Measurement of the Energy Loss of Germanium Atoms to Electrons in Germanium at Energies below 100 keV. II*

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The amount of energy lost in the production of hole-electron pairs by germanium atoms stopping in a germanium crystal has been measured at energies of 56.2 and 102.9 keV. Inelastic neutron scattering in a Ge(Li) gamma-ray detector was used to produce the 690-keV Ge⁷² state and the 596-keV Ge⁷⁴ state. The lines corresponding to the de-excitation of these states were broadened by summing with the hole-electron pairs produced by the recoiling Ge atom. A particular recoil energy was selected by requiring a coincidence with the outgoing inelastically scattered neutron, which was detected by a stilbene recoil counter placed at either 90° or 150° to the incident neutron beam. The results of the present experiment confirm a previous investigation by Chasman, Jones, and Ristinen and do not agree with the published results of Sattler, Vook, and Palms.

I. INTRODUCTION

W HEN an energetic germanium atom slows down and stops in a germanium crystal, energy is lost by atomic scattering and by electronic excitation. Measurements of the fraction of the energy lost to electronic excitation (production of electron-hole pairs) have been made by Sattler, Vook, and Palms¹ (SVP) and by Chasman, Jones, and Ristinen² (CJR). The results of the two experiments differ systematically by 15 to 25% in the energy range below 100 keV.

The techniques used in the two experiments were quite similar in that recoil germanium atoms were produced in a Ge(Li) gamma-ray detector by bombardment with fast neutrons. The detector is effectively a solid-state ionization chamber in which the number of collected hole-electron pairs (proportional to pulse height) is a measure of the energy lost by the stopping particle. CJR then observed the broadening of the 596.3 ± 1.0 gamma-ray line³ produced by inelastic scattering from Ge⁷⁴ and the broadening of the 690 ± 1 keV conversion electron line⁴ produced by inelastic scattering in Ge⁷². The lines are asymmetrically broadened because the ionization produced by the interaction of the gamma ray or electron in the germanium crystal is summed with the ionization produced by the recoil atom. In this method, the contribution from a single Ge isotope is uniquely identified by the energy of the radiation accompanying the recoil. There are several complications in the analysis of the data. In the case of the 596-keV

line, there is feeding of the state⁵ by a level in Ge⁷⁴ at 1.20 MeV which introduces a gamma ray at 0.60 MeV and a second lower energy recoil. For the 690-keV line in Ge⁷², feeding from higher states is less than 1% in the energy range covered, but the state has a half-life of 0.3 μ sec so that some care must be taken with the electronics to ensure that the energy of the conversion electron and recoil are summed. This can be done by using an integrating time constant which is long compared to the half-life of the level.

SVP observed the recoil spectrum directly by using a thin detector which was thick enough to stop the recoils, but which had a greatly reduced response to gamma rays. This technique has the disadvantage that it is difficult to distinguish clearly the recoils which are produced by elastic scattering from those produced by inelastic scattering.

The energy scale for the recoil atoms in both experiments was established by comparison with standard gamma and x-ray sources. The maximum recoil energy is known from kinematics, and hence the fraction of the recoil energy going into ionization can be found.

In their paper, SVP criticized at some length the published work of CJR. Certain points raised in their criticism were incorrectly inferred from the paper of CJR, but it is apparent that a salient difference between the work of the two groups of investigators concerns the method to be used to analyze a recoil spectrum. SVP contend that what they call the "knee" of the spectrum should be taken as the energy of the most energetic recoil. They define the "knee" to be the point toward the end of the recoil spectrum where the slope ideally should be zero. CJR took the half-maximum points of the broadened gamma or conversion electron line as that measure of the total broadening of the recoil spectrum, which allows satisfactorily for resolution effects.

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 [†] On leave from the University of Alberta, Edmonton, Canada.
 ¹ A. R. Sattler and J. M. Palms, Bull. Am. Phys. Soc. 10, 719 (1965); A. R. Sattler, F. L. Vook, and J. M. Palms, Phys. Rev. 143, 588 (1966).

