Positron annihilation in Ge(Li) detector from line shape of single-escape peak*

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The line shape of the single-escape peak of high-energy γ rays has been investigated. A 30-cc Ge(Li) detector was irradiated with 2614.5 and 2753.9-keV γ rays from ^{208}Tl and ^{24}Na, respectively. The observed broadening of the single-escape peaks was attributed to the Doppler shift of annihilation radiation, and, by unfolding the instrumental resolution the momentum distribution of the electrons in a germanium crystal was deduced. In addition, an upper limit of 5.4×10^{-3} for the ratio of the probabilities for positron annihilation with a K-shell electron to that with an electron from other atomic shells was determined.

INTRODUCTION

The study of positron annihilation in matter has been used as a tool to provide direct information on the momentum distribution of electrons in different materials. It has been shown that in the interior of the electronic structure of an atom the penetration of the positron wave function is small,[1–4] and that valence electrons give the largest contribution to the annihilation process. If both annihilating particles are at rest and free at the time of annihilation, the theory of the lowest-order process predicts the emission of two photons with equal energy, $E=m_0c^2$, in opposite directions. Since the electron-positron center-of-mass system is in general not at rest, conservation of momentum and energy requires a slight departure from collinearity and the unequal distribution of the energy between photons.

Quantitatively the Doppler shift in energy is given by the relation

$$\Delta E \approx \tfrac{1}{2} k_{\parallel} c\,, \tag{1}$$

where $k_{\parallel}$ is the pair center-of-mass momentum component parallel to the direction of emission. Since positrons are thermalized before annihilation,[5] $k_{\parallel}$ is that of the atomic electron. With recent improvements of high-energy-resolution Ge(Li) systems the Doppler shift of annihilation radiation has become a readily measurable quantity,[6,7] and the observation of the broadening of the annihilation line at 511 keV has been used in determining the electron momentum distribution in several materials.[7] The same mechanism of Doppler shift produces an observable broadening of the single-escape (SE) peaks seen when high-energy photons irradiate a Ge(Li) detector.[8]

When a photon with energy $E_0 \geqq 2m_0c^2$ enters the crystal, there is a certain probability that an electron-positron pair will be created in the Coulomb field of a nucleus. The available kinetic energy is shared between the two particles in a continuous way and is absorbed in the crystal in a short time, mostly through the Coulomb interaction with the surrounding electrons. In approximately 2.3×10^{-10} sec (the lifetime of positrons in germanium[9]) the thermalized positron annihilates with a nearby electron, creating two photons, the energies of which may be added to the already absorbed kinetic energies of the pair. The energy sum of the photons is constant and therefore, if both photons are absorbed in the crystal, or both escape, the total absorbed energies (E_0 or $E_0-2m_0c^2$), will also be constant and the Doppler shift ΔE cannot be observed. The linewidths of the corresponding full-energy (FE) and double-escape (DE) peaks are then the result of the finite-energy resolution of the detection system only.

If only one annihilation photon is absorbed, the total absorbed energy E_{abs} is equal to $E_0-(m_0c^2 \pm \Delta E)$ and the Doppler shift can be detected. As a result of many annihilation events on electrons with different momenta, the SE peak is broadened symmetrically about the energy $E_0-m_0c^2$. This energy distribution is distorted by the finite-energy resolution of the system, and the final line shape is broader than the line shape of a FE peak of the same absorbed energy.

By applying an unfolding program to remove the finite-resolution effects it is possible to find the momentum distribution of the valence electrons in germanium crystals under the operating conditions. In principle, by comparing the line shapes of FE, SE, and DE peaks, it is also possible to obtain some information on the higher positron momenta involved in the annihilation process and also on the space distribution of the positron wave function.

MEASUREMENTS AND RESULTS

In our experiment the energy spectra of 2753.9 and 2614.5-keV γ rays from ^{24}Na and ^{208}Tl have been investigated. The decay schemes of both nuclides are rather simple and the energies are sufficiently high so as to produce large numbers of electron-positron pairs.

