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GROWTH OF MERCURIC IODIDE (HgI_2) FOR NUCLEAR RADIATION DETECTORS

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1. INTRODUCTION

Mercuric Iodide is a material used for the fabrication of the sensing element in solid state x-ray and gamma ray detecting instruments. The advantage of the use of mercuric iodide in such instrumentation is that the systems are able to operate at room temperature, compared with systems based on germanium and silicon which need cooling to liquid nitrogen temperature.

The proper operation of the devices is determined to a large degree by the density of structural defects in the single crystalline material used in the sensing elements. Since there were strong indications that the quality of the material was degraded by the effects of gravity during the growth process (mechanical slippage and irregular convection) a research and engineering program was initiated to grow one or more crystals of mercuric iodide in the reduced gravity environment of space. A special furnace assembly was designed which could be accommodated in a Spacelab rack, and at the same time made it possible to use the same growth procedures and controls used when growing a crystal on the ground.

The space crystal, after the flight, was subjected to the same evaluation methods used for earth-grown crystals, so that comparisons could be made.

2. PREPARATORY GROUND-BASED RESEARCH

It is usually easy to suggest in general terms the possible benefits of an experiment in the reduced gravity of space flight. More effort is required to document with experimental results that the damaging effects of gravity on the structural quality of a crystal are indeed certain or at least highly probable.

Crystals of mercuric iodide are grown by physical vapor transport in a closed, evacuated ampoule. This means that mercuric iodide molecules sublime from a clump of heated source material and travel as vapor to the growing crystal. The transport in normal gravity is partially by diffusion and partially by convection, and as long as these transport mechanisms are constant with time, no negative effect on the crystal quality is expected to exist. Problems will arise when the convection transport becomes irregular with time, because the surface of the growing crystal will then be exposed to varying supersaturations of mercuric iodide vapor and growth irregularities will result. In the absence of gravity, transport will be exclusively by regular diffusion flow.

No technique has been developed at this point to measure in detail existing convection currents on the ground, and the only way to obtain information about the effects of the convection currents on the structural quality is by actually doing the experiment.

A different situation exists with respect to the statement that the crystal will slip by the force of its own weight, and thereby increase the density of defects. To obtain quantitative data concerning this effect the dislocation structure and the critical resolved shear stress (CRSS) of the crystalline material was studied in cooperation with a group at the University of California in Santa Barbara. This work has been reported in References 1 and 2. The results of the measurements indicated that, expressed on a macroscopic scale, a crystal of approximately 1 cm^3 can be subject to slippage perpendicular to the c-direction under the effect of its own weight when it is at the growth temperature. Figure 1 shows a series of yield strength measurements at different temperatures. The curves indicate how the CRSS of a sample changes with temperature and that no irregular hardening effects take place during the test. It should also be realized that the samples used for these measurements may already have been strain-hardened by the cutting and polishing process required to shape the samples.