² C. Chasman, K. W. Jones, and R. A. Ristinen, Phys. Rev. Letters 15, 245 (1965). The values quoted in this reference for ϵ and $\tilde{\eta}(\epsilon)$ are too large by a factor of $\sqrt{2}$ because of an error in the calculation of the atomic screening radius. See C. Chasman, K. W. Jones, and R. A. Ristinen, Phys. Rev. Letters 15, 684 (1965). ⁸ S. Johansson, Y. Cauchois, and K. Siegbahn, Phys. Rev. 82,

 ⁴ M. Nessin, T. H. Kruse, and K. E. Eklund, Phys. Rev. 125, 639 (1962).

⁵ The information on level schemes and properties of the gemanium isotopes are taken from the summary given in *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington, D. C., 1959).

Rather than discuss at length here the considerations involved in the analysis of the end points of a recoil spectrum, we present the results of an experiment which defines the energy of the recoil unambiguously by requiring a coincidence between the 690-keV conversion electron from Ge74 (or the 596-keV gamma ray from Ge⁷²) with the inelastically scattered neutron at a given angle. A sharp peak is then observed in the spectrum of coincidences versus detector pulse height which corresponds to the loss of the excitation energy of the excited nuclear state together with the uniquely defined energy loss of the recoiling atom. The difference between the pulse height of this peak and the pulse height obtained when the level is excited in such a way that the charge contribution from the recoil is negligible corresponds to the amount of energy lost to electron-hole production by the recoil atom when it stops in the germanium lattice. After presenting these new data, we compare further the two earlier experiments to demonstrate that the analysis given by SVP is apparently incorrect. A comparison with the theoretical calculations of Lindhard et al.⁶ is also given.

II. EXPERIMENTAL PROCEDURE

Neutrons were produced by means of the T(p,n)He³ reaction at a proton energy of 3217 ± 10 keV. The thickness of the tritium target, a water-cooled T-Zr type, was 105 ± 15 keV which thus gives a total neutron energy spread of the same amount, a proton energy at the center of the target of 3165 ± 13 keV, and a resultant mean neutron energy of 2381 keV at 0° with an estimated total uncertainty of perhaps ± 20 keV. The



FIG. 1. Schematic diagram of the experimental equipment in which the counters and shielding are shown for the 150° [Fig. 1(A)] and 90° [Fig. 1(B)] geometries.

⁶ J. Lindhard, V. Nielsen, M. Scharff, and P. V. Thomsen, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. 33, No. 10 (1963).



FIG. 2. Typical pulse-height spectrum produced by 2381-keV neutrons incident on a Ge(Li) gamma-ray detector for gamma-ray energies from 565 to 865 keV. Three prominent lines produced by inelastic scattering in germanium can be seen, as well as a line produced by inelastic scattering from Cu^{68} which is present in the detector container. The germanium lines are broadened by summing of the level de-excitation radiation energy with the energy of the recoiling germanium nucleus. The recoil energies range from close to 0 keV to a maximum of 107 keV depending on the angle of the scattered neutron. The energy dispersion is approximately 1 keV/channel.

spread in neutron energy corresponds to a spread in recoil energy of about 4 keV for inelastic scattering at 150° from Ge⁷⁴ (Q = -596 keV) or from Ge⁷² (Q = -690 keV), and 2.3 keV for the 90° inelastic scattering, and gives an uncertainty in the mean-recoil energy of about 1 and 0.5 keV, respectively.

The Ge(Li) gamma-ray detector with a cross-sectional area of 6 cm² and a depletion depth of 0.7 cm, was placed 39 cm from the tritium target at 0° to the incident proton beam. The full angle subtended by the detector was 4.6° which introduces a negligible spread (\sim 3 keV) in the incident neutron energy. A shielded stilbene neutron recoil detector in the shape of a right circular cylinder with a diameter of 5.08 cm and an altitude of 5.08 cm was placed 10 cm from the Ge(Li) gamma-ray detector to define the direction of the inelastically scattered neutrons. Two angles were used for the neutron detector, 90° and 150°, for which the corresponding recoil energies were about 56 and 103 keV. A schematic drawing of the two geometries used is shown in Fig. 1.

