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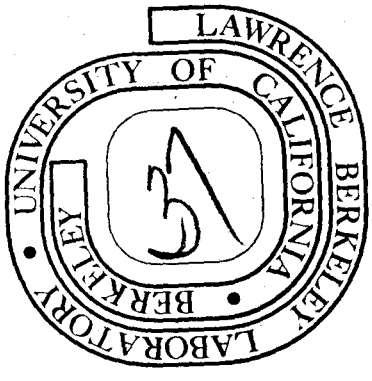

DISCLAIMER

HIGH-ENERGY PROTON RADIATION DAMAGE OF HIGH-PURITY GERMANIUM DETECTORS

LBL-7218

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Summary

Motivated by their applicability to gamma-ray spectroscopy experiments in space, quantitative studies of radiation damage effects in high-purity germanium detectors due to high-energy charged particles have been initiated with the irradiation by 6 GeV/c protons of two 1.0 cm thick planar detectors maintained at 88°K. The threshold for resolution degradation and the annealing characteristics differ markedly from those previously observed for detectors irradiated by fast neutrons.

Under proton bombardment, degradation in the energy resolution was found to begin below 7×10^7 protons/cm^2, and increased proportionately in both detectors until the experiment was terminated at a total flux of 5.7×10^8 protons/cm^2, equivalent to about a six year exposure to cosmic-ray protons in space. At the end of the irradiation, the FWHM resolution measured at 1332 keV stood at 8.5 and 13.6 keV, with both detectors of only marginal utility as a spectrometer due to the severe tailing caused by charge trapping. The two detectors displayed a significant difference in proton damage sensitivity, which is consistent with fast neutron damage effects. To ensure that detector variability did not influence the comparision of proton- and neutron-induced damage effects, one of the detectors had been used previously in a neutron damage experiment. The threshold for high-energy proton damage was found to be markedly lower, roughly 5×10^7 protons/cm^2, compared to 3×10^9 neutrons/cm^2 for fast neutrons.

Annealing these detectors after proton damage was found to be much easier than after neutron damage. A satisfactory level of recovery after high-energy proton damage can be achieved with in-situ annealing in the range of 100°C. The sharp contrast between the peak shapes observed during the annealing of proton and neutron irradiated detectors indicates that a different damage mechanism must account for at least a large part of the proton damage.

Introduction

Interest in the use of germanium high-resolution gamma-ray spectrometers for astronomical and planetological observations on extended space missions has grown recently.[1,2,3] Although several studies of the radiation damage effects of fast neutrons on germanium detectors have developed considerable empirical information,[4,5,6,7,8] no comparable measurements have been made to evaluate the effects of the high-energy cosmic-ray protons that will be encountered in space. Because the viability of many experiments conducted in space, as well as those using germanium-detector charged-particle telescopes in accelerator experiments, will be dependent on the radiation damage effects of high-energy protons, we have undertaken a program to study these effects in high-purity germanium detectors. The results of our initial experiment are reported here.

Experimental

Two high-purity planar germanium detectors fabricated from p-type material were irradiated with protons of 6 GeV/c momentum (5.1 GeV kinetic energy) at the Argonne Zero Gradient Synchrotron (ZGS) in May, 1976. The protons were polarized, which is irrelevant for the irradiation except that the lower intensity of protons in the accelerator ring during polarized operation will reduce the background level in the experimental area. The two detectors were mounted back-to-back in a single common vacuum cryostat cooled with LN_2. Allowing for the thermal gradient along the cold finger, the temperature of the detectors was about 88°K. Both detectors were 1.0 cm thick; the larger detector, designated 494-3.1, had an average diameter of 3.2 cm; the second detector, designated 285-1.0, was 2.7 cm in diameter. These detectors were made from germanium crystals grown at LBL. Detector 285-1.0 had previously been irradiated with 16.4 MeV neutrons and subsequently annealed to restore its spectrometer performance.[4] Resolutions for the 1332 keV ^{60}Co line, measured in the experimental configuration at the ZGS prior to the start of the irradiation, were 2.3 and 2.6 keV for 285-1.0 (2500 V bias, Ortec 452 amplifier, 2 μs shaping time) and 494-3.1 (2000 V bias, Tennelec TC200 amplifier, 1.6 μs shaping time), respectively. This two-detector cryostat and the associated preamplifiers were not designed for ultra high-resolution performance; when these detectors were tested in a better system they exhibited resolutions of about 1.8 keV for the 1332 keV line. The preamplifiers and amplifiers were located in the beam cave and long cables ran to the pulse height analyzer in the counting area.

The beam intensity, which ranged from 2×10^5 to 8×10^5 protons per pulse during the series of irradiation steps, was measured with two plastic scintillator paddles. The paddles (7.6 cm wide x 8.9 cm high) and detectors were oriented as shown in Fig. 1. The proton pulses were essentially flat-topped, extended 700 ms and recurred at 2.5 s intervals. The beam was tuned to an oval shape approximately 5 cm high and 3 cm wide. Unexposed Polaroid film was taped to the cryostat to monitor the beam shape and position during each irradiation. Measurements of the spatial distribution showed that about 80% of the flux traversed the two detectors.

