

Letters to the Editor

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Diffusion of Lithium into Germanium and Silicon

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 (Received May 7, 1953)

THE diffusion of lithium into *p* type single crystals of germanium and silicon¹ has been measured at temperatures between 450 and 1000°C by the *pn* junction method previously described.² The experiments were carried out by heating metallic lithium³ on the surfaces of specimens of germanium and silicon (0.125 cm thick) in a helium atmosphere. Hemispherical *n* type regions of low resistivity were observed to form, showing that lithium not only diffuses rapidly at the temperatures employed but also acts as a strong donor in germanium⁴ and silicon.

The diffusivity *D* and the surface concentration *C*₀ of Li in Ge and Si were calculated by means of Eqs. (1) and (2) given in reference 1. Corrections were introduced for the change in mobility with impurity concentration.⁵ The assumption was also made that each Li atom contributes one conduction electron.

The equations $D = 13 \times 10^{-4} \exp(-10700/RT)$ and $D = 94 \times 10^{-4} \exp(-18100/RT)$ were found to describe the dependence of diffusivity of Li in Ge and Si, respectively, for the temperature range studied, where *D* is given in cm²/sec and *R* = 1.98 calories. The maximum errors in the activation energies are estimated as ±1000 calories for Ge and ±3000 calories for Si. These results

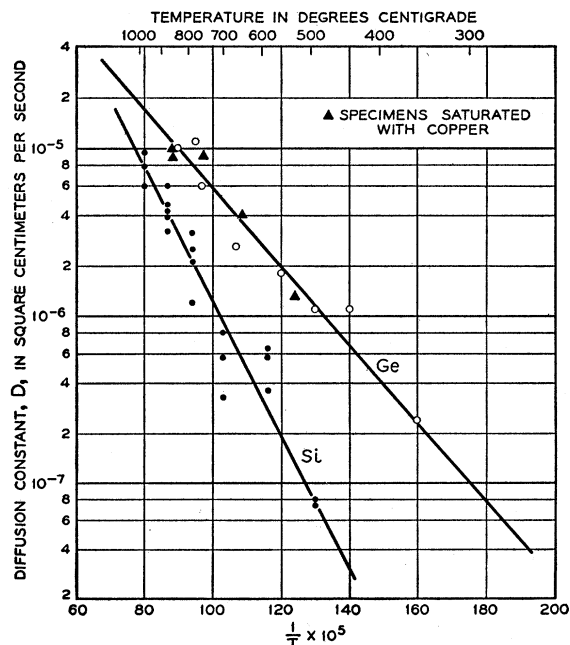


FIG. 1. Diffusion constant versus reciprocal of absolute temperature for Li in Ge and Si.

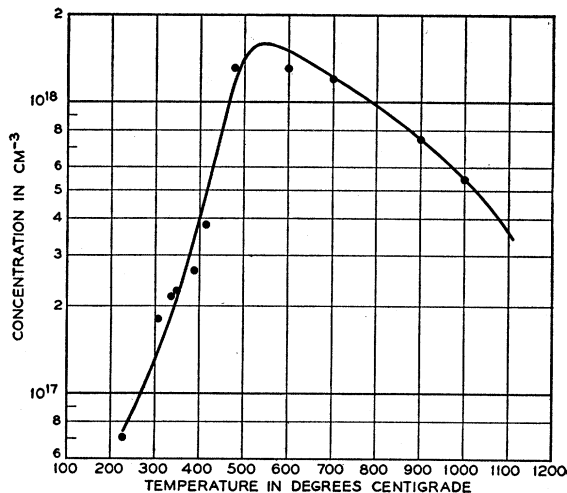


FIG. 2. Solubility of Li in Si as a function of temperature.

are shown in Fig. 1, where *D* is plotted against reciprocal of mean effective absolute temperature.⁶

Inasmuch as copper is also known to diffuse rapidly into germanium and silicon⁷ several specimens of germanium (see Fig. 1) were diffused in the presence of copper. No appreciable effect on the results was evident.

Calculated values of *C*₀ range from 6.1×10^{16} to 1.4×10^{18} cm⁻³ for Li in Ge and from 2.0×10^{17} to 8.0×10^{18} for Li in Si, but they are subject to large error. Determinations of the solubility of Li in Si have been made by measuring the change in resistivity upon saturation at a series of temperatures. These results are shown in Fig. 2. Similar measurements for Ge have not as yet been possible because of a rapid precipitation of Li at room temperature.

Resistivity and Hall measurements⁸ on Li doped Ge single crystals indicate a distribution coefficient between liquid and solid Ge for Li of greater than 0.01 and a donor level 0.01 eV below the conduction band. Precise determinations of distribution coefficient are difficult because of the precipitation previously mentioned.

¹ The authors are indebted to E. Buehler and G. K. Teal for supplying these crystals.

² C. S. Fuller, Phys. Rev. **86**, 136 (1952).

³ Spectrographic analysis made at Bell Laboratories showed only Ca, K, and Na present to greater than 0.005 percent.

