# SPACELAB 3 VAPOR CRYSTAL GROWTH EXPERIMENT

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## ABSTRACT

The Space Shuttle Challenger, with Spacelab 3 as its payload, was launched into orbit April 29, 1985. The mission, number 51-B, emphasized materials processing in space, although a wide variety of experiments in other disciplines were also carried onboard. This report describes one of the materials processing experiments on this flight, specifically the growth of single crystals of mercuric iodide by physical vapor transport.

## DISCUSSION

Mercuric iodide was chosen as the subject material for the space experiment for several reasons. In the form of a single crystal, it has high technological interest as a detector of X- and gamma radiation. Also, because of the relatively low growth temperatures used, the flight furnace could be built to operate with modest electrical power demands on the Spacelab resources. Finally, very few vapor crystal growth experiments have been done, to date, under microgravity conditions.

Single crystals of mercuric iodide can be used to detect nuclear radiation. This ability was first recognized in the early 1970's [1]. Since then, efforts at a number of different laboratories have succeeded in improving the overall quality of these detectors [2].

Two basic properties of the mercuric iodide make it attractive as a radiation detector material: First, the compound has relatively high atomic numbers (80, 53) for its constituents, which results in a large photoelectric cross-section for X- and gamma radiation. Thus, high absorption and good detector efficiency can be achieved in a relatively thin section of the material. The second feature of this semiconductor is its relatively large bandgap (2.1 eV). As a consequence, mercuric iodide detectors can be operated at room temperature, in contrast to the cryogenic temperatures which are required of other semiconductor nuclear radiation detectors.

While a steady and significant improvement in detector device characteristics has occurred over the past 10 years, it still remains that mercuric iodide has not yet achieved its full potential. As is generally true with all devices made from semiconductor single crystals, improved performance comes with advances in three key areas: purification and better control over the quality of the starting material, better understanding and control of the growth of the single crystals, and development of optimal processing techniques to fabricate the devices.

The space experiment focused on crystal growth, although parallel efforts were made in improving the other key areas. Two specific aspects of the crystal growth motivated the interest in growing crystals in microgravity.

First, on Earth in 1 g, convection occurs in the conventional growth furnaces that are used for growing mercuric iodide crystals. Early ground based research indicated that this occurred both outside and inside of the closed ampoule containing the polycrystalline mercuric iodide source material and the growing crystal. The occurrence of convection in crystal growing is not, in all cases, undesirable. There is, however, a risk that convective instabilities will occur, which would lead to growth irregularities, and these are highly undesirable. In our system, irregularities in the vapor transport and/or the temperature fields could occur. Such irregularities are known to produce crystal defects (vacancies, etc.).

A second aspect involved the extremely fragile nature of these crystals. Previous laboratory experience showed that they were very soft and easily damaged in handling, sometimes even "cold flowing," just by their own weight, over wires used for electrical leads. Testing indicated that at room temperature the critically resolved shear stress on well-grown crystals was indeed very low and decreased with increasing temperature. It seemed quite possible that the weight stresses in the crystal at growth temperatures might be sufficient to cause plastic deformation.

Regardless of the cause, flaws in a crystal's structure seem generally to result in degraded electronic properties for all semiconductor crystals. Growth in microgravity should help to alleviate both of these problems, thereby leading to a higher quality crystal. Gravity driven convection would be virtually removed, taking with it the problem of irregularities in the transport rate of the mercuric iodide vapor. Also, the crystal would be under no weight stress during its growth, and could be cooled down and optimally stored before the shuttle made its deorbit and landing.

Although a microgravity experiment was the eventual goal of this program, a large part of the total effort was done on the ground. Starting from apparatus and procedures developed for 1 g vapor crystal growth, studies were made on ways to reduce the power, size, weight and complexity of the crystal growth furnaces. As a result, several modified, "protoflight" furnaces were built and tested.

Attempts were also made to simulate the effect microgravity would have in changing the 1 g temperature fields in these furnaces (the absence of convection was simulated by testing the furnace under reduced pressure, in a bell jar). This testing led to several other furnace modifications that were used to "tailor" the simulated microgravity temperature profile. A schematic of a protoflight furnace is shown in Figure 1. The real flight furnace was very similar physically, and was functionally equivalent, to the protoflight unit.

The ground research phase also provided an opportunity to grow a number of 1 g control crystals and to develop new characterization methods for the crystals. In particular, methods were developed to measure mechanical properties of the crystals (critical resolved shear stress) and to examine the structural perfection, both on the surface and in the bulk of the crystal (X- and gamma ray diffraction "rocking" curves).