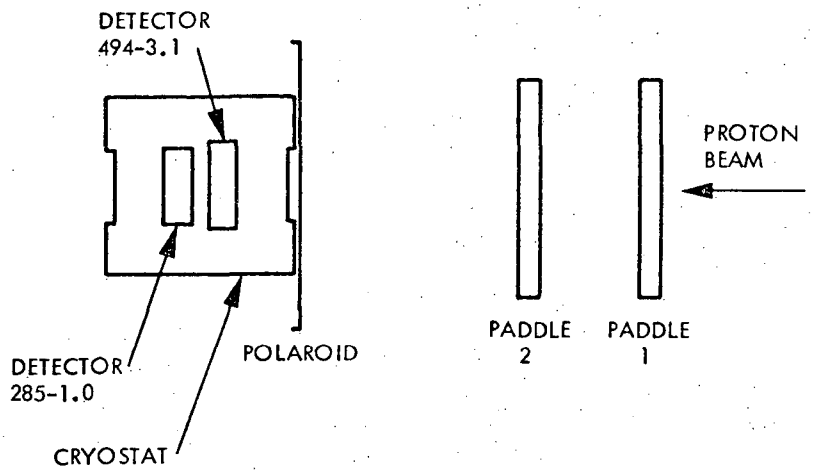

Fig. 1 Orientation of the detectors and monitoring paddles in the proton beam. The distance between the detectors and the paddles was small compared to the distance between the paddles and the beam focus.

Table I

FLUX (protons/cm^2)	RESOLUTION OF ^{60}Co 1332 keV LINE					
	285-1.0			494-3.1		
	2500 V FWHM(keV)	2000 V FWHM(keV)	1500 V FWHM(keV)	2000 V FWHM(keV)	1500 V FWHM(keV)	1000 V FWHM(keV)
0	2.3	2.4	2.8	2.6	2.7	2.8
7.3×10^7	2.6	2.6	2.9	3.2	3.4	3.4
1.9×10^8	3.6	4.3	4.7	5.3	5.5	5.7
2.6×10^8	3.9	4.7	5.2	6.8	7.2	8.8
5.7×10^8	8.5	9.1	12.0	13.6	17.8	22.6

The experimental plan was to irradiate the detectors with a given proton flux, then measure the resolution of the gamma-ray lines from a ^{60}Co source. Then a further irradiation would be made, and the resolution measured again. This procedure was followed until the detector resolutions were degraded to the point of negligible usefulness as gamma-ray spectrometers.

Results of Irradiation

Table I and Fig. 2 convey the crux of the measurements, giving the effect of proton flux on the FWHM resolution of both detectors. After receiving a cumulative flux of 5.7×10^8 protons/cm^2, the detector resolutions had degraded to 8.5 keV for detector 285-1.0 and 13.6 keV for detector 494-3.1. Although FWHM is a convenient expression to tabulate, when charge trapping becomes severe it is more meaningful to observe the spectral response over an extended energy region. Figure 3 demonstrates the change in the shape of the spectrometer response as a function of flux.

These results indicate that there exists a significant range of proton damage sensitivity between different high-purity germanium detectors. This observation is consistent with the finding of a wide range of fast neutron damage sensitivities (factor of ten) among detectors made from different germanium crystals.[4] Unfortunately, the crystal parameter(s) which is responsible for this wide range of damage sensitivities is still not known.

Annealing

When a germanium detector has been damaged by charged particles to the point where it is no longer deemed useful, one is faced with the question--can the detector be easily repaired? Although little basic physical knowledge of the recovery process is clearly understood, the results reported here allow us to answer the question affirmatively. To illustrate the general annealing behavior, data on detector 285-1.0 will be presented; detector 494-3.1 exhibited a similar annealing pattern.

To evaluate the extent of annealing at nominal operating temperature, the resolutions were measured over a period of two months, the detectors being continually maintained at 88°K. No significant change occurred during this time.

Also prior to heating the detectors to temperatures at which significant annealing was expected to take place, a "low temperature annealing" study was undertaken. This study was motivated by the fact that the space application with which we are most directly concerned will involve cooling by a passive

