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CRYSTAL GROWTH AND EVALUATION OF
MERCURIC IODIDE NUCLEAR DETECTORS*

(Reporting period November 15, 1975 to June 30, 1976)

by
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PREFACE

A complementary program to the ERDA/EG&G (Santa Barbara Operations) investigations in mercuric iodide crystal growth and evaluation of mercuric iodide nuclear radiation detectors is being performed by Professor M. Schieber and his students in the Department of Materials Science at Hebrew University of Jerusalem, Israel. Partial support for this program is provided by ERDA.

The crystal growth program at Hebrew University emphasizes horizontal growth techniques, and comparative data with EG&G's vertical growth investigations is developed. Crystal characterization includes gamma spectra evaluation, x-ray luminescence studies, thermally stimulated conductivity measurements, and the D.C. response to x-ray fluences. The results of the Hebrew University studies are reported here.

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1. INTRODUCTION

A research effort on mercuric iodide (HgI_2) was initiated in November, 1974 at the Hebrew University of Jerusalem to complement the crystal growth of HgI_2 now being studied as part of the SANDS Program at EG&G, Inc., at Santa Barbara, California. The program was supposed to carry on some crystal growth experiments in a horizontal furnace which, although was considered promising, had to be abandoned at EG&G, where the experiments concentrated on crystal growth in a vertical furnace. In addition, it was expected that certain crystal quality characterization and evaluation methods – interesting and not available at EG&G – would also be studied. This report describes the various experiments performed in Jerusalem.

In terms of crystal purification, because the Jerusalem group has used three times more sublimation cycles, resulting in better nuclear spectroscopic performance, it had induced the EG&G group to increase its number of purification cycles, resulting in better quality crystals. In terms of crystal growth, the horizontal furnace method was perfected and has allowed the growth of large (~50 g) good quality crystals. At the request of the ERDA-NVO patent office, a patent application has been filed to describe the HgI_2 crystal growth in a horizontal furnace.

The following crystal evaluation studies have also been performed: 1) scanning electron microscopy, 2) x-ray luminescence, 3) thermal stimulated conductivity (TSC) studies, 4) measuring the dc fluence x-ray irradiation, and 5) gamma nuclear spectral response of the grown crystals.

The various results have been communicated previously to Mr. R. Lynn at EG&G's Santa Barbara Operations through informal memos, and during a visit in March, 1976 of Mr. I. Beinglass, a Ph.D student of Prof. Schieber at Hebrew University. The cooperative research effort between EG&G and Schieber on HgI_2 has proved effective and promises to lead to further advances in the technology of HgI_2 nuclear detectors.

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2. PURIFICATION AND CRYSTAL GROWTH PROCEDURE

The present purification procedure uses the repeated sublimation method of commercially produced HgI_2 (Allied Chemicals). The equivalent EG&G purification notation 120X is used, of which 60X is in an open and 60X is in a closed ampoule (see Report No. EGG 1183-2333, SANDS R&D Program, FY75 Progress Report). Crystals are grown in a horizontal furnace originally developed at EG&G (see patent application by M. Schieber and W.F. Schnepple),¹ but which was discontinued at EG&G which presently uses only vertical furnaces.

To combine the advantages of the vertical furnace, which produces large but not so homogeneous crystals, with those of the horizontal furnace (which in its original EG&G version produced some small but "better" crystals), it was decided to attempt to build an improved horizontal furnace. It should contain some of the elements in the vertical Scholz-type furnace, which were missing in the horizontal unit. The new Jerusalem furnace, shown schematically in Fig. 1, contains a provision for a radial gradient, but some additional novel features have improved its design. (Note: for the readers convenience, all 36 figures in this report are grouped after Section 3.) To homogenize the local thermal asymmetries the ampoule is rotated at about 1 rpm. The horizontal furnace also uses larger ratios of length to inside diameter of the ampoule; thus, the ampoule is 30 cm long and 6 cm in diameter (12 inches by 2-1/4 inches), whereas the ampoule in the vertical furnace is about 25 cm long and 10 cm in diameter. Growing the crystal at a flat end of the ampoule in the horizontal furnace, the HgI_2 source-crystal distance, X , of 12 inches was found to be undesirable; therefore we decided to shorten X by blowing an axial air stream in different phases along the axial length of the ampoule (in effect, bringing the source material from the far end of the tube closer to the growing crystal). Cooling the source by air blowing shortens the cycling periods of growth and etching; blowing air at an axial distance of about 2 inches from the growing crystal produces a much sharper gradient, dT/dX , which stabilizes the growing interface and forces growth of a more perfect crystal. The sharper dT/dX is thus obtained by shortening dX rather than by increasing dT , which in turn may have favored undesired spurious nucleation on the growing crystal and caused

large overshoots in the electronic temperature controller. To keep the crystal vapor interface at a constant dT/dX , the axial blowing air tube is slowly moved inside the furnace in parallel with the movement of the growing crystal surface, as shown in Fig. 1. It should be noted that the radial gradient is being produced by a radial air blower. In order to flatten the temperature gradient and allow the growth of larger diameter crystals, the radial air blower is pulled away from the flat end of the tube while the air flow is increased from 350 to 450 cc/min.

