

FIG. 3. Temperature dependence of acousto-electric current.

Region 2 (15°K to 30°K). – The remarkably constant current in this range presumably results from a balance between  $e\varphi_0/kT$  decreasing with increasing temperature and the mobility increasing. <u>Region 3 (above 30°K)</u>. – The decrease in current is caused largely by an increase in  $\alpha_L$  due to the attenuation of the sound by thermal phonons.<sup>4</sup>

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## **RECOILLESS GAMMA-RAY EMISSION AFTER ALPHA DECAY\***

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Recoilless gamma rays are emitted after  $\alpha$  decay. We report the results of Mössbauer absorption experiments with the 59.6-keV gamma ray of Np<sup>237</sup> following both  $\alpha$  decay of Am<sup>241</sup> and  $\beta$  decay of U<sup>237</sup>.

The 59.6-keV gamma transition in Np<sup>237</sup> was chosen as a promising first candidate for Mössbauer absorption studies in the actinide region. The 59.6-keV nuclear level,<sup>1</sup> which has spin  $\frac{5}{2}^{-}$ and a half-life of  $6.3 \times 10^{-8}$  sec, decays by an E1 gamma transition to the ground state, which has spin  $\frac{5}{2}^{+}$ . The total internal conversion coefficient<sup>2</sup> for the transition is 1.0. The maximum resonance cross section,<sup>3</sup>  $\sigma_0$ , is  $0.34 \times 10^{-18}$  cm<sup>2</sup>. The 59.6keV level and the ground state of Np<sup>237</sup> are described by the same collective-model quantum numbers<sup>4</sup> as are the 26-keV level and the ground state of Dy<sup>161</sup>, for which the Mössbauer effect is known.<sup>5</sup>

In our experiments cubic NpO<sub>2</sub> was used both as the absorber and as the host lattice for the sources. A single thick polycrystalline absorber (250 mg/cm<sup>2</sup>) and two polycrystalline sources, each about 20 mg/cm<sup>2</sup>, were prepared. The sources consisted of  $Am^{241}$  and  $U^{237}$  as 1% solid solutions of AmO<sub>2</sub> and UO<sub>2</sub>, respectively, in isomorphous<sup>6</sup> NpO<sub>2</sub>. The 6.75-day  $U^{237}$  activity was obtained by neutron irradiation and subsequent ion-exchange separation of  $U_3O_8$  highly enriched in  $U^{236}$ .

Experiments were performed with a loudspeaker-type velocity spectrometer similar to that reported by Lynch and Baumgardner,<sup>7</sup> modified for cryogenic work. The system was calibrated with the Fe<sup>57</sup> Mössbauer effect. A Xe-filled proportional counter was used as the gamma-ray detector. Best resolution was obtained by counting the Xe escape peak of the 59.6-keV gamma radiation.

The velocity spectrum obtained for the absorber and  $Am^{241}$  source at 77°K is shown in Fig. 1(a). The resonance-absorption effect obtained after  $\alpha$  decay may be compared with the larger effects observed after  $\beta$  decay of U<sup>237</sup> with the source and absorber at 77°K [Fig. 1(b)] and at 4.2°K [Fig. 1(c)]. An experiment was also performed with the absorber and  $Am^{241}$  source at 4.2°K; a broad velocity spectrum similar to that of Fig. 1(c) was obtained, but with only a 1% change in count rate at zero velocity. In each case the

<sup>\*</sup>Most of this work was performed during a visit to the IBM Watson Research Center, Yorktown Heights, New York.



FIG. 1. Mössbauer absorption spectra of the 59.6-keV gamma ray of  $Np^{237}$  with a  $NpO_2$  absorber (250 mg/cm<sup>2</sup>). Sources were  $Am^{241}$  or  $U^{237}$  in  $NpO_2$ . For each velocity spectrum both source and absorber were at the temperature indicated.

absorption width is considerably greater than twice the natural linewidth  $(2\Gamma \approx 0.07 \text{ mm/sec})$ ; it should be noted that among the line-broadening mechanisms present are resonant self-absorption in the source and use of a very thick absorber.<sup>8</sup> Suggestions of poorly resolved hyperfine structure are apparent in the spectra; broad unresolved spectra were also observed in early work on dysprosium oxide.<sup>5</sup> A wide-range velocity spectrum for the absorber and U<sup>237</sup> source at 4.2°K is shown in Fig. 2.

The data shown in the figures are uncorrected



FIG. 2. Mössbauer absorption spectrum of the 59.6-keV gamma ray of  $Np^{237}$  at 4.2°K. The source was  $U^{237}$  in  $NpO_2$ .

for background. We estimate the ratio of pertinent counts to total counts to be 1:2 for Am<sup>241</sup> runs and 1:3 for U<sup>237</sup> runs. The result of such corrections would be to further accentuate the depth of the absorption resonance following  $\beta$ decay with respect to that following  $\alpha$  decay.

