INSTRUMENTATION

The Low-Temperature Scintillation Properties of Bismuth Germanate and Its Application to High-Energy Gamma Radiation Imaging Devices

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Bismuth germanate is a scintillation material with very high z, and high density (7.13 g/cm^3). It is a rugged, nonhygroscopic, crystalline material with room-temperature scintillation properties described by previous investigators as having a light yield ~8% of that of NaI(Tl), emission peak at ~480 nm, decay constant of 0.3 μsec, and energy resolution ≃ 15% (FWHM) for Cs-137 gamma radiations. These properties make it an excellent candidate for applications involving the detection of high-energy gamma photons and positron annihilation radiation, particularly when good spatial resolution is desired. At room temperature, however, the application of this material is somewhat limited by low light output and poor energy resolution. This paper presents new data on the scintillation properties of bismuth germanate as a function of temperature from −196°C to + 30°C. Low-temperature use of the material is shown to greatly improve its light yield and energy resolution. The implications of this work to the design of imaging devices for high-energy radiation in health physics and nuclear medicine are discussed.

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For many years there have been attempts to produce better detection materials for high-energy gamma radiations. Ideally such a scintillation material should have a high density, a high attenuation coefficient for gamma radiation, a good energy resolution, a high light output at wavelengths compatible with low-noise, low-cost photomultiplier tubes and inexpensive light-guide materials, and have a reasonably short decay time with minor amounts of afterglow. The material should also be very resistant to thermal and mechanical shock, scintillate over a wide temperature range, and should not require special hermetic encapsulation (i.e., be nonhygroscopic), as well as other factors dependent upon particular applications.

No known scintillation material meets all of these criteria, but certain materials such as sodium iodide (thallium activated), satisfy enough of them to be useful in a wide range of applications.

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In applications where the detector's physical size is critical and high attenuation coefficients are desired (such as imaging systems for radionuclides emitting high-energy gamma radiation), sodium iodide's modest density (3.67 g/cm^3) and photon attenuation coefficient (0.0417 cm^2/g at 0.5 MeV), along with its requirement for a rather bulky and delicate hermetically sealed envelope, have been serious disadvantages. The importance of the attenuation coefficients can be seen in Fig. 1, which demonstrates spatial resolution degradation for NaI(Tl) under various conditions of incident gamma radiation energy (and associated changes in attenuation coefficients) and crystal thickness when used in scintillation cameras of the Anger type (*1*). In Fig. 1, $\bar{r}$ is defined as the perpendicular distance from a primary gamma photon to the center of intensity of the light produced.

One material that promises great improvement over NaI(Tl) in high-energy applications is bismuth germanate ($Bi_4Ge_3O_{12}$) (BGO), and the material has recently become available commercially. The scintillation properties of this material at room temperature have