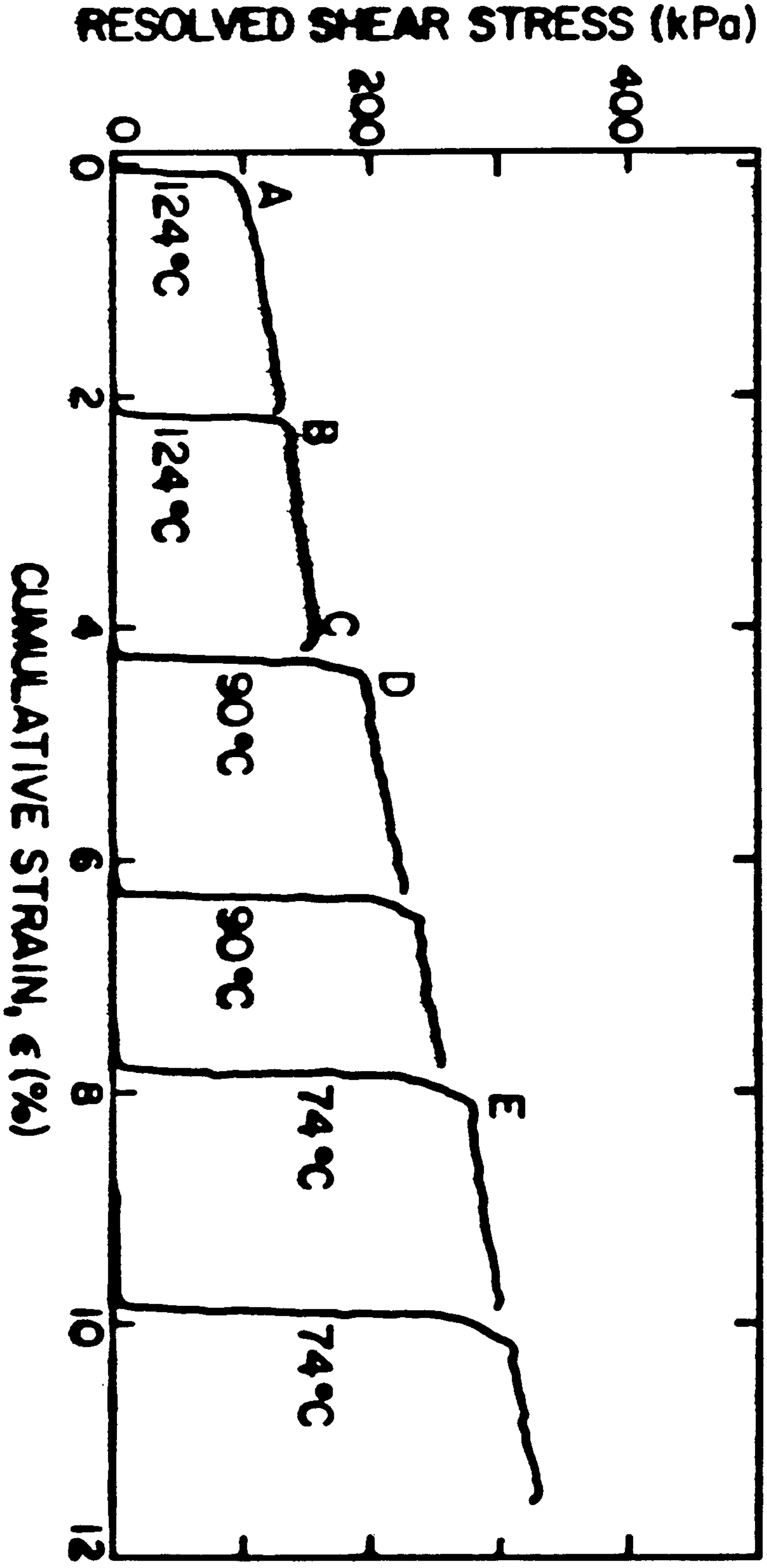
RESOLVED SHEAR STRESS (kPa)
0
200
400
CUMULATIVE STRAIN, ε (%)
0
2
4
6
8
10
12
A
B
C
D
E
124°C
124°C
90°C
90°C
74°C
74°C

Fig. 1

3. CRYSTAL GROWTH

Furnace and Growth Ampoule Development

To accommodate the Spacelab III facilities, a special furnace has been designed which reflects the basic elements of the usual ground-based furnaces (Ref. 3). A sketch of this prototype furnace is shown in Figure 2.

The crystal growing ampoule, located in the center of the furnace, has a cylindrical shape. The bottom surface of the ampoule is indented so that internally it forms a pedestal on which the crystal grows. The indentation fits over a metal support tube, inside of which a Peltier cooler is installed to provide cooling to the growing crystal.

