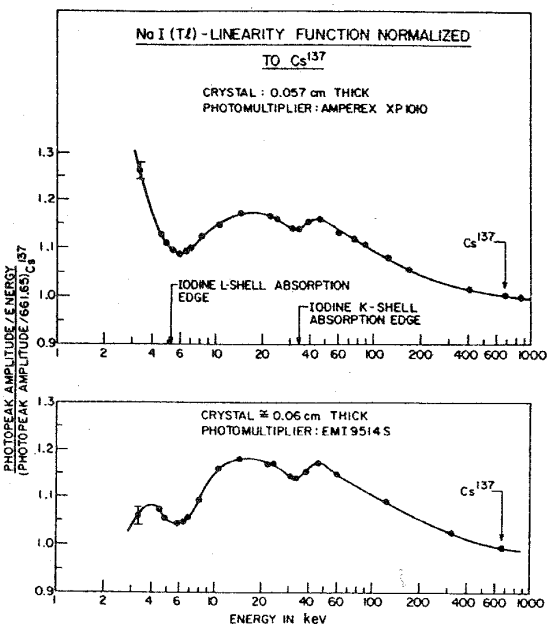

Fig. 1. Scintillation response curve for two thin NaI(Tl^+) crystal [20].

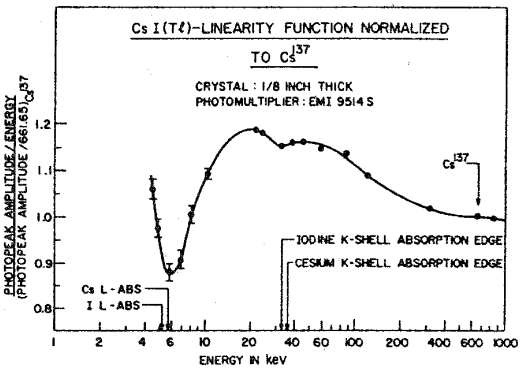

Fig. 2. Scintillation response curve of CsI(Tl^+) [20].

as reported by Averkiev *et al.* [34]. In all these figures, the light yield per unit energy is plotted relative to the light yield at the 662 keV gamma energy of a ^{137}Cs gamma source. For each scintillator, one observes a small drop in light yield near the K-edge of the heaviest ion. The drop in light yield near the L absorption edge for NaI(Tl^+) and CsI(Tl^+) is even more pronounced than near the K-edge.

It was demonstrated by Zerby *et al.* [35] and later by Hink [26] and Kaiser *et al.* [28] that the origin of nonproportionality especially near the K and L edges is related to nonproportionality in the response of the scintillator to energetic electrons. Even smooth changes with energy in the electron response may produce a significant drop in the γ-response curve near K and L absorption edge energies. To see this, one should consider the cascade of processes following the absorption of an X or γ -ray. Fig. 6, taken from the work of Kaiser *et al.* [28],

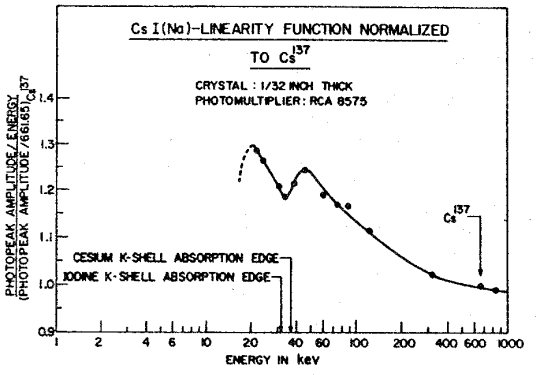

Fig. 3. Scintillation response curve for CsI(Na^+) [20].

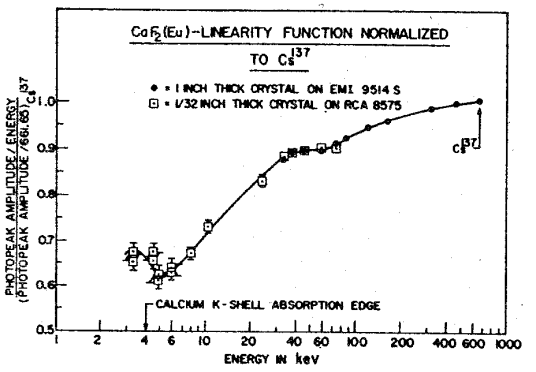

Fig. 4. Scintillation response curve for $CaF_2(Eu^{2+})$ [20].

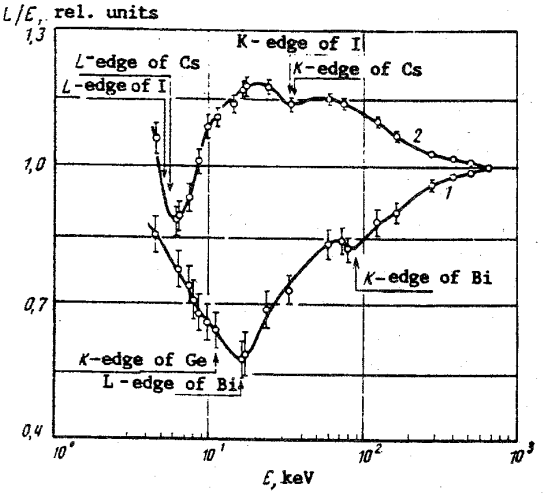

Fig. 5. Scintillation response curves for (1) $Bi_4Ge_3O_{12}$ and (2) CsI(Tl^+) [34].

shows a simplified cascade sequence for NaI(Tl^+). An X-ray of energy E_a interacts by means of photoelectric absorption in the K-shell of iodine, and produces a photoelectron of energy E_a minus the K-shell electron binding energy (33.2 keV). Further decay of the hole in the K-shell results in emission of a K_α X-ray which in Fig. 6 is absorbed by the photoelectric