A 30-cc true coaxial Ge(Li) detector with a high-stability pulse-height analysis system was used. The source-to-detector distances were adjusted to prevent an excessive counting rate from disturbing the energy resolution of the apparatus. The dead time of the analyzer was less than 5% in both cases. The stability of the system was checked regularly and using peak-centroid calculations it was found to be better than ±27 eV in long-term operation.

Nevertheless, to minimize possible errors caused by different measuring conditions, the line shapes of FE, SE, and DE peaks were measured simultaneously. To provide the necessary spread in channels the bias of the postamplifier was adjusted to just below the DE peaks in both cases. A 8192-channel pulse-height analyzer with a 100-MHz clock was used and the energy dispersion was adjusted to be 0.144 keV per channel. Zero and gain stabilizations were controlled with the DE and FE peaks, respectively. At the settings used on the stabilizer no measurable amount of line broadening has been observed. No change in gain of the main amplifier was made during the experiment, and only the bias of the postamplifier was changed when the sources were interchanged. All electronic units, except the preamplifier, were placed in a cabinet modified to provide temperature stability of ±0.1 °C.

Our results are shown in Fig. 1. The measured square of the full width at half-maximum, W^2, is plotted as a function of the absorbed energy. Full and open circles represent our data for FE and DE peaks, respectively, while triangles show W^2 for SE peaks. The full circles for energies below 2.6 MeV are the experimental points obtained with known γ transitions[10] from a ^{226}Ra source. The

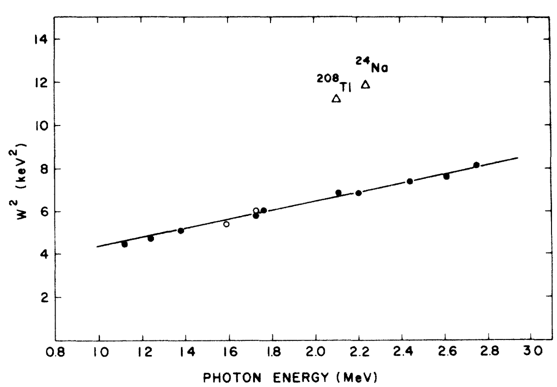

FIG. 1. W^2 as a function of energy. Triangles show W^2 for SE peaks, while open and full circles correspond to DE and FE peaks, respectively.

experimental conditions were the same as in the measurements of the ^{24}Na and ^{208}Tl spectra. The statistical errors are smaller than the dimensions of the circles and of the triangles.

The W^2 of a monoenergetic absorption line for a Ge(Li) detector can be expressed with good approximation as

$$W^2 = W_e^2 + 5.57\epsilon FE . \quad (2)$$

Here W_e is the electronic contribution to the line broadening and ϵ is the energy needed per charge pair produced. F is the Fano factor, defined as the variance of the number of pairs in the process of creation only. Up to the present, measurements have shown that within the experimental accuracies, the Fano factor is independent of the γ-ray energy E and therefore the W^2 should be a linear function of the energy. Using the least-squares method, we fitted the experimental points, omitting the SE points, with the straight line shown in Fig. 1.

In Fig. 1 it is clearly seen that the SE peaks show the broader distributions if compared with the FE peaks of the same absorbed energies. If the straight line is taken as a reference, then the mean value of the broadening of the SE peaks, ΔW, is 0.82 ± 0.04 keV. The error is estimated from the differences between two measurements. The error of the fit to the straight line has been neglected.

DATA ANALYSIS

The observed energy spectrum of any physical process is affected by the finite-energy resolution of the measuring system and is spread over all channels in amounts dictated by the response function of the detector. The resulting energy spectrum is broadened. Since the expected average broadening in our positron annihilation experiment and the system resolution are both of the order of 1 keV, a computer unfolding program had to be applied.