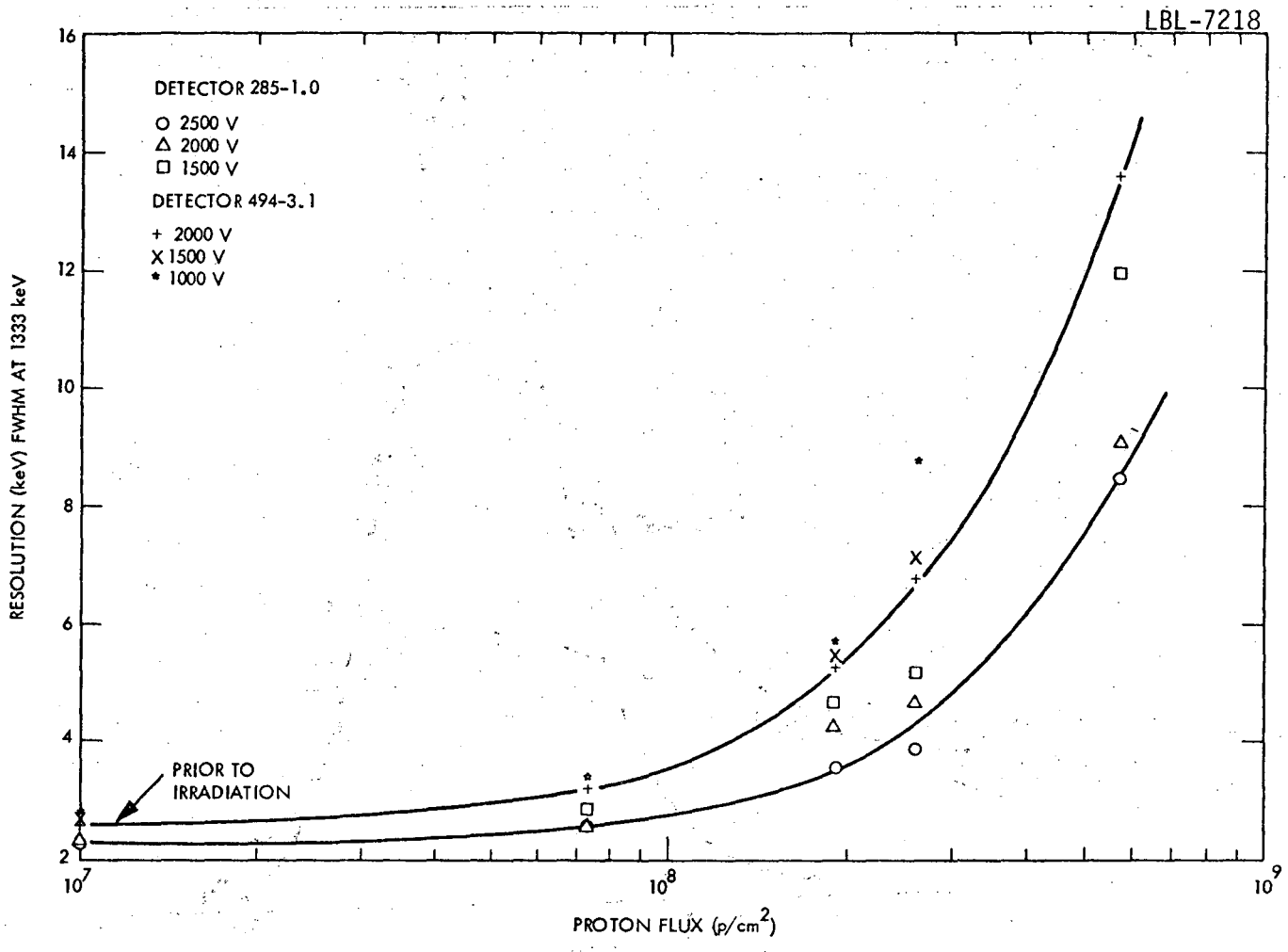

Fig. 2 Effect of proton flux on energy resolution. Each detector was measured at 3 bias values. Electronic noise has not been subtracted but contributes a constant value of 1.8 keV to 285-1.0 and 2.2 keV to 494-3.1.

radiator to a detector temperature of 120°K, rather than the colder temperatures obtained in liquid nitrogen-cooled cryostats.

To vary the detector temperature above LN_2 for the low temperature annealing, a device was built to direct evaporating N_2 gas into the emptied LN_2 reservoir of the dewar. A thermocouple mounted in the dewar against the cold finger controlled the flow of N_2 gas to maintain the desired temperature. When the thermocouple temperature was raised to 120°K (corresponding to detector temperature about 131°K) the resolution suffered a remarkably abrupt and severe degradation within 20 minutes, so bad that no full-energy peak was visible in the spectrum; just the leading edge of the maximum energy deposited could be seen. (This degradation may have occurred even more quickly and at a lower temperature. Unfortunately, measurements at smaller temperature and time increments were not initially made because such dramatic changes were not anticipated.) No further significant change occurred when the detectors were left at this temperature for 25 hours. This was followed by 5 hours at a detector temperature of 160°K. Then the dewar was filled with LN_2 and the detectors allowed to come to thermal equilibrium. At this point the resolution of detector 285-1.0 at 1332 keV was 19 keV. Figure 4 compares the spectra of detector 285-1.0 prior to, during and after the first low-temperature anneal. An additional week at nominal operating temperature resulted in no significant change in detector resolution.

Another series of low temperature anneals was then undertaken. In the first cycle, the detector temperature was raised to 106°K for 2 hours then returned to 88°K. In the second cycle, the temperature was raised to 125°K for 1.5 hours, then returned to 88°K. In the final cycle, the temperature was raised to 125°K (1.5 hours duration), 137°K(1 hour), 154°K(1 hour), 190°K(1 hour), and returned to 88°K.

The response of the detectors to the low-temperature anneal cycles was noteworthy in two respects. At each equilibrium temperature above the nominal 88°K, the collapse in energy resolution was essentially total, but after each cycle, the energy resolution was restored to approximately 20 keV when the detector temperature returned to 88°K. Secondly, the energy resolution measured at 88°K showed marked degradation after the first cycle, but negligible degradation thereafter, though the detector temperature was taken as high as 190°K. An explanation of these results in terms of microscopic detail

