## PRESENCE OF DEPLETED ZONES IN PLATINUM\*

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In the bombardment of materials with heavy particles a large amount of energy can be deposited in a very small region by a primary knock-on atom, and the local atomic arrangement can be thereby drastically disrupted.<sup>1</sup> Various measurements of physical properties of such irradiated materials indicate the presence of distributions of defects which are removed in a steplike manner by annealing.<sup>2</sup> One of the more interesting physical property changes accompanying fast-particle irradiation is the attendant change in mechanical properties of irradiated crystals.<sup>3,4</sup> The defect which is responsible for the mechanical property changes of irradiated crystals is only removed at high temperatures, temperatures corresponding to self-diffusion.<sup>4</sup> This observation, as well as others, has led to the model of a depleted zone<sup>5</sup> as being responsible for the changes of mechanical properties of irradiated crystals. A depleted zone is envisioned as a region of crystal where a high local concentration of point defects exists - a belt of interstitials surrounding a multiply connected complex of vacancy clusters.<sup>5</sup> We would like to present here some evidence which lends support to the existence of such defects.

The experimental technique used to observe directly the irradiated material was field-ion microscopy, which has been extensively described elsewhere.<sup>6</sup> The irradiated platinum (99.999% pure) was annealed prior to irradiation at 1300°C by resistively heating in vacuo of  $<10^{-7}$  Torr. This material was examined, prior to irradiation, in a field-ion microscope<sup>7</sup> operated at 4.2°K in order to characterize the material. The annealed wire was then irradiated to an integrated neutron flux of  $1 \times 10^{18} nvt$  (E > 1.4 MeV), and this wire examined in the field-ion microscope. Repeated examination of such irradiated materials revealed the presence of large disturbances of the lattice which are apparently introduced by isolated damage events. A micrograph of one is shown in Fig. 1.

As stated above, a large number of specimens in the unirradiated condition were examined to characterize the material. From these examinations it can be stated that no such disturbances of the atomic arrangement are observed in well-annealed platinum. Furthermore, the specific effect of impurities<sup>6</sup> as well as other damage products<sup>8</sup> have been previously observed, and they are distinctly different from those reported here.

We would like to present two other pieces of evidence that indicate that these disturbances of the regularity of the lattice are strikingly similar to Seeger's depleted zone.<sup>5</sup> Firstly, by careful field evaporation of one atomic layer at a time, the spatial distribution of a dis-



FIG. 1. Micrograph of a disturbed region in Pt irradiated to  $10^{18}$  *nvt*, E > 1.4 MeV.



FIG. 2. The spatial distribution of defects in a disturbed region in irradiated Pt. Circle: vacancy. Square: interstitial.

turbed region could be mapped out, Fig. 2, from which it can be seen that we are dealing with a multiply connected region in the crystal. This is quite similar to the computer-simulated damage distribution calculated by Beeler.<sup>9</sup> Secondly, the examination of a large number of irradiated specimens allowed us to establish a density for such disturbed regions in order to compare the results with the density predicted theoretically. This calculation predicts  $\sim 5 \times 10^{16}$  disturbed regions/cm<sup>3</sup>,<sup>10</sup> while our measured density is approximately  $6 \times 10^{15}$ disturbed regions/cm<sup>3</sup>.

Another interesting aspect of these observations is the detailed shape of the depleted zones in the irradiated platinum. The shapes of a few individual disturbed regions were specifically examined, by field evaporation, to determine if the disturbed region had any characteristic anisotropy of shape. Within the limits of the field-evaporation technique, no such anisotropy of shape was observed. The exterior shape of the zone was generally found to be highly irregular, and the volume constituting the zone was found to be multiply connected. This is roughly what one expects from the depleted-zone model.

Finally, in Fig. 3, we present a measured size distribution of disturbed regions in the irradiated platinum. As shown, there is a smooth decrease in the number of disturbed regions



FIG. 3. (a) Measured size distribution of disturbed regions in Pt. (b) Plot of number of disturbed regions versus reciprocal of the volume of the region.

with increasing size of the disturbed region. In the measurements of the size of any individual disturbed region, the maximum size was obtained from field evaporation through the whole disturbed region. The maximum size we have used is the largest diameter across the disturbed region being measured.

From this measurement an additional argument can be made. It is expected that the energy distribution of the neutrons used in this experiment (the Brookhaven graphite reactor —hole E7) would approximate a 1/E distribution of neutron-fission spectrum. If we accordingly assume that the volume of the cluster is proportional to the energy of the neutrons used, we should get a linear plot of the number of disturbed regions versus 1/volume. This is shown in Fig. 3(b).