Inelastic scattering events were defined by requiring a fast $(2\tau \sim 50 \text{ nsec})$ coincidence between the two detectors. Pulse-shape discrimination was used on the output of the stilbene crystal to reject pulses caused by gamma rays thus making gamma-gamma coincidences between the two detectors of negligible importance. An auxiliary window on the linear signal from the neutron counter was used to eliminate low-energy neutrons produced by inelastic scattering to states with excitation greater than 1200 keV. The use of this window eliminated feeding of the 596-keV state by higher states and ensured that a single coincidence peak was observed. Pulses from the Ge(Li) gamma-ray detector that satisfied the coincidence and neutron selection criteria were then stored in a 1024-channel pulse-height analyzer.

The energy calibration of the Ge(Li) gamma-ray detector was made with well-known gamma-ray sources:



FIG. 3. Coincidence spectrum obtained with the Ge(Li) gammaray detector when the inelastically scattered neutron is observed at 150°. The peaks produced by inelastic scattering in the ger-manium are labeled with the isotope and level involved. The coincidence peaks of interest are at channels 614 and 710 and are superposed on a spectrum which consists of accidental coincidences, which have the same shape as the singles spectrum shown in Fig. 1, and true coincidences between neutrons and Compton scattered gammas from the decay of the Ge²⁸ 35-keV level, which form a continuous background of true coincidences below about channel 640. Typical statistical uncertainties are shown on several points. The curve drawn through the points is not a theoretical fit. The energy dispersion is approximately 1 keV/channel.

annihilation radiation at 511.006±0.002 keV,7 Ba¹³⁷ at 661.595±0.076 keV,8 and Fe56 at 846.79±0.09 keV.9 The linearity of the entire electronic system was checked carefully with a precision pulser.

All runs were made at a detector bias of 680 V which corresponds to a field of ~ 100 V/mm. Check runs were made at different bias voltages to see if any fielddependent effects were present, but no such effects were observed.

III. EXPERIMENTAL RESULTS AND ANALYSIS

A typical pulse-height spectrum for the gamma-ray energy region from 565-865 keV produced by the bombardment of a Ge(Li) gamma-ray detector by 2381-keV neutrons is shown in Fig. 2. Broadened peaks corresponding to the de-excitation of the 690- and 835-keV states in Ge72 and the 596-keV state in Ge74 are indicated. Results of coincidence runs with the neutron detector at 150° and 90° to the incident beam are shown in Figs. 3 and 4, respectively. Peaks which correspond to the summing of the 596- or 690-keV radiations with the particular recoil energy defined by the outgoing neutron are clearly visible superposed on a spectrum of accidental coincidences which has the same shape as the singles spectrum. There is also a contribution from true coincidences from Compton scattered gamma rays from the Ge⁷² 835-keV level. The widths of the peaks are consistent with kinematics.

Analysis of the data shown in Figs. 3 and 4 was performed in a straightforward manner. For the 150°

run, an estimate was first made of the continuous part of the spectrum and this was then subtracted from the total spectrum. The remainder then constitutes the sum of true and accidental coincidences associated with the 690-keV Ge72 state and the 596-keV Ge74 state. A correction for accidental coincidences was made by use of the singles spectrum. In order to obtain an estimate of the line shape for the 596- and 690-keV lines a smooth background spectrum was also subtracted from the singles spectrum. In the case of the 596-keV line, this procedure is complicated by the presence of summing of the 596- and 610-keV lines with recoils from the 596and 1.20-MeV states in Ge74. A correction was made for the presence of the other recoils so that a reasonable estimate of the line shape of the 596-keV line alone was obtained. The resulting line shapes from the singles runs were then normalized to the peaks at channels 581 and 677 in Fig. 3 and subtracted from the total coincidences to find the true coincidence peak. A summary of this procedure is shown in Fig. 5 for the 690-keV Ge⁷² level. The points given in Fig. 5(a) show the spectrum of coincidences obtained in the experiment. In Fig. 5(b), the continuous part of the spectrum has been subtracted to give the line shape for the 690-keV level alone. The contribution of accidental coincidences is shown by the curve which is a singles spectrum with the continuous spectrum also subtracted. Figure 5(c) shows the coincidence spectrum after subtraction of the accidentals. The centroid of the peak shown in Fig. 5(c) represents the sum of the energy of the level de-excitation plus the energy of the recoil associated with a neutron inelastically scattered at 150° to the incident beam. The energy corresponding to the peak is found by comparison with the annihilation radiation and Ba137 gamma-ray calibration lines. The same procedure was used for the 596-keV Ge⁷⁴ level and for both levels shown in the 90° data.

