Mobility of positrons in polyethylene

A. P. Mills, Jr., E. M. Gullikson, L. Pfeiffer, and W. S. Rockward*
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
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We have measured the positron drift velocity versus electric field in polyethylene by observing the Doppler shift of the annihilation photons. We find that the average positron mobility computed using the internal electric field (dielectric constant $=2.26$) is $\mu=10.3\pm1.5$ cm^2 V^{-1} sec^{-1}. This value agrees with the measurement of Brandt and Mourino [Bull. Am. Phys. Soc. **24**, 72 (1979)], but disagrees with that of MacKenzie and Ghorayshi [Solid State Commun. **55**, 125 (1985)], possibly indicating a sensitivity to the precise composition of the sample.

Knowledge of how positrons move in solids is useful for understanding the trapping of positrons at defects and the emission of positrons and positronium from surfaces.[1] The positron diffusion constant may be obtained from studies of positron annihilation in small particles of powders[2] and alloys[3] or from positron beam experiments.[4] Alternatively, the positron mobility μ may be obtained by measuring the displacement of positrons[5] or their drift velocity[6] in an electric field. If the positrons are at a temperature T, the diffusion constant and the mobility are related by $D=\mu kT/e$.[7] At low electric field E, the drift velocity is $v_d=\mu E$.

The positron mobility has been measured in Ge,[6] Si,[8] and compared in the case of Ge to surface experiments using slow positron beams.[9,10] The positron mobility has also been measured in polyethylene[11–14] and in a number of other solid and liquid insulators.[14,15] In order to extend the mobility measurements to insulators with small diffusion lengths[16] we have assembled a Doppler shift apparatus capable of detecting drift velocities ten times smaller than before. We have made our first measurements on polyethylene, $(-CH_2-)_n$, because of the discrepancy that exists among the experiments of Refs. 11–14. The mobility of positrons in polyethylene is useful for estimating the size-to-lifetime ratio of the positronium-forming spur[17] based on measurements of the positronium formation probability versus electric field.[18]

The energy of the 511-keV annihilation photons was measured with an intrinsic Ge detector (35% efficiency) having a 1.65-keV resolution and a 29.5% photo fraction at 514 keV. The pulses from the detector were amplified and shaped with a 1.5-μsec time constant and sent to two single channel analyzers. The energy windows, A and B, from the latter were about 2 keV wide and were set on the lower and upper halves of the 511-keV photopeak, respectively (see inset of Fig 1). The lower level of B was set below the upper level of A and the window output of B was required to be in coincidence with the upper level of A. The amplifier gain was adjusted automatically by a long time constant (>100 sec) feedback loop that sensed the difference in the A and B count rates. These two rates were recorded in a two-input 4096 channel multiscaler. The channel number at which the counts were being added swept continuously up and down with a 1-msec dwell time at each channel. An analogue voltage corresponding to the channel number, a symmetric sawtooth wave with a 4.096-sec period, was amplified to a ±1000-V signal and its inverse, which were applied to the two sides of the sample.

The polyethylene sample was two pieces $2\times2\times0.092$ cm painted with 0.001 cm thick spots of aquadag (colloidal carbon) in the center of each side. A 20-μCi ^{22}Na source sealed between 0.0007 cm thick Al foils was placed between the two samples. The samples were clamped between two thin stainless-steel plates with 1-cm-diameter holes in the centers. (The holes ensured that the aquadag would not be scraped off the sample by the clamping plates.) The two plates served as the contacts to the outer aquadag spots on the samples, and the two inner spots were grounded by a wire attached to the Al foil. The source was located on the axis of the cylindrical Ge detector crystal, (57 mm diam by 60 mm long), 80 mm from its face. The detector was behind a Pb collimator hole 38 mm diameter by 50 mm long. Solid-angle corrections to our Doppler shift measurements are less than a few percent and will be neglected.

A Doppler shift of the 511-keV annihilation photons causes equal and opposite changes in the count rates A and B. The instrument was calibrated by turning off the gain control feedback loop, introducing $\pm0.067\%$ changes in the amplifier gain using a fixed resistor attenuator, and noting the corresponding changes $\Delta N_{\rm cal}$ in the count rates A and B. Figure 1 shows a calibration run taken after the mobility experiment was over. The positron drift velocity is computed from the count rates N_A and N_B using

$$v_d = \tfrac{1}{2}\alpha[(1-N_A/\langle N_A\rangle)-(1-N_B/\langle N_B\rangle)], \quad (1)$$

where $\langle N\rangle$ is the average number of counts. The proportionality constant is given by

$$\alpha = 2\times c\times6.7\times10^{-4}(N/\Delta N_{\rm cal}), \quad (2)$$

where c is the velocity of light and the factor of 2 arises because only the positron is moving while the electrons are stationary on the average. The data of Fig. 1 give