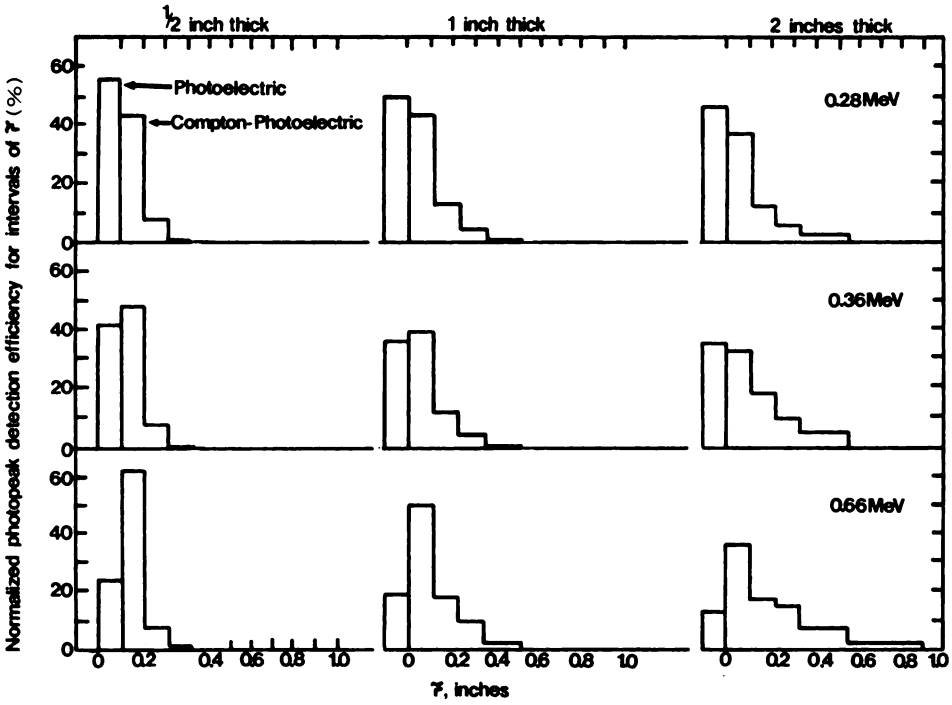

FIG. 1. Loss of position resolution due to multiple gamma ray photopeak interactions in 1/2-, 1-, and 2-in.-thick sodium iodide at 0.28, 0.36, and 0.66 MeV (*1*).

been reported by Weber and Monchemp (*2*) and Nestor and Huang (*3*) (see Table 1).

The advantages of bismuth germanate are most pronounced in its density (7.13 g/cm^3), its photon attenuation coefficient (0.0735 cm^2/g at 0.5 MeV), its shock resistance, and its nonhygroscopic properties. This material enables one to achieve high detection efficiencies for high-energy photons with a crystal volume much smaller than that of NaI(Tl) and without a hermetically sealed detector enclosure. However, the application of bismuth germanate is limited by its low light yield, its moderately longer decay time, its poor energy resolution, and its relatively high cost.

In an attempt to investigate methods for minimizing certain of these disadvantages of BGO, a study of its low-temperature scintillation properties was carried out. The usefulness of any scintillator is strongly dependent on the efficiency with which its scintillation light can be transmitted and detected; consequently, potentially compatible photomultiplier (PM) tubes were evaluated based on their manufacturers' published photocathode quantum efficiencies at room temperature, their noise at a fixed gain, and their response to cooling (Fig. 2) (*4*).

TABLE 1. COMPARISON OF SCINTILLATOR MATERIALS (*3*)

	NaI(Tl)	BGO
Density (g/cm^3)	3.67	7.13
Wavelength (nm) of maximum scintillation intensity	420	480
Refractive index at intensity maximum	1.85	2.1
Decay constant (μsec)	0.25	0.30
Resolution at Cs-137 photopeak (percent)	7	15
Cs-137 photopeak pulse height (relative)	100	8

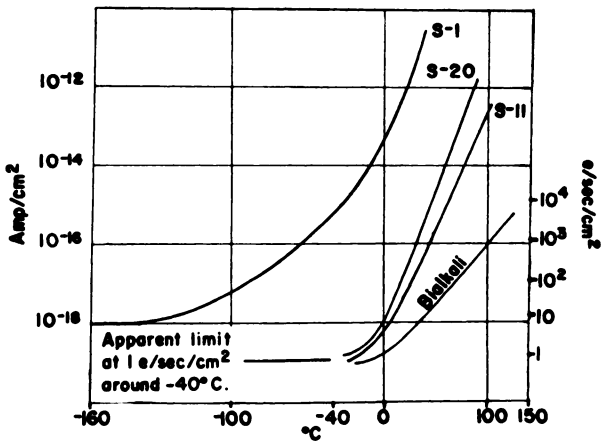

FIG. 2. Apparent temperature variation of cathode dark current for typical photomultiplier tubes.

On the basis of these factors, a PM tube was selected for use in these experiments, coupled to a BGO sample, and operated at the same temperature as the BGO.

MATERIALS AND METHODS

Measurements of relative spectral irradiance. These measurements were made, at a given sampling position and as a function of sample temperature, using BGO sample No. 1. The sample was coated on its back and edges with reflective paint* and mounted on the sample block of the refrigerated cold finger in a cryogenic sample chamber (see Figs. 3 and 4), in such a way that x-rays incident on the sample produced scintillation quanta that were received and analyzed by the spectroradiometer. The sample chamber is operated at a pressure of ~1 mm Hg to insulate the cold finger from the surrounding materials and to prevent condensation on the sample.

Spectral irradiance measurements were made between 300 and 700 nm in 10-nm increments using a 10-nm FWHM bandpass monochrometer setting. The spectroradiometer was calibrated at the time of its use by placing it in a light field of known spectral irradiance produced by a quartz-halogen tungsten-filament lamp standard. Calibration factors were determined for each wavelength setting used in subsequent measurements.

