Measurement of Spectral Distribution of Positron Flux in an Infinite Copper Medium Containing Cu^{64}†

W. H. WILKIE* AND R. D. BIRKHOFF
Health Physics Division, Oak Ridge National Laboratory,‡ Oak Ridge, Tennessee
(Received 25 March 1964)

The spectral distribution of the flux of positrons inside a beta radioactive medium was experimentally determined using an anthracene coincidence scintillation spectrometer. Positrons leaving a copper cavity or plane source containing Cu^{64} were absorbed in a thin anthracene crystal. The light pulses from the anthracene were analyzed and recorded if a NaI scintillation spectrometer nearby recorded simultaneously a count under the total absorption peak for annihilation radiation. Thus anthracene pulses due to absorption of negatrons and secondary electrons were not recorded. The spectra were corrected for the nonlinear pulse-height-energy characteristic of anthracene by using a semiempirical theory of Birks. This correction was shown to change an observed nonlinear Fermi plot of Pm^{147} to the accepted linear shape. The positron spectra were compared with the theoretical continuous slowing-down model. Over the range measured (20–650 keV) it was concluded that the continuous-slowing-down model gives the correct shape for the primary slowing-down spectrum for the cavity source, but that the plane source exhibited a deficiency of low-energy positrons.

I. INTRODUCTION

KNOWLEDGE of the spectral distribution of the primary and secondary electron flux generated in an infinite medium by ionizing radiation is of basic importance in understanding the effects of radiation on matter.

Most measurements in the past on the interaction of electrons with solids have been made either with an anisotropic situation and/or in a medium containing boundaries, e.g., secondary emission experiments and stopping power measurements in thin foils. In such cases, electron scattering tends to obscure the basic physical processes wherein the electron loses energy to the medium. If, however, a medium is chosen which is larger in size than the range of the highest energy electron present, and a small vacuum cavity is introduced within this medium, the electron flux within the cavity is representative of the flux within the medium. Then if a channel connects the cavity with an electron spectrometer, a sample of this flux may escape into the electron spectrometer for energy analysis. The cavity source concept is similar to the concept of a blackbody cavity with the substitution of electron flux for electromagnetic flux. This experimental technique was used by Birkhoff *et al.*[1] to determine the spectral distribution of both the primary and secondary electron flux in a medium composed of P^{32} uniformly dispersed in Bakelite. Their results compared favorably with the theoretical total flux as calculated using the Spencer-Fano-Attix theory. The understanding of the electron-flux spectrum is basic to an understanding of the response of gas-filled dosimeters. This subject has been extensively reviewed recently.[2] The electron flux described in the present research is actually the primary part of the "Greening Effect" described in this reference. It seems obvious that accurate descriptions of the electrons traversing a vacuum cavity must be obtained before their interactions with a gas may be calculated with precision.

In the present study, use of a positron emitter premitted the measurement of the primary component of the slowing-down flux separately. This is possible because the positron retains its identity from birth until annihilation, whereas a primary electron is indistinguishable from a secondary of the same energy. A scintillation spectrometer was used for the flux measurements, with anthracene being chosen as the scintillator because of its high relative pulse height, high efficiency for beta detection, and low gamma-ray response. It was also an obvious choice because previous research concerning its response to electrons provided correction factors for the slightly nonlinear, pulse-height-energy relationship. Although the anthracene produced pulses from absorptions of negative beta rays, secondary electrons, and positrons, only the latter were recorded because only they were in coincidence with the annihilation gamma rays detected by a nearby NaI spectrometer.

II. THEORY

A. Beta Spectra: Continuous-Slowing-Down Theory

A beta particle born in an infinite, homogeneous medium, travels through the medium dissipating energy by inelastic nuclear collisions with the subsequent emission of bremsstrahlung and by excitation and ionization of atoms in the medium. For beta particles born in copper with energies below 1 MeV, the energy loss by inelastic nuclear collisions is small. Collisions in which a substantial fraction of the energy is transferred are

† A more extensive report on this research is available as Oak Ridge National Laboratory Report No. ORNL-3469 (unpublished).
* This research was submitted to Vanderbilt University by W. H. Wilkie in partial fulfillment of the requirements for the degree of Master of Science.
‡ Operated by Union Carbide for U. S. Atomic Energy Commission.
[1] R. D. Birkhoff, H. H. Hubbell, Jr., J. S. Cheka, and R. H. Ritchie, Health Phys. 1, 27 (1958).
[2] Natl. Bur. Std. (U. S.), Handbook 79 (1961).

