

## STATUS OF PHOTON COUNTING USING SOLID SCINTILLATORS

R.L. Heath  
Idaho National Engineering Laboratory  
EG&G Idaho, Inc.  
Idaho Falls, Idaho, U.S.A. 83415

The purpose of this summary paper will be to present a review of recent developments in solid scintillation detectors, associated measurement techniques, and apparatus as applied to photon counting. Important advances in scintillator materials, optical sensors, and examples of application of solid scintillators in applied gamma-ray spectrometry will be presented as an aid to researchers and applied spectroscopists.

One of the most promising developments in recent years has been the appearance of Bismuth Germanate ( $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ ) as a scintillator. Advantages of this material are high density (7.13), stable mechanical and thermal properties, and resistance to radiation damage. Disadvantages, as compared to NaI for photon spectrometry, include low light output, high index of refraction which complicates optical coupling and a significant temperature coefficient for light output. At the present state-of-the-art in material and detector design, energy resolution of commercial detectors appears to be 8 to 12% (FWHM) for the .661 MeV  $^{137}\text{Cs}$  line. Considerable interest is being shown for the use of this scintillator material in commercial field applications and for photon spectrometry in nuclear structure and high-energy physics research. Design considerations and examples of applications in research will be presented in some detail.

### INTRODUCTION

The objective of this plenary paper is to present a review of recent developments and applications of solid scintillation detectors. This will include developments in inorganic phosphors and optical devices employed in scintillation detector design. In order to provide perspective, the characteristics of solid phosphor materials and optical sensors will be discussed in terms of detector performance

in a variety of applications.

The past decade has seen the development of multiple uses for scintillation detectors for the detection and characterization of x and  $\gamma$ -radiation. The requirements for radioisotopic assay in the environment, photon and particle spectroscopy in high energy and astrophysics and advanced medical imaging techniques have provided the incentive for the continued development of specialized solid scintillation detectors to meet scientific needs. Although the development of high-resolution solid-state detector devices using single-crystal Ge and Si resulted in superior performance for photon spectroscopy, solid scintillation detectors in many forms meet many specialized needs for the detection and characterization of particle and photon radiations.

#### Inorganic Phosphors

To meet the many needs for nuclear radiation detection systems, a variety of inorganic crystal scintillator materials are currently available.

Table 1 lists major inorganic phosphor materials which are currently available in commercial quantities and the important physical properties of these materials. A recent summary and discussion of the important properties of these materials has been presented by Farukhi<sup>1</sup>.

First, considering NaI(Tl), which is still by far the major solid scintillator in general use, considerable progress has been made, both in material technology and detector performance. NaI(Tl) is presently produced in single-crystal ingots up to 30" in diameter. Such large volumes of detector-grade material are used in high-energy physics research as total energy absorption spectrometers in fundamental particle experiments. A second important movement in NaI production technology has been the development of techniques to produce NaI(Tl) in polycrystalline form which can be extruded to produce detectors in many exotic shapes<sup>2</sup>. This material exhibits improved resistance to thermal and mechanical shock with negligible optical degradation.

The major characteristics of importance in detector response include density and atomic number, decay life time, and light output.

Table 1. Characteristics of Important Solid Scintillation Phosphors

Material	Wavelength of Maximum Emission (nm)	Decay Constant (μs)*	Scintillation Cutoff Wavelength (nm)	Index of Refraction	Density (g/cm <sup>3</sup> )	Hygroscopic	γ-Scintillation Conversion Efficiency (%)
NaI(Tl)	410	23	320	1.85	3.67	Yes	100
CaF <sub>2</sub> (Eu)	435	.94	405	1.44	3.18	No	50
CsI(Na)	420	.63	300	1.84	4.51	Yes	85
CsI(Tl)	565	1.0	330	1.80	4.51	No	45
ε <sub>l</sub> Li(Eu)	470-485	1.4	450	1.96	4.08	Yes	35
TlCl(Be-I)	465	0.2	390	2.4	7.00	No	2.5
CsF	390	.005	220	1.48	4.11	Yes	5
BaF <sub>2</sub>	325	.63	134	1.49	4.88	No	10
Bi <sub>4</sub> Ge <sub>3</sub> O <sub>12</sub>	480	.30	350	2.15	7.13	No	15
KI(Tl)	426	.24/2.5	325	1.71	3.13	Yes	24
CaWO <sub>4</sub>	430	.9-20	300	1.92	6.12	No	14-18
CdWO <sub>4</sub>	530	.9-20	450	2.2	7.90	No	17-20

Data courtesy of Harshaw Chemical Co.

Density and atomic number of phosphors determine the efficiency for photon detection and the relative probability for total energy absorption in a given detector volume. Phosphor lifetime is an important experimental parameter in the use of scintillation detectors. In the scintillation process, optical emission follows an exponential decay law with typical decay constants in the range 0.1 to several microseconds. In pulse counting systems, the decay constant determines resolution time in coincidence circuitry, limiting counting losses at high input pulse rates. Differences in lifetime between materials or response to different types of radiation provide the basis for differentiating between different types of radiation falling on the detector.