The x-ray source was operated at 90 kV (p), 10 mA, for a period of 2.5 sec for each measurement point. Reproducibility of the sample exposure appeared to be approximately ±3% based on five data points immediately before and after collection of a given set of data

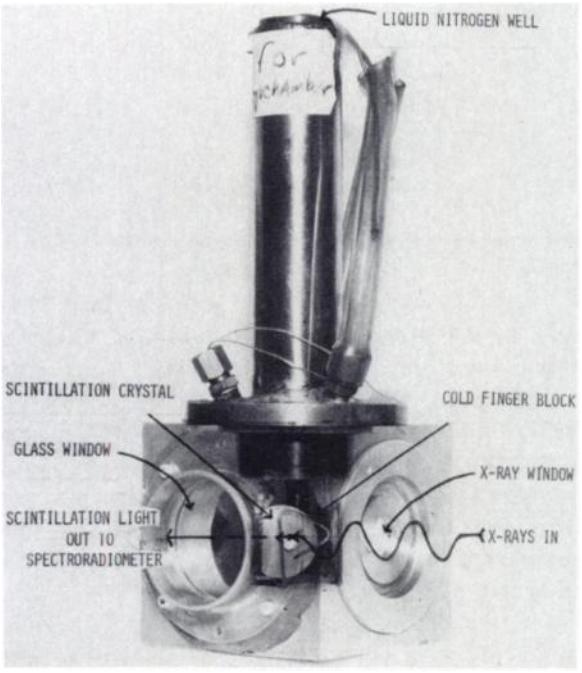

FIG. 3. Cryogenic sample chamber (radiographic view).

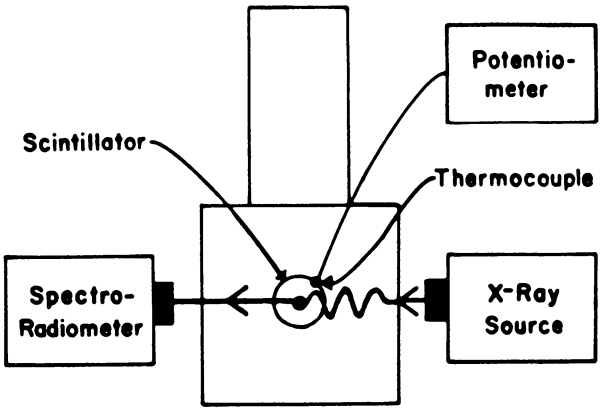

FIG. 4. Diagram of apparatus for measuring emission spectra of scintillators.

(data points from 300 to 700 nm for a given sample temperature).

Overall error for the measurement of relative spectral irradiance was estimated to be ±12%. This includes both systematic and random errors involved in calibration sources, wavelength settings, x-ray source instabilities, and sample temperature stability.

Measurements were made at temperatures of +30, 0, −50, −100, −150, and −196°C (liquid-nitrogen temperature). These temperatures were determined by use of a thermocouple mounted on the sample-holder block and read with a thermocouple potentiometer.

Test gamma-energy spectra and decay-time measurements. Examples of gamma energy spectra and decay-time measurements were produced by coupling BGO sample No. 2 to a PM tube† using RTV-615 silicone-rubber optical coupling medium. Gamma energy spectra measurements were made at +30 and −78.6°C (dry-ice temperature) for Am-241 (0.060-MeV) and Cs-137 (0.662-MeV) radiation sources.

Data were gathered using a multichannel analyzer. Decay-time measurements were made at +30 and −78.6°C by routing the PM tube's current pulses into a 500-ohm load and using an oscilloscope to observe pulse-shape characteristics. A Polaroid camera recorded the data. Decay constants (T) were determined by observing the time required for decay of the pulse to 1/e of its maximum value.

A complete listing of materials used is included in Appendix 1.