The amount of energy lost by the germanium recoil to the production of electron-hole pairs is then the energy corresponding to the coincidence peak less the energy corresponding to nuclear de-excitation under conditions of negligible nuclear recoil energy. We stress



FIG. 4. Coincidence spectrum obtained with the Ge(Li) gammaray detector when the inelastically scattered neutron is observed at 90°. The peaks produced by inelastic scattering in the germanium are labeled with the isotope and level involved. Typical statistical uncertainties are shown on several points. The curve drawn through the points is not a theoretical fit. The energy dispersion is approximately 1 keV/channel.

⁷ E. R. Cohen and J. W. M. DuMond, Rev. Mod. Phys. 37, 537 (1965). ⁸ R. L. Graham, G. T. Ewan, and J. S. Geiger, Nucl. Instr.

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Isotope and level	θ	E (keV)	É	$ar{oldsymbol{\eta}}(E)^{ a}\ ({ m keV})$	$ar{m{\eta}}(E)/E$	$ar\eta(\epsilon)$
Ge ⁷² (690) Ge ⁷⁴ (596)	90 150 90 150	56.4 ± 0.9 103.1 \pm 1.3 56.1 \pm 0.9 102.7 \pm 1.3	$\begin{array}{c} 0.200 \pm 0.003 \\ 0.366 \pm 0.005 \\ 0.197 \pm 0.003 \\ 0.360 \pm 0.005 \end{array}$	$19.5 \pm 1.2 \\ 37.6 \pm 1.2 \\ 17.8 \pm 1.2 \\ 35.5 \pm 1.2$	$\begin{array}{c} 0.346 {\pm} 0.022 \\ 0.365 {\pm} 0.012 \\ 0.317 {\pm} 0.022 \\ 0.346 {\pm} 0.012 \end{array}$	$\begin{array}{c} 0.069 {\pm} 0.004 \\ 0.134 {\pm} 0.004 \\ 0.062 {\pm} 0.004 \\ 0.125 {\pm} 0.004 \end{array}$

TABLE I. Summary of results, where θ is the angle of the scattered neutron and the remaining parameters are defined in the text.

^a The values given for $\overline{\eta}(E)$ are directly dependent on the particular level energies used. For the 150° data the accidentals spectrum can be used to find the channel corresponding to the low-energy end of the recoil spectrum, i.e., that channel is the midpoint of the low-energy end of the recoil spectrum. The energy calibration of the pulse-height analyzer is known and $\overline{\eta}(E)$ can then be found directly from the spectrum shown in Fig. 3. If this is done, a value of $\overline{\eta}(E)$ equal to 36.5 ±0.7 keV is found for both the Ge¹² (690) level and the Ge¹⁴ (596) level. From this it would appear that the energy of the Ge¹² level should be somewhat greater than 690 keV and the energy of the Ge¹⁴ level should be less than 596 keV. The adjustments needed are about equal to the quoted uncertainties in the determination of the level energies. Such an adjustment would also improve the agreement for the 90° data.

TABLE II. Summary of uncertainties for 150° run, 690-keV Ge72 level.