known to be rare; thus, most secondary electrons are born with energies of less than a few hundred electron volts. In 1954 Spencer and Fano[3] published a theory of electron slowing-down which treated most of the secondaries on a continuous slowing-down model; i.e., a primary electron is considered to lose energy in infinitesimal increments and therefore may be thought of as losing energy continuously; however, large losses were included as a source of new "primary" electrons. More recently Schneider and Cormack[4] have found very good agreement between the flux spectrum calculated from the continuous slowing-down model and the spectrum determined using the Monte Carlo technique.

If one considers a spherical probe with 1-cm^2 cross-sectional area in an infinite beta-radioactive medium, then the flux at energy E is defined as the number of beta particles which cross the sphere per second from any direction with energies between E and $E+dE$. If one multiplies the flux $\Phi(E)$ by the energy increment dE and then by the stopping power at energy E, $(dE/dx)_E$, which is the average spatial rate of energy loss, the result is the energy loss per cm^3 per sec by electrons with energy in dE. That is, the average energy loss per cm^3 per sec is

$$\Phi(E)dE(dE/dx)_E. \tag{1}$$

In the continuous-slowing-down model, one neglects large energy losses and assumes that every electron born with energy greater than E will at some time during its life pass through energy E and will therefore contribute to the flux at this energy. In equilibrium conditions $P(E)$ electrons per cm^3 per sec are born with energies between E and $E+dE$, and all electrons with initial energies above E will pass through the energy interval dE at E. The energy deposition in the medium from these electrons as they pass through dE will be

$$dE\cdot\int_E^{E_{\max}} P(E')dE'. \tag{2}$$

By equating Eqs. (1) and (2) and solving this identity for $\Phi(E)$, one has a method for calculating the slowing-down flux in simplest approximation.

$$\Phi(E)=\int_E^{E_{\max}} P(E')dE'/(dE/dx)_E. \tag{3}$$

The physical significance of this expression may be visualized in another way. The flux between E and $E+dE$ is directly proportional to the number of electrons born which can contribute to it; i.e., those born with energy greater than E. It is inversely proportional to the distance the electrons move in losing the energy dE; i.e., long tracks contribute more to the flux than short tracks.

[3] L. V. Spencer and U. Fano, Phys. Rev. **93**, 1172 (1954).
[4] D. O. Schneider and D. V. Cormack, Radiation Res. **11**, 418 (1959).

The nuclear beta spectrum $P(E)$ may be calculated from the Fermi function[5] for radionuclides having an allowed beta spectrum. The flux $\Phi(E)$ may then be determined by Eq. (3) using the stopping power calculated from the Rohrlich-Carlson expression.[6]

B. Experimental Procedure

The object of this experiment was to determine the primary slowing down spectrum of Cu^{64} positrons in an infinite copper medium. Positrons are required rather than negatrons because a primary negatron is indistinguishable from a secondary electron of the same energy.

The experimental method consisted of allowing positrons from a cavity source to strike and be absorbed in a thin anthracene crystal. After losing essentially all of their initial kinetic energy, the positrons were annihilated, and each emitted two 0.511-MeV gamma rays.[7] If a NaI (Tl) spectrometer positioned near the anthracene crystal (but shielded from the cavity source) detected under the photopeak one of the oppositely directed annihilation gammas, it electronically gated a multichannel pulse-height analyzer (Nuclear Data Model-101 analyzer and Nuclear Data Model-500 dual amplifier discriminator) connected to the anthracene spectrometer. The multichannel analyzer then allocated the coincident pulse produced by the anthracene spectrometer to a channel appropriate to the pulse size. The coincidence technique was necessary in order to sort the pulses produced by positrons slowing down in the anthracene from pulses from negatrons, secondary electrons, and gamma rays.