Although most excited states in a scintillator appear to exhibit the same lifetime, some phosphors are characterized by the presence of long-lived states. The existence of long-life-time components in the decay of a phosphor is referred to as "afterglow". All inorganic scintillator materials exhibit this characteristic to a degree. As indicated in Table 1,  $\text{CaF}_2(\text{Eu})$  and  $\text{Bi}_4\text{Ge}_3\text{O}_{12}$  exhibit minimal residual long-lived states. This effect is important in applications where integrated current output is used and in coincidence timing where prompt return of the signal to the base line influences the ability to trigger on the first few photoelectrons produced in a given event.

Other properties of phosphors and coupled light sensors which are important in the design and operation of solid scintillation detectors include temperature stability and mechanical properties. This is particularly important in field applications requiring operation in an extreme temperature and mechanical environment.

#### Bismuth Germanate

One of the more interesting developments in solid scintillators has been the discovery, development and application of  $\text{Bi}_4\text{Ge}_3\text{O}_{12}$  (Bismuth Germanate) as a scintillator. Luminescence of this material (termed BGO) was first studied by Weber and Monchamp<sup>3</sup> in 1973. A year later, Nestor and Huang<sup>4</sup> demonstrated the use of the material as a gamma-ray spectrometer. Since that time, it has seen a phenomenal growth in acceptance, in spite of many technical limitations. The

principal driving force behind this is high detection efficiency for energetic photons resulting from its density (7.13) and high Z. This is illustrated in Figure 1, which is a plot of the linear absorption coefficient,  $\mu$ , vs photon energy. For comparison, data are shown for BGO, NaI(Tl), and CsI(Tl).

Observing Table 1 again, BGO has minimal "afterglow" is non-hygroscopic, and mechanically rugged. This combined with high stopping power led to its becoming the detector of choice for x-ray CT instruments, replacing NaI(Tl) in this application. Although CdWO<sub>4</sub> is now coming into use for this application, this commercial success greatly enhanced the development of BGO as a scintillator material.

As large volumes of this material became available, interest grew in its potential use for gamma-ray spectrometry. Evans<sup>5</sup>, in 1980, studied the gamma-ray response of a 1-1/2" x 1-1/2" cylindrical BGO detector, demonstrating its potential for general use in gamma-ray spectrometry. The energy resolution of this device, which was fabricated from state-of-the-art material at that time, was about 15% (FWHM). Figure 2 compares the pulse-height spectra obtained with 1-1/2" x 1-1/2" cylindrical detectors of BGO and NaI(Tl) for a 15.0 hr Na<sup>24</sup> radioactive source. The major observation is the enhanced photo peak and pair-production escape peak-response of the BGO detector relative to NaI. This results from increased probability for multiple Compton and Compton-photoelectric events with increased electron density and higher Z value of Bismuth Germanate. Inspection of the relative absorption curves for NaI and BGO in Figure 1 indicates a total cross section for BGO between 2 and 3 times that for NaI from 0.1 to 10 MeV. The improved total-energy-absorption in BGO is shown in Figure 3 as a plot of full-energy peak efficiency vs. energy for 1-1/2" x 1-1/2" NaI and BGO detectors. These data are from the Evans study in Reference 5. From these data, we may conclude that BGO offers advantages for high-energy gamma-ray spectroscopy.

At the present time, progress continues to be made in material purity. By successive re-crystallization of material, considerable improvement has been realized in light output, resulting in improved energy resolution. Farukhi<sup>6</sup> has reported a value of 8.9% (FWHM) for the <sup>137</sup>Cs line for a 1" x 1" sample of material twice regrown. These