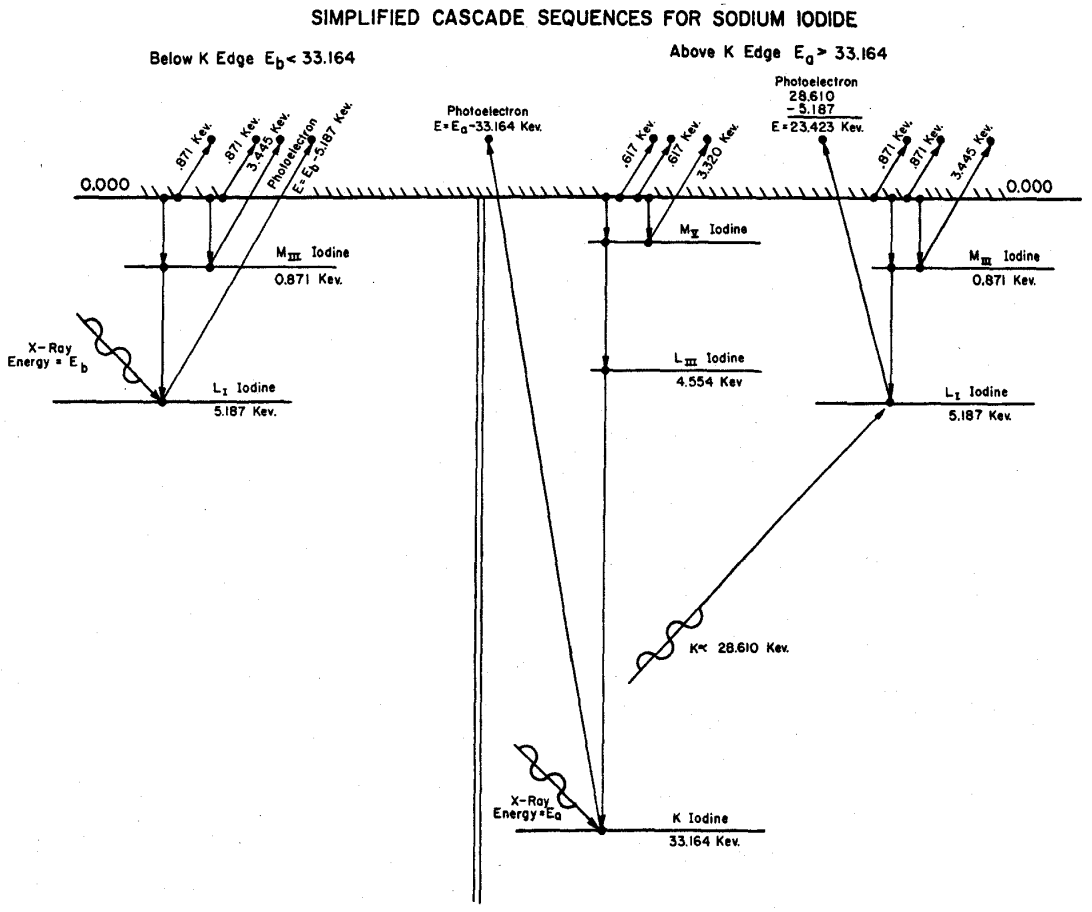

Fig. 6. Simplified cascade sequence following the absorption of a gamma ray in NaI(Tl$^+$). The left side shows the situation if the γ-ray energy is lower than the K-edge of iodine, the right side if it is larger [28].

effect in the L_I iodine shell resulting in a photoelectron of energy 23.4 keV. The holes in the L_{III} and L_I shells and eventually in the M-shells decay under emission of low energy (< 4 keV) Auger electrons. If the incident X-ray energy is smaller than the K-shell absorption edge, the cascade sequence on the left side of Fig. 6 may result. An X-ray with an energy just above an absorption edge produces relatively more low energy electrons than if its energy is just below the edge.

Based on such simplified cascade sequences Zerby *et al.* [35] and later Hink [26] and Kaiser *et al.* [28] deduced electron response curves for NaI(Tl$^+$) at energies up to 50–70 keV from the gamma ray response curves. At energies above 100 keV the situation is complicated by the fact that the total energy of a gamma ray can be transferred to the crystals by several means: 1) the photoelectric effect creating one energetic photoelectron, 2) Compton scattering followed by photoelectron absorption of the created secondary γ -quantum, 3) multiple Compton scattering. In the last two cases several secondary electrons are created. The distribution in number and energy of secondary electrons is therefore also determined by the way in which the γ-quantum transfers its energy to the crystal. Zerby *et al.* [35] simulated these effects by means of Monte Carlo calculations and calculated the electron response curve from 100 keV to beyond 1 MeV (see Fig. 7).

Electron response curves for NaI(Tl$^+$) have also been measured directly by means of electron excitation of the scintillator by Porter *et al.* [36]. This experimental response curve is shown in Fig. 7 together with ones derived from γ-response curves. All are smooth functions of the electron energy with a maximum near 10 to 20 keV and without anomalies near K and L shell binding energies. Below 10 keV, the discrepancies between different measurements on the electron response are very large and reliable values are not available. Clearly this is related with the inaccuracies in determining the X-ray response curve in this energy region. Prescott *et al.* [37] suggest that, since the range of electrons of energy less than 10 keV is only a few microns, this may be connected with surface effects. An elegant method to determine electron response curves and to avoid surface effects was proposed by Valentine and Rooney [38]. Using a Compton spectrometer configuration and a collimated γ-ray beam incident on a scintillator, the energy deposited

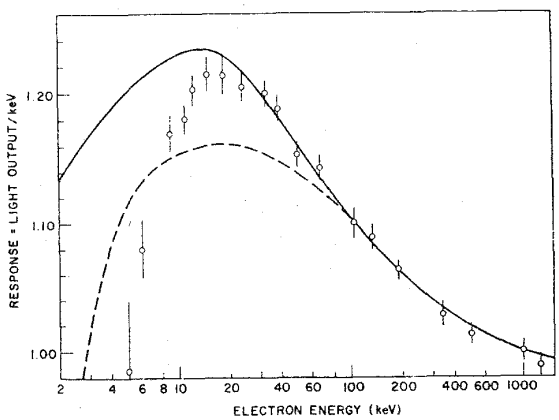

Fig. 7. Response of NaI(Tl^+) to electrons as a function of electron energy normalized to unity at 1 MeV. The data points were obtained by means of electron excitation [36]. The solid curve was calculated in [35] and the dashed curve in [32] from gamma ray response curves [36].

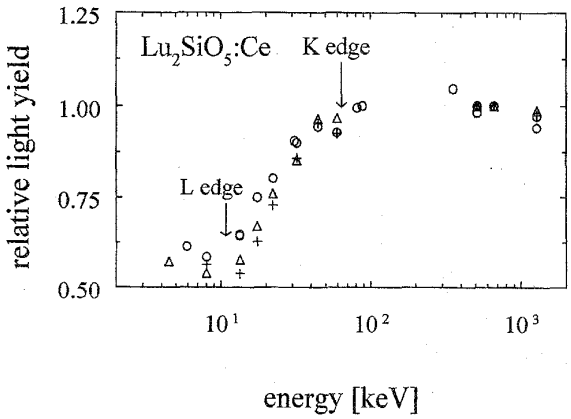

Fig. 8. Scintillation response curve of three difference $Lu_2SiO_5(Ce^{3+})$ crystals. +, △, and ○, data points for a crystal with an absolute light yield at 662 keV of 22 000, 25 000, and 13 000 ph/MeV, respectively [39].

by a Compton scattered electron in that scintillator can be calculated. The light output of the scintillator versus the calculated energy can provide electron response curves over a wide range of energies.

Most detailed studies have been performed on NaI(Tl^+), much less is known for the other "classical" scintillators. A few reports have appeared on the electron response curve of CsI(Tl^+) [32], [33]. It also shows a smoothly varying function with a maximum near 10-20 keV. For the other "classical" scintillators and also the newly discovered ones, to our knowledge, electron response curves have not been determined.

B. $CdWO_4$ and "Modern" Scintillators (New Results)

Our research on nonproportionality was initiated by the results on the recently discovered scintillator $Lu_2SiO_5(Ce^{3+})$ [13], [16]. We observed a light yield of 25 000 photons/MeV at 662 keV excitation energy whereas excitation with an X-ray tube (Cu-anode operated at 35 kV and 25 mA) yielded 15 000 photons/MeV [39], [40]. Furthermore initial reports on the energy resolution of $Lu_2SiO_5(Ce^{3+})$ at 662 keV excitation were somewhat disappointing, i.e., 10.3% [15], and we suggested that it might be related to nonproportionality [39].