The thermal profile around the ampoule necessary for crystal growth is provided by two independently controlled heating coils. The lower part of the ampoule is heated by means of a circular coil which is covered with a metal heat equalization ring. A helically wound vertical coil provides basically an equally distributed amount of heat along the vertical walls of the ampoule. The combined heat inputs create a temperature profile which has a minimum at approximately half the height of the ampoule. At this level the crystal growth source material accumulates when the center of the pedestal is not cooled by the cold sting. Nucleation and crystal growth can be initiated by a combination of a reduction in the ampoule bottom temperature and activation of the Peltier cooler.

On the tope of the helical coil a reflective shield is installed to minimize heat losses. This furnace assembly is covered with a bell jar of high optical quality, so that the growing crystal can be observed through a microscope.

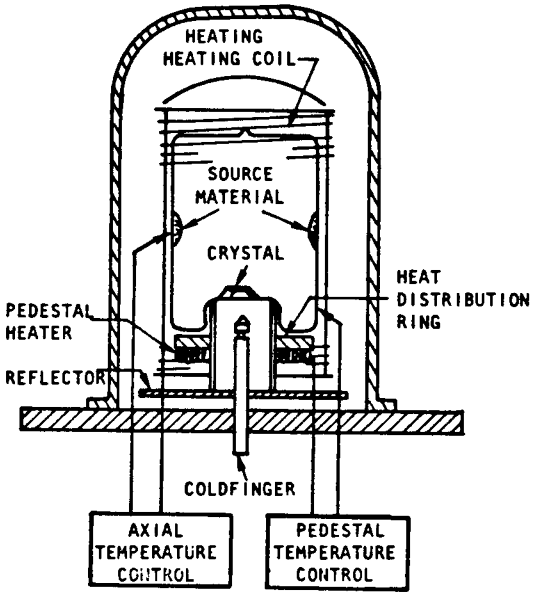

Fig. 2

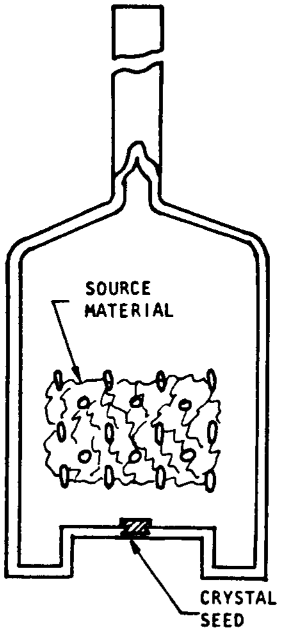

Fig. 3

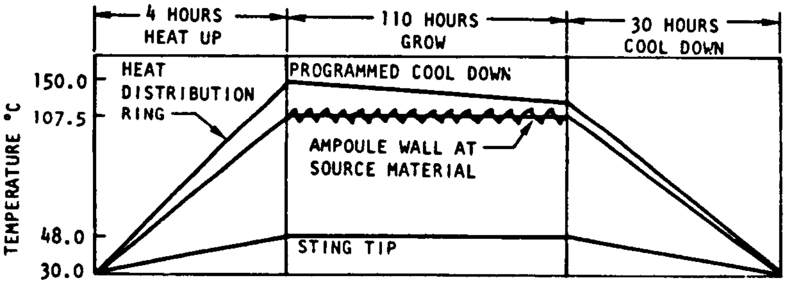

Fig. 4

The temperature profile along the walls of the ampoule used to grow the crystals on the ground is created by a combination of radiation and convection effects. During an experiment in the Spacelab, the driving force for convection in the air between the bell jar and ampoule will be reduced. In addition, the furnace assembly will assume different orientations with respect to the prevalent acceleration vector. In order to evaluate the effects of changed gravity conditions a series of experimental simulations was performed whereby the space between the bell jar and ampoule was evacuated and the whole furnace assembly was positioned upside down. These experiments showed that the heat supply around the top of the ampoule is not sufficient when there is not a continuous convective supply of hot air from the lower part of the furnace which collects in the upper part of the bell jar and keeps the top of the ampoule hot. This problem was corrected by installation of a lamellar heater on the inside surface of the reflecting dome (not shown in Figure 2). The power input to this heater is slaved to the power controls for the helical heating coil.