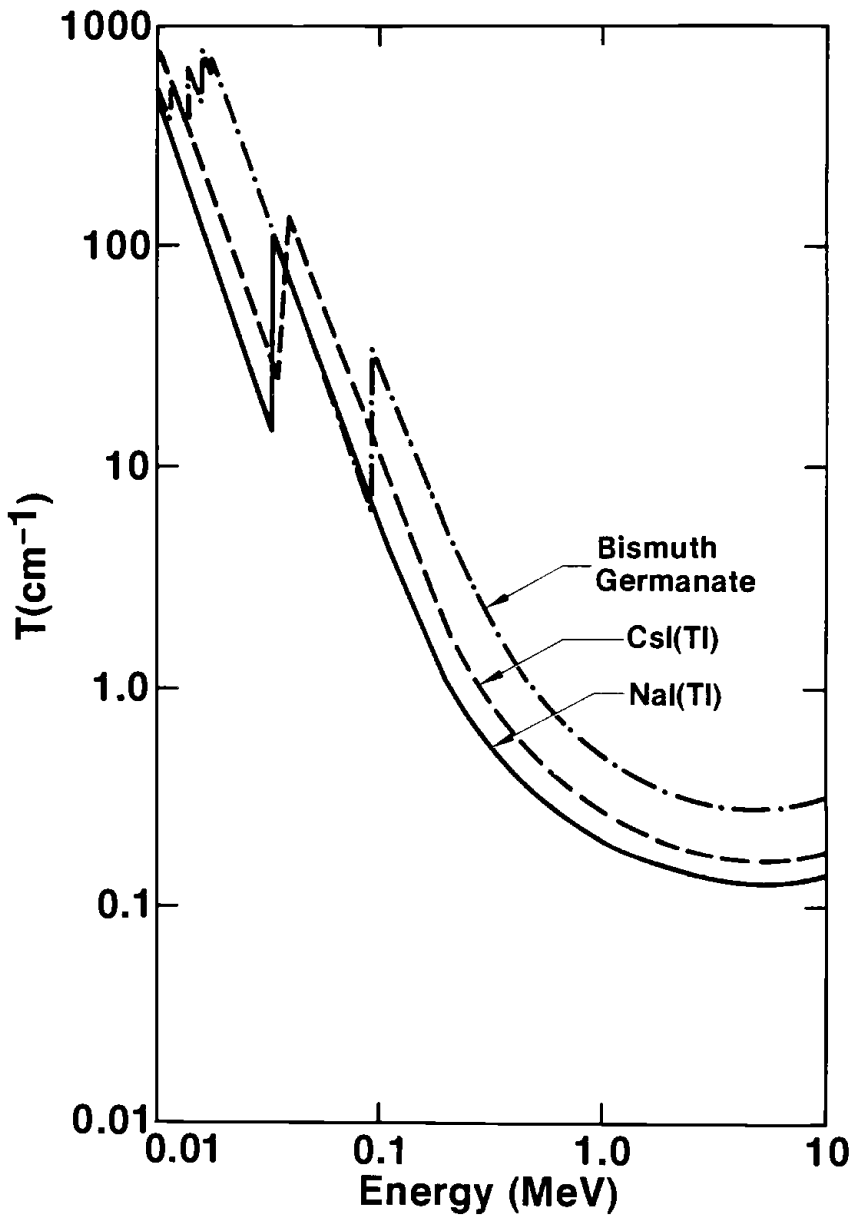


Figure 1. Plot of the Photon Absorption Coefficient,  $\mu$ , as a Function of Photon Energy for Bismuth Germanate (BG), CsI(Tl) and NaI(Tl).