In terms of the temperature oscillation method (TOM) the crystals are grown by both the periodic oscillation of the source temperature, POST, and the periodic oscillation of the crystal temperature, POCT method.² The nucleation of only one single crystallite is being done using the POCT method, but use of the POST variation only has also been successful.

An example of a growth experiment is as follows. An ampoule is cleaned with sulphochromic acid, washed with detergent, and rinsed in high purity methanol. At the end of the each rinse, between the successive cleaning operations, the ampoule is washed with deionized water and degassed for two hours at 150° at a vacuum of 10^{-6} torr. Chemically pure HgI_2 (produced by Allied Chemicals, Morristown, N.J.) is further purified by repeated sublimation in a continuously evacuated ampoule. The material is gradually transferred from one end to the other two times and then repeated twice more in a pinched-off closed ampoule (not continuously pumped) evacuated to 10^{-6} torr. (This corresponds to the "120x" notation used at EG&G, i.e., "60x" open and "60x" closed.) The purified material is introduced mainly in the growth ampoule. The furnace is maintained so that the source zone is at 119°C and the crystal zone at 122°C. The temperature oscillation is obtained by switching the air flow of the axial air blower between 1700 to 4500 cc/min for growth and re-evaporation, respectively. The periods of growth and reevaporation (etching) are 4 1/2 and 1/2 minutes, respectively, and the temperature differentials, T , for growth and reevaporation are 1°C and 3°C, respectively.

At the beginning, a single seed is caught by flowing 480 cc/min of air on the flat end of the tube, and when one single seed (which should be as small as possible) crystallizes the flow is decreased to 320 cc/min. During growth of the crystal the air flow is increased gradually, and the distance between the end of the air tube and ampoule is increased about 1 to 2 mm/day

so that the crystal interface can advance in all directions, i.e., radial and axial according to the crystallograph structure of the crystal and the respective temperature gradient. During crystal growth the axial air tube blower is also moved by about 1 to 2 mm/day, so that the distance between the crystal interface and the source material is kept constant, and therefore the temperature gradient at which the crystal grows is kept constant. The growth procedure is finished when all the source material is gone. The radial flow of air at the end is dependent on the amount of material that is charged in the ampoule. For example, in growing a 40-g crystal the flow at the flat end substrate was 480 cc/min. A typical crystal weighing 50 g grown in the horizontal furnace is shown in Fig. 2.

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3. EVALUATION OF HgI_2 CRYSTALS

The HgI_2 crystals have been studied by various methods detailed below:
1) optical and electron microscopy, 2) x-ray luminescence, 3) TSC studies,
4) x-ray irradiation, and 5) nuclear detector studies.

3.1 OPTICAL AND ELECTRON MICROSCOPIC STUDIES

Optical microscopy is the best method to study the growth rings or etch pit density in the HgI_2 crystals. Figure 3 shows the distribution of growth rings in one of our earlier crystals; the same rings at a larger magnification are shown in Fig. 4. These growth rings are revealed by etching HgI_2 with 0.2% bromine solution in methanol. The growth is the source of inhomogeneities of the crystal, where the distribution of impurities and etch pit density is different than in the bulk. A major effort is being expended to overcome the occurrence of these growth rings.

Figure 5 shows the distribution of etch pits on two top faces of an "as grown" crystal after etching in a 0.2% bromine solution in ethanol. The etch pit density varies from 10^4 cm^{-2} for the (110) to values of about 10^6 cm^{-2} in the (001); the density tends to decrease towards the edge. Occasionally, one can find faces which have even fewer etch pits, of the order of 10^3 cm^{-2} . At the site of the growth rings the etch pit density may reach 10^9 cm^{-2} . Scanning electron microscopic (SEM) studies have produced thermal etching on the top surface of the crystals. To avoid the thermal etching the crystals were attached to a liquid air-cooled stage. Figure 6 shows a typical (001) face of a HgI_2 crystal showing (probably) a two-dimensional island growth. Figure 7 shows an SEM picture of a cleaved crystal showing how uneven the "as-cleaved" surface is. To prepare a "good" detector surface the crystal has to be polished to remove the incomplete cleaved areas.

3.2 X-RAY LUMINESCENCE STUDIES

The x-ray luminescence studies were performed at the Raccah Institute of Physics of the Hebrew University under the direction of Prof. A. Halperin, with the aid of a graduate student, E. Pinsky. The results are still preliminary, and more results will be given in the FY77 report.