RESULTS

Table 2 shows the relative spectral irradiance from scintillation light in the crystal, measured at the input receptor of the spectroradiometer. Values for the relative irradiance integrated from 350 to 580 nm as a function of temperature are given in Fig. 5. Plots of relative spectral irradiance against wavelength for various temperatures are given in Fig. 6. Figure 7 shows the

TABLE 2. EMISSION SPECTRA OF BGO FOR 90-kV(p) X-RAYS [RELATIVE SPECTRAL IRRADIANCE (I_λ)]

Nanometers	Temperature (°C)					
	+30	0	−50	−100	−150	−196
350	0.020	—	0.041	0.041	0.061	0.061
360	0.030	—	0.045	0.045	0.061	0.075
370	0.046	0.069	0.053	0.046	0.038	0.038
380	0.145	0.237	0.215	0.184	0.130	0.107
390	0.430	0.227	0.798	0.704	0.493	0.383
400	1.69	2.68	2.69	2.94	2.33	1.84
410	6.37	8.75	10.4	10.9	9.23	7.94
420	11.4	14.88	17.8	19.3	17.3	15.2
430	15.5	20.6	25.1	28.1	26.8	24.8
440	19.0	24.8	32.0	36.8	37.2	35.9
450	22.8	30.1	39.1	46.3	48.8	48.2
460	25.2	33.5	44.5	53.6	57.0	57.2
470	27.6	34.7	49.6	58.6	64.7	66.1
480	28.8	35.7	53.4	68.3	72.7	76.0
490	28.8	36.1	54.2	67.3	77.8	81.8
500	28.0	35.9	52.9	68.9	81.0	87.6
510	26.5	33.1	49.5	64.5	79.3	87.6
520	24.1	30.2	45.5	58.4	73.5	83.3
530	23.5	29.4	43.4	57.0	71.9	82.7
540	23.1	28.9	43.6	56.9	72.0	83.7
550	21.0	25.9	38.4	51.3	64.4	73.9
560	17.6	22.3	32.9	43.6	54.6	63.6
570	18.8	20.9	31.1	40.7	52.2	59.1
580	7.22	9.02	13.1	17.1	22.0	25.1
590	3.19	3.94	6.05	8.16	10.1	11.4
600	1.97	2.31	3.60	4.83	5.91	6.63
610	1.80	2.30	3.44	4.50	5.65	6.63
620	1.72	1.80	2.75	3.86	4.64	5.23
630	1.19	1.40	2.00	2.49	3.39	3.99
640	0.640	0.640	1.18	1.40	1.93	2.26
650	0.450	0.446	0.550	0.890	1.00	1.12
660	0.225	0.225	0.338	0.338	0.564	0.564
670	0.118	0.118	0.236	0.236	0.354	0.473
680	0.164	—	0.164	0.164	0.328	0.328
$\sum_{350}^{680} I_\lambda =$	386	490	699	875	1023	1099

relative response of several PM tube photocathodes convolved with the emission spectrum of BGO. Gamma energy spectra for Cs-137 and Am-241 using the BGO and photo multiplier tube at +30 and −78°C are shown in Fig. 8. Decay-time data for the scintillation light are shown in Fig. 9.

DISCUSSION

It can be seen from Table 2 and Figs. 5 and 6 that cooling of the BGO scintillator alone results in an increase in light output of the scintillator by ~1.7% per deg. C from +30 to −100°C. It is also apparent that the emission spectrum of the scintillation light remains relatively constant over that temperature range, with four emission peaks being resolved: a main one at 490 nm and lesser peaks at 530, 570, and 610 nm. (This is in contrast to data reported by Nestor and Huang, which show a broad single peak at 480 nm (*3*).) Figure 10 shows the emission spectrum of BGO at −196°C in terms of relative photon flux compared with wavelength.

Measurements of the gamma spectra of Cs-137 and Am-241, using BGO coupled to a photomultiplier tube† at +30 and −78.6°C (Fig. 8 and Table 3), resulted in a

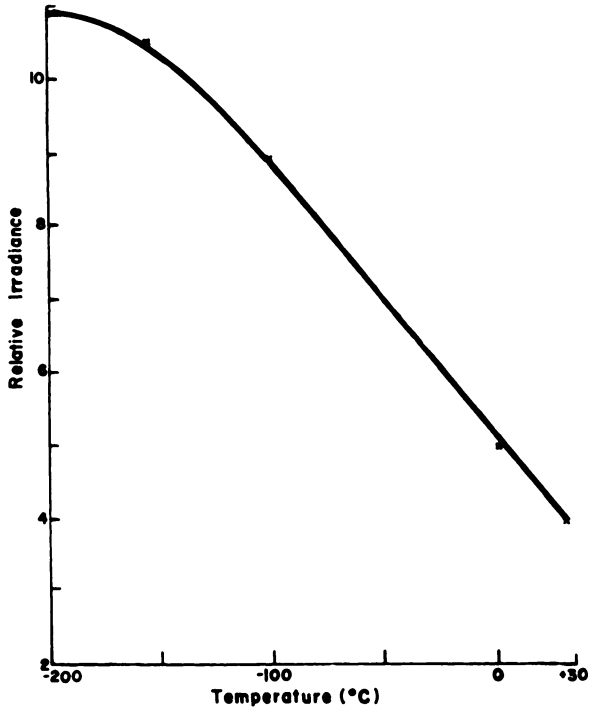

FIG. 5. BGO scintillation yield as function of temperature.