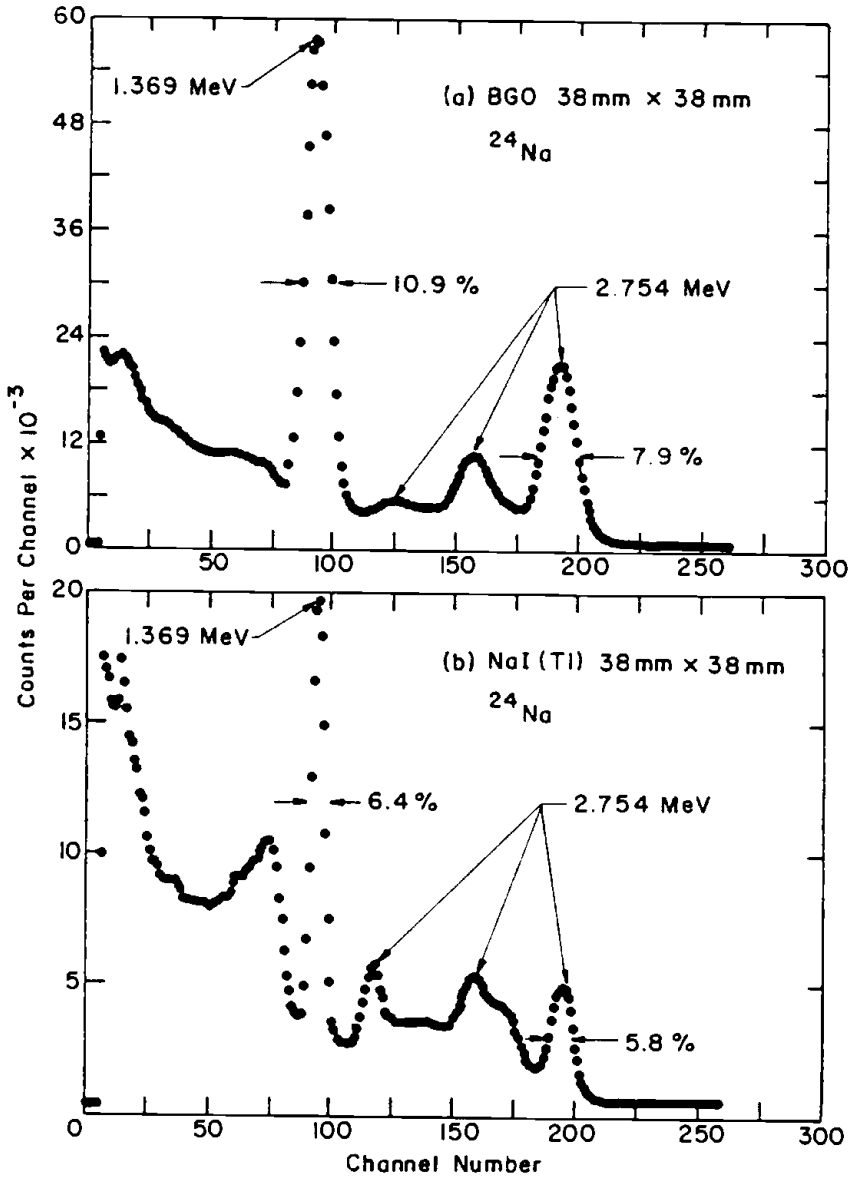


Figure 2. Pulse-height Spectra Obtained Using a 38 mm x 38 mm Cylindrical NaI(Tl) and BGO Detectors for Gamma Radiation from a  $^{24}\text{Na}$  Source.

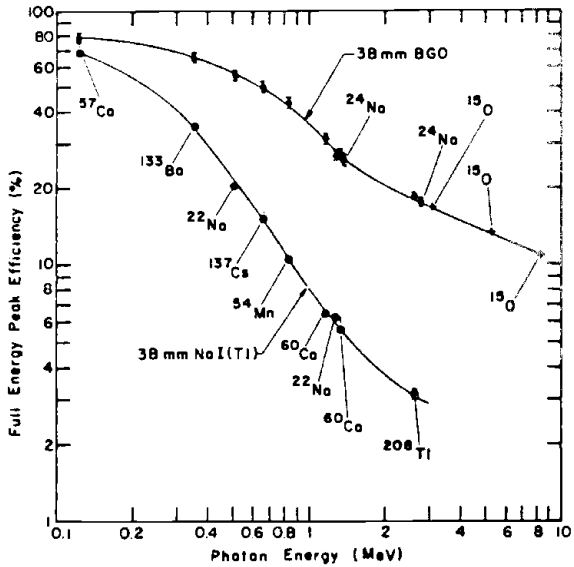


Figure 3. Experimental Full-energy Peak Efficiency as a Function of Gamma-ray Energy for 38 mm BGO and NaI(Tl) Detectors. (Data from Evans<sup>5</sup>)

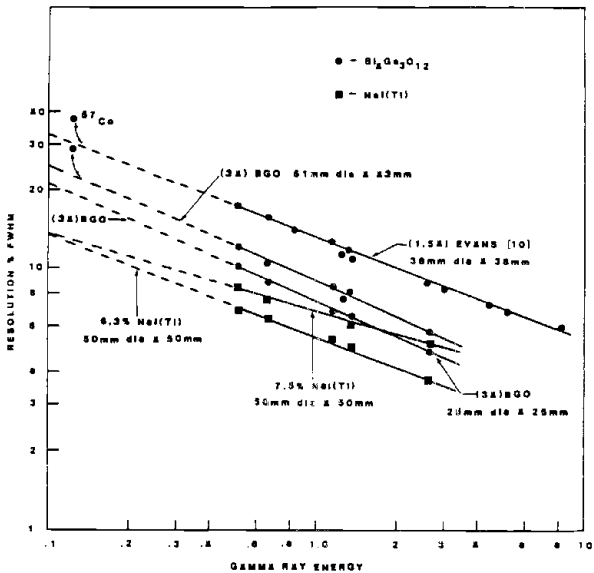


Figure 4. Measured Energy Resolution as a Function of Gamma-ray Energy for BGO and NaI(Tl) Detectors. (Data from Farukhi<sup>6</sup>).

results were obtained using a selected 2" diameter Hamamatsu R1306 phototube. These tubes achieve an energy resolution of 6.3 - 6.6% FWHM with 2" NaI(Tl) crystals. A summary of energy resolution obtained in various studies is presented in Figure 4, compared with NaI(Tl) results on 2" crystals.

### Optical Properties

Important optical properties of solid scintillators include: light output, phosphor lifetime, absorption of fluorescent radiation in the crystal and index of refraction. BGO, when compared with NaI(Tl) has a number of optical deficiencies. The light output, or quantum conversion efficiency, measured under the same conditions as for a conventional NaI detector is reported to be 15% relative to NaI. The fluorescent radiation is peaked at 480 nm, which is a reasonable match to the best electron multiplier tubes. The high index of refraction (2.15) presents serious problems in removing the light from the scintillator volume. The high index of refraction decreases the critical angle for internal reflections. At the present time, much work remains to optimize optical system design to achieve maximum light transfer from phosphor to optical sensor. Factors in design which influence light output include crystal geometry, optical coupling to the photocathode of the electron multiplier and reflective surface treatment on the scintillation crystal.

Figure 5 is a plot of fluorescent radiation as a function of temperature, as reported by Weber and Monchamp<sup>3</sup>. Both parameters are observed to be highly temperature dependent, which presents problems in many experimental applications. Recent lifetime studies of highly refined material indicate that BGO exhibits an initial decay component of 60 ns., representing 10% of the total light output. This results in an initial fast rise time component which compares favourably with NaI. These results, reported by Moszynski<sup>7</sup>, indicate that this characteristic, coupled with the absence of "afterglow" will promote triggering off the first few photoelectrons. This should facilitate coincidence requirements in the application of BGO for Compton-rejection detector designs and positron scanners.