Source of uncertainty, X_i	ΔX_i	Corresponding uncertainty in recoil energy and $\bar{\eta}(E)$
A. Recoil energy Mean neutron energy, including Van de Graaff energy calibra- tion and target thickness un- cortainties	± 20 keV	±1.1 keV
Mean scattering angle	$\pm 0.5^{\circ}$	± 0.7 keV
Total uncertainty in E		± 1.3 keV
B. $\bar{\eta}(E)$ Centroid channel for Ge ⁷² (690) level Channel for Ba ¹³⁷ calibration line Energy calibration of analyzer, keV/channel Calibration line energy, Ba ¹³⁷ 690-keV Ge ⁷² level energy Total rms uncertainty in $\bar{\eta}(E)$	\pm 0.5 \pm 0.2 \pm 0.5% \pm 0.1 keV \pm 1.0 keV	$\pm 0.5 \text{ keV}$ $\pm 0.2 \text{ keV}$ $\pm 0.2 \text{ keV}$ $\pm 0.1 \text{ keV}$ $\pm 1.0 \text{ keV}$ $\pm 1.2 \text{ keV}$
C. Total rms uncertainty in $\overline{\eta}(E)/E$		$\pm 3.5\%$ or ± 0.012

that this procedure requires no interpretation of the spectra in terms of end points of the recoil spectra and hence represents an independent check on the results obtained in the earlier experiments of CJR and SVP.

A summary of the results obtained from the analysis of the data shown in Figs. 3 and 4 is given in Table I. Table II gives a summary of the experimental un-

TABLE III. Comparison of the present results for $\bar{\eta}(E)/E$ with the experiments of Sattler, Vook, and Palms (Ref. 1) and Chasman, Jones, and Ristinen (Ref. 2). The values given for the present experiment are the averages of the values given in Table I.

Experiment	E (keV)	$ar{m{\eta}}(E)/E$
Sattler, Vook, and Palms	54.3 107	0.235 ± 0.021 0.274 ± 0.030
Chasman, Jones, and Ristinen	56.3 100.3	0.320 ± 0.014 0.352 ± 0.008
Present experiment	56.2 102.9	0.331 ± 0.022 0.355 ± 0.012

certainties for the case of 150° inelastic scattering to the Ge⁷² 690-keV state. The relevant results obtained by CJR and SVP are compared with the present results in Table III. It can be seen that the results of the present experiment are in excellent agreement with the earlier work of CJR and disagree with the work of SVP.

IV. DISCUSSION AND CONCLUSIONS

The explanation for the disagreement between the experiments in the lower energy region is the apparently incorrect analysis of recoil spectra by SVP. An illustration of this error can be seen by comparison of Fig. 1 of Ref. 1 with Fig. 5 of this paper. The point chosen by



FIG. 5. The method used to analyze the coincidence data is depicted here for the Ge⁷² 690-keV level. The spectrum shown in A is the measured curve. B shows the contribution from the 690-keV level after subtraction of a smooth background. The line shows the magnitude of accidental coincidences which are assumed to have the same shape as the singles spectrum and were normalized at channels 690-700. The net peak which gives the energy of the 690-keV conversion electron from de-excitation of the level summed with the recoil energy is shown in C.

Sattler et al. as the end point of the recoil spectrum would be at about channel 705 of Fig. 5 while the centroid of the peak for recoils from 150° neutron scattering is at channel 710.4. The position of the peak for 180° scattering calculated from kinematics is 2.8 channels greater than that for 150° and thus should be at channel 713.2 which is in agreement with the position of the midpoint of the high-energy edge of the singles spectrum which was the reference point used by CJR. Indeed, a corrected value of $\bar{\eta}(E)/E$, where $\bar{\eta}(E)$ is the energy loss to electron-hole production by the recoil germanium atom and E is the recoil energy, for the data shown by Sattler et al. in their Fig. 1, can be calculated using the midpoint of their spectrum. The corrected value of $\bar{\eta}(E)/E$ is about 0.294 \pm 0.035 for a recoil energy of 42.9 keV which is in agreement with the results of CJR.