The increased width of the spectra at  $4.2^{\circ}$ K relative to that at 77°K is probably related to magnetic hyperfine splitting due to the onset of antiferromagnetism in NpO<sub>2</sub> at 25°K, as indicated by heat-capacity data.<sup>9</sup> The resonance area is larger at the lower temperature; this is consistent with the usual behavior of recoilless fractions.

Recoil energy due to  $\alpha$  decay of Am<sup>241</sup> is about 93 keV, which is sufficient to remove the recoiling atom from its lattice site and to disrupt local crystalline binding forces.<sup>10</sup> Very little is known about lattice dynamics following  $\alpha$  decay; thus, the probability of subsequent recoilless gammaray emission can not as yet be accurately described. One would expect the energetic recoil to greatly reduce the recoilless emission fraction, yet the observed Mössbauer absorption is a large fraction of that obtained after  $\beta$  decay. The existence of a sizable resonance absorption after  $\alpha$  decay offers evidence that the  $\alpha$ -recoil stopping time and the dielectric relaxation time of NpO<sub>2</sub> are short compared to the lifetime of the 59.6keV level of Np<sup>237</sup>. Further study of the Mössbauer effect after  $\alpha$  decay will lead to a better understanding of the dynamics of crystalline lattices undergoing radiation damage.

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## NEW VALUE FOR THE FINE-STRUCTURE CONSTANT $\alpha$ FROM MUONIUM HYPERFINE STRUCTURE INTERVAL\*

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We have remeasured the ground-state hyperfine structure interval,  $\Delta \nu_M$ , of muonium ( $\mu^+e^-$ ) to a much higher precision than previously.<sup>1,2</sup> The result is important for obtaining an independent measurement of the fine-structure constant  $\alpha$ , and hence also for an understanding of proton structure.<sup>3</sup>

The experiment was very similar to that described previously.<sup>1</sup> The Nevis synchrocyclotron of Columbia University provided a longitudinally polarized beam of positive muons, which were stopped in high-pressure argon gas, and formed polarized muonium. A microwave-induced transition between the high-field states  $(m_J, m_{II}) = (\frac{1}{2}, \frac{1}{2})$ and  $(\frac{1}{2}, -\frac{1}{2})$  (*m<sub>J</sub>* and *m<sub>II</sub>* are the magnetic quantum numbers of the electron and the muon) changed the angular distribution of the decay positrons from the muons, notably near  $0^{\circ}$  (forward) and  $180^{\circ}$  (backward) where we observed the positrons. For fixed microwave frequency, the ratio of decay positrons to stopped muons, as a function of magnetic field, has a resonant behavior as shown in Fig. 1. A great increase in precision as compared to the original resonance experiment<sup>1</sup> was achieved by (1) obtaining much narrower resonance lines through use of a more homogeneous static magnetic field and low microwave power, (2) improving the counting statistics very substantially in a long experiment in which positrons were observed in both the forward and backward directions, (3) improving the reliability and stability of all components in the experiment.

The argon gas perturbs the muonium atoms and produces a fractional change in the measured value of  $\Delta \nu_M$  which is proportional to the argon density to first order. This density shift (or pressure shift) has been measured<sup>4</sup> accurately for the isotopes of hydrogen in argon at pressures up to 40 mm Hg. From the results of a run carried out by us in December 1962, with argon pressures of 68 atm and 35 atm, it was clear that the pressure shift was present; another run which extended the measurements to argon pressures as low as 10 atm was carried out in January and February 1964. The results of both runs are shown in Fig. 2. The errors in the individual points come about equally from statistical counting uncertainties and from uncertainties in the magnetic field measurements.

These points have been fitted to both a straight line and a parabola. The quadratic term was found to be very small, and consistent with zero, so all results are based on the linear fit. We found a fractional pressure shift of  $(-4.05\pm0.49)$  $\times 10^{-9}$  (mm Hg)<sup>-1</sup> at 0°C for muonium in argon, which is in good agreement with the corresponding values of  $(-4.77\pm0.12)\times10^{-9}$  for hydrogen,  $(-4.52\pm0.40)\times10^{-9}$  for deuterium, and  $(-5.05\pm0.15)\times10^{-9}$  for tritium in argon determined by optical pumping.<sup>4</sup> We obtain

 $\Delta \nu_{M}(\exp t) = 4463.15 \pm 0.06 \text{ Mc/sec} (\pm 13 \text{ ppm})$  (1)