In the present work we investigated the luminescence emitted by HgI_2 , especially under x-ray excitation. Crystals of the same batch were examined both for their characteristics as detectors and their x-ray luminescence, with the aim of finding correlations between the two phenomena. It is believed this information should help in the production of improved HgI_2 detectors. Although the photoluminescence of HgI_2 has been described by earlier investigators³ very little, if anything, has been reported on the emission of these crystals under x-ray excitation. The crystals used in the present work were grown by the TOM in a horizontal furnace.² Measurements were carried out over a wide range of temperatures, from below 10°K to about 350°K. In a typical emission spectrum obtained at 77°K (Fig. 8) three main bands appear: at about 5400Å (2.30 eV), 5650Å (2.192 eV), and at 6200Å (2.00 eV). The full widths at half maximum (FWHM) at 77°K were about 0.016, 0.05, and 0.17 eV, respectively, for the three bands. The band peaks are fairly close to those reported by Bube³ for P photoluminescence, except for their relative intensities. These intensities were found to depend not only on the method of excitation but also on the thermal history of the crystals.

The bands in Fig. 8 show some structure; thus, we were able to resolve at least two more bands: at about 5450Å (2.275 eV) and 6920Å (1.79 eV). At temperatures below 77°K the structure is much more complicated. The above bands differ from each other in behavior; thus, the different bands have different thermal ionization energies. Figure 9 shows on a log-intensity versus reciprocal temperature plot the behavior of the 5400Å band. A straight line is obtained for the 100-200°K temperature range, which gives a thermal ionization of about 40 MeV. Above 200°K, quenching seems to set in, which causes a deviation from the straight line.

The thermal activation energy for the 5650Å band was found to be 65 MeV and that for the 6200Å band was about 120 MeV. The shift with temperature of the emission band was also measured. The shift for the 5400Å band (Fig. 10) was -6.8×10^{-4} eV/deg. The shift of the 5650Å band was much smaller (about -2×10^{-4} eV/deg), while the 6200Å band showed a positive shift (to higher energies with increasing temperature) of about 4×10^{-4} eV/deg. It should be noted that the shift of the 5400Å band is equal to that in the absorption edge, which was found to be -7×10^{-4} eV/deg.³

The dependence of the intensities of the various emission bands on the x-ray beam current at its peak voltage, as well as other characteristics of the emission and its relation to the photoluminescence, were also investigated and will be discussed in the FY77 report.

3.3 THERMALLY STIMULATED CONDUCTIVITY (TSC) STUDIES

A study of imperfections of HgI_2 by the TSC method⁴⁻⁶ was performed at the Raccah Institute of Physics under the guidance of Prof. Y. Yacoby and a post-doctoral research fellow, Dr. U. Gelbert. These results are also preliminary, and more results will be reported in FY77.

In this paper we describe experiments made on crystals grown at Hebrew University from the vapor phase in a horizontal furnace by both temperature oscillating methods.² It was not possible until now to take advantage of the promising detector properties of HgI_2 because of its poor collection, non-uniformity, and polarization effects. The purpose of this work was to find the impurity energy levels and their capture cross-section, which may be useful in finding a way to improve detector performance.

The experiments reported here were performed on an ingot of HgI_2 : No. HR-I-11. Two touching samples, 200-300 μm thick, were cleaved. A nuclear detector^{7,8} was prepared from one, and the TSC measurements were performed on the other. A typical gamma-ray spectrum of the first sample is shown in Fig. 11. The detector was biased under 800 V and irradiated with an ^{241}Am source. The ordinate represents the number of counts, and the abscissa delineates the channel energy. The dominant peak corresponds to the 59.6 keV line of ^{241}Am . The full width half maximum (FWHM) of this peak is 2.35 keV, in comparison to the 1.8 keV of the pulser (not shown in the figure). No severe polarization degradation effects were observed on this sample.

A typical TSC spectrum obtained from the second sample is given in Fig. 12, where the TSC current versus temperature is plotted (on a semilog scale). The spectrum, at the temperature range 90°-210°K, shows many structures which correspond to seven different TSC maxima (A to F) superimposed on each other. At higher temperatures (220°-280°K) no definite maximum is seen, and the spectrum shows a broad plateau. To derive the energy levels corresponding to each maximum, one should separate the various TSC peaks. We achieved that by using the "thermal cleaning" technique,⁹ and got a series of "cleaned" TSC curves, each of them giving information on the lowest, yet uncleaned level.

An example for such a curve is given in Fig. 13 (TSC current plotted versus the reciprocal of temperature). Here, the sample was illuminated at 145°K (excitation temperature), which is slightly above the maximum corresponding to level D (see Fig. 12). After illumination, the sample was kept at that temperature in the dark, then cooled and reheated in the dark through the whole TSC cycle. With such a procedure we "cleaned," as shown in the graph, the lower temperature side of the TSC curve of level E. The activation energy can now be derived from the exponential "initial rise" slope. This gives the value of 0.48 eV for the energy.