## Applications of Bismuth Germanate (BGO)

In addition to the use of BGO in x-ray and positron CT scanners, considerable interest is being shown in the use of this material for gamma-ray spectroscopy in medium and high-energy physics research. With the availability of this material in large volumes, the combination of improved full-energy peak efficiency and stopping power offers many advantages. In high-energy physics, fundamental particle research currently involves the use of large NaI(Tl) array counter systems for measuring particle energy and direction. Total energy absorption of multiple particles may involve a total energy deposition greater than 1 GeV. This implies large volumes of scintillator material. BGO offers a number of advantages in the design of such detectors, including higher energy absorption per unit length, resistance to radiation damage, and improved mechanical properties.

Perhaps the most impressive of the large detectors being developed are those proposed for use on the colliding-beam electron-positron machines such as SLC, CESR, and LEP. Typical is the BGO Crystal Ball detector proposed for use on the Linear Collider Facility at Stanford<sup>8</sup>. An extension of the NaI Ball Detector now in use, this counter would employ 9200 BGO segments, hexagonal in shape, 1.0 cm on a side and 20 cm in length. This requires a total of 6000 Kg of BGO in the array. The array is being considered to study the neutral weak vector boson ( $Z^0$ ) at energies approaching 100 GeV. A second large detector is being proposed for the LEP storage ring at CERN to study the  $Z^0$  particle and to investigate physics in the 100 GeV range. In this detector, a BGO calorimeter is being considered which would consist of 12000 individual BGO segments each 25 cm in length. The total amount of BGO which would be required is approximately 1500 liters in volume. The estimated cost of the BGO calorimeters for this device is in excess of \$6M. As will be discussed in a later section, the complexity and environmental constraints rule out the use of phototubes in these detector assemblies.

## Compton-rejection Spectrometers

In many areas of nuclear physics research there is a need for gamma-ray spectrometers which combine high energy resolution with a

response characterized by a large peak-to-total ratio. Enhanced full-energy peak fraction implies total energy absorption and large detector volumes. At the present state of technology, high-energy resolution is achieved with high purity germanium detectors which can be fabricated in sizes up to 3" in diameter. The requirement for enhanced full-energy peak response can be achieved by surrounding the Ge detector with a large volume high-density detector which can be arranged to detect the secondary Compton scattered photons from events occurring in the central detector. A coincidence requirement is then used to reject such events in the spectrum of the central detector. Such Compton-rejection detectors have been fabricated until recently using NaI(Tl) for the mantle counter. Bismuth Germanate, with an absorption coefficient twice that of NaI, permits a much more compact

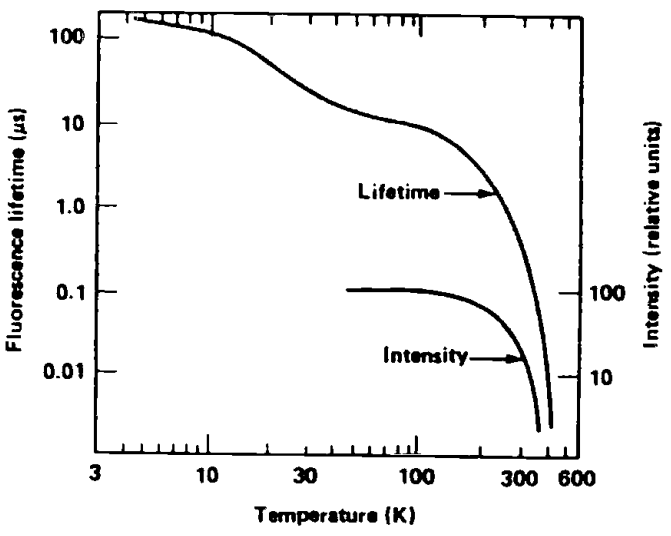


Figure 5. Temperature Dependence of Observed Lifetime and Intensity of Luminescence in BGO.

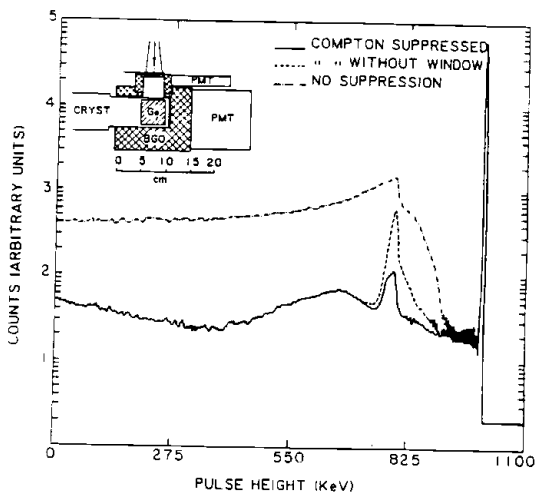


Figure 6. Calculated response of  $100 \text{ cm}^3$  Ge Detector to 1 MeV Gamma-ray with BGO Compton Rejection Mantle. Spectra are shown with and without Compton Rejection. Effect of adding thin detector to entrance window to eliminate  $180^\circ$  scattered photons which produce sharp peak at maximum Compton electron energy is also illustrated.