If the detector response function is known, the broadening of the ideal spectrum can be calculated from the expression[11]

$$f(n') = \frac{\sum_n I(n) g[n, w(n), n']}{\sum_m g[n, w(n), m]} . \quad (3)$$

Here $f(n')$ is the number of counts in the channel n' observed in the experiment, $I(n)$ is the number of counts in the channel n of an ideal distribution, and g is the detector response function whose value is measured at only a finite number of discrete points. $w(n)$ represents all the energy-dependent parameters which determine the final line shape of the absorption line.

If one does not know the ideal distribution $I(n)$, the procedure begins with some arbitrary distribution $I'(n)$ and then the calculated $f'(n')$ can be compared with the observed one using the standard χ^2

test

$$\frac{\chi^2}{i-1} = \sum_{n'} \frac{[f(n') - f'(n')]^2}{f(n')}, \tag{4}$$

where i is the number of fitting points. New values of $I'(n)$ can then be calculated by multiplying the old ones by the ratio $f(n')/f'(n')$. This iterative procedure is continued until a reasonable $\chi^2/(i-1)$ value is found.

The analytic expression for germanium-detector response function has been found to be rather complicated,[12] but fairly good results were obtained by fitting the experimental line shapes with the sum of a Gaussian and an arctangent function. The latter has to be used to fit the flat nonzero plateau on the low-energy side of the peak, which seems to be indeed an integral part of the line shape. To eliminate uncertainties, the response function is usually determined experimentally by observing the shape of the full-energy absorption peak of the monoenergetic photons. Unfortunately, this procedure could not be used in our case since the physical processes contributing to the FE peak and to the escape peaks are not the same.

Let us consider the simplest case when Doppler shift ΔE is equal to zero, which means that annihilation quanta have the same energy. Created inside the crystal, they can be either completely absorbed via photoelectric effect or they can escape. In the latter case, prior to their escape, photons can be Compton scattered and the recoil electron will be absorbed in the crystal and will be added to the kinetic energy of the created electron-positron pair. For forward-scattering processes the amount of energy transferred to the crystal in a Compton collision can be of the order of the energy resolution of the detector and, therefore, this process can cause tails on the high-energy sides of the escape peaks. The ratio of the heights of the tails of DE and SE peaks will be approximately 2 : 1, and one would expect that, for given detector dimensions, their heights relative to the corresponding escape peaks do not change with energy. The final detector response function for escape peaks can be approximated by a Gaussian function superimposed on both the low- and the high-energy tails.

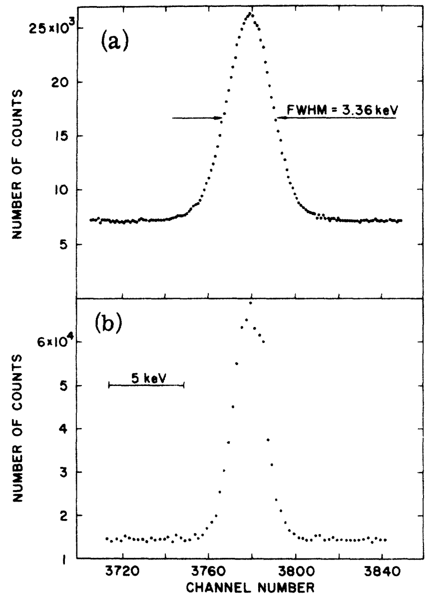

FIG. 3. (a) Pulse-height distribution of 2614.5-keV SE peak. (b) Ideal pulse-height distribution of 2614.5-keV SE peak after the unfolding computer program has been applied.

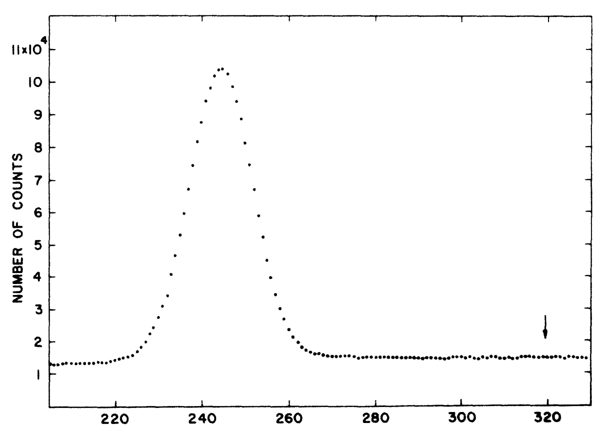

FIG. 2. Pulse-height distribution of 2614.5-keV DE peak. The arrow points to the expected position of annihilations with K-shell electrons.