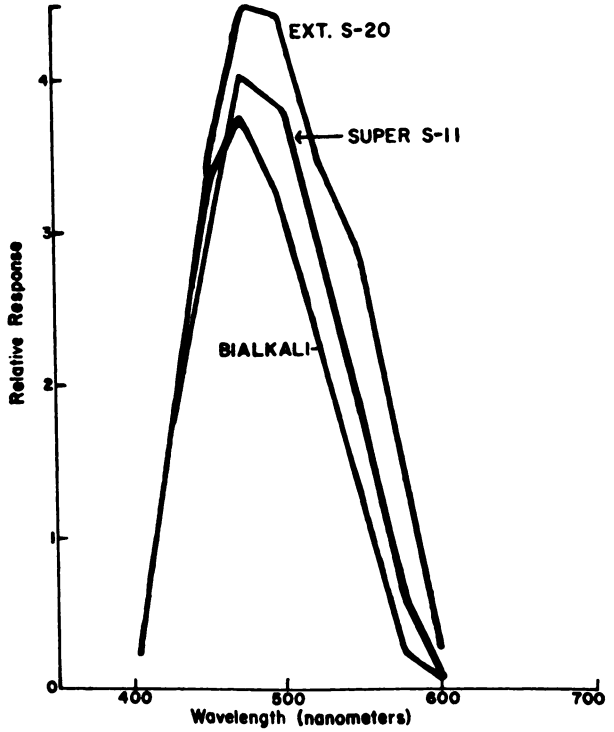

FIG. 7. BGO emission spectrum, corrected for photomultiplier response.

room-temperature energy resolution of 23% for BGO with Cs-137 (0.662 MeV photons), as compared with the 15% value of Nestor and Huang. This difference in resolution is felt to be due to the methods of mounting the crystal, light reflectors used, and properties of the individual crystal samples. (An earlier mounting of the same crystal on a different PM tube resulted in a resolution of ~17% at room temperature). However, because the study was designed to examine relative performance of the scintillator at various temperatures, this difference was not felt to be important. Cooling the BGO and PM tube to −78.6°C resulted in an increase in pulse height by a factor of ~2.9, with a resolution of 14.1% (FWHM) at 0.662 MeV. Calculation shows that an effective change by a factor of ~1.12 in the quantum efficiency of the PM photocathode would account for both the increased pulse height and improvement in energy resolution over that expected due to the increased light output of the scintillator alone. It also appears that an optimized choice of PM type and operating temperature could increase this low-temperature gain in quantum efficiency even more (6).

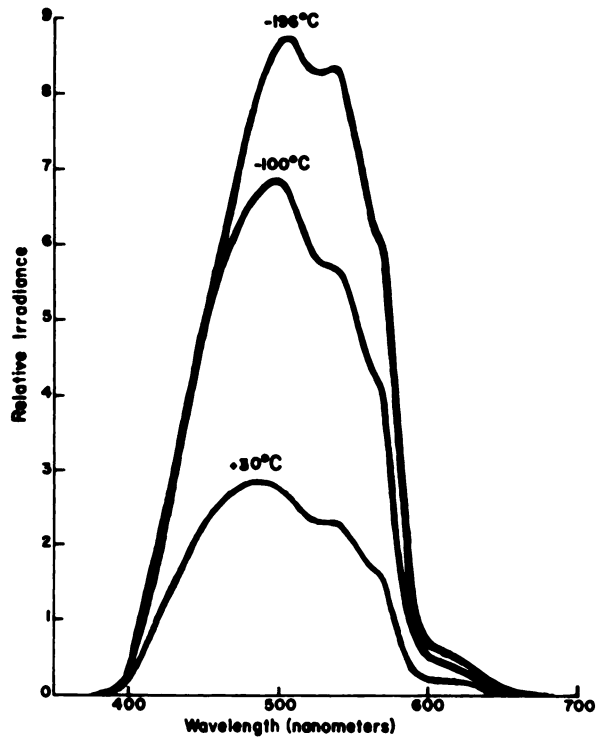

FIG. 6. Emission spectrum of BGO as function of temperature.