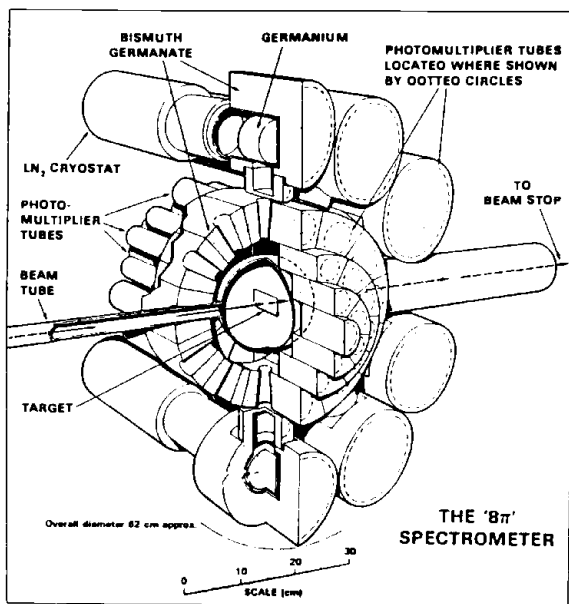


Figure 7.  $4\pi$  BGO Detector Array including Compton-suppression. Ge spectrometers proposed for heavy-ion Columb excitation studies on CRNL Super-conducting Cyclotron.

design to achieve the same result. A typical arrangement for such a counter is shown as an insert in Figure 6. In this configuration, a collimated beam of photons enters the central detector through an aperture at the top of the figure. To illustrate the improvement in detector response which can be achieved using this technique, the calculated response of a 100 cm<sup>3</sup> Ge detector to 1 MeV gamma rays is shown. Spectra are presented with and without the Compton-rejection requirement. Also shown is the result of adding a thin detector covering the entrance window to eliminate 180° scattered photons which would otherwise escape the detector. This addition eliminates the sharp peak in the Compton electron distribution.

As an example of the application of these detector systems in physics research, Figure 7 is a view of a 4π BGO detector array proposed for heavy-ion coulomb excitation studies on the CRNL super-conducting cyclotron<sup>9</sup>. In this system, the target being studied is surrounded by a segmented BGO array to determine multilicity. High-resolution spectroscopy of discrete lines is obtained with 10 Ge spectrometers, each surrounded with a Compton-suppression mantle counter of BGO.

Table II. Applications of solid scintillation detectors.

Application	Preferred phosphor	Important considerations
Physics Research		
High energy calorimeters	BGO	Density, Z, readout
Compton suppression	BGO	Density, Z, $\tau$
Neutron spectroscopy	LiI (Eu)	Li(n, $\alpha$ )
Applied Science		
X-ray CT	CdWO <sub>4</sub> , BGO	Spectral output, low afterglow
Positron CT	BGO, CsF	$\tau$ , A
Borehole scanning	BGO	Density, Z
Nuclear fuel scanning	BGO	Density, Z
Environmental assay	BGO	Density, Z

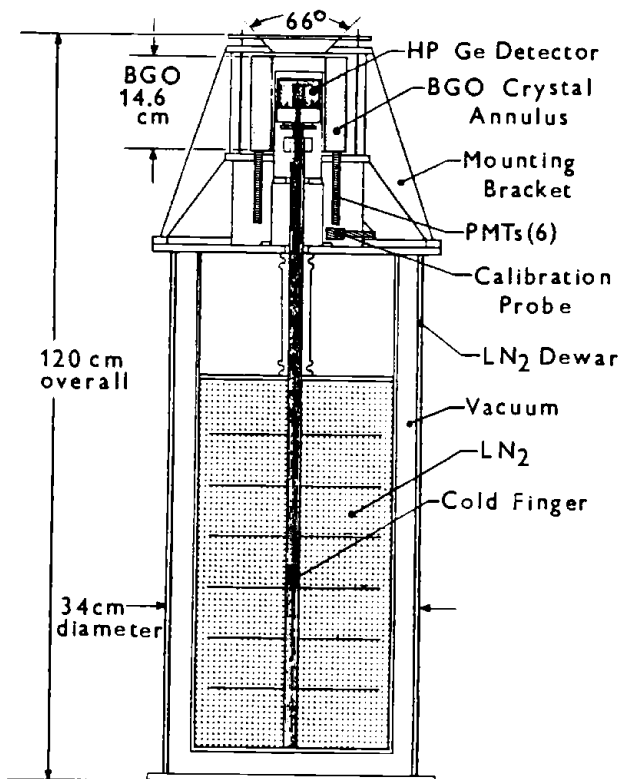


Figure 8. Experimental Compton-suppression Ge Spectrometer Developed for Space Shuttle Experiment. A BGO annulus Detector surrounds n-type Ge detector.