The normally used ground-based growth ampoules have smooth walls and a flat pedestal. In the initial phase of the crystal growth procedures, the poly-crystalline charge is evaporated from the bottom of the ampoule to the minimum temperature zone on the vertical wall, and subsequently a seed is nucleated by reducing the pedestal temperature and activating the Peltier cooler. In order to maximize the crystal growth time in space it was decided to perform these activities on the ground before the flight. The resulting ampoules with source and seed in place were subjected to vibration tests, simulating take-off and re-entry vibrations, to ascertain the adherence of source and seed to the glass surface. Both the grown seed and parts of the source material started to come loose at certain critical vibration intensities. The ampoule design was therefore modified to incorporate glass fingers on the part of the inside wall where the source is located and a dovetail cavity in the pedestal to anchor the seed (see Figure 3). The ampoule is positioned in the furnace with the section with glass fingers at the rear. The resistance of the furnace windings has been adjusted in that area so that an asymmetric cold region is created for the source material to collect. The seed is nucleated inside the dovetail cavity and is grown until it completely fills and extends somewhat beyond the cavity. The ampoules prepared in this way are again subjected to qualification testing before flight.

The growth sequence is fully automated and consists of three essential parts: heat-up, growth and cool-down (see Figure 4). During the heat-up phase the temperatures at the different control points in the furnace (source, heat distribution ring, cooling sting) are increased in a controlled way to values which are optimal for growth. The growth phase is then started by gradually decreasing the pedestal temperature, primarily by reducing the heat input to the base heater. During the growth phase it is possible, when necessary, to oscillate the temperature of the source material so that periodic growing and etching of the crystal occurs to obtain more stable crystal growth and to avoid spurious nucleation. This is a technique usually employed during the early stages of the growth of mercuric iodide crystals (Ref. 4 and 5).

When the source material has almost been depleted or when the experiment time has elapsed, the system is gradually cooled down to ambient temperature and is returned to its storage container for re-entry.

4. SPACE EXPERIMENT

Notwithstanding the efforts made to predetermine the proper growth conditions on the ground (elimination of convection around the ampoule by evacuation of the space between the ampoule and the bell jar), the temperatures again had to be adjusted to obtain crystal growth in space. This accounts for the fact that during the first 14 hours in space no growth was observed. The temperature difference between the source and the pedestal had to be increased by 1.6°C to initiate growth.

As growth continued, it became clear that much higher temperature gradients could be applied than is usually done on the ground, without disturbing the stability of the crystal surfaces or without causing any new nucleation. Using the higher temperature gradients it was therefore possible to obtain higher growth rates by diffusion transport alone than is usually seen on the ground with both diffusion and convection transport, as shown in Figure 5.