A sectional view of the coincidence spectrometer is shown in Fig. 1. It was necessary to separate the source from the anthracene and to provide lead shielding of the NaI(Tl) crystal to minimize accidental gating. No significant loss in positrons due to magnetic deflection by the earth's field was observed at a source to crystal distance of up to 12 in. The vacuum chamber was an aluminum tube 2 in. in diameter. The cavity source was a truncated hollow copper cone with walls and base 0.018 in. thick, a base diameter of 0.75 in., and an open top 0.200 in. in diameter. The fractional solid angle subtended at the center of the base by the open truncated top was 0.10. It was important that this be as small as possible so that the emission of positrons from the base would be characteristic of the flux in a medium of infinite extent. That is, the interior surface of the source which "looks out" through the hole into the spectrometer should be surrounded by wall material as much as possible so that its emission is characteristic of that within a medium of infinite extent. The shape of the cavity is not important in contrast to the situa-

[5] Natl. Bur. Std. Appl. Math. Ser. **13**, pp. 21–61 (1952).
[6] F. Rohrlich and B. C. Carlson, Phys. Rev. **93**, 38 (1954).
[7] P. R. Wallace, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1960), Vol. 10.

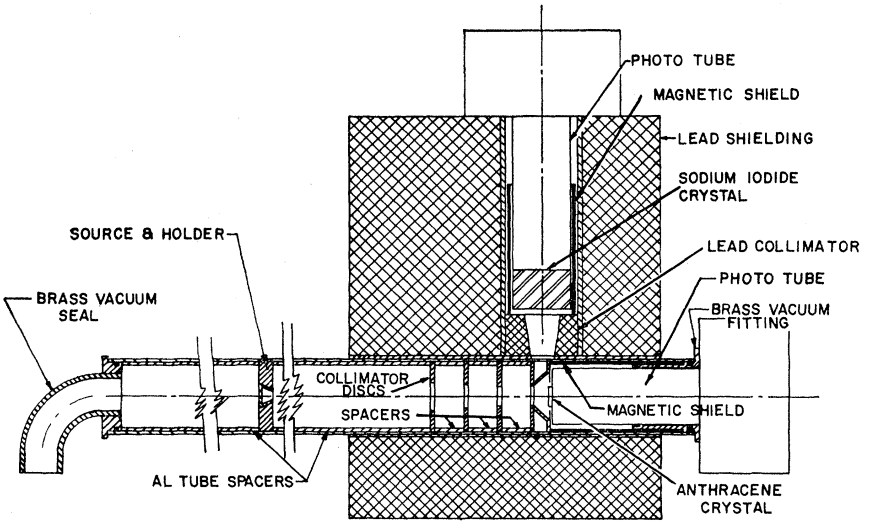

FIG. 1. Sectional view of anthracene coincidence spectrometer showing cavity source, vacuum chamber, anthracene crystal and photomultiplier and sodium iodide crystal and photomultiplier.

tion where the cavity is filled with gas.[2] The flat source was a copper disk 0.018 in. thick and 0.5 in. in diameter. After fabrication the sources were irradiated with thermal neutrons to produce Cu^{64}.

The linearity of the spectrometer was checked by measuring the response of anthracene to surface incident negatrons from Pm^{147}. This source was prepared by drying a drop of Pm^{147} in solution on a very thin sandwich of Formvar and aluminum with a total thickness of about 15 $\mu g/cm^2$.

C. Response of the Anthracene Scintillation Spectrometer

The response of anthracene to surface incident monoenergetic electrons has been investigated by several experimenters, and the nonlinearity is well known. This nonlinear response is most apparent at low energies, i.e., less than 100 keV. Not so well recognized is the fact that simple corrections for the nonlinearity can be made which then make an anthracene beta spectrometer feasible.

The pulse-height-energy relationship for externally incident electrons above 20 keV in energy may be explained with the aid of the theory of Birks[8] on the scintillation process. Below this energy, surface effects become important and corrections to Birks' theory,[9] are required unless a well-type crystal geometry is employed or the electrons are born inside the crystal. Let S be the number of photons emitted by the scintillating crystal during the complete absorption of an energetic electron. In Birks' theory the spatial rate of production of photons along the electron track, dS/dx, depends on the spatial rate of energy loss or stopping power dE/dx as follows:

$$\frac{dS}{dx}=\frac{AdE/dx}{1+(kBdE/dx)}, \tag{4}$$

where A and B are constants of proportionality and k is the differential capture probability of a quencher molecule relative to a fluorescer molecule. The constant A may be identified with the light producing efficiency of electrons in anthracene and kB is related to the effective local quenching.[10]