Figure 8 shows another example of a Compton-rejection spectrometer designed to be applied in a space shuttle experiment. In this arrangement, a cylindrical annulus of BGO surrounds an n-type Ge central detector. This system was developed to study solar radiation and induced background radioactivity in spacecraft.

To summarize recent trends in the application of solid scintillators, Table II lists areas of application, the phosphor being utilized, and considerations important to this choice of scintillator material.

## Recent Advances in Optical Sensors

Although the electron-multiplier phototube continues to be the major optical sensor employed in scintillation detector design, considerable progress has been made in the development of alternative optical devices. Advantages offered by the phototube include high signal gain, broad bandwidth (time response), and low noise. Typical quantum efficiencies of modern cathode surfaces are in the range of 30%. Disadvantages or problem areas, include physical size, cost, mechanical stability, sensitivity to magnetic fields, and gain instabilities related to a variety of experimental parameters including input signal rate. Current trends in high-energy physics experiments involve very large arrays of solid scintillators arranged in  $4\pi$  geometry for the measurement of energy and direction for charged particles and photons. This implies hundreds of detectors which must be operated in high magnetic fields. This requirement has led to the replacement of phototubes in these applications with silicon photodiodes which offer the advantage of physical and electrical stability, reduced size and cost, and insensitivity to operation in magnetic fields. Considerable effort is currently being made in this area, aimed at improving diode characteristics.

## Silicon Photodiodes

Solid-state devices currently being used for this application are P-I-N silicon photodiodes with surface areas up to  $3 \text{ cm}^2$ . Although these devices have unity gain, requiring electronic gain to amplify the signals to usable levels, they do exhibit high quantum efficiency when compared with typical phototube cathodes. Figure 9 compares quantum efficiency for silicon devices with alkali photocathode surfaces as a function of incident wavelength. Also shown is the spectrum of emitted light from BGO, indicating an excellent match in response. The normal experimental arrangement for a scintillation detector employing a photodiode as optical sensor is shown in Figure 10. To achieve high quantum efficiency with low noise, diodes have been fabricated with a very thin p layer (0.2-0.5 micron) with an overlay metallic grid deposited to reduce resistance. Typical noise from state-of-the-art diode-amplifier combinations is in the range

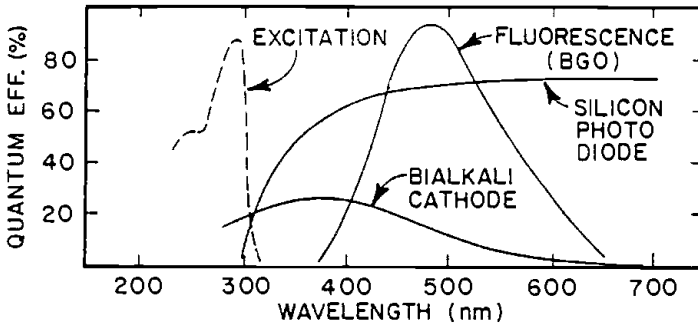


Figure 9. Quantum Efficiency of Silicon Photodiodes and Bi-alkali Cathode vs. Wavelength. For Comparison Emission Spectrum of BGO is shown.

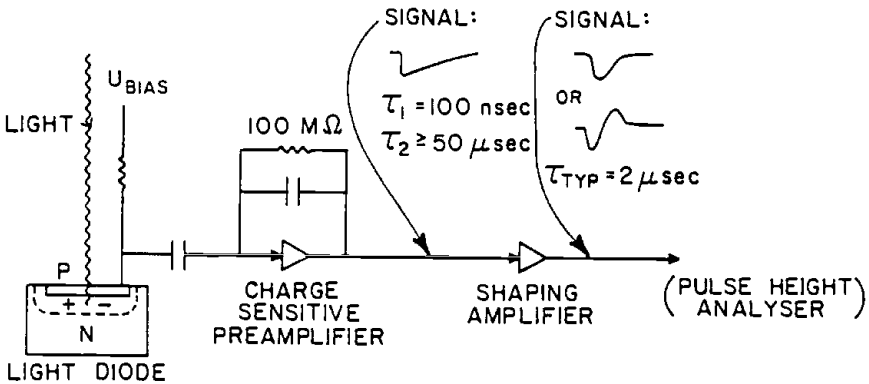


Figure 10. Experimental Arrangement for Operation of Photodiode Coupled to Scintillator.

from 0.5-1 MeV. This limits the use of diode detectors to high-energy spectroscopy in calorimeters where this noise limitation does not limit energy resolution. Improved reliability, with reduced space/cost makes this approach quite attractive.