In Fig. 2 the line shape of the 2614.5-keV DE peak is shown and the high-energy tail can be seen. The SE peak of the same γ ray is presented in Fig. 3(a) and no severe asymmetries can be observed. Since the final shape of the SE peak is the result of the superposition of detector response functions for the different energies arising from Doppler broadening, we can conclude that these response functions themselves are also symmetrical. This situation occurs when the low-energy and the high-energy tails have the same height. In that case, the detector response function for the differential energy interval of the ideal SE peak can be well approximated by a Gaussian function added to a constant background. If this is assumed, the set of parameters $w(n)$, introduced in Eq. (3), reduces to the full width at half-maximum, W, and a constant factor only. W can be found with the help of the straight line shown in Fig. 1. Over any particular SE peak region we used a constant value for W, since it changes very little in the small energy interval where the fitting procedure was performed.

We began the iterative procedure by taking in Eq. (3) for the $I(n)$ the values of $f(n')$ for the same channel numbers. After 12 iterative steps $\chi^2/(i-1)$ became almost constant, in both cases. For ^{24}Na and the ^{208}Tl values of $\chi^2/(i-1)$, 1.12, and 0.89 were obtained. The ideal SE curve resulting from the stripping procedure can be seen in Fig. 3(b). For statistical reasons, each point is obtained by adding the counts in two adjacent channels. With this procedure the small oscillations, which are frequently generated by deconvolution procedures, and which appeared at the far end of our distribution, were averaged out.

The resulting spectrum in Fig. 3(b) was converted to the electron-space momentum density function $\rho(k)$ by the following relation[7]:

$$\rho(k)=\frac{\text{const}}{k}\left(\frac{dI(n)}{dn}\right)\frac{dn}{dk}\,. \qquad (5)$$

The final results are shown in Fig. 4(a). The experimental points, represented with open and full circles, are obtained from the analysis of ^{208}Tl and ^{24}Na SE peaks, respectively. Statistical errors are smaller than the dimensions of the circles, and the total error of our fit can be roughly estimated from the values of $\chi^2/(i-1)$. The full line is a visual fit to the experimental points and the dashed line is taken from the data of Stewart,[13] normalized to the same area. Stewart has shown that it is useful sometimes to express the results within the function

$$N(k)=4\pi k^2\rho(k)\,, \qquad (6)$$

which serves to magnify the higher electron momenta contributing to the annihilation process. The plot of this function is shown in Fig. 4(b).

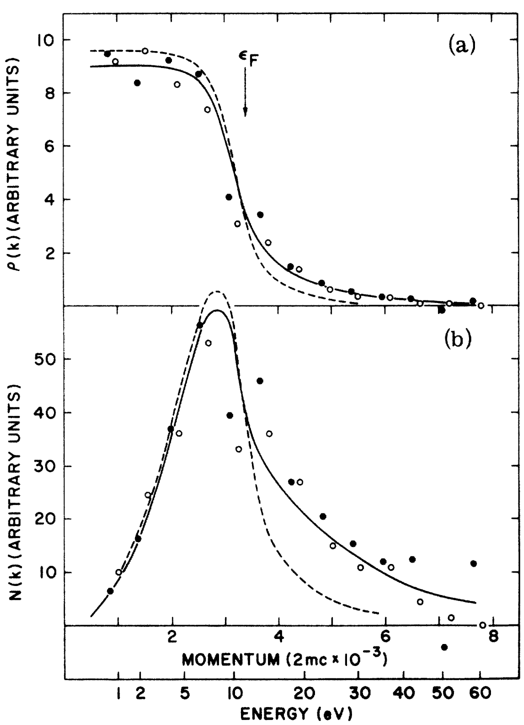

FIG. 4. (a) Momentum space density of germanium electrons involved in annihilation processes. Open and full circles are the experimental data derived from the line shapes of 2753.9- and 2614.5-keV SE peaks from ^{24}Na and ^{208}Tl, respectively. Statistical errors are smaller than the dimensions of the circles. Full lines are the visual fit on our experimental data while the dashed line is taken from the experiment of Stewart and normalized to the same area. ϵ_F is the Fermi cutoff energy. (b) Momentum distribution of germanium electrons.