The mission timeline was developed to allow the crystal growth experiments to be activated as early as possible in order to maximize growth. For the Vapor Crystal Growth (VCG) experiment, its timeline was divided into three phases: heatup, crystal growth, and cooldown. The durations were approximately 4, 118, and 8 hours, respectively, with the initial turn-on occurring about 10 hours after orbit was achieved.

Two members of the payload crew were responsible for the VCG on alternating 12 hour shifts, with duties that included stowing and unstowing of the crystal growth ampoule, setting up and conducting the experiment, and holding periodic two-way dialogs with the science team at Johnson Space Center. Members of the science team at the JSC Payload Operations Control Center had available to them several data displays, including all of the key furnace temperatures and, at times, a TV image of the growing crystal.

The experiment made use of an ampoule containing a small seed (approximately 3 mm on a side) which had been grown on Earth. During the initial heatup phase, some thermal etching of the seed occurred, as planned, in order to remove any stray particles of mercuric iodide "dust" that might have come onto the seed's surfaces.

After landing at Edwards Air Force Base, the crystal, protected within its growth ampoule, was taken from the Spacelab and transported to the Marshall Space Flight Center, where a series of photographs were made of it. The size of the crystal was approximately 1.2 cm x 1.2 cm x 0.8 cm, and its weight was about 7.2 grams. During the experiment, the linear growth rate of the crystal varied, reaching a maximum of about 3 mm/day. This compares with growth rates on earth that range from about 0.5 to 2 mm/day, with an average of about 1 mm/day. (On Earth it is difficult to achieve a rate of 3 mm/day without the rapid occurrence of undesired spurious nucleation).

The ampoule containing the crystal was next carried to the University of Missouri for gamma ray rocking curve testing. In this testing, a collimated, 1 mm diameter monoenergetic beam of gamma rays impinges on the crystal. Much of the highly penetrating beam simply goes straight on through the sample; however, a fraction is Bragg-diffracted at a small angle and, after passing through and exiting the sample, it is then detected. An advantage to using gamma rays is their ability to penetrate materials. This allowed testing the crystal while it was still enclosed in its glass ampoule. In addition, the measurement of the sample in transmission gave a good indication of its bulk quality.

By orienting the crystal at its appropriate Bragg angle relative to the incident beam, the signal from the diffracted beam will yield information about the crystal's structural quality. For a perfect structure, when the crystal is rotated, or rocked, through the neighborhood of its Bragg angle, the intensity of the diffracted beam will go through a sharp maximum. If defects occur, however, the Bragg diffraction peak can be broadened, or even split into multiple peaks. For example, a crystal containing two subgrains, one slightly misoriented from the other, can produce two separate Bragg diffraction peaks that are slightly displaced in angle, as the crystal is "rocked," or rotated, in the beam.

The gamma ray rocking curve tests showed the space crystal to be of high quality. The results were approximately equal to the very best obtained from samples grown on Earth, and noticeably superior to the average. Figures 2 and 3 show rocking curves for space and Earth crystals. The space crystal, with a single peak, shows the region to be free of these defects. The multiple peaks in Figure 3, which are a usual occurrence for Earth-grown crystals, are an indication that nearby regions of the crystal are slightly misoriented to one another (subgrains).

Rocking curve testing of a similar nature, but using less penetrating X-rays, was performed at the University of California at Los Angeles. Again, the results showed the space material to be of high quality, and with a quality gradient, confirming the gamma ray results. Because of the lower energy, the X-ray beam penetration was only a few micrometers into the sample, in contrast to the gamma ray measurements. Because it was essentially a surface measurement, this test did provide a sensitive means of monitoring damage due to handling and detector processing. Such damage was expected, and found, although attempts were made to minimize it.

Damage did occur to the first slice that was cut from the crystal. About 2 mm thick, the slice was sawed from the crystal, then temporarily mounted with silicone rubber for additional X-ray rocking curve testing. In demounting the slice from the rubber, damage occurred. After fabrication into a detector, the unit was tested for its performance as a gamma ray spectrometer using Americium 241 and Cesium 137 isotope sources. Only average results were obtained.

Another slice was then cut and used for detector fabrication, however the steps of silicone rubber mounting and X-ray rocking curve testing were omitted. In this case, the performance of the detector proved to be much better than that of the first slice. If similar data from a population of several thousand detectors made from Earth-grown crystals is compared with it, only about the top 6 percent of the Earth detectors would perform as well.