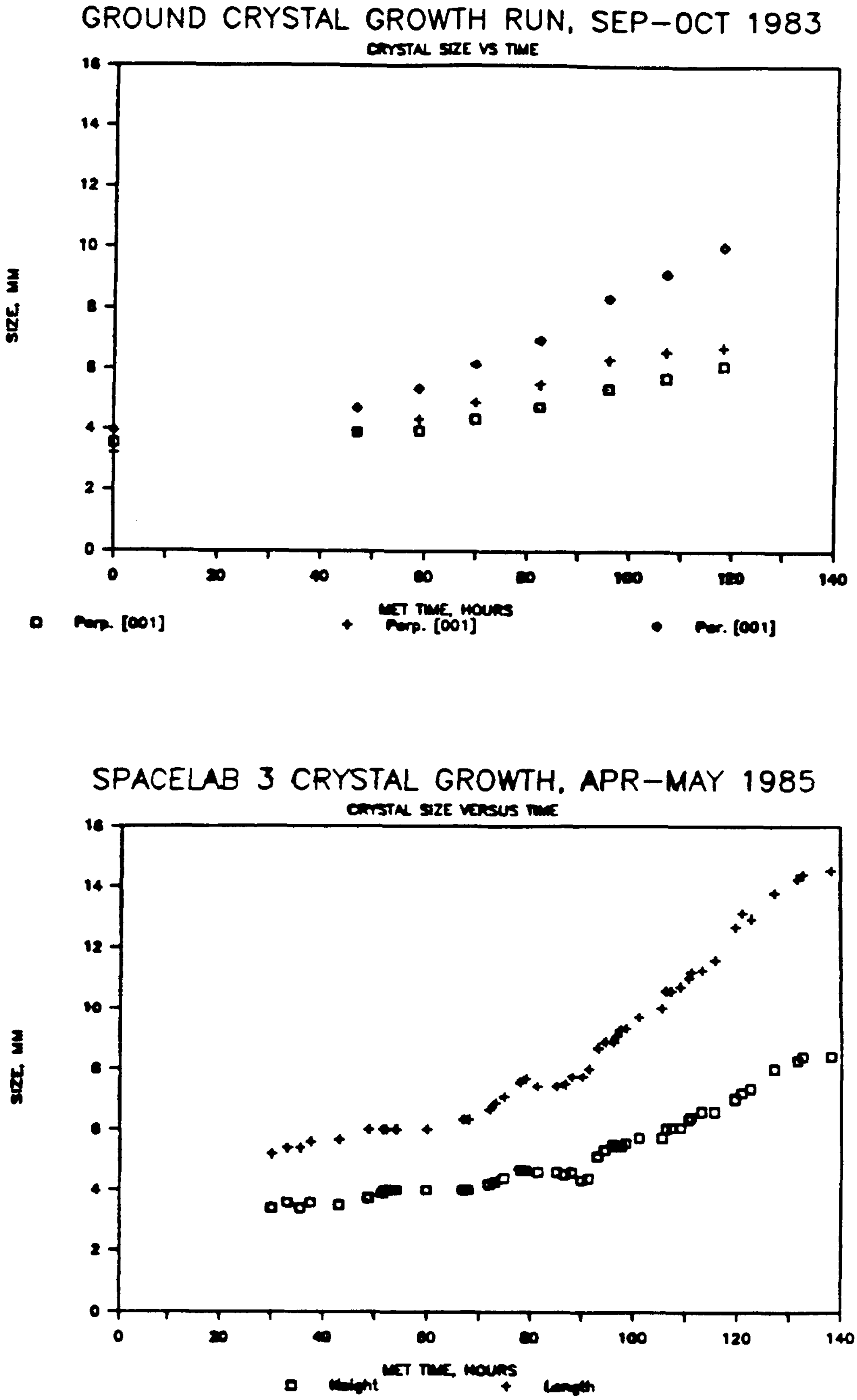

Fig. 5

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5. CRYSTAL ANALYSIS

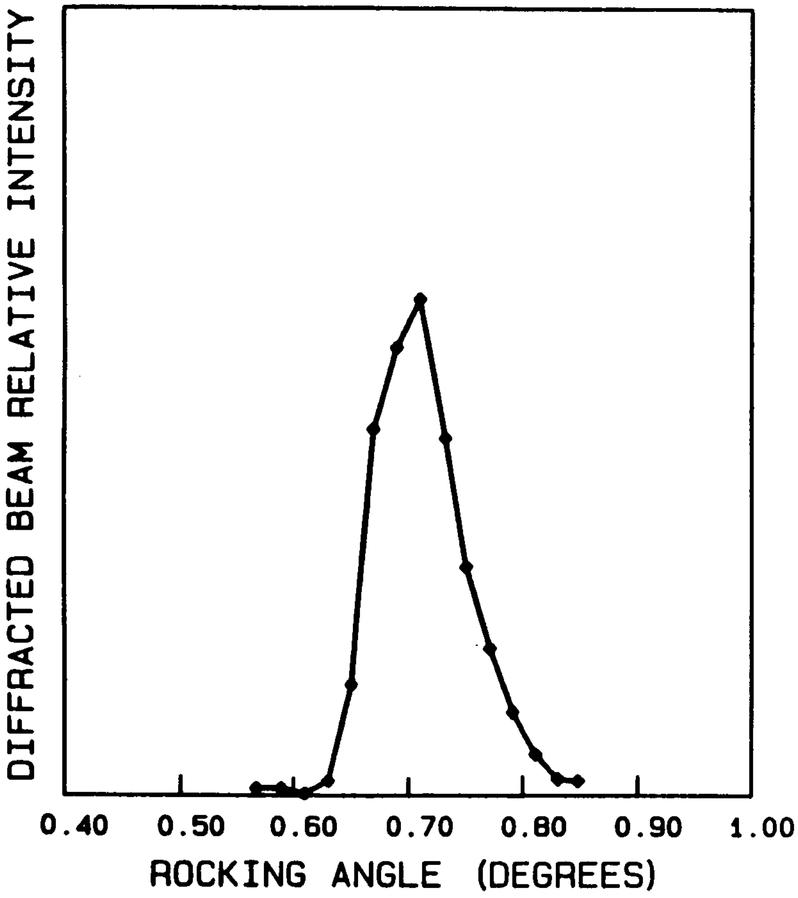
Gamma Ray Rocking Curves

Since mercuric iodide is selected for radiation measurements because of its high absorption, it is impossible to use regular x-rays to do transmission measurements on pieces thicker than a few hundred um. To evaluate the structural quality of a crystal approximately 1 cm thick it is necessary to use higher energy gamma rays emitted by artificial sources. The advantage of these kind of sources is that the radiation emitted by them is highly monochromatic, so that the diffraction peaks have a Gaussian shape.

Rocking curve measurements on the space crystal have been performed at the University of Missouri Research Reactor in Columbia, Missouri, where high intensity isotope sources can be produced and the equipment to do this type of measurements is available (Ref. 6). The results are shown in Figures 6a and 6b. The rocking curve of an earth-grown crystal shows several peaks, indicating that the crystal really consists of three sections at slight angles with respect to each other. The rocking curve of the space crystal on the other hand shows only the one peak expected of a single crystal without major grain boundaries. The space crystal however shows signs of internal strain as evidenced by the width and asymmetrical shape of the peak.