At higher neutron energies the data presented by SVP become more difficult to interpret. In Fig. 1 of Ref. 1, the tail of the recoil spectrum falls in an energy interval of 6 keV corresponding to about twice their



FIG. 6. A comparison of the experimental data with the theory of Lindhard, et al. (Ref. 6). The quantities $\bar{\eta}(\epsilon)$ and ϵ are defined in the text. The upper line for $\bar{\eta}(\epsilon) = \epsilon$ represents the limit in which all energy is lost to electron-hole production. The lower line is calculated from the theory of Lindhard *et al.* (Ref. 6). The experimental points are taken from the present experiment (indi-cated by arrows) and the results presented by CJR and SVP. The values shown for SVP were calculated from the values given by them for $\bar{\eta}(E)/E$ and E. The CJR results are for Ge^{72} (690-keV level) only since the points shown by them for Ge⁷⁴ (596-keV level) were subject to relatively large uncertainties because of feeding of that state by decays from higher excited states. The two or three lowest energy points of CJR tend to be systematically high because, for those recoil energies, the width of the recoil energy distribution is only slightly larger than the detector resolution. Hence, the approximation of the end points of the recoil spectrum by the use of the midpoints of the edges starts to break down and an exact unfolding of the detector resolution effects is required. The estimated size of the resolution effect is not enough to account for the discrepancy between CJR and SVP at these recoil energies, \sim 20–35 keV.

energy resolution of 3 keV while in Fig. 2 of Ref. 1, which is for a higher neutron energy, the rise requires about 20 keV. A 6-keV interval is understandable since, using a crude picture, the rise should take just twice the full width at half-maximum of a triangular resolution function folded into a flat recoil spectrum. This will not be true exactly here since the resolution function of the Ge(Li) detector is not triangular and the angular distributions of the scattered neutrons (related to the recoil energy distribution) are probably not isotropic, but it is certainly a sufficiently good approximation. The reason that the rise shown in Fig. 2 of Ref. 1 is so slow may be that at 2 MeV the spread of maximum recoil energies for elastic scattering and for inelastic scattering to just the available states in Ge⁷² and Ge⁷⁴ is about 60 keV, or about 20 keV after correcting for the proper values of electron-hole production by the recoils. The data shown in Fig. 2 are thus a sum of recoil spectra for many states. To our knowledge, adequate experimental data do not exist to unfold reliably the resolution and the sums of the various recoil spectra. The choice of end point given by SVP is thus hard to correlate with the end point of a particular recoil spectrum and leads to a large uncertainty in the recoil energy.

The experimental method of SVP should be reliable at energies less than about 50 keV, but their interpretation of the recoil spectrum end point is probably incorrect. At higher energies, 107 keV say, the effects of inelastic scattering become large and the interpretation of their data, while not as dependent on resolution effects, is rendered highly uncertain. In contrast, the techniques described by CJR and in the present work uniquely define a recoil energy by observing the summation of the recoil energy with the accompanying radiation characteristic of a particular level in a single isotope, although in both these experiments careful consideration must be given to the feeding of a particular level by higher levels. Resolution will, of course, be a limiting factor at the very low recoil energies, i.e., where the line broadening is comparable to the detector resolution.

A comparison of our data and the data of CJR and SVP with the theory of Lindhard *et al.*⁶ is given in Fig. 6. We have used here the notation of Lindhard *et al.* The quantity ϵ is a dimensionless measure of the recoil energy *E* given by,

$$\epsilon = E \frac{am_2}{Z_1 Z_2 e^2(m_1 + m_2)}$$

where $m_1 \approx m_2 \approx 72$, $Z_1 = Z_2 = 32$, and $a = 0.6260 a_0 Z^{-1/8}$ = 1.04×10^{-9} cm. The fractional energy loss of the recoil to electron-hole production is given by $\bar{\eta}(\epsilon)/\epsilon$ and the fractional atomic energy loss is $\bar{\nu}(\epsilon)/\epsilon$. It is assumed that $\bar{\eta}(\epsilon) + \bar{\nu}(\epsilon) = \epsilon$. The limiting case for all the recoil energy producing electron-hole pairs, $\bar{\eta}(\epsilon) = \epsilon$, is shown in Fig. 6 by the upper line. The theoretical predictions of Lindhard *et al.*⁶ for $k = 0.157 = 0.133 Z_2^{2/3} A^{-1/2}$ are given by the lower line. It can be seen that the theoretical predictions give an excellent fit to the experimental data of CJR and the present work in the energy range covered. The excellence of the fit is indeed surprising when one considers that Lindhard et al. made simplifying assumptions about the nature of the energy loss process in order to make the calculation feasible.