For determining the absolute and relative light yield, a crystal is mounted with an optical coupling compound (Viscasil 60 000 cSt from General Electric) to the window of a Philips XP2020Q photomultiplier tube (PMT) with a CERN type 4238 voltage divider. The crystal was covered with 0.1 mm thick ultraviolet reflecting Teflon tape. We used ^{241}Am, ^{109}Cd, ^{57}Co, ^{203}Hg, ^{133}Ba, ^{137}Cs, and ^{22}Na γ-ray sources for exciting the crystals at energies between 59 keV and 1.27 MeV. An Amersham (code AMC.2084) variable X-ray source was employed to excite the crystals at energies between 8 and 44.5 keV. In this source, ^{241}Am produces characteristic K_α and K_β X-rays from Cu, Rb, Mo, Ag, Ba, and Tb targets. X-rays of 5.9 keV were obtained from a ^{55}Fe source, and a ^{55}Fe source combined with a Ti target was used for the characteristic 4.5 keV Ti X-rays.

Pulse height spectra of the above excitation sources were recorded using standard spectroscopic techniques [39], [41]. The relative light yield was obtained by comparing the position of the photopeak at energy, say, E with the position of the photopeak at energy 662 keV in the pulse height spectrum of ^{137}Cs. Drift in the gain of the PMT was monitored by measuring single photoelectron pulse height spectra, and if necessary a correction was made.

Depending on the type of excitation source used or crystal studied, the photopeak of interest was accompanied by satellite peaks. These peaks are either due to escape of characteristic X-rays from the crystal or because the source emits more than one type of X-ray, e.g., K_α and K_β X-rays. Therefore, in order to determine the location of the photopeak of interest, we occasionally fitted the composite photopeak with a sum of several Gaussian shaped peaks. In cases of low energy excitation ($\approx$10 keV) of relatively low light output crystals, the energy resolution of the photopeak becomes rather poor and a Poisson fit was performed. The error in the data points caused by errors in the determination of the relative gain of the PMT and the position of the photopeak is typically 3%. Since this error is also present in the normalization point at 662 keV, there is also a systematic error of 3%. Sometimes the photopeak was not symmetrical in shape, probably caused by the presence of unresolved X-ray escape peaks on the low-energy side of the photopeak or the contribution of multiple Compton scatter events to the total absorption peak. Effectively this causes a small shift of the position of the peak maximum, and introduces an additional error in the data points.

Fig. 8 shows response curves of three different roughly polished $Lu_2SiO_5(Ce^{3+})$ crystals. Decreasing from 1 MeV, the response remains approximately constant down to 80 keV. It shows a small drop near the K-edge of Lu ions (63.3 keV) followed by a decrease to 55% near the L absorption edges (9.2–10.8 keV). Below 8 keV, the response tends to increase again. Melcher [42] reports for some polished $Lu_2SiO_5(Ce^{3+})$ crystals a constant response between energies from 20 keV to beyond 1 MeV, therefore quite different from the results presented in Fig. 8. The absolute light yield of the crystals

TABLE I

PHOTON YIELDS (N), PHOTOELECTRON YIELDS ($\overline{Np}$), AND ENERGY RESOLUTIONS (R) DETERMINED AT 662 keV GAMMA EXCITATION OF SCINTILLATORS OPTICALLY COUPLED TO PHOTOMULTIPLIER TUBES WITH BIALKALI PHOTOCATHODES. R_M AND R_s ARE THE PHOTOMULTIPLIER AND SCINTILLATOR RESOLUTIONS CALCULATED FROM THE PHOTOELECTRON YIELD AND RESOLUTION COMPILED IN THE THIRD AND FOURTH COLUMNS, RESPECTIVELY. THE REFERENCE IN COLUMN NINE PERTAINS TO THE DATA IN COLUMNS THREE THROUGH EIGHT. SEE THE TEXT FOR A DISCUSSION ON THE PHOTON YIELDS COMPILED IN THE SECOND COLUMN

crystal	N 10^3 ph/MeV	$\overline{Np}$ phe/662 keV	R %	R_M %	R_s %	dimension mm^3	PMT	ref.
NaI(Tl$^+$)	38-43	6,230	6.5	3.1	5.7±0.2	Ø25×12.5	R1306	[50]
		4,900	7.1	3.7	6.1±0.2	10×10×20	R1306	[44]
CsI(Tl$^+$)	65	3,240	7.3	4.3	5.9±0.3	Ø18×18	XP2020Q	[53]
		3,640	5.7	4.2	3.9±0.4	10×10×7	R1306	[44]
CsI(Na$^+$)	42	5,740	7.4	3.3	6.6±0.3	10×10×7	R1306	[44]
$CaF_2(Eu^{2+})$	24							
$CdWO_4$	≈28	2,380	6.8	5.2	4.4±0.4	Ø25×12.5	R1306	[50]
		1,200	8.0	7.3	3.3±0.6	10×10×20	R1306	[44]
$Bi_4Ge_3O_{12}$	9	960	9.3	8.1	4.2±0.6	24×24×15	R1306	[57]
		960	9.0	8.1	3.9±0.7	10×10×10	R1306	[44]
BaF_2	11	1,590	7-8	6.2	4±1	Ø24×10	XP2020Q	[58]
		1,590	7.7	6.2	4.6±0.5	Ø20×10	XP2020Q	[59]
$Gd_2SiO_5(Ce^{3+})$	8.5-10	1,250	7.8	7.3	2.7±1.0	10×10×10	R878	[61, 62]
		1,480	7.8	6.6	4.2±0.5	10×10×10	R1306	[44]
$YAlO_3(Ce^{3+})$	14,3	1,900	7.2	5.7	4.4±0.5	21×21×21	R2059	[69]
		1,740	7.5	5.9	4.6±0.5	Ø10×1	XP2020Q	[67]
$Lu_2SiO_5(Ce^{3+})$	23	3,360	7.9	4.4	6.6±0.4	10×10×10	R878	[42]
$Lu_3Al_5O_{12}(Sc^{3+})$	23	2,320	6.5	5.1	4.0±0.4	11×5×1	XP2020Q	[17, 18]
$K_2LaCl_5(Ce^{3+})$	28	3,900	5.1	3.9	3.3±0.4	Ø7×2.5	XP2020Q	[19]

with a constant response is about a factor of two lower (12 000 photons/MeV) than that of the best crystals grown to date. Whether the improved proportionality and the lower light yield are related with properties of the bulk crystal or with the surface conditions of the crystals is still not clear.