## REFERENCES

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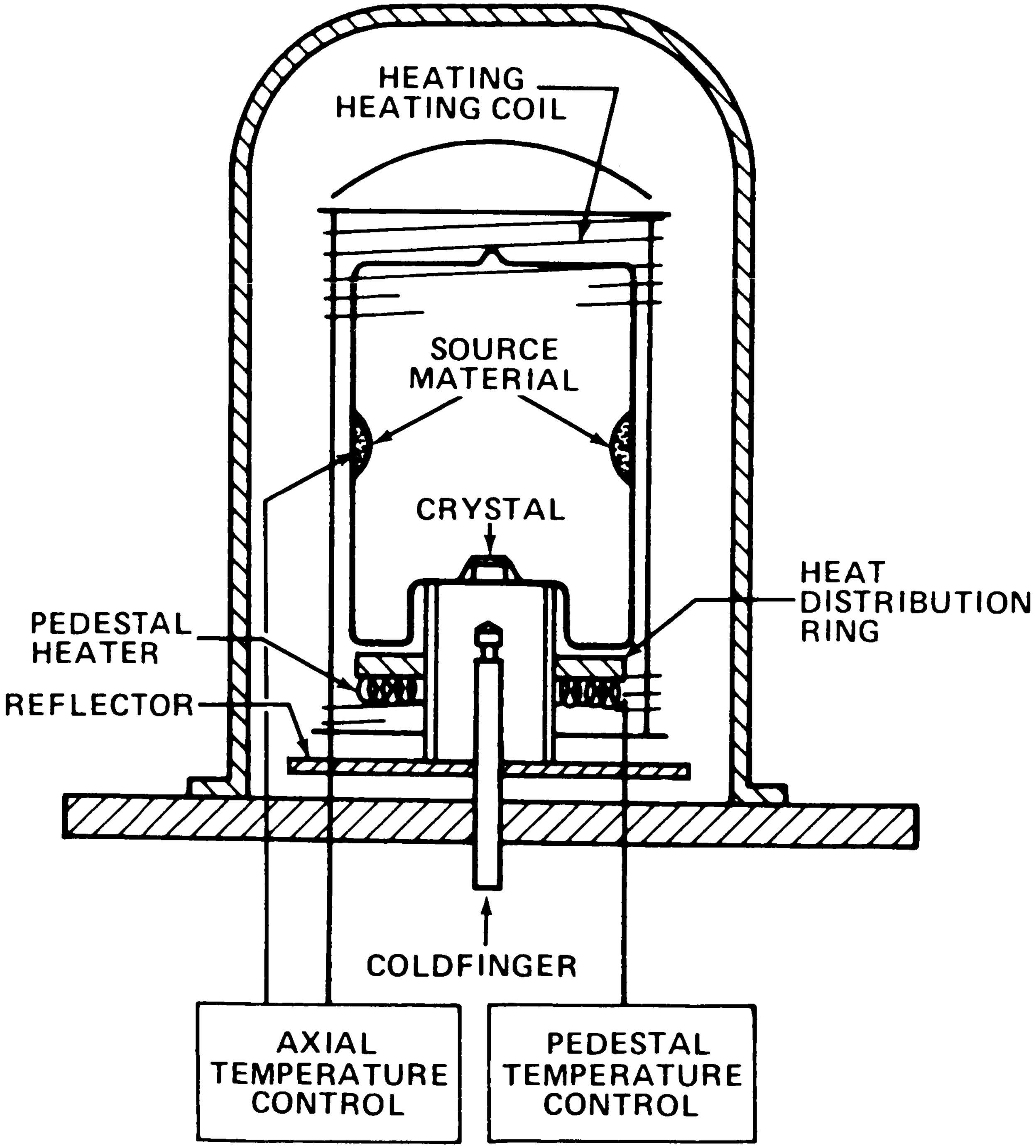
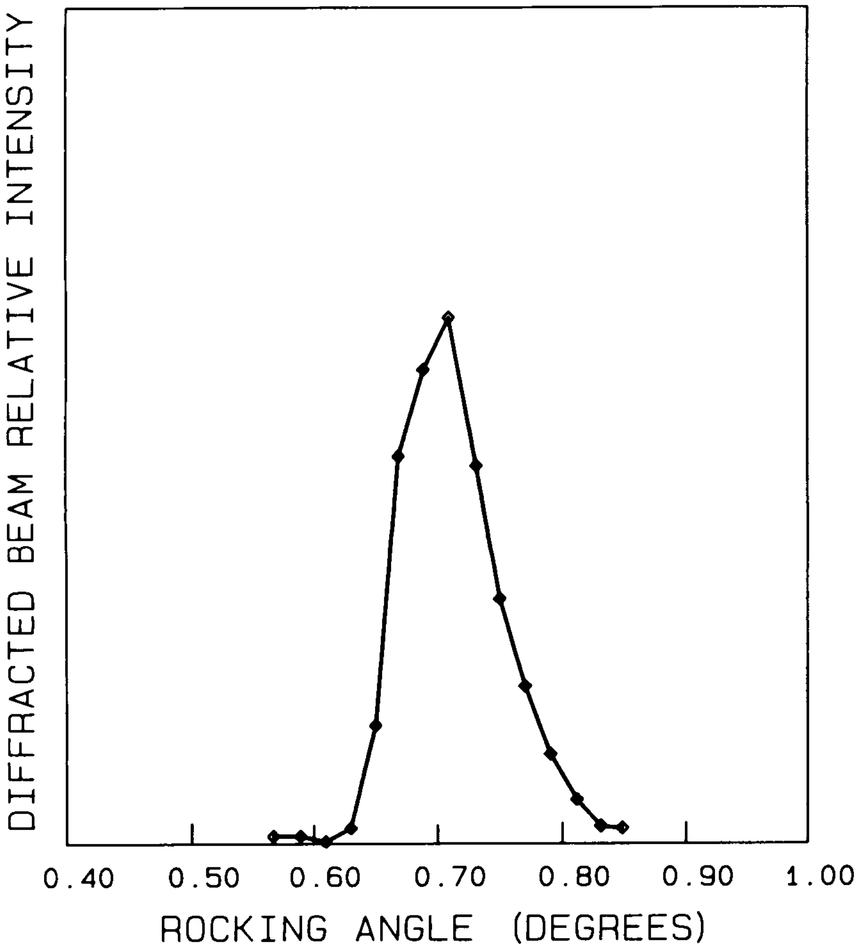