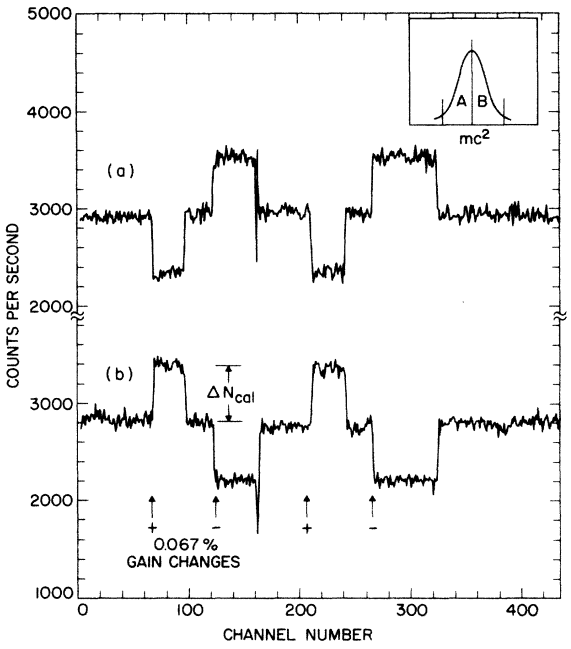

FIG. 1. Count rates in the lower and upper halves of the window set on the 511-keV annihilation line (see inset). The step changes were caused by switching an attenuator in steps of 0.067%. The gain control was disabled for this calibration run.

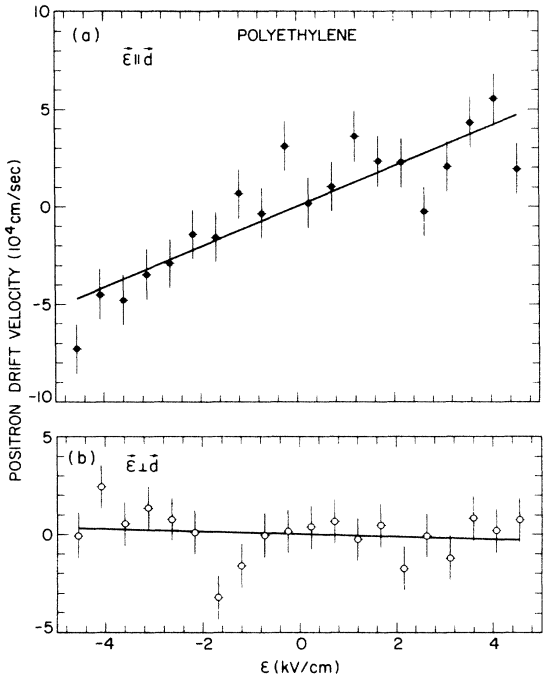

FIG. 2. Positron drift velocity measurements vs applied electric field in polyethylene. Note that the electric field is defined to be the voltage difference across the sample divided by the product of its thickness and the dielectric constant. (a) Electric field parallel to the line d joining the sample and the detector. (b) Independent set of data obtained with the electric field perpendicular to d.

$\alpha=2.0\times10^{-8}$ and a second measurement gave $\alpha=2.2\times10^{-8}$. We use the average of these two values multiplied by 1.05 to account for 5% of the positrons annihilating in material other than polyethylene.

Because of the common power supplies and ground leads in the apparatus, there might be some systematic effects in our measurement of $\Delta N/N$ vs E. To eliminate such effects odd in the electric field E, we reversed the two high-voltage leads on the sample before each $\approx$1-day-long run. We corrected for effects due to changes in the photopeak count rate, caused for example by the positronium formation probability varying with electric field,[18] by combining our v_d measurements from the two halves of the photopeak, A and B [see Eq. (1)]. The data from the several runs (10 days of data) were averaged together, taking into account the sign of E, and this average is shown in Fig. 2(a). The electric field computed using the 0.092-cm sample thickness and 2.26 for the dielectric constant varied from about -5 to $+5$ kV/cm. The Doppler effect due to the positron motion in the electric field is obvious. To check that we are indeed measuring the positron mobility in Fig. 2(a), we obtained an equal amount of data with the sample electric field directed perpendicular to the line joining the sample and the detector. The result shown in Fig. 2(b) is consistent with zero. The least-squares-fitted slope in Fig. 2(a) (the line is constrained to pass through the origin) is $\mu=(10.34\pm1.04)$ cm^2 V^{-1} sec^{-1}, with a χ^2 per degree of freedom $\chi^2/\nu=29.97/19$. Allowing for the possibility of a $\pm10\%$ calibration error, our measurement becomes $\mu=(10.3\pm1.5)$ cm^2 V^{-1} sec^{-1}. The fitted slope in Fig. 2(b) is -0.66 ± 0.92 with $\chi^2/\nu=21.51/19$.