Besides $Lu_2SiO_5(Ce^{3+})$, we studied the response curve of $CdWO_4$ and the "modern" scintillators BaF_2, $Gd_2SiO_5(Ce^{3+})$, $YAlO_3(Ce^{3+})$, $Lu_3Al_5O_{12}(Sc^{3+})$, and $K_2LaCl_5(Ce^{3+})$. The response curve of $Gd_2SiO_5(Ce^{3+})$, see Fig. 9, resembles the one of $Lu_2SiO_5(Ce^{3+})$. The decrease near the L-edges of Gd ions (7.3–8.4 keV) is somewhat less than for $Lu_2SiO_5(Ce^{3+})$. Fig. 10 shows the response curve of $CdWO_4$. The response curves of BaF_2 in Fig. 11, $YAlO_3(Ce^{3+})$ in Fig. 12, $Lu_3Al_5O_{12}(Sc^{3+})$ in Fig. 13, and $K_2LaCl_5(Ce^{3+})$ in Fig. 14 are between 10 keV and 1 MeV constant within about 10%. For these four scintillators, a drop in the response near the K edge is not observed.

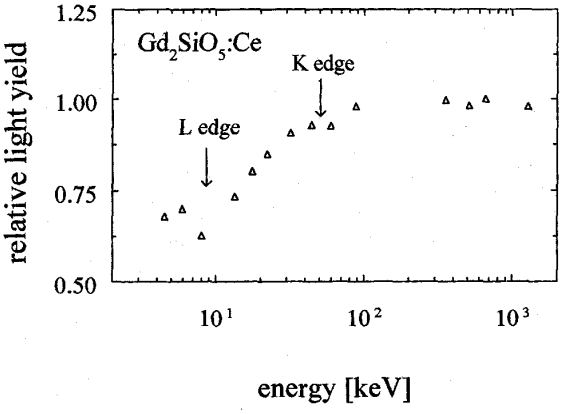

Fig. 9. Scintillation response curve of $Gd_2SiO_5(Ce^{3+})$.

IV. LIGHT YIELDS AND ENERGY RESOLUTIONS OBTAINABLE WITH SCINTILLATOR-PMT COMBINATIONS

Below and in Table I, a review is presented on the energy resolution, photoelectron yield, and light yield obtainable with scintillators optically coupled to a photomultiplier tube. The review and discussion is limited to the case of full absorption of 662 keV gamma quanta in the scintillator. Often there have been improvements in crystal quality in the years following the discovery of a new scintillator. Consequently improvements in energy resolution have been reported. We therefore selected those papers for which we believe they report on the best crystals manufactured to date. Because the energy resolution may depend on crystal size, its dimensions are of importance for the interpretation of the data. If available, results are compiled on crystals with volumes of a few cm^3. The energy resolutions and light yields reported in this work were either determined with Philips XP2020, XP2020Q and XP2262 type or Hamamatsu R1306, R878, and R2059 type photomultiplier tubes. All these tubes have a bialkali photocathode and a multistage electron multiplier.

A source of light yield data is the work by Holl *et al.* [43] who carefully determined the absolute photon yield of several of the scintillators discussed in this work. Data on the light

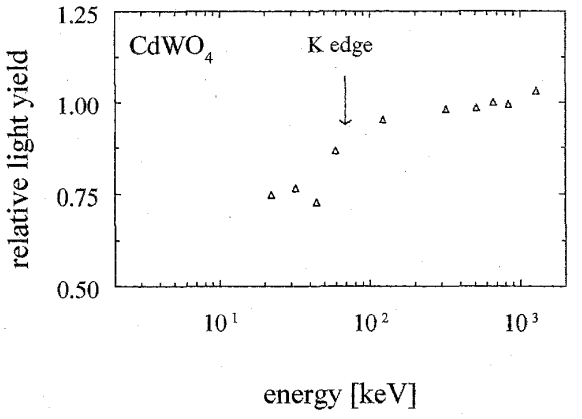

Fig. 10. Scintillation response curve of $CdWO_4$.

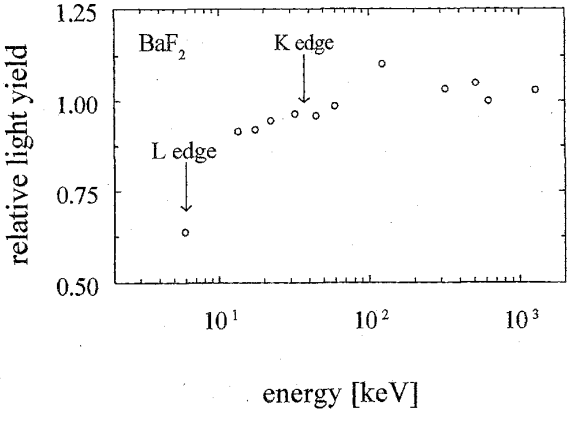

Fig. 11. Scintillation response curve of BaF_2.

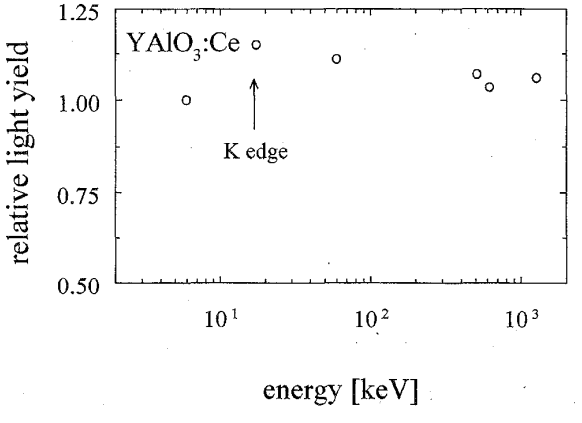

Fig. 12. Scintillation response curve of $YAlO_3(Ce^{3+})$.

yield, photoelectron yield, and energy resolutions of many scintillators have also been reported by Sakai [44].

The interpretation of photoelectron yields reported in the literature requires some explanation.

Usually photoelectron yields are determined by the method of Bertolaccini *et al.* [45] in which the pulse height of

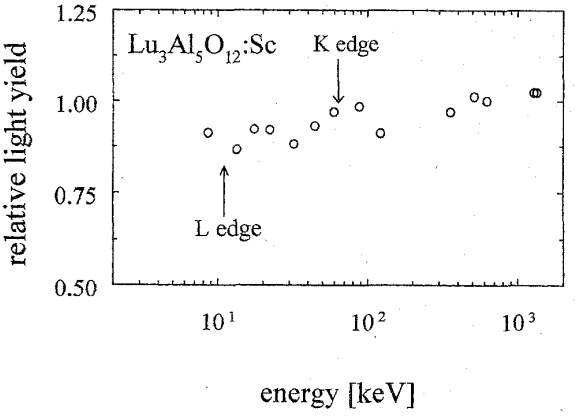

Fig. 13. Scintillation response curve of $Lu_3Al_5O_{12}(Sc^{3+})$.