### Silicon Avalanche Photodiodes

Low noise, large area ( $5 \text{ cm}^2$ ) silicon avalanche photodiodes have been reported by G. Entine, et al<sup>10</sup> which offer credible performance as an optical sensor for scintillation detectors. When coupled to a 1" x 1" NaI(Tl) crystal an energy resolution of 9.5% has been obtained for the 0.66 MeV line of  $^{137}\text{Cs}$ . The limitation on performance of avalanche diodes in the past has been attributed to poor optical efficiency as illustrated in Figure 11. As indicated in the figure, new devices have been developed with improved photoresponse in the wavelength region of light emitted by inorganic scintillators. This has been achieved by reducing the thickness of the entrance window. The ability to fabricate larger area devices than have been possible is attributed to improvement in the uniformity of silicon material. Improvement in noise level in the avalanche region of operation has been possible through the use of transmutation doped silicon material.

When operated to achieve optimum noise performance, a device gain in excess of 100 is possible, requiring minimum electronic gain for spectroscopy applications. The possibility exists for further improvement in noise performance by reducing the reflection at the surface of the device. Such devices are compact and rugged and offer advantages over phototubes for many field applications of photon spectroscopy.

### Mercuric Iodide Photodetectors

Photodetectors fabricated from  $\text{HgI}_2$  have been successfully applied as sensors for light from scintillators. J. Iwaczyk et al<sup>11</sup> have reported the use of such devices coupled to CsI(Tl) to achieve an energy resolution of 10% for annihilation radiation (0.51 MeV). Single crystal  $\text{HgI}_2$  has been used to fabricate photosensitive devices with a sensitive surface area up to  $100 \text{ mm}^2$ . A semitransparent electrode is formed by deposition of Pd in thicknesses below 100 Angstrom units. Operating at a bias of several hundred volts, these devices exhibit very low noise current. If the surface area can be increased these devices should be of interest in many scintillator applications.

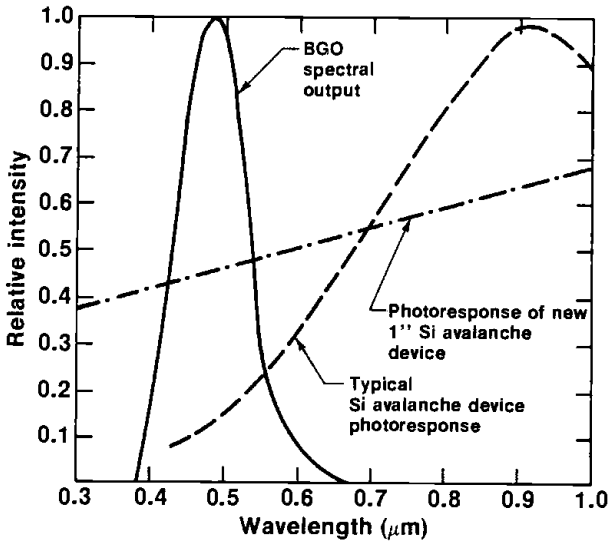


Figure 11. Comparison of BGO Spectral Output to Response of Silicon Avalanche Photodiodes.

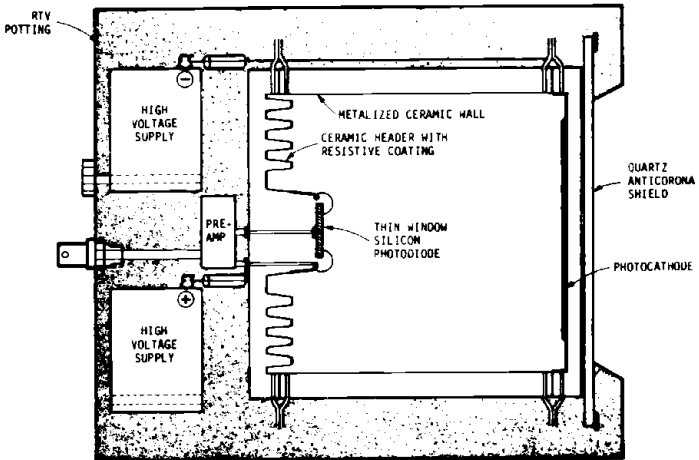


Figure 12. Design Concept for light amplifier for scintillation detectors. Photoelectrons from photocathode are Accelerated and Focused on a Silicon Diode.

## Silicon Photoelectron Converter

An interesting light amplifier concept reported by Orphan et al<sup>12</sup> could offer advantages for use with solid scintillators. This concept couples a conventional photocathode surface to a solid silicon photodiode as shown in Figure 12. Photoelectrons produced at the photocathode are accelerated through a 15 keV field and focused onto the surface of a reverse-biased P-I-N junction diode. Considering that 3.6 eV is required to produce a hole-electron pair in silicon, this results in a single-stage gain of 4000. Electronic gain, using a low-noise charge-sensitive preamplifier then produces the equivalent gain to an electron multiplier. Improved photoelectron statistics, gain stability, and mechanical properties are indicated. It has been reported that prototype units produced an energy resolution of 7.6% for the 0.66 MeV line of <sup>137</sup>Cs using NaI(Tl).

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