HgI₂ has a very low "dark current" in the range of the temperature mentioned above (see Fig. 14); thus, the "initial rise" method for evaluating the activation energy (known as Garlick and Gibson method¹¹) was successfully adapted for all the energy levels. In the TSC "dark current" measurement in Fig. 14 the sample was heated in the dark up to 270°K, where the "cleaning" process took place, and was then cooled and the TSC cycle was performed. When comparing Fig. 14 with Fig. 13, one can see that the "dark current" is much smaller than the TSC currents at all temperatures of interest.

Analysis of data corresponding to levels which are too close (like C and D in Fig. 12) is more complicated. This analysis is shown in Fig. 14, where the TSC curve consists of two different slopes corresponding to two different levels (H and I). One can obtain the lower temperature TSC curve by subtracting the higher slope from the data. The dotted line is the result of such a calculation. These two levels (H and I) were not labeled in Fig. 12. Levels C and D of Fig. 12, which are poorly resolved, were calculated in a similar way.

Additional information on a given level can be derived if one can determine whether the carriers thermally emitted from that level correspond to slow or fast retrapping.^{6,11} For some of the levels, such a decision was possible. The level energy calculated from the "initial-rise" method is the true energy, provided the capture cross-section is approximately temperature independent. The energy calculated from the Bube equation¹¹ ($E_{B'}$) corresponds to fast retrapping.

$$E_{B'} = kT_m \ln \frac{qN_c(T_m) \mu(T_m)}{J_m} E \quad (1)$$

The energy calculated from the Grossweiner equation^{1,2} ($E_{G'}$) corresponds to slow retrapping.

$$E_{G'} = \frac{1.5 k T_m T^1}{T_m - T^1} \quad (2)$$

where

T_m = temperature of the TSC maximum

μ = mobility

N_c = conduction band density of states

J_m = current density at the peak

E = electric field

T^1 = temperature at half maximum of the lower side of the TSC curve

If the "initial rise" value falls near one of the curves, then the capture and retrapping mechanism of that level can be determined.

The capture cross-section can be derived directly only for levels exhibiting slow retrapping. Assuming a slow variation with temperature of the capture cross-section σ , one can derive the equation¹¹

$$\sigma = \frac{V_{th} E \beta}{T_m^2 k N_c} \exp (E/kT_m) \text{ (cm}^2\text{)} \quad (3)$$

where β is the rate of temperature rise in °K/sec, V_{th} is the thermal velocity, and E is the "initial rise" energy.

For fast retrapping levels no direct information can be deduced on the capture cross-section. Instead, one can calculate the product $N_i \tau$, where N_i is the impurity concentration and τ is the overall recombination life time at T_m .¹¹

$$N_i \tau = \frac{T_m^2 N_i k}{\beta^2 E} \exp (-E/kT_m) \quad (4)$$

Table 1 summarizes the results derived for levels A-I, where E_{ir} is the energy derived by the "initial rise" method, and h and e stand for values calculated for holes and for electrons respectively.

Table 1. Trapping levels detected by TSC measurements

T_m (°K)	$N_i \cdot \tau$ (sec/cm ³)	(T_m) (cm ²)		Mechanism of Retrapping	E_G (eV)	E_B (eV)		E_{ir} (eV)	Level
		Holes	Electrons			Holes	Electrons		
90-95	2.5×10^8	5×10^{-18}	10^{-17}	Slow	0.13-0.17	0.22	0.23	0.16	A
115-120				Fast if holes	0.18-0.20	0.25	0.28	0.245	B
128-133		6×10^{-23} to 5×10^{-24}	10^{-22} to 10^{-23}	Slow		0.29		0.115-0.14	C
140-145				Probably fast		0.32	0.35	0.27 -0.36	D
165-168						0.36	0.40	0.485	E
180-185						0.43	0.47	0.51	F
204-207				Fast if holes	0.43	0.49	0.54	0.49	G
236-240				Slow				0.13	H
$T_m > 280$								0.62	I

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3.4 X-RAY IRRADIATION OF HgI₂

Most studies performed with HgI₂ and reported in the literature deal with the spectroscopic behavior of HgI₂ detectors irradiated with radioisotopes which emit gamma- or x-rays having low fluences of up to 10^4 photons per sec⁻¹ cm⁻² (pps). We measured dc current collected from a HgI₂ nuclear detector irradiated directly with an x-ray generator capable of producing high fluences of up to 10^{11} pps and energies up to 40 keV. The experiments were performed with continuous pulsed x-ray irradiation.