In order to obtain a pulse-height-energy relationship from this expression, it is necessary to invoke a stopping power or range-energy equation for materials of low atomic number. The range-energy relationship of electrons has been studied by Glocker,[11] Libby,[12] and by Lane and Zaffarano,[13] and according to them the range x in g cm^{-2} is related to the electron energy E in MeV by the expression

$$x=bE^{5/3}, \tag{5}$$

where b is proportionality constant, and the expression holds for energies between 1 and 300 keV. This relationship may be differentiated and substituted into Eq. (4) to obtain the differential fluorescence efficiency:

$$\frac{dS}{dE}=\frac{A}{[1+(3kB/5b)E^{-3/2}]}. \tag{6}$$

Birks obtained an experimental value of 7.15 cm-air per MeV for kB, and the average value of b from the references listed is 0.65 $g/cm^2(MeV)^{-5/3}$. One may then

[8] J. B. Birks, Proc. Phys. Soc. (London) **A64**, 874 (1951).
[9] J. B. Birks and F. D. Brooks, Proc. Phys. Soc. (London) **B69**, 721 (1956).
[10] E. Newman, A. M. Smith, and F. E. Steigert, Phys. Rev. **122**, 1520–1524 (1961).
[11] R. Glocker, Z. Naturforsch. **3a**, 147 (1948).
[12] W. E. Libby, Anal. Chem. **19**, 2 (1947).
[13] R. O. Lane and D. J. Zaffarano, Iowa State College Report ISC 439, 1953 (unpublished).

evaluate the quantity $\alpha=3kB/5b$

$$\alpha=\frac{3\times 7.15\text{ cm-air/MeV}\times 0.001293\text{ (g/cm}^2\text{)/cm-air}\times 273/288\times(1000\text{ keV/MeV})^{2/3}}{5\times 0.65\text{ (g/cm}^2\text{)/(MeV)}^{5/3}}$$
$$=0.81\text{ (keV)}^{2/3}. \tag{7}$$

The number of photons S may be found immediately by integrating[14,15] Eq. (6) and making the substitution for $\alpha=3kB/5b$; thus

$$S=AE\left[1-\frac{3}{\mu^2}+\frac{3}{\mu^3}\tan^{-1}\mu\right], \tag{8}$$

where

$$\mu=E^{1/3}\alpha^{-1/2}. \tag{9}$$

At high energies the sum of the second and third terms in the bracket in Eq. (8) approaches zero and

$$S\rightarrow AE. \tag{10}$$

Thus, a quantitative explanation is obtained for the apparent linear response of anthracene to high electron energies. A plot of the low-energy portion of Eq. (8) is given in Fig. 2. Also plotted is a straight line passing through the origin, and the pulse height of Eq. (8) at 624 keV.

The slope of Eq. (8) may be taken and extrapolated to determine the energy intercept E_i. A plot of E_i as a function of the energy at the point of tangency E_1 is shown in Fig. 3 and given by Eq. (11):

$$E_i=E_1\{1-[1+\mu_1^{-2}][1-3\mu_1^{-2}(1-\mu_1^{-1}\tan^{-1}\mu_1)]\}, \tag{11}$$

where

$$\mu_1=E_1^{1/3}\alpha^{-1/2}.$$

It may be seen that E_i varies from about 4 keV to 20 keV in agreement with results published by Johnston *et al.*[16] and Hopkins,[17] respectively.

One may relate the number of counts appearing in the analyzer channels $N_1(C)$ to the number of electrons detected per unit energy $N(E)$ by

$$N(E)=N_1(C)dC/dE \tag{12}$$

by using the slope of the C versus E response curve. Thus, any point on a pulse-height spectrum must be corrected in abscissa by use of the pulse-height-energy curve for anthracene and in ordinate by Eq. (12) before it may represent a point on the energy spectrum.

The transmission and energy calibration of the spectrometer was examined using a Pm^{147} source. Figure 4 shows a Fermi plot of the uncorrected data and the same with the data corrected for the nonlinearity of anthracene and for changing bandwidth. Note that the momentum distribution $N'(P)$ necessary for the ordinate has been obtained from the experimental energy distribution $N(E)$ by multiplying the latter by the electron velocity v.