⁴ It has been called to the authors' attention that W. C. Dunlap, Jr., and R. N. Hall at the General Electric Research Laboratory have independently reported lithium to act as a donor element in germanium.

⁵ Haynes, Pearson, Debye, and Prince, unpublished data [Bull. Am. Phys. Soc. **28**, No. 2, 10 (1953)]; Esther M. Conwell, Proc. Inst. Radio Engrs. **40**, 1327 (1952).

⁶ J. L. Ham, Trans. Am. Soc. Metals **31**, 849 (1943).

⁷ C. S. Fuller and J. D. Struthers, Phys. Rev. **87**, 526 (1952).

⁸ J. A. Burton and F. J. Morin, unpublished results.

The Mobility of Slow Electrons in Polar Crystals

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 (Received May 13, 1953)

WE have calculated the mobility of slow electrons in polar crystals using the same basic approximation as in our calculation (with T. D. Lee) of the effective mass.¹ The mobility has previously been calculated by Fröhlich and Mott² using perturbation theory. The dimensionless constant α which measures the strength of the electron lattice interaction is, however, between 1 and 6 for typical polar crystals, whereas the validity of perturbation theory requires $\alpha < 1$. The calculation of LLP for the effective mass was found to have considerable validity (~ 20

percent) for α as large as 6, so that we were encouraged to apply a similar method to this problem.

We describe the polaron (the electron plus its associated phonon cloud) by the same wave function as LLP, and calculate $1/\tau$, the probability per unit time that the polaron be scattered through an angle ϑ by the thermal phonons. We proceed by finding an equivalent Hamiltonian for transitions between bare polaron states, to which we apply the general scattering theory of Lippmann and Schwinger³ using the approximation that over and above the bound phonons, the state vector contain at most one additional phonon. The transition probability (and thus the mobility) can then be found in closed form in terms of the roots of a fairly complicated transcendental equation. The calculation is enormously simplified provided $(P^2/2m^*) \ll \hbar\omega$, where P and m^* are the polaron momentum and mass, and ω is the frequency of the optical mode phonons. (This is the case for temperatures such that $kT \ll \hbar\omega$.) In this temperature range $1/\tau$ is essentially independent of the polaron momentum. It should be emphasized that results which apply in this range *cannot* be extrapolated to higher temperatures.

We state our results. The mobility u is given by

$$u = \frac{1}{2\alpha\omega} \frac{e}{m^*} \left(\frac{m}{m^*}\right)^2 f(\alpha) \exp(\hbar\omega/kT),$$

where m is the effective crystal mass of the electron (owing to its interaction with the periodic field of the lattice), $\alpha = (e^2/2\hbar c) \times (2mc^2/\hbar\omega)^{1/2} (1/n^2 - 1/\epsilon)$, m^* is the polaron effective mass [$m^* = m(1 + \alpha/6)$ according to LLP], and $f(\alpha)$ is a slowly varying function of α which may be taken as $5/4$ for $3 < \alpha < 6$. n^2 and ϵ are the high-frequency and low-frequency dielectric constants of the crystal. The perturbation theory limit of m^* is m , and that of $f(\alpha)$ is 1, so that in the limit of weak coupling our result reduces to the correct perturbation-theoretic value.⁴

Details will be described in a forthcoming paper. One of us (D.P.) would like to acknowledge the partial support of the Office of Ordnance Research, U. S. Army, during this work.

¹ Lee, Low, and Pines, *Phys. Rev.* **90**, 297 (1953), hereafter referred to as LLP.

² H. Fröhlich and N. F. Mott, *Proc. Roy. Soc. (London)* **A171**, 496 (1939). Some corrections to the original theory were made by Fröhlich, Pelzer, and Zienau, *Phil. Mag.* **41**, 221 (1950).

³ B. A. Lippmann and J. Schwinger, *Phys. Rev.* **79**, 469 (1950).

⁴ It should be noted that this perturbation-theory limiting value is smaller by a factor of 3 than that quoted by Fröhlich *et al.*, reference 2. Conyers Herring (private communication) was the first to point out that such a correction factor needs to be applied to the previously published perturbation-theoretic calculations.

Radiations of Neutron-Deficient Barium and Cesium Nuclides*

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(Received May 5, 1953)

IN the study of spallation reactions of cesium with high-energy protons, radiations of nuclides Ba^{128} , Ba^{129} , and Cs^{128} were investigated. Previously it has been reported¹ that 2.4 ± 0.1 -day Ba^{128} generates a 3.2-minute cesium daughter, accompanied by emission of 3-Mev positrons from the equilibrium mixture. Existence of 2.0-hour Ba^{129} with emission of positrons of unknown energy has also been reported,^{2,3} with assignment by mass spectrograph.² Recently Lindner and Osborne⁴ have reported that Ba^{128} decays purely by orbital-electron capture leading to a 3.8-minute Cs^{128} daughter which decays with emission of 3-Mev positrons. Wapstra *et al.*⁵ have studied Cs^{128} and report gamma-rays of 455 ± 5 keV in about 20 percent of the decays, and 980 ± 30 keV present to about 2 percent of the intensity of the 455-keV line.