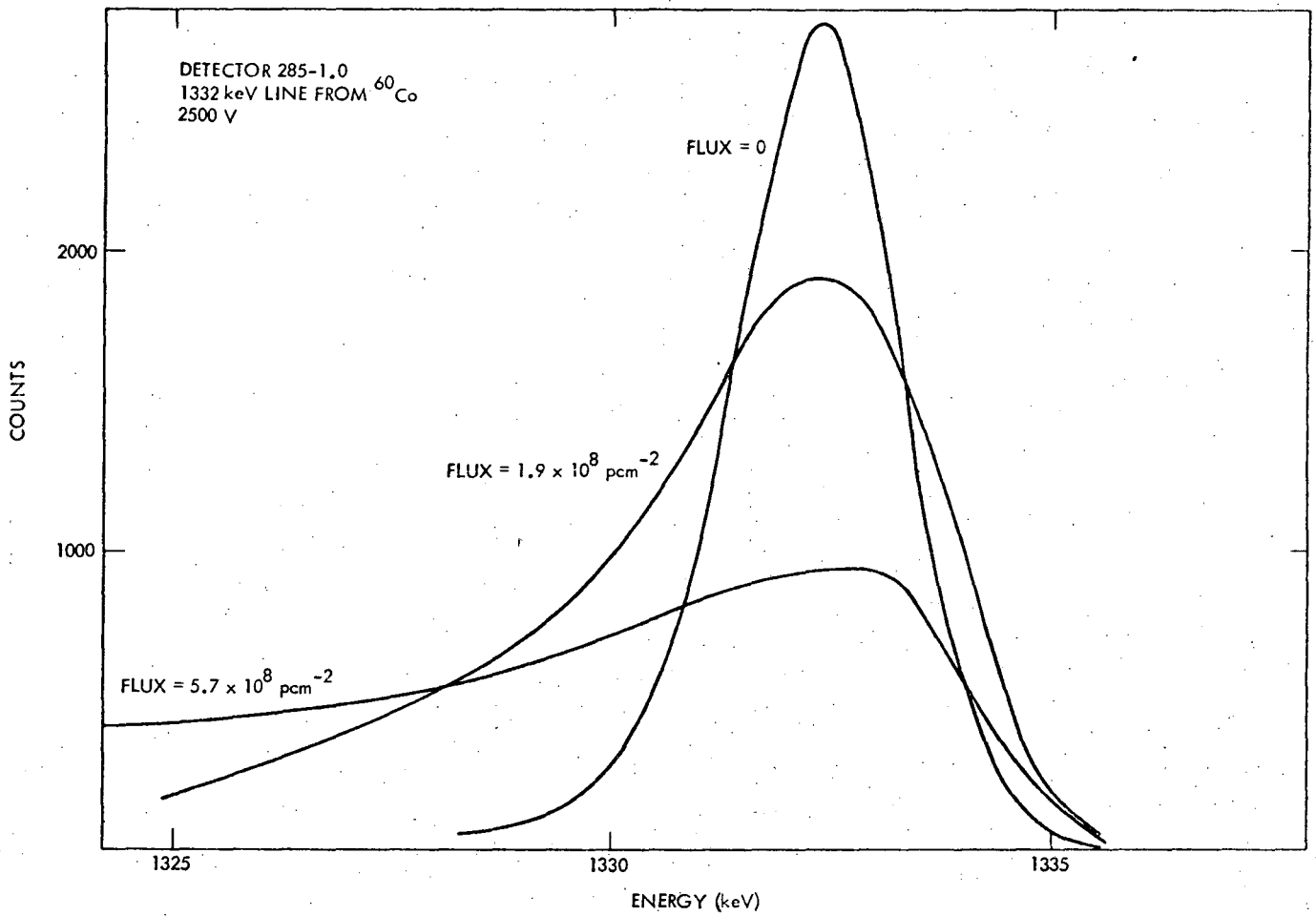

Fig. 3 Change in peak shape with proton flux for detector 285-1.0. The behavior of detector 494-3.1 was similar.

is beyond the scope of this paper and our present knowledge of the dynamics of collision-produced vacancies and defect clusters.

The degradation in resolution when the detector temperature was raised indicates that the effect of radiation damage will be much more pronounced if the detector is operated above the temperature of LN_2. Furthermore, the resolution degradation from 8 keV to 19 keV caused by the first low-temperature anneal cycle should serve as a warning against thermal cycling any germanium detector that has been exposed to a significant charged-particle flux even if the detector has not shown any degradation.

The detectors were then annealed at progressively higher temperatures. Figure 5 outlines the spectrometer performance of detector 285-1.0 as a function of annealing treatment. ^{60}Co spectra at various stages of annealing are presented. The peaks have been displaced and the energy scale varied for clarity of illustration; peak position is not relevant.

Stage 1

This represents the situation at the completion of the "low temperature annealing."

Stage 2

This represents the situation after the detector had been annealed at room temperature for 17 hours. The resolution has improved from 19 keV to 10 keV with a corresponding decrease in the tailing. In light of the annealing results following the 16.4 MeV neutron irradiation of detector 285-1.0, this is a surprisingly large improvement for such a short length of time at this temperature.

Stage 3

An additional 160 hours at room temperature had now elapsed. Again we observe a surprisingly large improvement to a FWHM of 7 keV.

Stage 4

An additional 32 hours at room temperature, 66 hours between 55°C and almost 100°C, and 12 hours at almost 100°C had now transpired. Once more we observe a surprisingly large improvement to a FWHM of 3.0 keV. For many applications this detector would now be considered acceptable, especially if significant overvoltage could be applied.