The corrected Fermi plot appears linear except at energies near the endpoint where the resolution effect[18] becomes apparent. The data were not corrected for the statistical shift of the photomultiplier response[19,20] at low energies because only a small number of photoelectrons were ejected at the photocathode, nor for electrons scattered from the collimators and source

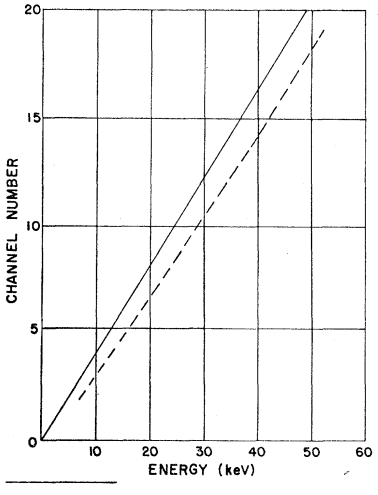

FIG. 2. Theoretical pulse-height-energy curve for anthracene (dashed line) according to Birks. Solid straight line passes through the origin and the same channel as the dashed line at 624 keV.

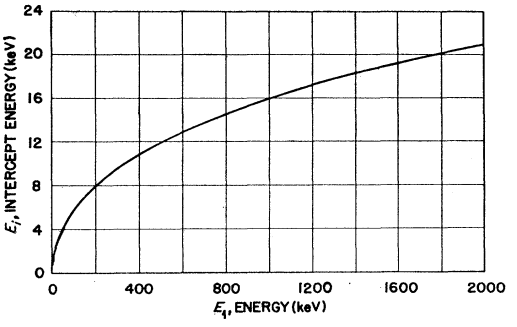

FIG. 3. Theoretical energy intercept of the extrapolated slope of the pulse-height-energy curve as a function of the energy at which slope is taken.

[14] L. W. Johnston, R. D. Birkhoff, J. S. Cheka, H. H. Hubbell, Jr., and B. G. Saunders, Oak Ridge National Laboratory Report No. ORNL-2298, 1959 (unpublished).

[15] Oak Ridge National Laboratory, Health Physics Division Annual Progress Report ORNL-2806, 1959 (unpublished), p. 153.

[16] L. W. Johnston, R. D. Birkhoff, J. S. Cheka, H. H. Hubbell, Jr., and B. G. Saunders, Rev. Sci. Instr. **28**, 765 (1957).

[17] J. I. Hopkins, Rev. Sci. Instr. **22**, 29 (1951).

[18] Von Klaus-Werner Hoffmann, Nachr. Akad. Wiss. Göttingen, II. Math. Phys. Kl. No. 13, 273 (1960).

[19] G. T. Wright, Phys. Rev. **96**, 569 (1954).

[20] E. Brannen and G. L. Olde, Radiation Res. **16**, 1 (1962).

backing, nor for backscattering[21] of electrons from the crystal surface. These phenomena have compensating effects, and the linearity at the lowest energies is somewhat fortuitous but, fortunately, indicates a constant transmission spectrometer.

D. Results of the Flux Measurements

The spectral distribution of positrons from Cu^{64} was compared with the flux predicted by the continuous-slowing-down model by means of the relation

$$\Phi(E)=N(E)\frac{1}{G_1}\frac{1}{\epsilon}\frac{1}{G_2}\frac{1}{\bar{A}}, \tag{13}$$

where $\Phi(E)$ is the flux in units of positrons cm^{-2} keV^{-1} sec^{-1} normalized to one positron born per cm^3 per sec; $N(E)$ is the average number of positrons detected per keV per sec during the counting period; G_1 is the NaI(Tl) crystal geometry factor (8×10^{-3}); ϵ is the NaI(Tl) crystal photoelectric efficiency for 0.511-MeV gamma radiation (0.17); G_2 is the anthracene geometry factor (1.7×10^{-3}) and $\bar{A}$ is the average number of positrons born per sec per cm^3 during the counting period.

Figure 5 shows a corrected spectrum of positrons from plane and cavity copper sources compared with the continuous slowing-down theory. The spectrum from the plane source was multiplied everywhere by a constant to make it agree with the continuous slowing-down theory at 300 keV. The absence of emitting walls adjacent to the plane source resulted in a loss of positrons originating in the walls and backscattering into the detector from the plane source. This missing backscattered contribution to the flux from the plane source has a degraded energy spectrum. The data for the cavity source were multiplied by a normalization factor of 1.6 in order to bring the data into agreement with the theory at 300 keV. No significance was given to this factor as it seemed to vary from source to source over the range of 0.75 to about 2.0. It is possible that nonreproducible source self-shielding in the reactor was responsible.