We have measured the maximum positron energy of 2.0-hour Ba^{129} produced by irradiating CsCl with 60-Mev protons followed by chemical separation similar to that reported previously.¹ At

this bombardment energy, the yield of Ba^{128} is extremely small while that of Ba^{129} is at a maximum. This eliminates masking due to the energetic positrons from Ba^{128} . A low-resolution beta-ray spectrometer indicated a maximum positron energy for Ba^{129} of 1.6 ± 0.2 Mev, consistent with a total predicted positron decay energy of 1.8 ± 0.3 Mev computed from the beta-decay systematics of Coryell.⁶ These positrons decayed with a half-life of about 2 hours, so that their assignment to Ba^{129} is unequivocal. Conversion electrons of 0.13 Mev also were present in the decay of Ba^{129} . Conversion electrons of 0.24 Mev presumably were due to the presence of Ba^{130m} .

The equilibrium mixture Ba^{128} - Cs^{128} was studied by the method of coincidence absorption using two anthracene scintillation detectors with associated equipment having a measured resolving time of 0.23 microsecond. Coincidence absorption curves in aluminum indicate that there exists a lower-energy positron group which has a maximum energy of 1.1 ± 0.7 Mev based on the range in aluminum and which is in coincidence with one or more gamma-rays. Since no positron-gamma-ray coincidences were observed from the 3-Mev group, the latter must proceed directly to the ground state of Xe^{128} . Computation from Coryell's systematics indicates a total Cs^{128} - Xe^{128} disintegration energy of 3.5 ± 0.5 Mev, consistent with the maximum energy of 3.1 ± 0.2 Mev observed. Lead absorption curves showed no gamma-rays of energy greater than 0.9 Mev. Conversion electrons of 0.2 Mev were observed on the spectrometer. The number of positrons exceeds the number of electrons by a factor of 3 or 4.

That Ba^{128} decays primarily if not entirely by electron capture has been shown by repurification experiments in which a fresh barium sample exhibited a growth due to positrons from daughter Cs^{128} . This growth curve had a t_{max} of about 30 minutes in agreement with that expected on the basis of a 2.4-day parent growing into a 3.2-minute daughter. A calculation from Coryell's systematics indicates that Ba^{128} should have insufficient decay energy for positron emission. Decay of the positron peak from a young sample of Cs^{128} , isolated carrier-free by leaching solid $BaCl_2 \cdot H_2O$ with concentrated HCl-ethyl ether (5:1) mixture, was followed on the spectrometer. A half-life of 3.5 ± 0.6 minutes was found.

To study the gamma-emission from Ba^{128} - Cs^{128} equilibrium mixture, a fresh barium sample mounted as $BaCrO_4$ from a bombardment of CsCl with 240-Mev protons on the Rochester 130-inch cyclotron was taken to Brookhaven National Laboratory,⁷ where measurements were made by Mr. Alois Schardt using a gray-wedge scintillation spectrometer fitted with a NaI(Tl) crystal. Those gamma-ray lines assigned to the decay of Ba^{128} - Cs^{128} mixture are, in keV: 30 (K x-ray), 135 ± 5 (strong), 285 ± 10 , 455 ± 5 , and 965 ± 20 (weak). Weak lines at about 220 and 380 keV are attributed to the presence of a certain amount of Ba^{131} . The line at 135 keV is about three times as intense as that at 455 keV. The gray-wedge equipment was calibrated using a Na^{22} source.

Further work is required to establish the exact decay scheme of the Ba^{128} - Cs^{128} chain. No detailed work was done on gamma-gamma coincidences other than a survey with the gray-wedge instrument which demonstrated the existence of gamma-gamma coincidences, indicating a decay involving a gamma-ray cascade.

We wish to thank Professor Sidney W. Barnes, Mr. Kurt Ennslein, and Mr. William Coombs of the Cyclotron Laboratory for assistance in bombardments and design of certain equipment used in this study.

* This work was performed under contract with the U. S. Atomic Energy Commission. Part of the data is taken from the Doctoral Thesis of R. W. Fink submitted in partial fulfillment of the requirements for the Ph.D. degree, January 20, 1953.

† U. S. Atomic Energy Commission Predoctoral Fellow in Physical Science, 1951-53.

¹ R. W. Fink and E. O. Wiig, *J. Am. Chem. Soc.* **73**, 2365 (1951).

² R. W. Fink and D. H. Templeton, *J. Am. Chem. Soc.* **72**, 2818 (1950).

³ C. C. Thomas, Jr., and E. O. Wiig, *J. Am. Chem. Soc.* **72**, 2818 (1950).

⁴ M. Lindner and R. N. Osborne, *Phys. Rev.* **83**, 1422 (1952).

⁵ Wapstra, Verster, and Boelhouwer, *Physica* **19**, 138 (1952).

⁶ C. D. Coryell, *Annual Reviews of Nuclear Science* (Annual Reviews, Inc., Stanford, 1953), Vol. II, p. 305.

⁷ The hospitality of Brookhaven Laboratory was extended by Dr. G. Friedlander of the Department of Chemistry.