One may question why the resolution did not strictly follow the pulse height for a decrease in temperature from +30 to −78.6°C (a resolution improvement factor of 1.63 measured against 1.69 calculated). Probably this was due largely to a temperature gradient across the BGO crystal and/or changes in the relative sensitivity distribution of the PM photocathode.

It is also apparent in Fig. 8 that the cooling of the detector assembly has allowed the measurement of 60-keV radiations of Am-241, whereas the PM noise at room temperature caused this peak to be obscured.

Measurements of time constant, T, for the decay of

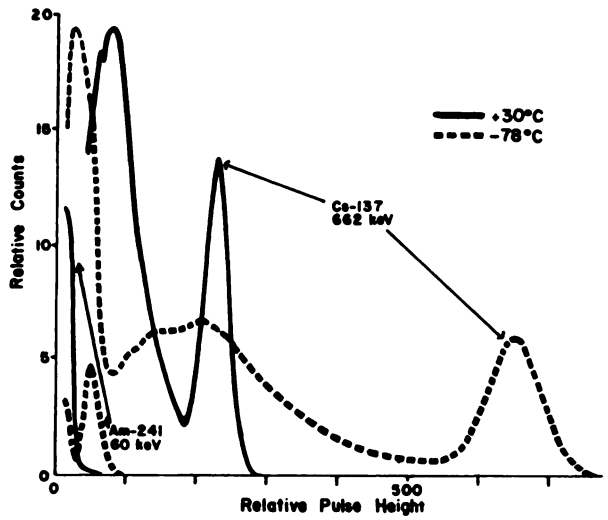

FIG. 8. Pulse height spectra from Cs-137 and Am-241 using BGO and photomultiplier tube at +30 and −78°C.

the scintillations at +30 and −78.6°C are shown in Fig. 9 to be approximately 450 and 2000 nsec, respectively. Use of the ratio of T to relative light yield, as an index of the possible root-mean-square resolving time for the scintillator, results in a factor of 2.1 increase in resolving times between +30 and −78.6°C. Use of this ratio is approximated by calculations developed by Post and Schiff (7) when the average number of photoelectrons (R) is substituted by either the scintillator light output or the PM tube's output pulse height. Since use at −78.6°C would most likely be in conjunction with a cooled PM tube, that ratio would probably become 1.6 or better. The significance of this is implied in the comments of Cho and Farukhi (5) who found the time "jitter" related to the resolving time to be 6.8 nsec FWHM (and 14.5 nsec FWTM) for BGO at room temperature. They felt that "given present-day electronic components and circuitry, coincidence resolving times of 10-20 nanoseconds seem well within acceptable limits" for their proposed ring types of positron cameras. Consequently, it would seem that if one assumed an increase of time

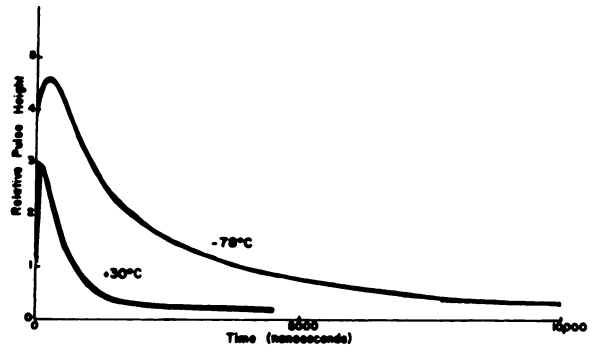

FIG. 9. Decay-time measurements of BGO as function of temperature.