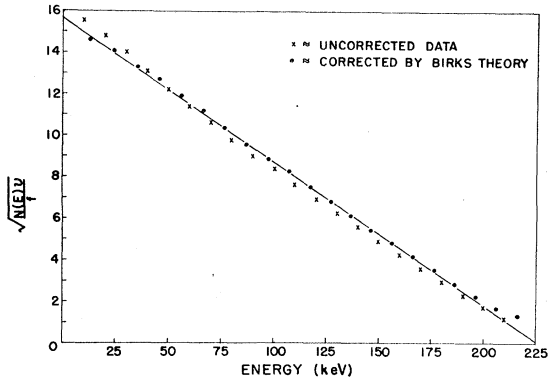

FIG. 4. Fermi plot of Pm^{147} beta spectrum showing effect of correction of data by Birks' theory.

It may be concluded that a plane anthracene crystal-photomultiplier combination is suitable for precise beta spectroscopy, and that the spectral corrections described above adequately correct the experimental data for the effects of anthracene nonlinearity and variable energy bandwidth. The shape of the positron slowing-down spectrum in copper is in good agreement with the

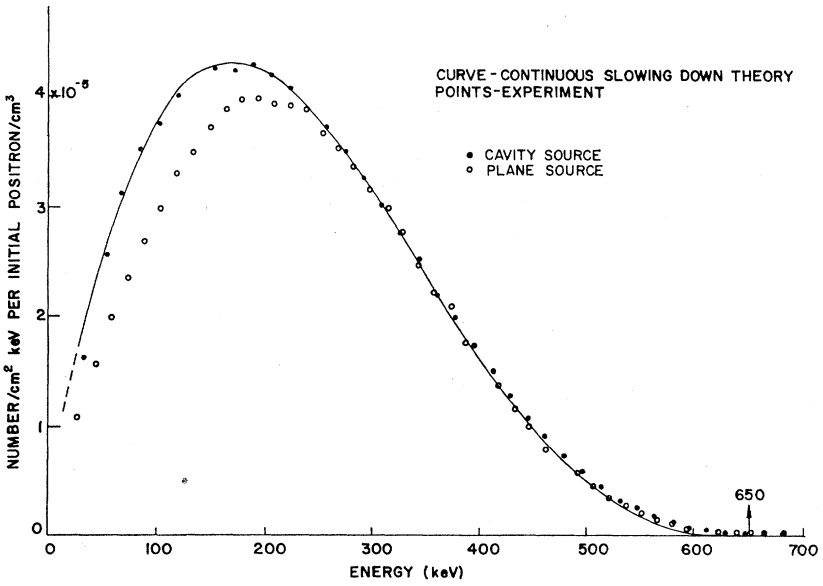

FIG. 5. Theoretical positron flux in Cu using the continuous-slowing-down model, and data for cavity and flat sources. The two sets of data are multiplied by constants to make them agree with theory at 300 keV.

[21] G. Bertolini, F. Cappellani, and A. Rota, Nucl. Instr. Methods 9, 107 (1960).

continuous slowing down model. The spectrum of the positrons escaping the surface of a plane source thicker than the positron range is not identical with the slowing-down spectrum in an infinite medium due to a deficiency in the number of positrons with low energies.

ACKNOWLEDGMENTS

Appreciation is extended to H. H. Hubbell, Jr., for his many valuable comments and suggestions on all phases of the research. The authors wish to thank J. A. Harter, P. W. Reinhardt, and F. J. Davis for their helpful advice and assistance.

Credit is due to P. N. Hensley and G. H. Hager for their help with the engineering aspects of the research. Further appreciation is extended to the Oak Ridge Institute of Nuclear Studies which made this research possible for one of us (W. H. Wilkie) through the AEC Special Fellowship in Health Physics.