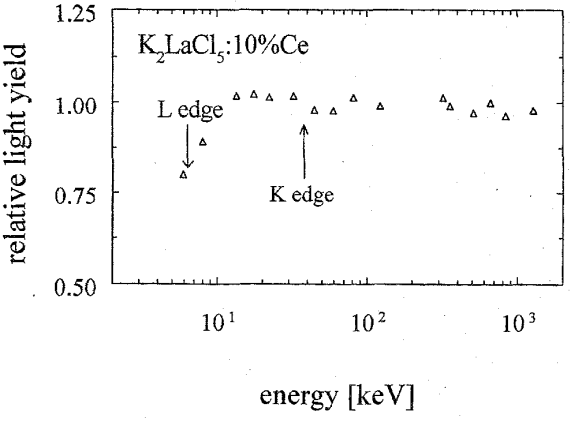

Fig. 14. Scintillation response curve of K_2LaCl_5:10% Ce^{3+}.

the photopeak is compared with the pulse height of single photoelectrons from the photocathode. A typical single photoelectron pulse height spectrum is shown in Fig. 15. The wide peak near channel 350 is due to photoelectrons which have experienced the full multiplication in the electron multiplier. In PMT's, a considerable fraction (10–20%) of the incident photoelectrons are inelastically scattered from the first dynode [46]. For these electrons there is no multiplication at the first dynode, and they appear as very small signal pulses in the pulse height spectrum. In the spectrum of Fig. 15 they are located below channel 50. With the method of Bertolaccini *et al.* one determines therefore the number of photoelectrons which undergo the full multiplication process in the PMT, i.e., one determines the value of $\overline{Np}$ in (8).

Sakai [44] employs another method for determining the photoelectron yield. The photomultiplier is used in photodiode mode. In the photodiode mode, the grid, the first and the second dynodes of the PMT are connected together with the input of a charge-sensitive preamplifier. Since there is no multiplication in the PMT, the inelastically scattered photoelectrons also contribute to the number of photoelectrons. Furthermore, in this method, there is a more efficient collection of photoelectrons than in the method of Bertolaccini, *et*

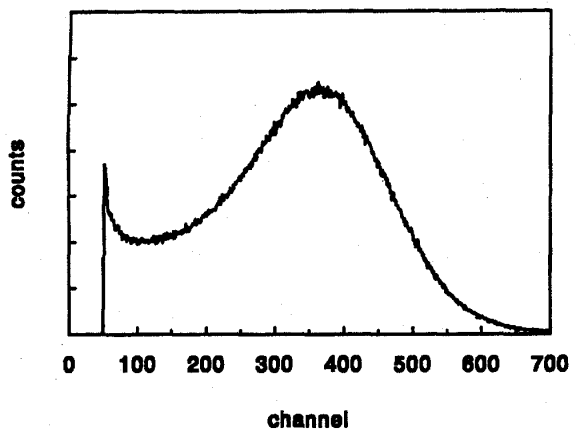

Fig. 15. Typical single photoelectron pulse height spectrum obtained with an XP2020Q tube at a gain of 3×10^7.

al.. Although the photodiode method gives a more accurate estimate of the intrinsic scintillation light output, it does not determine the value of $\overline{Np}$ in which we are interested. Based on comparison of the results by Sakai with photoelectron yield data reported elsewhere employing the method of Bertolaccini, we estimate the values of $\overline{Np}$ to be 20% lower than the photoelectron yields reported by Sakai.

In order to obtain absolute photon yields of the scintillators from reported photoelectron yields one needs to know the mean transfer efficiency. It is largely determined by the quantum efficiency of the PMT averaged over the emission spectrum of the scintillator. It is also determined by the light collection efficiency in the scintillator and the photoelectron collection efficiency in the PMT. Quantum efficiencies and collection efficiencies are often not accurately known which makes it difficult to determine the photon yield.

In Table I, the light yields, photoelectron yields and energy resolutions of several scintillators are compiled. Below we will give for each scintillator some comments about how the data were obtained.

NaI(Tl^+): NaI(Tl^+) shows a broad emission band near 415 nm with a main decay time of 230 ns. Coupled to modern PMT's with bialkali photocathodes, energy resolutions down to 6.5% at 662 keV can be obtained (6.8% (R878 PMT) [49], 6.5% (R878 PMT) [50]). Photoelectron yields of 9000 phe/MeV ((XP2020) [51]), 10 350 ((R878) [49]), and 11 970 ((R1306) [44]) have been reported. Considering the overestimation of the photoelectron yield in the photodiode method used by Miyajima, *et al.* and Sakai, we obtain that typical photoelectron yields of 9400 phe/MeV are obtainable with PMT's with a bialkali photocathode. For the photon yield of NaI(Tl^+), Holl *et al.* [43] report values between 38 000 and 43 000 photons/MeV. Results by Sakai on a NaI(Tl^+) crystal with an estimated photoelectron yield of 7400 phe/MeV and an energy resolution of 7.07% are given in Table I.

CsI(Tl^+): CsI(Tl^+) shows a broad emission band extending from 350 to 700 nm with a maximum near 560 nm [52]. Schotanus, *et al.* [53] report a yield of 4900 phe/MeV and an energy resolution of 7.3% employing an XP2020Q PMT and 6 μs shaping time. There have appeared several reports on the scintillation decay of CsI(Tl^+) [52]. Schotanus *et al.* report a fast component of about 600 ns and a slow component of 3.4 μs with intensity ratio fast/slow = 1.2. Valentine *et al.* [54] report a fast decay component of 800 ns and a slow one of 6 μs with intensity ratio of 1.5. From the work of Sakai, who reports an energy resolution of 5.7% employing a shaping time of 4 μs, we estimate a photoelectron yield of 5500 phe/MeV (see Table I). Because the emission of CsI(Tl^+) extends to beyond 600 nm where bialkali photocathodes show a diminishing quantum efficiency, the mean transfer efficiency is relatively low (0.09–0.1) and may differ depending on the type of PMT used. Holl *et al.* [43] report for the total photon yield a value of about 52 000 ph/MeV, and Valentine *et al.* [54] report a yield of 64 000 $\pm$ 3200 ph/MeV. The about 25% larger output reported by Valentine, *et al.* was attributed to the ballistic deficit.

CsI(Na^+): The scintillator CsI(Na^+) shows an emission band near 425 nm. Brinckmann [4] reports a decay time of 650 ns. We observe a 850 $\pm$ 150 ns component with an additional 6 $\pm$1 μs slow component [55]. The photon yield is about equal, i.e., 42 000 ph/MeV, to that of NaI(Tl^+) crystals [43], [44]. We did not find many papers on the energy resolutions and photoelectron yields obtainable with CsI(Na^+); the results in Table I were obtained from the work of Sakai [44].

$CaF_2(Eu^{2+})$: This material, which was discovered as a scintillator by Menefee *et al.* in 1966 [5], has a 50 nm wide emission band near 435 nm and a decay time of 730$\pm$50 ns [55]. For the light output a value of 24 000 photons/MeV reported by Holl *et al.* [43]. Unfortunately data on the energy resolution at 662 keV are not available; due to the very small photoelectric absorption coefficient of $CaF_2(Eu^{2+})$; a clear 662 keV photopeak is not present in the pulse height spectrum of ^{137}Cs.