The crystals were cleaved, and a detector having a surface of 10×8 mm² and a thickness of 0.45 mm was fabricated. Aquadag contacts with a contact area of 8×6 mm² were applied. The detector was irradiated with a Philips PW 1140/00/60 x-ray generator, having as a target a copper tube which can reach a maximum power of 2000 W. The maximum current of the copper x-ray tube is 40 mA at an energy output of 40 keV. The beam width of the x-rays was 2×12 mm and the distance between the detector and the focus of the tube was 5 cm. The dc current produced from the HgI₂ nuclear detector was measured with a PAR model 134 Electrometer,

and the output registered with a recorder. The bias on the detector was 450 V, i.e., the electrical field acting on the detector was 10^4 V/cm. The $\mu\tau$ product (mobility μ times trapping time τ) of the HgI₂ crystal investigated here was 2×10^{-6} and 5×10^{-5} cm²/V for holes and electrons, respectively, measured from an uncollimated beam of ²⁴¹Am irradiated at an angle of 170° to the detector. Due to the low values of the $\mu\tau$ product, the collection efficiency of this particular detector was poor. Nevertheless, the results of the dc current measurement gave interesting information on the polarization problems of this crystal.

Figures 15 and 16 show the dependence of the detector current with the tube current, which in turn is proportional to the fluence (30 mA $\sim 10^{11}$ pps). In Fig. 15 this dependence is shown for several energy outputs taken in the same day; Fig. 16 shows this dependence for various times of irradiation at a constant x-ray energy output of 40 keV. One can see that the detector current is linear with the fluence, but that there is a certain time decay of the intensity of the detector current. Despite the time decay the current stays linear with the x-ray fluence.

Figure 17 shows the time decay of the detector current immediately after irradiation and that the current output reaches a steady value after eight hours of irradiation. The decay time at this fluence of 10^{11} pps is 30 minutes. The time dependence of the detector current caused by polarization can be explained by the equilibrium condition reached between the filling of new carriers and the release of the old carriers from the traps. The equilibrium condition thus minimizes the effect of the polarization.

In addition to the experiments performed with continuous irradiation, we investigated the possibility of measuring the dc response of HgI₂ to both collimated and uncollimated x-ray pulses having millisecond duration and fluences of 10^{11} pps. In order to obtain pulses of about 3 milliseconds, a mechanical chopper was specially built and placed between the x-ray tube and the HgI₂ crystal, as shown in Fig. 18. The chopper was standing at a distance of 25 mm from the collimator, and the electrode of the detector was exposed to the x-ray radiation and connected to a 7603 Tektronix oscilloscope by a 120-cm coaxial cable. The input impedance of this oscilloscope is 1 M Ω . Capacitance of the detector is 10 pF and of the coaxial cable is 110 pF. Total capacitance, including stray capacitance, was about 150 pF. The integration time of the electronic system was $150 \text{ pF} \times 1 \text{ M}\Omega = 0.15 \text{ msec}$. The chopper rise time was negligible.

For collimation purposes we used 2.3-mm-thick lead with a 1.6-mm-diam hole. The shape of the dc current pulses, both ideal and measured, are shown schematically in Fig. 19. Each pulse has two slopes, one due to the mechanical chopper and which gives electronic rise and decay times, and the other due to the trapping-detrapping detector response and which gives the detector rise and decay times. Since we collect mainly electrons, the pulse voltage is negative.

Three detectors were investigated: HR-I-11-8, HR-I-1-3, and HR-I-11-11, which had dimensions of $6 \times 6 \times 0.3$ mm, $5 \times 5 \times 0.5$ mm, and $5 \times 5 \times 0.3$ mm, respectively, and all had contact areas of 5×5 mm. Some of the detectors have been investigated with collimated and uncollimated x-ray pulses. Figure 20 compares the dc response between collimated and uncollimated x-ray pulses up to 40 keV and tube current of 16 mA, taken with the detector HR-I-11-8 biased in both cases at 750 V. One can see that by collimating the x-ray pulses the fluence is reduced from a pulse height of 400 mV for the uncollimated to about 20 mV for the collimated beam on the other side. The rise and decay times due to the detector are much shorter in the collimated pulses; for example, the uncollimated pulse slope in Fig. 20b is about 15%, whereas the same slope (Fig. 20a) in the collimated experiment is much smaller. To measure the small slope of the collimated detector, we intend to use a dc amplifier connected to the multichannel analyzer rather than to the oscilloscope. Alternatively, we may use a logarithmic amplifier.

The dc uncollimated pulses obtained by irradiation with 40-keV x-rays of detector HR-I-11-8 are shown in Figs. 21a, b, and c; the value of the tube currents were 10, 20, and 40 mA, which correspond to different fluences of about 10^9 photons $\text{sec}^{-1} \text{cm}^{-2}$. The linearity of the x-ray fluence with the tube current was shown in Fig. 15. Figures 22a and b show the uncollimated dc pulses obtained by irradiation of detector HR-I-11-8 biased with -1000 V. The tube currents in Figs. 22a and b were 10 and 40 mA, respectively.