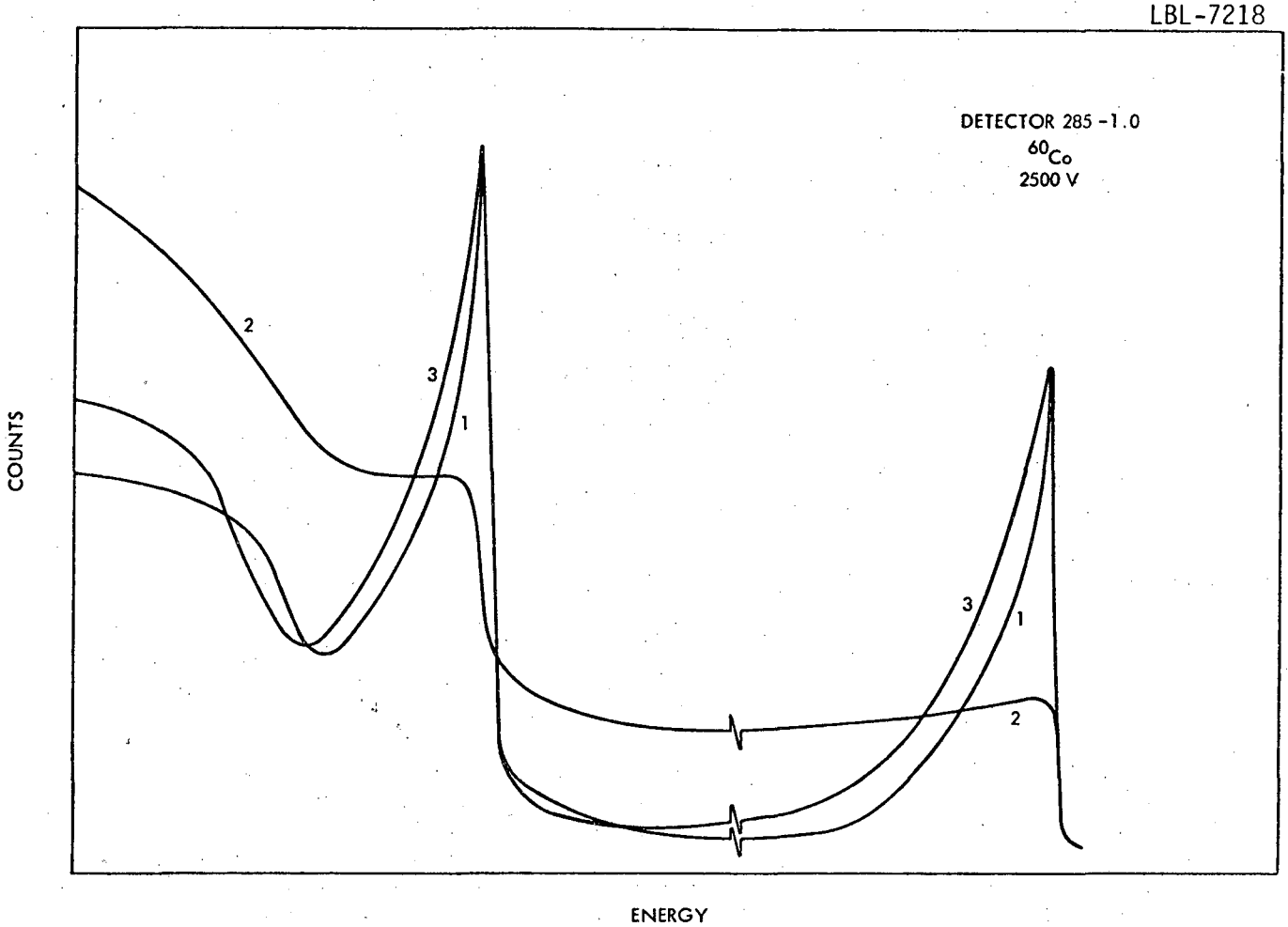

Fig. 4 ^{60}Co spectra of detector 285-1.0 before (1), during (2), and after (3), the first low temperature anneal cycle. Spectra 1 and 3 were obtained with the detector at its nominal temperature of 88°K, spectrum 2 at 131°K. Each spectrum was obtained using slightly different gain settings; note the break in the energy scale.

Stage 5

An additional 48 hours between 50°C and 85°C, 144 hours at 85°C and 120 hours at 100°C had now elapsed. When the detectors were cooled after this anneal cycle the leakage current increased significantly, consequently we were unable to go to as high a bias as previously but there clearly had been another improvement.

Up to this point the detector had remained in the same cryostat throughout all the annealing cycles. After being left in this cryostat at room temperature for another 204 days the detectors were removed, given a new surface treatment, and mounted into a cryostat with a preamplifier capable of higher resolution. The overall detector performance was approximately the same as prior to the proton irradiation.

Stage 6

The detector was annealed at 150°C for 50 hours, and now exhibits a spectrometer performance equal to the best previously observed for this detector, 1.8 keV. (Immediately prior to the proton irradiation the spectrometer performance of detector 285-1.0 was slightly inferior to its best.)

From these data one can conclude that in-situ annealing of high-purity germanium spectrometers after proton-induced damage is possible. Although complete recovery may not be obtainable with annealing in the range of 100°C, the spectrometer performance should be acceptable for nearly all applications.

Proton-Neutron Comparison

Detector 285-1.0 had previously been irradiated with 16.4 MeV neutrons and subsequently annealed to restore its spectrometer performance.[4] Thus a direct comparison between proton and neutron damage sensitivity can be made. Likewise, a direct comparison can be made of the annealing characteristics of a germanium detector following proton and neutron irradiation. In light of the wide range of radiation damage sensitivities among detectors made from different crystals, these proton-neutron comparisons should not be made between detectors fabricated from different crystals.

After detector 285-1.0 had been irradiated by a fluence of 10^{10} neutrons/cm^2, the resolution of the 1332 keV line of ^{60}Co was 3.5 keV. As shown in Fig. 2