$CdWO_4$: The luminescence properties of $CdWO_4$ have been known since the work of Kröger [2] in 1948. It shows a broad emission band near 480 nm with a decay curve that can be characterized by a slow and a fast component (5 and 20 μs [56], 2 and 15 μs [50]). Despite the long history of $CdWO_4$, improvements in crystal quality are still being made. Kinloch *et al.* [50] report on $CdWO_4$ crystals showing less self absorption of scintillation light than $CdWO_4$ crystals reported on before. Employing shaping times of 12 μs, they obtain photoelectron yields of 38% relative to a good quality NaI(Tl^+) crystal; i.e., 3600 phe/MeV (see Table I). From the results of Sakai [44] performed on a poorer quality $CdWO_4$ crystal and with a shaping time of 4 μs, we estimate a photoelectron yield of 1800 phe/MeV (see Table I). Taking into account that part of the $CdWO_4$ emission spectrum falls beyond the sensitivity of bialkali photocathodes and the ballistic deficit, we estimate an absolute photon yield of 28 000 ph/MeV. Note, that due to the improved quality of the $CdWO_4$ this is significantly larger than the value of 20 000 ph/MeV reported by Holl, *et al.* [43].

$Bi_4Ge_3O_{12}$: $Bi_4Ge_3O_{12}$ shows a 150 nm (FWHM) wide emission band at a wavelength of 490 nm [6], [7]. There is an initial decay of 60 ns followed by a main component of 300 ns decay time [51]. Due to improvements in crystal quality, the light yield and energy resolution have improved over the

years. Photoelectron yields of 15–16% relative to high quality NaI(Tl^+) crystals [57], [44], i.e., 1450 phe/MeV, and energy resolutions of 9% [44] and 9.3% [57] can be obtained. With an estimated transfer efficiency of 0.16, a photon yield of 9000 ph/MeV is obtained which agrees with results reported by Holl, *et al.* [43].

BaF_2: The interest in BaF_2 as a scintillator stems from its very fast (<1 ns) decay component [8], [9]. A few years ago we reported for BaF_2, studied by means of an XP2020Q PMT, a total yield of 2400 phe/MeV corresponding with a photon yield of 11000 ph/MeV [41]. The best energy resolutions have been reported by Sperr [58] and Zhu, *et al.* [59], see Table I.

$Gd_2SiO_5(Ce^{3+})$: $Gd_2SiO_5(Ce^{3+})$ shows a 70 nm wide emission band near 440 nm, and it was first reported by Takagi, *et al.* [10] in 1983. At the optimum Ce^{3+} concentration near 0.5 mol% [60], [61], a fraction of 85–90% of the total light yield shows a fast decay of 56 ns; the remaining fraction has 600 ns decay time [61]. Photoelectron yields of 20% relative to that of NaI(Tl^+), i.e., ≈1900 phe/MeV, were reported by Ishibashi *et al.* [60] and Melcher *et al.* [62]. From the results reported by Sakai a photoelectron yield of 2200 is estimated. Assuming a mean transfer efficiency of 0.22, we estimate photon yields of 8500–10 000 ph/MeV.

$YAlO_3(Ce^{3+})$: Photo excited luminescence properties of $YAlO_3(Ce^{3+})$ crystals have been studied for many years because of their potential application as a laser crystal [63], [64]. Cathode luminescence properties were reported by Takeda *et al.* [11] in 1980 and later by Autrata *et al.* [12] in 1983. The optimum Ce^{3+} concentration is near 0.2 weight% [65]. The emission of $YAlO_3(Ce^{3+})$ at 360 nm and the light yield depends slightly on the method of crystal growth [66]. $YAlO_3(Ce^{3+})$ shows a main decay component of 36 ns, but also a slow component of about 10 μs which constitutes 5–10% of the total emission intensity [67]. Ziegler *et al.* [68] reports, employing 1 μs integration time, a yield of 2670 phe/MeV and a resolution of 6.7% for 511 keV γ-ray excitation [68]. Kierstead *et al.* [69] reports, employing 100 ns integration time, 2900 photons/MeV and 7.1% resolution at 662 keV. Employing 0.5 μs shaping time, we observe a quite similar yield of 2650 phe/MeV and a resolution of 7.5% for 662 keV γ-ray excitation. A yield of 3300 phe/MeV was found if shaping times of 4 μs were used [67]. This corresponds with a yield of 14 300 photons/MeV.

$Lu_2SiO_5(Ce^{3+})$: $Lu_2SiO_5(Ce^{3+})$ as a powder was studied in 1969 for its cathode luminescence properties by Gomes de Mesquita, *et al.* [70]. Scintillation properties on crystals were first reported in 1990 by Melcher [13] and Shul'gin *et al.* [16]. $Lu_2SiO_5(Ce^{3+})$ scintillates at 420 nm and it has the unique combination of a high density (7.4 g/cm^3), fast response (40 ns), and high light yield (25 000 ph/MeV) [62], [39]. The material is still actively being studied. An energy resolution of 10.3% was reported by Melcher *et al.* in 1992 [15]. Due to an improved crystal growth technology, the energy resolution has been increased to 7.9% for a 1 cm^3 crystal with a photoelectron yield of 54% relative to that of NaI(Tl^+) ; with smaller crystals resolutions down to 7.5% have been observed [42]. Recently we reported for crystals manufactured by Schlumberger Doll Research a yield of 4900 phe/MeV [39], [40] employing an XP2020Q PMT. Ludziejewski *et al.* [71] report yields of 4200 phe/MeV with an XP2020Q PMT on crystals manufactured at the Lebedev Physical Institute in Moscow.

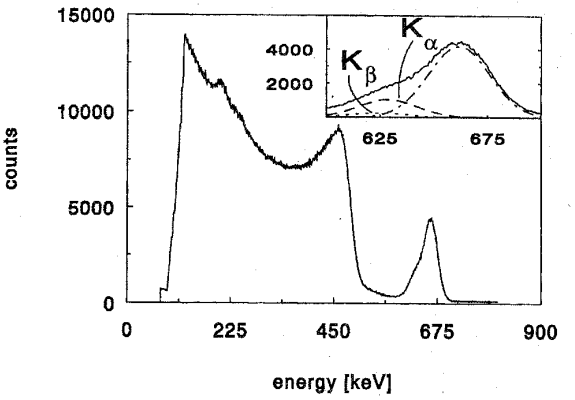

Fig. 16. ^{137}Cs pulse height spectrum obtained with a K_2LaCl_5 : 10 mol% Ce^{3+} crystal of dimensions Ø7 ×2.5 mm^3 optically coupled to an XP2020Q PMT. The inset shows a fit of the photopeak and escape peak with three Gaussian peaks representing the total absorption peak at 662 keV and the K_α and K_β escape peaks [19].

$Lu_3Al_{5-x}Sc_xO_{12}$: Luminescence properties of these materials were first reported by Batygov *et al.* [72]. The first detailed studies of its scintillation properties under X-ray and γ-ray excitation were recently reported by our group [17], [18]. The optimum Sc^{3+} concentration in terms of photon yield and speed of decay is near $x = 0.2$. $Lu_3Al_5O_{12}(Sc^{3+})$ scintillates near 275 nm with a main (89%) decay component of 610 ns and the rest has a relatively slow decay of 3.3 μs. The best resolution (6.5%) was observed with 2 μs shaping time yielding 3500 phe/MeV. 4150 phe/MeV were observed with 10 μs shaping time from which a photon yield of 23 000 ph/MeV is estimated.