It is interesting to note that the pulsed dc output of the x-ray irradiated detector is apparently unrelated to the quality of the HgI_2 crystal. For example, detector HR-I-1-3, which had a poor nuclear spectral resolution of ^{241}Am (uncollimated FWHM of 12 keV), showed a similar dc behavior to detector HR-I-11-8, which showed a good nuclear spectral resolution of ^{241}Am (uncollimated FWHM of 4 keV). Figure 23 shows the uncollimated response of HR-I-1-3 at a bias of -450 V

and irradiated with 40 kV x-rays at a tube current of 20 mA. Comparing Fig. 23 to Fig. 21b, which shows the dc response of detector HR-I-11-8, one can see that the rise time and decay time were unchanged, i.e., of about 1.3 msec, but the total height of the pulse is much lower despite the fact that the two detectors received the same x-ray fluence.

Figure 24 shows the dc pulses obtained by irradiation with collimated x-rays at 30 keV of detector HR-I-11-8. The shape of the pulse resembles much more the idealized case shown in Fig. 19, but this occurs at the lower x-ray energy of 30 keV, rather than the 40 keV shown in Fig. 20a.

Figure 25 shows similar dc pulses obtained by irradiation of detector HR-I-11-11 with collimated x-rays. At this stage our instrumentation does not allow us to compare the quality of detectors HR-I-11-8 and HR-I-11-11. Comparing Figs. 25a and c one can see that the pulse height in 25c is apparently twice the one shown in 25a, reflecting that the tube current in 25c is twice the one shown in 25a. Figure 26 shows the results with 40 keV x-rays.

3.5 NUCLEAR SPECTROSCOPIC DATA

In this section we report some ^{241}Am and ^{55}Fe nuclear spectra performed at UCLA on the HgI_2 crystals grown in Jerusalem, and some ^{137}Cs spectroscopic data using a pulse shape discriminator.

3.5.1 ^{241}Am and ^{55}Fe Spectroscopy

Two HgI_2 crystals grown in the horizontal furnace were measured. Crystal HR-I-11 weighed 10 g and was grown under conditions of continuous automatic increase in the air blowing, and HR-II-4 weighed 5 g and was grown by heating the air pipe leading from the air compressor to the furnace entrance. Detectors were fabricated from cleaved sections of the crystal and are shown schematically in Figs. 27a and b. Detector HR-I-11-13 measured $5 \times 5 \times 0.3$ mm, with a 3-mm-diam contact area of Aquadag, and was free of growth rings. Detector HR-II-4-2 was $3 \times 8 \times 0.5$ mm, with a 5-mm-diam contact area of Aquadag, and had two growth rings on the outer limits of the surface, as shown in Fig. 27.

The detector was fabricated in March 1976 by Dr. Slapa at UCLA. The detector crystal was first cleaved to a thickness of about 1 mm under the microscope, and was etched and polished on the bottom of the glass beaker manually in

5% KI solution until it obtained the desired thickness. The crystal was never touched by tweezers and was handled by two pieces of filter paper. The surfaces prior to contacting were microscopically flat without any cleavage steps. The Aquadag solution was dropped on the crystal with a syringe needle (for two minutes without touching the crystal) and allowed to dry for an hour.

The nuclear spectroscopic results we obtained follow. Figure 28 shows a collimated spectrum of ^{241}Am detected with the HR-I-11-13 detector under a bias of 800 V. The FWHM is 3.5 keV. The μ_{eTe} measured with ^{55}Fe was $6.1 \times 10^{-4} \text{cm}^2 \text{V}^{-1}$. No good hole spectrum was obtained, and the μ_{hTh} was measured. Detector HR-I-11-10, fabricated at Hebrew University, showed a much better nuclear spectrum, and FWHM of 5 keV and of 2.3 keV for an uncollimated and collimated source of ^{241}Am . Figure 29 shows an uncollimated spectrum of ^{57}Co detected with HR-I-11-10 at Hebrew University.

Much better nuclear spectroscopic data have been obtained at UCLA with Jerusalem detector, HR-II-4-2. Figures 30a and b show the collimated and uncollimated spectrum of ^{55}Fe at a bias of 1100 V. The FWHM was 560 eV for both detectors. (The gross FWHM of the detector was 900 and that of the pulser was 700 eV.) Figure 31a shows the collimated low energy region of a mixed ^{57}Co and ^{109}Cd and reveals photopeaks of 6.46, 14.36, 22.10, and 25 keV, with respective gross FWHM values of 1.2, 1.02, 1.15, and 1.25 keV, respectively. Figure 31b shows the respective collimated high and low energy spectra at a bias of 1100 V, and reveals the escape photopeaks and the 122-keV photopeak of ^{57}Co . Figure 32a shows the collimated and Figs. 32b and c the uncollimated spectra of ^{241}Am detected with HR-II-4-2 at biases of 1100 and 1700 V, respectively, with FWHM of 2.0 and 3.0 keV. The uncollimated spectrum in Fig. 32b was taken immediately after applying the bias, and the spectrum of Fig. 32c was taken after irradiating the detector with ^{241}Am for 3.5 hours. No measurable shift in the uncollimated 59.6-keV photopeak was observed during the 3.5 hours of radiation.