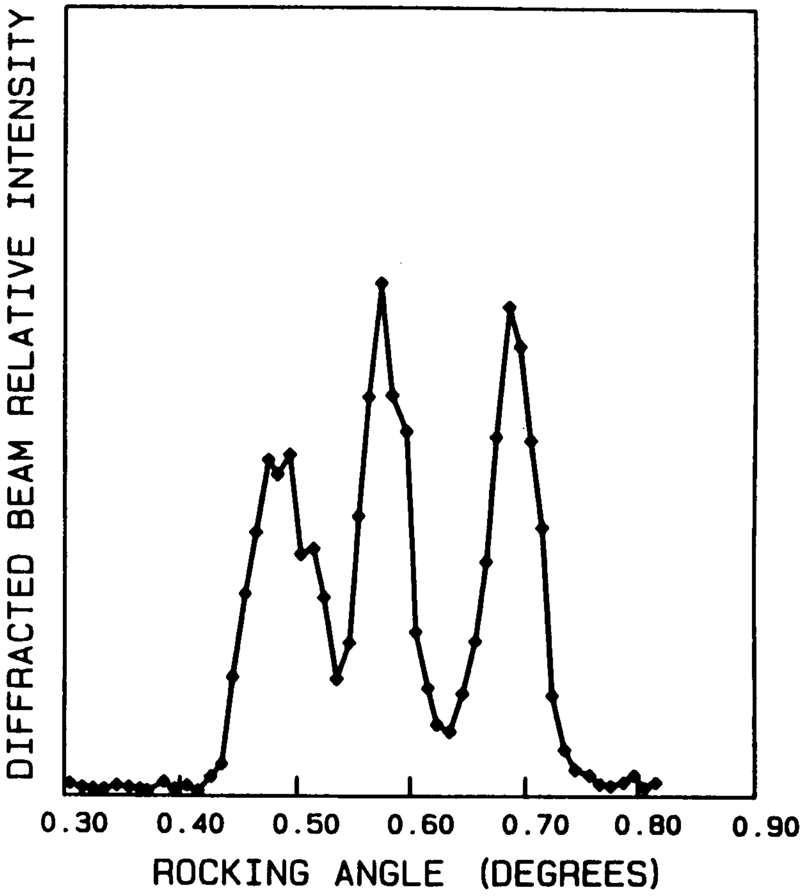
Electronic Measurements

The operation of radiation detectors is critically dependent on the transport properties of the electronic charge carriers created by the incident radiation. A good measure of the quality of the material is therefore provided by the values of the electron and hole mobilities and lifetimes (trapping times). Because of the high resistivity of the material these values can most adequately be determined by evaluation of the nuclear spectral response of a detector structure as a function of applied bias. Details of this procedure have been described in the literature (Ref. 7).



GAMMA DIFFRACTION ROCKING CURVE
FOR THE SL-3 CRYSTAL

Fig. 6a



GAMMA DIFFRACTION ROCKING CURVE
FOR AN EARTH GROWN CRYSTAL

Fig. 6b

The data obtained for detectors made from the space crystal and the maximum values found for ground-based detectors are as follows:

	μ_e (cm^2/Vsec)	μ_h (cm^2/Vsec)	$\mu_e\tau_e$ (10^{-4} cm^2/V)	$\mu_h\tau_h$ (10^{-6} cm^2/V)
Ground-based Crystals (max.)	125	4.2	20	6.2
Space Crystal	250	10.7	21	42

One can see that the electronic properties of the space crystal are significantly improved over ground-based crystals, especially with respect to the holes, which is the most critical factor for detector operation. When considering these results it should be realized that the maximum values quoted for the ground-based crystals are for different detectors and are not found together in one single detector.

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SUMMARY AND CONCLUSIONS

The experiment to grow a crystal of mercuric iodide in space performed exceptionally well, so that the stated experimental objectives were fulfilled. That the experiment was successful the very first time around is, in our opinion, for a large part due to the extensive pre-flight ground-based research and operational experiments.

The space growth conditions showed that the stability of the crystal surfaces is much higher than on the ground where convection is present. As a result the temperature gradient in the furnace could be increased so that the growth rate with diffusion transport alone was still larger than on the ground.

The structural quality of the space crystals, as measured by gamma ray rocking curves, is superior to ground-based crystals. As a result, the electronic transport properties of the material were significantly improved.

ACKNOWLEDGMENTS

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FUTURE ACTIVITIES

THE VAPER CRYSTAL GROWTH (VCG) EXPERIMENT IS PRESENTLY SCHEDULED TO BE REFLOWN ON THE IML-1 MISSION. THE PURPOSE OF THE IML-1 EXPERIMENT IS PRIMARILY TO REPEAT THE SPACELAB 3 EXPERIMENT SO THAT BETTER STATISTICS ABOUT THE QUALITY OF SPACE-GROWN CRYSTALS AND THE PERFORMANCE OF THE EQUIPMENT WILL BE AVAILABLE. USING THE EXPERIENCE GAINED IN SPACELAB 3, IT WILL BE POSSIBLE TO MAKE THE GROWTH PROCEDURE MORE EFFICIENT BY STARTING ACTUAL GROWTH SOONER. IN ADDITION, SOME NEW ASPECTS OF THE CRYSTAL GROWTH PROCEDURE WHICH ARE ALREADY BEING USED ON THE GROUND WILL BE INCORPORATED IN THE FLIGHT PROCEDURES.