PHYSICAL REVIEW VOLUME 135, NUMBER 4A 17 AUGUST 1964

Relative Energy Loss to Optical and Acoustic Modes of Electrons in Avalanche Breakdown in Ge

E. M. CONWELL
General Telephone and Electronics Laboratories Inc., Bayside, New York
(Received 4 March 1964)

Expressions are derived for the rates of loss of high-energy electrons in a many-valley band structure to acoustic and optical modes. Evaluating these for germanium, for which all the parameters are now reasonably well known, we find that, contrary to what is usually assumed, the rate of loss to acoustic modes is greater than that to optical modes for electron energies of the order of the energy gap.

IT has been customary to assume, rather arbitrarily, in calculations on avalanche breakdown in germanium and silicon that, just below the threshold for ionization, the predominant energy loss process for electrons and holes is optical phonon emission.[1–3] It has been realized for some time that for both electrons and holes in germanium, contrary to earlier expectations, the rate of loss to optical phonons exceeds that to acoustic phonons over a wide range of fields starting from low ones.[4,5] However, in the limit of high enough fields, or high enough carrier energies, the loss to acoustic modes must once again predominate.[5,6] It is the purpose of the present note to demonstrate that in Ge, for electrons with energies of the order of the band gap and larger, the rate of loss to acoustic modes is larger than that to optical modes.

Using the usual perturbation theory, we may write for the rate of energy loss to acoustic modes of an electron with wave vector $\mathbf{k}$, measured relative to the band edge,

$$\frac{d\mathcal{E}}{dt}(\mathbf{k})=\frac{2\pi}{\hbar}\sum_{\mathbf{q}}\sum_{\alpha}[\hbar\omega_{q\alpha}|(\mathbf{k}+\mathbf{q},N_{\mathbf{q}\alpha}-1|H_\alpha'|\mathbf{k},N_{\mathbf{q}\alpha})|^2 \times\delta(\mathcal{E}_{\mathbf{k}+\mathbf{q}}-\hbar\omega_{q\alpha}-\mathcal{E}_{\mathbf{k}})-\hbar\omega_{q\alpha}|(\mathbf{k}-\mathbf{q},N_{\mathbf{q}\alpha}+1 \times|H_\alpha'|\mathbf{k},N_{\mathbf{q}\alpha})|^2\delta(\mathcal{E}_{\mathbf{k}-\mathbf{q}}+\hbar\omega_{q\alpha}-\mathcal{E}_{\mathbf{k}})], \quad (1)$$

where $\hbar\omega_{q\alpha}$ and $N_{\mathbf{q}\alpha}$ are the energy and steady-state number of phonons with wave vector $\mathbf{q}$ and polarization α. The argument of the δ function is the difference between the initial and final energies of the system. The first term of (1) gives the rate of energy gain due to phonon absorption, while the second term gives the rate of loss due to emission. The matrix elements for acoustic phonon interaction in the many-valley band structure are given by[7]

$$|(\mathbf{k}\pm\mathbf{q}|H_\alpha'|\mathbf{k})|^2=\frac{\Xi_\alpha^2\hbar q}{2V\rho u_\alpha}\left(N_{q\alpha}+\tfrac{1}{2}+\frac{\delta N_{q\alpha}}{2}\right), \quad (2)$$

where for longitudinal waves $\alpha=l$, $\Xi_l=\Xi_d+\Xi_u\cos^2\theta$, and for transverse waves $\alpha=t$, $\Xi_t=\Xi_u\sin\theta\cos\theta$, θ being the angle between $\mathbf{q}$ and the z (longitudinal) axis of the constant-energy ellipsoids. The quantity u_α is an average velocity for acoustic waves of polarization α, and V and ρ are the volume and density of the material, respectively.

[1] P. A. Wolff, Phys. Rev. **95**, 1415 (1954).
[2] W. Shockley, in *Proceedings of the International Conference on Semiconductor Physics, Prague, 1960* (Czechoslovakian Academy of Sciences, Prague, 1961), p. 81.
[3] G. A. Baraff, Phys. Rev. **133**, A26 (1964).
[4] T. N. Morgan, Bull. Am. Phys. Soc. **2**, 265 (1959).
[5] E. M. Conwell, Phys. Chem. Solids **8**, 234 (1959).
[6] This was first pointed out by F. Seitz, Phys. Rev. **76**, 1376 (1949). Also, W. Shockley, Bell System Tech. J. **30**, 990 (1951), notes that for very high-energy electrons the loss to acoustic modes again "makes an appreciable contribution."
[7] C. Herring and E. Vogt, Phys. Rev. **101**, 944 (1956).