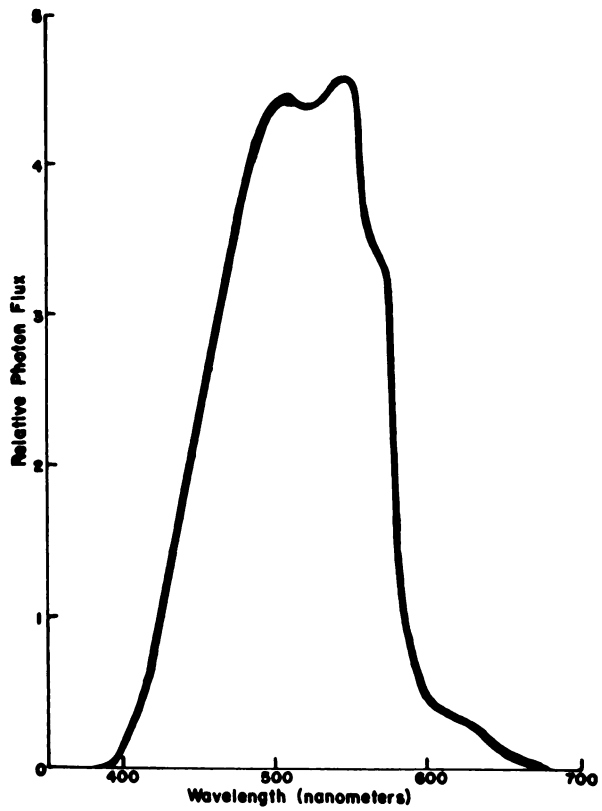

FIG. 10. Relative photon flux emission spectrum of BGO (−196°C).

"jitter" from 6.8 nsec FWHM to 10.9 nsec, it would still be "within acceptable limits" in that type of application.

While the cooling of large arrays of closely packed detectors may pose special engineering problems, use of recirculated chilled air in the −50 to −80°C range seems to hold promise both technically and economically for cooling such arrays.

CONCLUSIONS

It appears that the cooling of BGO, and under some conditions its associated light detectors, provides a means for greatly increasing its light output and detection efficiency, and the consequent energy and spatial resolutions, when applicable. This could be an important factor in the future design of imaging devices such as positron cameras or conventional gamma cameras using either a single- or multidimensional Anger type of logic system. It could also be valuable in detection systems designed for mixed high- and low-energy photons where improved energy resolution and signal-to-noise ratio are desired, along with BGO's other favorable qualities.

APPENDIX I: MATERIALS

The following is a list of materials used in the evaluation:

TABLE 3. MEASURED PROPERTIES OF BGO AS A FUNCTION OF TEMPERATURE

	Temperature (°C)						
	+30	0	−50	−78	−100	−150	−196
Relative light emission [% emission of NaI(Tl)]	8	10	14	17	18	21	23
$\frac{1}{\sqrt{\text{Relative light emission}}}$	0.35	0.32	0.27	0.24	0.23	0.22	0.21
Relative gamma-radiation pulse height (BGO + PM Tube)	76			218			
Decay time to 1/e (T, nsec)	450			2000			
T/relative light yield	56			117			
T/relative pulse height	5.92			9.17			
FWHM resolution at 662 keV	23%			14.1%			

1. Sample BGO crystals, Harshaw Chemical Co.
   a. Sample No. 1, disk 3 cm diameter × 3 mm thick with slice removed from edge
   b. Sample no. 2, 4 mm × 5 mm × 2 cm
2. Products for Research Inc., Model TE-200 refrigerated PM tube housing with EMI Model 9659B PM tube.
3. Cryogenic sample chamber
4. Honeywell Model 2732 Potentiometer
5. Technique Associates Model 11B1 reference junction (Cu/C)
6. Spectroradiometer consisting of:
   a. Oriel Model 724 Grating Monochrometer with 280-nm blaze grating
   b. Detector system using EMI Model 9781R PM tube with electrometer, power supply, and Data Precision Model 245 digital voltmeter readout
7. Tektronix Inc., Model 7844 oscilloscope with Model 7A11 amplifier
8. Nuclear Data Series 2200 multichannel analyzer
9. Quartzline quartz-halogen tungsten filament lamp, Spectral Irradiance Standard Q1 (calibration traceable to the National Bureau of Standards)
10. 3M Company #202-A10 White Velvet paint
11. Weber Dental Mfg. Co., Ultron 90 dental x-ray machine
12. Victoreen Inst. Co., Model 555 dose integrator
13. General Electric Co., RTV-615 silicone rubber

FOOTNOTES

* 3 M Company.
† EMI Model 9659B.

ACKNOWLEDGMENTS

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NOTE

The mention of any commercial products in this article does not constitute the endorsement of that product by the U.S. Government Food and Drug Administration.

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