DISCUSSION

For the free-electron-gas theory, $\rho(k)$ is expected to be constant out to near the Fermi cutoff energy ϵ_F, where it falls to zero. Such a distribution has been found to be characteristic of many metals. Positron-annihilation experiments, performed on semiconductor crystals,[2,13-15] yielded angular correlation curves very similar to those obtained for metals. This indicates that the energy gap of less than 1 eV between the valence and the conduction band is sufficiently small that the electron wave functions can be well approximated with the Bloch waves of metallic electrons. The flat shape of our experimental curve supports this theory and is in agreement with the data reported by others.

As can be seen from Fig. 4(a) our results differ from the data of Stewart[13] in that the relative contribution of the high-momentum component is greater in our case. In more recent experiments performed on oriented germanium and silicon crystals[14,15] the data were interpreted by considering the valence electrons in detail and no claim was made for precision on treating the core component. The tailing we have observed on the high-momentum side is rather large and one could wonder if it is not of somehow instrumental origin. In principle, the instability of the system could produce such tailings. However, in our experiment the stability of the system has been controlled during the time of data collection, and it was found to be better than ±27 eV. The effects of this electronic drift are negligible. The obtained Fermi cutoff energy ϵ_F is in agreement with that reported by Stewart, which shows that the electronic drifts did not alter the line shape of the single-escape peak.

The method applied in the procedure of unfolding the SE peaks for the instrumental resolution is based on the assumption that the detector response function is Gaussian. If another function were used, the resulting total spectrum would be different. Therefore it is necessary to check the reliability of our assumption. Since we were not able to do this experimentally we fitted the ^{208}Tl full-energy peak with a Gaussian function imposed on a constant background. In this case an arctangent function has to be added in order to fit the flat plateau on the lower-energy side. For the energy interval $E_0 \pm 4\sigma$ we obtained a value of 1.46 for $\chi^2/(i-1)$, which shows that our assumption of the Gaussian shape of the response function is valid. We therefore believe that the obtained total spectrum is mainly the result of the electron-momentum distribution in the germanium lattices and that the instrumental effects are negligible. The observed tailing is probably due to annihilations with core electrons. The relative contribution of core annihilations is influenced substantially by the density of certain types of lattice defects, most notably vacancies and dislocations. These defects can trap positrons and cause a large reduction in core annihilations. In the case of the good Ge(Li) detector, we are dealing with a sample of extremely low lattice-defect density, and the core fraction is certainly maximized. The dislocation densities in Stewart's and in our samples might differ by an appreciable amount, which could explain the difference in the fraction of core annihilations.

We also tried to obtain some information on positron annihilation with deeply bound atomic electrons. If the annihilation takes place on K-shell electrons, the two emitted photons will share the energy $2m_0c^2 - B_K$, where B_K is the binding energy of K electrons in germanium. The energy balance occurs as the emitted x ray, which is absorbed in the crystal, since there is a small probability for it to escape. Therefore, the other peak is expected at the energy $E - 2m_0c^2 + B_K$. For germanium $B_K \approx 11$ keV and this peak should be well resolved from the DE peak, but during the time of data collection we did not observe any structure in the expected energy region. Therefore, for the ratio of the annihilations on K-shell electrons and on other atomic electrons we were able to give only an upper limit 3.6×10^{-4}. That means that the ratio of the probabilities for the positron annihilation with a K-shell electron and with an electron from other shells is $\leq 5.4\times10^{-3}$.

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