$K_2LaCl_5(Ce^{3+})$: We are currently studying the scintillation properties of $K_2LaCl_5(Ce^{3+})$. The crystals scintillate near 370 nm caused by luminescence of the Ce^{3+} ions. Fig. 16 shows the pulse height spectrum of a ^{137}Cs source obtained with a crystal doped with 10 mol% Ce^{3+}. A fit of the 662 keV photopeak and the accompanying 629 keV escape peak with two Gaussian peaks reveals an excellent energy resolution of 5.1 ± 0.2% at 662 keV. The escape peak is a result of the escape of 33 keV characteristic X-rays of La^{3+} ions from the crystal. After 100 ns, the intensity of the scintillation pulse decays as $t^{-1.6}$ with time t and 90% of the light output occurs in 900 ns. With 10 μs shaping time a photoelectron yield of 5900 phe/MeV [19] is observed which corresponds with a photon yield of 28 000 ph/MeV. The optimum Ce concentration has not been determined yet and the optical quality of the crystals can still be improved. We therefore expect that the performance of $K_2LaCl_5(Ce^{3+})$ will be enhanced in the future.

V. Discussion

For the idealized case of normal scintillation variance and constant transfer efficiency one expects an energy resolution as given by (8). Values for $\overline{Np}$ at 662 keV excitation reported

TABLE II
THE CONTRIBUTIONS TO THE ENERGY RESOLUTION (FWHM) OF THE 662 KEV TOTAL ABSORPTION PEAK IN PULSE HEIGHT SPECTRA OF ^{137}Cs MEASURED WITH NaI(Tl$^+$) SCINTILLATORS OPTICALLY COUPLED TO PHOTOMULTIPLIER TUBES. ALL RESOLUTIONS ARE IN %. R_{np} IS DEFINED AS $\sqrt{R_C^2 + R_\delta^2}$

R_C	R_δ	R_{np}	$\sqrt{R_{inh}^2 + R_p^2} = \sqrt{R_s^2 - R_{np}^2}$	$R_s = \sqrt{R^2 - R_M^2}$	R_M	R	Reference
4.0	–	4.0	4.6	6.1	4.9	7.8	Zerby *et al.* [35]
4.1	3.2	5.2	1.5	5.4	5.5	7.7	Iredale [73]
–	–	6.6	1.5	6.8	3.7	7.8	Narayan *et al.* [74, 75]
5.8	3.1	6.6	–	–	–	–	Prescott *et al.* [37]
3.3	3.1	4.5	3.5	5.7	3.1	6.5	from Table 1

in the literature can be found in the third column of Table I. For calculating the gain variance $v(M)$ we have assumed that each PMT in Table I has been operated at its typical gain and with a voltage divider as recommended by the manufacturer [47], [48]. Employing (4) and (5), we obtain for R878, R1306, R2059, and XP2020Q gain variances of 0.19, 0.14, 0.10, and 0.09, respectively. Note, $v(M)$ can also be determined experimentally from the width of the single electron pulse height spectrum. For example, the XP2020Q PMT shows a typical single photoelectron pulse height width of 70% [48] (see also Fig. 15). Using the relation $(FWHM) = 2.35\sqrt{v(M)}$, one obtains $v(M) = 0.088$, which agrees with the theoretical value of 0.09. With the above values for $v(M)$, the resolutions R_M were calculated, see Table I. R_s was obtained from $\sqrt{R^2 - R_M^2}$. The errors in R_s were calculated assuming relative errors of 3, 30, and 5% for R, $v(M)$, and the photoelectron yield at 662 keV, respectively.

A. The Energy Resolution of NaI(Tl$^+$)

The energy resolution obtainable with NaI(Tl$^+$) has been studied extensively in the 20 years after its discovery in 1948. It was soon realized that the resolution is not simply determined by the photomultiplier resolution alone. Contributions due to nonnormal variance in the photon production and due to the transfer variance are also important. One important contribution stems from the nonproportional response of the scintillator to electrons [see Fig. 7 and (7)]. The creation of secondary electrons, i.e., photoelectrons, Compton electrons, and Auger electrons, upon total absorption of the energy of a γ-quantum is a statistical process. Principally, the number of photons from the crystal N is given by

$$N = \sum_{i=1}^{n} Y(E_i)E_i \tag{9}$$

where the summation runs over all n secondary electrons. E_i is the energy of the ith electron and $Y(E_i)$ is the value of the electron response curve at that energy expressed in photons/(unit of electron energy). The nonconstancy of $Y(E_i)$ and the statistics in the n and E_i manifest themselves as a variation in the light output for totally absorbed γ-rays resulting in a broadening of the pulse height spectrum [35].

Based on the electron response curve, solid curve in Fig. 7, Zerby, *et al.* [35] calculated the different contributions to the observed resolution of $R = 7.8\%$ for a Ø2.5 × 2.5 cm^3 NaI(Tl$^+$) ; their results are compiled in Table II. They calculated a 4% contribution to the intrinsic resolution by multiple Compton scatter processes. This Compton contribution, which is denoted as R_C in Table II, depends on the crystal size. For very small crystals, multiple Compton scatter events become unlikely and R_C goes to zero. R_C also becomes very small at energies below 100 keV when γ-quanta are predominantly absorbed by the photoelectric effect.

Besides photoelectrons, Compton electrons and Auger electrons, one may also consider the creation of δ-rays in the crystal. The electron response curve calculated from the γ-response curve will then be somewhat different [37]. Iredale [73] calculated the effects of such secondary δ-rays which contributes an extra $R_\delta = 3.2\%$ to the intrinsic resolution. More elaborate studies by Narayan and Prescott [37], [74], [75] taking into account Compton, photoelectric, Auger and δ-ray creation processes confirmed the results of Zerby *et al.* [35] and partly those of Iredale [73]. Their results on the intrinsic line width are shown in Fig. 17. The authors found that the value of R_C depends quite critically on the electron response function. Small changes in the chosen response may lead to variations in the calculated R_C by a factor of 3 to 4 [37]. Delta rays increase the linewidth at low energy (<100 keV), where their probability of occurrence becomes relatively high, and again at high energies (>500 keV) where the range of energy available for the delta-rays is greater [37]. Although R_δ is not a dominant contribution to R_i it may become important for small crystals when R_C becomes very small.

The difference in energy resolution of 7.8% at 662 keV reported about 30 years ago and the resolution of 6.5% obtainable currently can be attributed to the better quality of photomultiplier tubes resulting in lower values for R_M and R_p. The values for the resolutions R_s, R_M, and R in the last row of Table II pertain to the Ø25 × 12.5 mm^3 NaI(Tl$^+$) crystal of Table I. The value of R_C for this crystal is an estimation based on the work of Zerby *et al.* [35]. Adopting the calculated results of Prescott *et al.* [37], we assumed a value of 3.1% for R_δ. These results indicate that for NaI(Tl$^+$), the main contribution to the energy resolution stems from its nonproportional response to electrons.