The $\mu\tau$ products for electrons and holes measured with ^{55}Fe on the detector HR-II-4-2 were $1.8 \times 10^{-4} \text{cm}^2 \text{V}^{-1}$ and $1.2 \times 10^{-6} \text{cm}^2 \text{V}^{-1}$, respectively. The most impressive nuclear spectroscopic results measured with this detector were the uniformity rather than the relative high μ value. The better uniformity is evident in comparing Figs. 30a and b with Figs. 32a, b, and c for collimated and uncollimated nuclear spectra.

3.5.2 ^{137}Cs Spectroscopy

In view of interesting spectroscopic results obtained with CdTe crystals using a pulse shaped discriminator¹³ we decided to try similar experiments with HgI₂ crystals. We used a slightly different electronic variation to replace the discriminator. The experimental setup, shown in Fig. 33, used an amplifier (CAV-N-3 Elscint) pileup rejector, which was originally designed to eliminate pileups of two pulses by measuring the time distance between the beginning and top of the pulse peak. In our setup the amplifier discriminates between pulses having short and long rise times; thus, the multichannel analyzer (MCA) will accept only pulses with short rise time. This is done by a valid pulse emitted by the CAV-N-3 which opens the gate of the MCA and allows the CAV-N-3 pulse to enter the MCA input. Each of the HgI₂ crystals showed a different optimum range of short rise times to give good spectra without losing too many counts. This optimum range depends on the $\mu\tau$ product and the detector thickness. The average optimum rise time was 200 nsec.

Figure 34 shows one of the best ^{241}Am spectra detected with the setup shown in Fig. 33. The FWHM of the 59.6 keV photopeak was 2.35 keV, compared to the pulser FWHM of 1.8 keV, which gives a net FWHM of 1.9 keV. The average number of counts reaching the MCA is 10 pulses per sec. The detector had a contact area of 5×5 mm, a thickness of 0.25 mm, and was biased with 800 V.

Figure 35 shows a spectrum of ^{241}Am of poor quality, taken with and without the discriminated pulse shape, measured with a $6 \times 6 \times 0.3$ mm detector which had a contact area 5×5 mm. The average number of counts entering the MCA was 31 pulses per sec in the coincidence mode and 144 pulses per sec in the normal mode. Some unexpected and unexplained results were obtained with the discriminator on the ^{137}Cs spectrum shown in Fig. 36. The detector was $6 \times 6 \times 0.3$ mm, with 5×5 mm contact area and biased with 800 V. The ^{137}Cs gamma radiation passed a 1-mm-diam, 10-mm-long collimator and penetrated the detector at an angle of 25° . The photopeak shown in Fig. 34 corresponds to 348 keV, which is much lower than the 662-keV photopeak of ^{137}Cs or the 582-keV mercury escape peak. The average number of counts entering the MCA was 3 pulses per sec. A possible explanation of the gamma photopeak at a much lower energy than expected is the fact that the detector was too thin (300μ), and the charge carriers did not have the time to transfer their energy so that hot electrons reached the electrode.

Fig. 1. Schematic representation of horizontal furnace for vapor growth of HgI_2 by temperature oscillation method developed at the Hebrew University of Jerusalem

2. Typical 50-g HgI_2 crystal grown in horizontal furnace

4424

Fig. 3. Growth rings revealed by etching in a 2% bromine solution in methanol (actual size)

4425

Fig. 4. Growth rings at larger magnification (200X)

4426

Fig. 5. Etch pit density in a freshly cleaved crystal (magnification 200X)

4427

Magnification 50X

Magnification 100X

Magnification 200X

Magnification 500X

Fig. 6. SEM picture of growth islands photographed while the crystal was in liquid air-cooled stage of the microscope

4428

Magnification 50X

Magnification 100X

Magnification 200X

Magnification 500X

Fig. 7. SEM picture of a cleaved surface of HgI_2 showing incomplete cleaved layers

4429

Fig. 8. X-ray excited emission of HgI_2 crystal at 77°K

4430

Fig. 9. Temperature dependence of the intensity of the $5400\text{\AA}$ band

4431

Fig. 10. Shift with temperature of the 5400Å emission band

4432

Fig. 11. Gamma-ray spectrum of ^{241}Am detected with $5 \times 5 \times 0.3$ mm cleaved sample (HR-I-11-8) biased under 800 V

4466

Fig. 12. TSC spectrum obtained from $16 \text{ mm}^2 \times 0.25 \text{ mm}$ cleaved sample (HR-I-11-9), at heating rate of 1°K/sec , after excitation and "cleaning" at 82°K

4434

Fig. 13. A "cleaned" TSC spectrum obtained from sample HR-I-11-9, at heating rate of 0.33°K/sec , after excitation and cleaning at 146°K . The energy calculated from the initial rise slope is indicated.