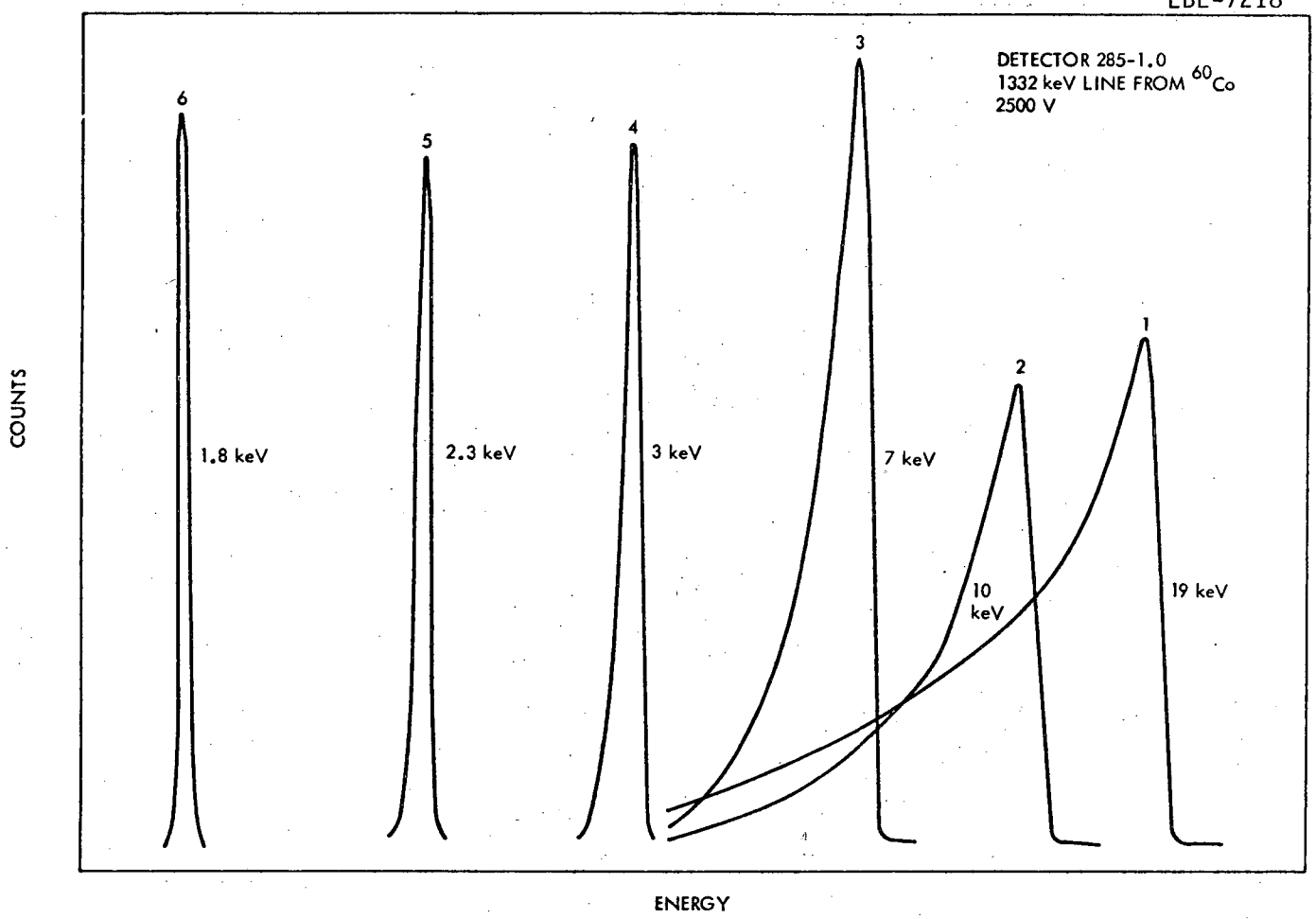

Fig. 5 ^{60}Co spectra at various stages of annealing following the proton irradiation of detector 285-1.0. The number on each curve refers to the stage listed in the text. No relevance should be attached to the peak positions since the peaks have been displaced and the energy scale varied to clarify the observation of peak shapes.

an equivalent resolution was measured when the proton flux was only 1.5×10^8 protons/cm^2. These data indicate that 5.1 GeV protons cause about 60 times more damage than 16.4 MeV neutrons. This difference is roughly consistent with the calculated number of lattice defects expected.

Although the spectrometer performance of detector 285-1.0 was considerably worse following the proton irradiation than it was following the neutron irradiation--8.5 keV vs 3.5 keV--the detector annealed far more easily, indicating a different damage mechanism for at least a large part of the damage.

For comparison Figs. 6a and 6b outline the spectrometer performance of detector 285-1.0 as a function of annealing treatment following the neutron irradiation. ^{60}Co spectra at various stages of annealing are presented. Again the peaks have been displaced and the energy scale varied for clarity of illustration; peak position is not relevant.

Stage 1

This represents the situation after being irradiated by 16.4 MeV neutrons to a fluence of 10^{10} neutrons/cm^2; the detector had remained at its normal operating temperature, a few degrees above LN_2 temperature, for 5 days before these measurements were made. This detector appeared to improve slightly during LN_2 anneal. However, this is not a characteristic of all neutron-irradiated detectors.[4]

Stage 2

This represents the situation after the detector had been annealed at dry ice temperature for about 15 hours. No peak from ^{60}Co can be seen--or even imagined. A substantial increase in the acceptor concentration was determined from a measurement of detector capacitance, as was found for all the neutron-irradiated detectors. A scan with collimated ^{241}Am 60 keV gamma rays from contact to contact revealed severe hole trapping.

Stage 3

At this point the detector had been annealed at dry ice temperature an additional 43 hours. The ^{60}Co