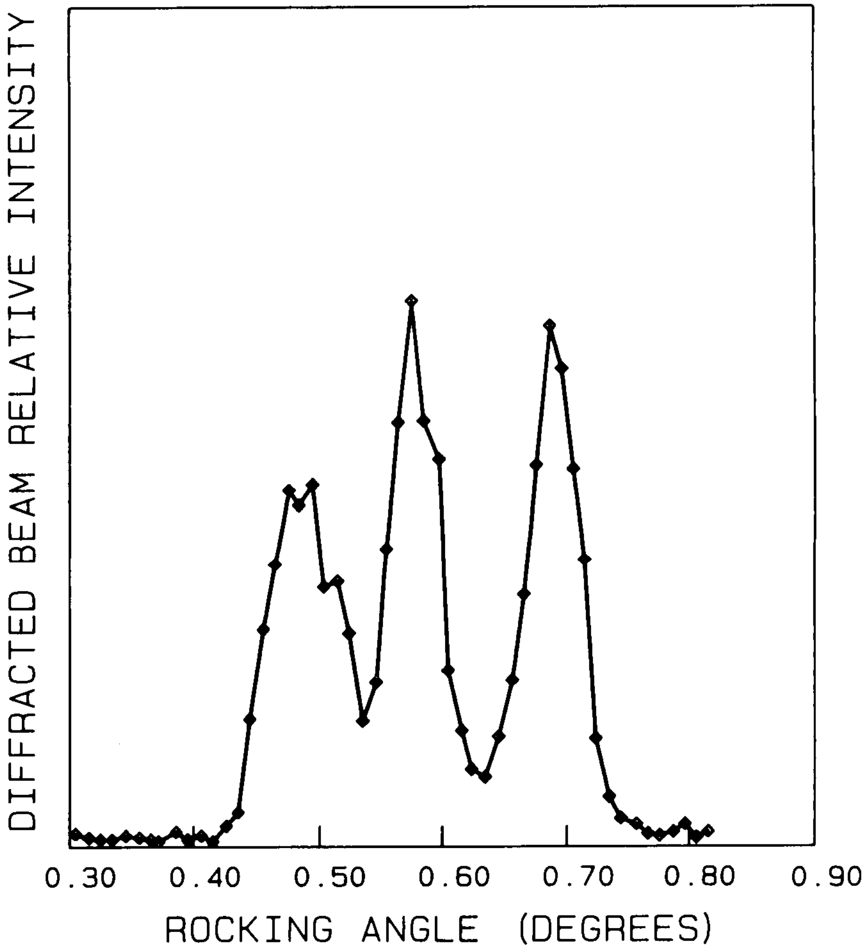
HEATING
HEATING COIL
SOURCE MATERIAL
CRYSTAL
HEAT DISTRIBUTION RING
PEDESTAL HEATER
REFLECTOR
COLDFINGER
AXIAL TEMPERATURE CONTROL
PEDESTAL TEMPERATURE CONTROL

FIG. 1



GAMMA DIFFRACTION ROCKING CURVE
FOR THE SL-3 CRYSTAL

FIGURE 2



GAMMA DIFFRACTION ROCKING CURVE
FOR AN EARTH GROWN CRYSTAL

FIGURE 3