Fig. 14. "Dark current" TSC spectrum after a long "cleaning" process at 273°K . Dotted line curve is obtained by subtracting the higher slope from the data. The energy calculated from the initial rise slopes is indicated.

Fig. 15. Dependence of continuous dc current of the detector as a function of the x-ray tube current at different energy outputs when detector was irradiated the same day.

4457

Fig. 16. Dependence of continuous dc current of the detector as a function of the x-ray tube current at different times of irradiation but at a constant energy output of 40 keV.

4458

Fig. 17. Decay of fresh continuous dc current of the detector irradiated with 40-keV x-rays as a function of time.

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Fig. 18. Experimental setup for measuring pulsed dc current emitted from an x-ray generator.

4440

Fig. 19. Schematic representation of an idealized and measured shape of dc pulses obtained by x-ray irradiation of a HgI_2 detector.

4441

2 msec/division

a) Collimated

2 msec/division

b) Uncollimated

Fig. 20. Pulsed dc response of the detector HR-I-11-8 at a bias of 750 V irradiated with x-rays of up to 40 keV and a tube current of 16 mA.

4442

1 msec/division

a) 10 mA tube current

1 msec/division

b) 20 mA tube current

1 msec/division

c) 40 mA tube current

Fig. 21. Pulsed dc response of detector HR-I-11-8 at a bias of 750 V irradiated with x-rays emitted by a tube current, with energy of up to 40 keV.

4443

2 msec/division

a) 10 mA tube current

1 msec/division

b) 40 mA tube current

Fig. 22. Pulsed dc response of detector HR-I-11-8 at a bias of 100 V irradiated with tube current

4444

2 msec/division

Fig. 23. Pulsed dc response of detector HR-I-1-3 at a bias of 450 V irradiated with x-rays emitted from a tube current of 20 mA with energies of up to keV

4445

1 msec/division

a) 16 mA tube current
(collimated)

1 msec/division

b) 20 mA tube current
(collimated)

Fig. 24. Pulsed dc response of detector HR-I-11-8 at a bias of 750 V, irradiated with x-rays with energies of up to 30 keV.

4446

1 msec/division

a) 20 mA tube current

5 mV/division

2 msec/division

b) 30 mA tube current

10 mV/division

2 msec/division

c) 40 mA tube current

Fig. 25. Pulsed dc response of detector HR-I-11-11 at a bias of 800 V irradiated with collimated x-rays of up to 30 keV.

1-117

Fig. 26. Pulsed dc response of detector HR-I-11-11 at a bias of 800 V irradiated with collimated x-rays of up to 40 keV and tube current of 40 mA.

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Fig. 27. Schematic representation of the shape of Jerusalem detector plate HgI_2 crystals investigated at UCLA.

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Fig. 28. Collimated spectra of ^{241}Am under a bias of 800 V of HR-I-11-13. Crystal is 0.3 mm thick.

4450

Fig. 29. Uncollimated spectra of ^{57}Co detected with HR-I-11-10.

4451

a) Collimated

b) Uncollimated

Fig. 30. Spectrum of ^{55}Fe

4452

a) Low energy spectrum

b) High energy spectrum

Fig. 31. Mixed spectrum of ^{57}Co and ^{109}Cd (collimated).

4453

a) Collimated

b) Uncollimated immediately
after irradiation

c) Uncollimated after 3.5 hours
of irradiation

Fig. 32. Spectrum of ^{241}Am

4454

Fig. 33. Experimental setup using a pulse shape discriminator. Amplifier CAV-N-1 is connected to input, whereas valid output of the CAV-N-3 amplifier is connected to coincidence gate of MCA.

Fig. 34. Pulse shape discriminated spectrum of ^{241}Am . Time constant of CAV-N-1 was 6.4 msec biased under 800 V. Positive electrode of detector irradiated with gamma source. FWHM of 59.6-keV photopeak is 2.35 keV. The detector is 250 μm thick.

Fig. 35. Pulse discriminated and undiscriminated spectrum of ^{241}Am taken with a "poor" detector, 250 μm thick. Spectrum taken under same condition as in Fig. 2.

Fig. 36. Pulse discriminated spectrum of ^{137}Cs . Radiation enters detector through 1-mm-diam by 10-mm-long lead collimator at an angle of 25° . Detector thickness was 300 μm .

4421

4. CONCLUSIONS

The HgI_2 group headed by Prof. M. Schieber at the Hebrew University of Jerusalem has expanded its work during FY76. In addition to a novel crystal growth method in the horizontal furnace patented by ERDA, the group has also developed some complementary methods of evaluating the quality of HgI_2 crystals. These include TSC, x-ray luminescence, and dc measurements of x-ray irradiated HgI_2 detectors. These evaluation studies will be helpful in understanding better the carrier trapping-detrapping mechanism and may lead to the growth of better HgI_2 nuclear detectors in the future.

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