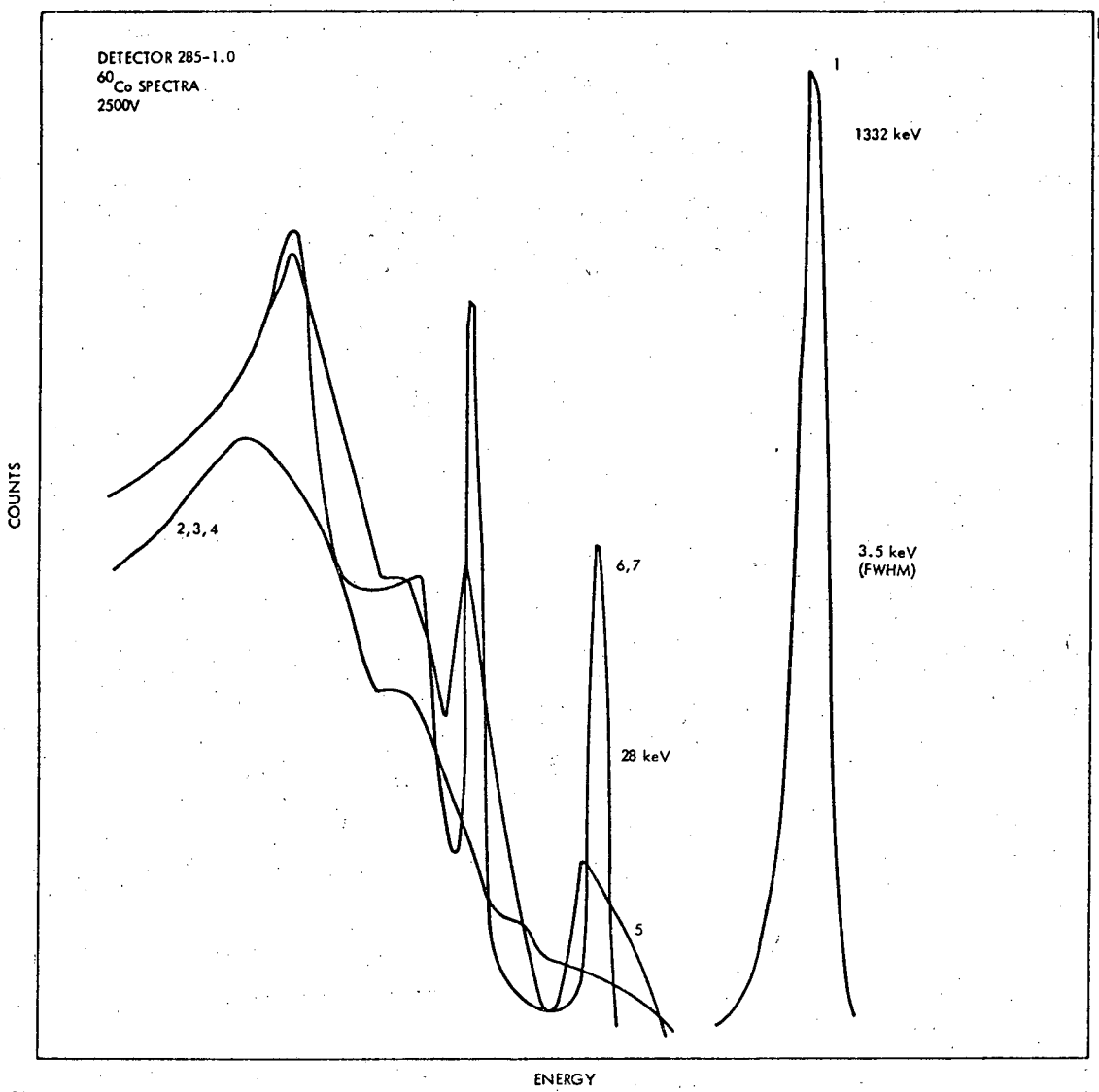

Fig. 6a ^{60}Co spectra at various stages of annealing following the neutron irradiation of detector 285-1.0. The number on each curve refers to the stage listed in the text. No relevance should be attached to the peak positions since the peaks have been displaced and the energy scale varied to clarify the observation of peak shapes.

spectrum appears to be identical to that observed following the first anneal at dry ice temperature. The acceptor concentration did not change measurably during these additional 43 hours at dry ice temperature.

Stage 4

The detector had now gone through a brief room temperature anneal; about 30 minutes at room temperature in addition to the warm up and cool down periods. No change in the ^{60}Co spectrum was apparent, and the acceptor concentration had not changed measurably.

Stage 5

An additional 69 hours at room temperature had now transpired. Although still very poor, the spectrometer performance is markedly improved. The acceptor concentration had decreased significantly. To fully anneal this detector at room temperature in a reasonable time, if ever, is clearly not feasible.

Stage 6

This represents the situation after a "boiling water" anneal that resulted in the detector spending 40 hours at 80°C, 12 hours at 70°C, 12 hours at 50°C, and 17 additional hours at room temperature. Although still poor, the spectrometer has shown marked improvement again, and the acceptor concentration has also decreased significantly once more.

Stage 7

The detector system was transported warm from Brookhaven to Berkeley, and an additional 8 days at room temperature elapsed. This lengthy room temperature anneal produced negligible change in either the spectrometer performance or acceptor concentration.

Stage 8

The detector was annealed in a furnace at 100°C for 22 hours prior to these measurements. Marked improvement in spectrometer performance is observed, and the acceptor concentration has also decreased once more.