B. Energy Resolution of Other Crystals

In a Compton scatter event of a 662 keV γ-quantum followed by photoelectric absorption of the inelastically scattered γ-quantum, secondary electrons of energies typically

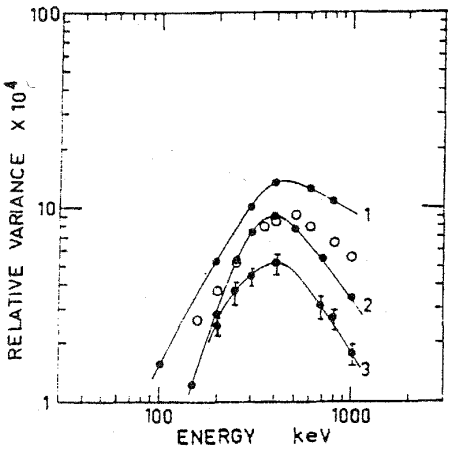

Fig. 17. The intrinsic variance of NaI(Tl^+) as a function of gamma-ray energy. Curves 1, 2, 3 are computed employing different electron response curves: open circles show the effect of including delta-rays in calculation 2 [37].

200–450 keV are created. The Compton contribution to the energy resolution is therefore sensitive to the electron response curve in this energy range. The Compton contribution is also dependent on the relative amount of events in the 662 keV photopeak originating from multiple Compton scattering. Its contribution decreases with 1) a decrease of the sample size and 2) increasing atomic number of the elements in the scintillator. A high effective atomic number provides the crystal with a large photoelectric absorption coefficient.

Inspection of the calculated scintillator resolution R_s in Table I shows that its value is relatively large (6–6.6%) for NaI(Tl^+), CsI(Na^+), and $Lu_2SiO_5(Ce^{3+})$. For these scintillators the contribution of R_s to the energy resolution R dominates over the contribution from R_M. The response curve of CsI(Na^+) and $Lu_2SiO_5(Ce^{3+})$ to electrons is not known and calculations on R_C and R_δ have not been reported yet. Considering that the response of $Lu_2SiO_5(Ce^{3+})$ is reasonably constant between 100 and 662 keV and it has high density and large effective atomic number, the Compton contribution to the resolution is expected to be quite insignificant. CsI(Tl^+) and CsI(Na^+) show a nonconstant response at energies between 100 and 662 keV, and for these crystals one may expect a significant Compton contribution.

For the relatively low light output crystals $CdWO_4$, $Bi_4Ge_3O_{12}$, BaF_2, $Gd_2SiO_5(Ce^{3+})$, and $YAlO_3(Ce^{3+})$, the energy resolution is dominated by the contribution from the Poisson statistics in the number of photoelectrons, i.e., in R_M. The scintillator contribution is typically 4%.

$Lu_3Al_5O_{12}(Sc^{3+})$ shows the same energy resolution as NaI(Tl^+) despite its almost three times lower photoelectron yield. The energy resolution obtainable with $K_2LaCl_5(Ce^{3+})$ is even significantly better. For these two crystals, R_M is larger than for NaI(Tl^+), but this is compensated by a smaller value for the scintillator resolution. Because of the very good proportionality in the scintillation response of both $Lu_3Al_5O_{12}(Sc^{3+})$ and $K_2LaCl_5(Ce^{3+})$ see Figs. 13 and 14, we expect the contributions of R_C and R_δ to be quite small. Consequently, for these crystals, R_s is mainly determined by the transfer variance.

C. Possible Origins of Nonproportionality

With (9), it is in principle possible to calculate both the light output of a scintillator under γ-ray excitation and the contribution of R_{np} to the energy resolution. The light yield is determined by the value of N averaged over many γ-ray absorption events. The resolution is determined by the spread (FWHM) in N. To employ (9) one needs accurate electron response curves $Y(E_i)$ which at present are not available. Furthermore, one needs to know for a large number of γ-ray absorption events the energies E_i of the collection of n secondary fast electrons created in the scintillator. This latter information can be obtained from Monte Carlo simulations of the events following the absorption of a γ-quantum. Currently we are working on developing a program to perform such calculations. Experimental data is still needed on the electron response curves. In this respect, the method proposed by Valentine and Rooney [38] seems to be promising. Once electron response curves are known, its influence on R_{np} and γ-ray response curves can be calculated.

What is the origin of nonproportional response to electrons? From the theory of interaction of high energy electrons with matter, it is known that the ionization density in the track increases with decreasing primary electron energy. This phenomenon was used by Murray *et al.* [76] in order to explain the nonproportional response observed for NaI(Tl^+) and CsI(Tl^+). They proposed that a free electron and a free hole in the ionization track first need to form an exciton like state. Next the exciton is trapped by a Tl^+ center followed by luminescence. The probability of forming an exciton increases with the concentration of free electrons and holes, i.e., with the ionization density in the track. As a result the scintillation light yield increases with a decrease of electron energy.

One can also speculate about a relationship between light yield and the density of free electrons and holes in the ionization track compared with the density of luminescence centers. Suppose, for example, the electron and hole density is larger than the density of luminescence centers. One can then imagine that some of the free electrons and holes do not recombine at a luminescence center and are lost in the scintillation process. In this case, a decrease of light yield with decreasing electron energy results. It is clear that one needs very detailed knowledge about the scintillation mechanism to explain nonproportional response based on such arguments. Unfortunately for almost all scintillators, this information is not available.

A completely different origin of nonproportional response may be related with surface effects [31]. If the mean penetration depth of low energy X-rays in the scintillator becomes very small, part of the luminescence would be created near the surface of the crystal. Suppose that near the surface the scintillator is less efficient then, effectively the light yield decreases. The penetration depth of X-rays is inversely proportional to the photoelectric absorption coefficient of the scintillator material. Generally, this coefficient increases with

decreasing energy and shows a discontinuous drop at the K-shell and L-shell binding energies. As a result, this model predicts a decrease of scintillation efficiency with decreasing energy except just below K-shell and L-shell binding energies where the efficiency is expected to increase slightly. Note, that such behavior can be observed in several of the response curves presented in this work. Whether this is really related with surface effects is still being studied.

VI. Summary and Conclusion

A review and new data have been presented on the proportionality of the scintillation response as a function of γ-ray energy. These response curves behave quite unpredictably. Starting at 662 keV, the response curves of NaI(Tl^+), CsI(Tl^+), and CsI(Na^+) increase by 20–30% upon lowering the γ-ray energy to 20 keV. Instead of an increase, $CaF_2(Eu^{2+})$, $Bi_4Ge_3O_{12}$, $Lu_2SiO_5(Ce^{3+})$, and $Gd_2SiO_5(Ce^{3+})$ show a decrease by 20 to almost 50%. For both series of crystals, one observes a drop in the light yield near the K-shell and L-shell absorption edges of the most heavy ions in the crystals. BaF_2, $YAlO_3(Ce^{3+})$, $Lu_3Al_5O_{12}(Sc^{3+})$, and $K_2LaCl_5(Ce^{3+})$ show a more constant response; variations are at most 15%. Furthermore, anomalies near the K-shell absorption edge are not observed.

From the experimental energy resolution at 662 keV and the photoelectron yield, the photomultiplier resolution R_M and scintillator resolution R_s were determined. The results show that the scintillator resolution contributes significantly to the overall energy resolution, and in some cases like NaI(Tl^+) it even dominates.

There appears to be a close relationship between the nonproportionality of scintillators to γ-quanta and to electrons. Detailed knowledge on the scintillation mechanism and accurate electron response curves of scintillators are needed in order to explain quantitatively the causes of nonproportionality. Unfortunately both of these are not available yet.

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