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<sup>&</sup>lt;sup>1</sup>An extensive review of the theoretical aspects of

this problem have been given by D. K. Holmes in <u>The</u> <u>Interaction of Radiation with Solids</u>, edited by R. Strumane, J. Nihoul, R. Gevers, and S. Amelinckx (North-Holland Publishing Company, Amsterdam, 1964).

<sup>2</sup>H. G. Van Bueren, <u>Imperfections in Crystals</u> (North-Holland Publishing Company, Amsterdam, 1960).

<sup>3</sup>T. H. Blewitt, R. R. Coltman, R. W. Jamison, and J. K. Redman, J. Nucl. Mater. <u>2</u>, 277 (1960).

<sup>4</sup>J. Diehl in <u>Radiation Damage in Solids</u> (International Atomic Energy Agency, Vienna, 1962), Vol. 1, p. 129.

<sup>5</sup>A. K. Seeger, in <u>Proceedings of the Second United</u> <u>Nations International Conference on Peaceful Uses of</u> Atomic Energy (United Nations, Geneva, Switzerland, 1958), Vol. 6, p. 250.

<sup>6</sup>E. W. Müller, <u>Advances in Electronics and Electron</u> <u>Physics</u> (Academic Press, Inc., New York, 1960), Vol. 13, p. 83.

<sup>7</sup>M. J. Attardo and J. M. Galligan, to be published.

 $^{8}\mathrm{M.}$  J. Attardo and J. M. Galligan, Phys. Rev. Letters <u>16</u>, 641 (1965).

<sup>9</sup>J. R. Beeler, to be published.

<sup>10</sup>G. Leibfried in <u>The Interaction of Radiation with Sol-</u> <u>ids</u>, edited by R. Strumane, J. Nihoul, R. Gevers, and S. Amelinckx (North Holland Publishing Company, Amsterdam, 1964).

## BRILLOUIN-SCATTERING DISPERSION IN FERROELECTRIC TRIGLYCINE SULFATE

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Dispersion has been observed in the Brillouin components of the light-scattering spectrum of triglycine sulfate. It is attributed to the Landau-Khalatnikov relaxation of fluctuations in the spontaneous polarization.

We have observed the light-scattering spectrum of triglycine sulfate (TGS) in the temperature range 34-54°C and found an anomalous change in the shifts of both the longitudinal and transverse Brillouin components. TGS is a transparent monoclinic crystal which is ferroelectric at room temperature and undergoes a second-order phase transition to a nonpolar phase at the Curie temperature  $(T_c \approx 49^{\circ}\text{C})$ .<sup>1</sup> In the neighborhood of  $T_c$ , both the magnitude and the relaxation time of fluctuations in the spontaneous polarization  $\vec{P}$  become very large. According to Landau and Khalatnikov, the relaxation time  $\tau$  is given by<sup>2</sup>

$$\tau = A (T_c - T)^{-1}$$
 sec. (1)

Thus, if phonons can couple to polarization, a temperature-dependent relaxation is expected, which will produce a dispersion in the phonon velocity of the form<sup>3</sup>

$$V^{2} = V_{\infty}^{2} - (V_{\infty}^{2} - V_{0}^{2}) / [1 + (\omega \tau)^{2}].$$
 (2)

The ultrasonic attenuation produced by this relaxation has been studied by O'Brien and Litovitz in the frequency range 15 to 125 MHz.<sup>4</sup> Their measurements give a temperature dependence for the relaxation time which fits Eq. (1) with  $A = 2.25 \times 10^{-10}$  sec °C. Since in our experiment the Brillouin shifts are proportional to the phonon velocity, we believe that the changes in the Brillouin shifts which we have observed are due to the L-K relaxation.

Our apparatus, which has been described elsewhere,<sup>5</sup> employs a Spectra-Physics model 125 He-Ne laser (approximately 80 mW at  $\lambda$ 6328) as the light source, and a pressure-swept Fabry-Perot interferometer with photoelectric detection for spectral analysis of the scattered light. The sample is placed in a large paraffin-oil bath which acts as the thermostat and also matches the appropriate refractive index of the sample, thus eliminating scattering from the crystal surfaces and refraction of the beam. Silver electrodes painted onto the *b* faces permit the sample to be electrically polarized between runs.

A typical spectrum is shown in Fig. 1. The measured Brillouin shifts for longitudinal and transverse phonons are shown in Fig. 2 for scattering angles of 90 and 135°, i.e., for two different phonon wavelengths. The orientation in the crystal of the scattering vector  $\mathbf{q}$  was the same for the two scattering angles and is shown in the inset in Fig. 1. The scattering plane was parallel to the crystal (010) plane, and the polarization of the incident and scattered light was perpendicular to this plane. The transverse phonon giving the Brillouin component is polarized in the (010) plane. The refractive



FIG. 1. Micrograph of a disturbed region in Pt irradiated to  $10^{18} nvt$ , E > 1.4 MeV.