Stage 9

An additional 69.5 hours of annealing at 100°C

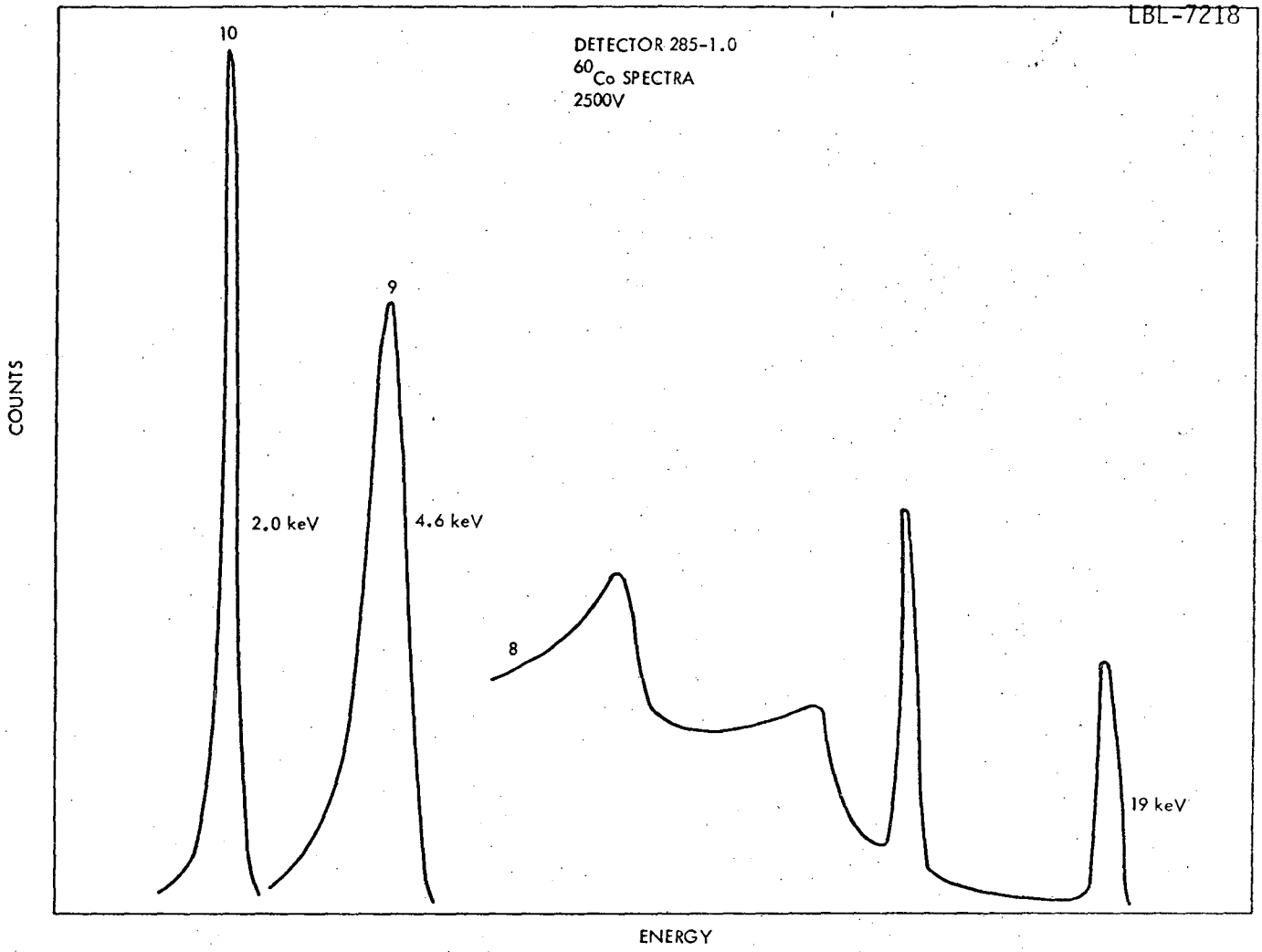

Fig. 6b ^{60}Co spectra at various stages of annealing following the neutron irradiation of detector 285-1.0. The number on each curve refers to the stage listed in the text. No relevance should be attached to the peak positions since the peaks have been displaced and the energy scale varied to clarify the observation of peak shapes.

had now transpired. The spectrometer performance continues to improve significantly, and the acceptor concentration continues to decrease.

Stage 10

An additional 164.5 hours of annealing at 100°C, an accumulated total of 256 hours had now transpired. The detector is now acceptable for nearly all applications although the spectrometer performance is not yet equal to the initial quality. As expected, the degradation relative to initial conditions is seen especially at lower bias, and when one looks carefully at peak symmetry.

After remaining at room temperature for an additional 31 months, the detector was mounted into the cryostat used for the proton irradiation. The dramatic difference between the peak shapes observed during the annealing following neutron irradiation compared to those observed during the annealing following proton irradiation of this detector must be noted. Figure 7 compares the peak shapes when the resolution (FWHM) is equal. The relatively square-topped peak exhibited by the neutron-irradiated detector corresponds to the preferential trapping of a single carrier. At no time during the annealing of the proton damaged detectors was anything resembling a square-topped peak observed. These observations provide additional evidence that a different damage mechanism must account for at least a large part of the proton damage.

Conclusion

We have conducted quantitative studies of high-energy proton radiation damage and subsequent annealing of high-purity germanium gamma-ray detectors. These are the first studies in which high-energy proton bombardments have been used to simulate the effects of cosmic rays on the operation of germanium gamma-ray detectors in space. The threshold for resolution degradation was found to be lower in the case of proton bombardment compared to neutron irradiation, but the detectors were annealed more easily. This is encouraging for the development of spectrometers for extended space missions because annealing high-purity germanium detectors in space appears feasible. Current and projected experiments will extend these measurements to coaxial detectors and include the parameters of detector temperature, irradiation rate, electrode configuration, and particle charge and energy.

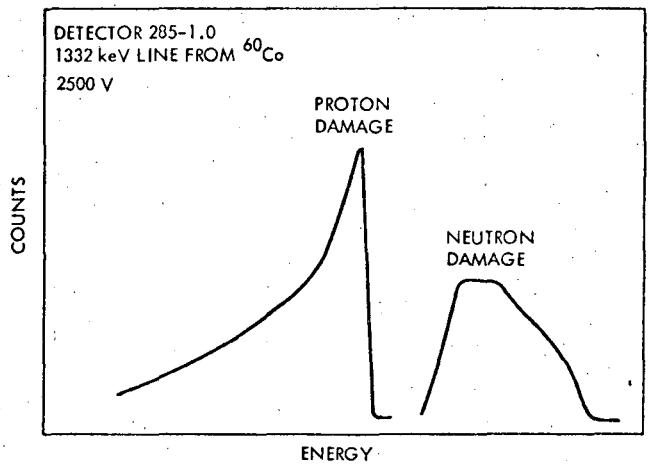

Fig. 7 Comparison between the peak shapes observed during the annealing following neutron and proton irradiation. The resolution of both peaks is 19 keV (FWHM).

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