Further precise measurements of $\bar{\eta}(E)$ at lower and higher energies than considered here would be of interest to see if the agreement with the theory is as good as that in the present energy range. The results of SVP may be obscured by incorrect treatment of resolution

effects and by the extreme difficulties in properly accounting for inelastic scattering so that accurate measurements are still needed for comparison with theory in the region above 100 keV. Since it is possible that the results quoted for $\bar{\eta}(E)$ for silicon are also systematically low because of improper analysis of the end point of the recoil spectrum, further consideration should also be given to acquiring more data on $\bar{\eta}(E)$ for silicon atoms stopping in a silicon lattice so that the earlier work of Sattler¹⁰ on this element can be confirmed or corrected.

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Paramagnetic Resonance of F_2^- Ions Trapped in Electron-Irradiated Potassium Bifluoride*

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Electron-spin resonance (ESR) spectra attributed to F_2^- molecular ions located at two inequivalent anion sites were observed at 9.2 Gc/sec after electron bombardment of single crystals of potassium bifluoride (KHF2) at 77°K. The molecular axis of the F2⁻ ion lies along [110] or along [110], and the spectrum for each site consists of four hyperfine resonances. They derive from a hole interacting equally with two nuclei of spin $\frac{1}{2}$, giving an effective nuclear spin of 0 or 1 and a highly anisotropic A tensor. For $z \parallel [110], x \parallel [001], z \parallel [001],$ and $y \parallel [110]$, components of the g and A tensors for 77°K are $g_z = 2.0020 \pm 0.001$, $g_z \approx g_y = 2.0168 \pm 0.001$, $A_z = 955.4 \pm 1$ G, and $A_x \approx A_y = 21 \pm 4$ G. At room temperature $g_z = 2.0024 \pm 0.001$, $g_x \approx g_y = 2.0180 \pm 0.0015$, $A_z = 928.8 \pm 1$ G, $A_x = 35.5 \pm 1$ G, and $A_y = 25.5 \pm 3$ G. The crystal changed from colorless to intense green upon electron irradiation, with optical absorption bands at 2.0 and 4.1 eV. As the sample was brought to room temperature, the 2.0-eV band rapidly annealed, whereas the 4.1-eV band showed annealing behavior similar to the spin resonances, remaining about two days. The ESR spectra resulted from electron irradiations with electron energies as low as 0.15 MeV, but not from a 106-R dose of Co60 gamma rays.

I. INTRODUCTION

 $\mathbf{E}^{\mathrm{LECTRON-SPIN}}$ resonance (ESR) investigations were initiated to study the effects of ionizing radiation on single crystals of potassium bifluoride (KHF₂). This work was prompted, in part, by the similarity of the crystal symmetry of KHF₂ and that of KN₃, whose radiation-induced ESR spectra have been reported previously by this laboratory^{1,2} and other laboratories.^{3,4} In contrast to KN₃, which has defects generated by gamma rays, or even by ultraviolet light, the KHF₂ was found to be resistant to change by gamma-ray irradiation. When no spin resonances were detected in a KHF₂ sample which had received a 10⁶-R Co⁶⁰ gammaray dose at 77°K, electron bombardment was employed for further study of this material's radiation stability. Electron irradiation resulted in a paramagnetic species which has been identified as F_2^- molecular ions trapped in two inequivalent anion sites; the observed ESR spectra have shown similarities to the spectra of H centers and V centers in the alkali flourides.⁵⁻⁷ The characteristics of trapped F_2^- ions in KHF₂ are the subject of this paper.

Crystalline potassium bifluoride has been studied previously by x rays,⁸⁻¹⁰ infrared spectroscopy,¹¹⁻¹⁵ neutron

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