The χ^2 per degree of freedom for the fit in Fig. 2(a) is not very good; a random sample of points distributed about the line gives no better fit only 3.7% of the time. Possibly we are seeing a line center shift proportional to the absolute value of the electric field (even in E) superimposed on the (odd in E) drift velocity Doppler effect. In Fig. 3 we display the odd and even parts of the data. Figure 3(a) contains the odd part of the parallel data of Fig. 2(a) and should be due to only the drift velocity Doppler shift. The fitted line has the same slope as in Fig. 2(a), but with a good $\chi^2/\nu=8.20/9$. The perpendicular data of Fig. 3(b) also has the same slope as in Fig. 2(b), consistent with zero, with $\chi^2/\nu=12.05/9$. The even part of the data of Fig. 2 is displayed in Fig. 3(c). A two-parameter fit to the average of the parallel and perpendicular data gives a slope $\mu=-1.48\pm1.38$ and $\chi^2/\nu=9.25/8$. We conclude that the even effect due to the changing total annihilation energy of the positrons is small, amounting to less than a 0.1-eV shift at $E=5$ kV/cm. According to Bisi *et al.*[18] at low E, the positronium formation probability f decreases linearly with a slope that would make $f=0$ at about 60 kV/cm. Assuming a 30% Ps formation probability, this shift implies a less than 3-eV difference between the average annihilation photon energy of Ps and of free positrons in polyethylene.

We now compare our value for the positron mobility in polyethylene with earlier measurements. Brandt and

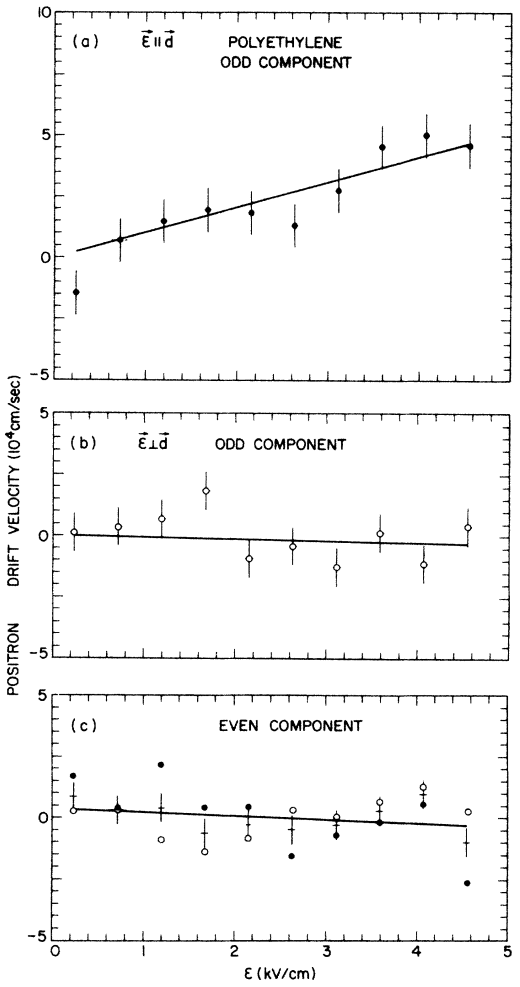

FIG. 3. Odd and even components of the data of Fig. 2.

Mourino[11] measured the displacement of the positrons caused by an electric field in polyethylene. They give a value of ≈ 10 cm^2 V^{-1} sec^{-1} and state that the mobility increases with increasing crystallinity of the sample. The density of our sample was found to be 0.924(1) g cm^{-3}. Such a low density is characteristic of a branched chain polymer with an amorphous content of about 40%.[19] On the other hand, MacKenzie and Ghorayshi[14] measured the Doppler shift versus E in polyethylene and find $\mu = (27.7 \pm 2.4)$ cm^2 V^{-1} sec^{-1} for their low-density sample. The agreement between our result and that of Brandt and Mourino allows us to conclude that there is no large systematic difference between mobilities measured using the displacement[5] and the Doppler shift[8] methods. Some caution is required, however, in interpreting the positron-drift experiments[5] because of the unexplained negative drift results of Ref. 13. Further caution is also required in comparing these results with each other because in Refs. 14 and 15 and possibly in other places the electric field is computed neglecting the dielectric constant of the sample.[20] By using such a convention, our mobility value would become (4.6 ± 0.7) cm^2 V^{-1} sec^{-1}. Finally, in Refs. 14 and 15 the positron mobility has been corrected for the fraction of the positrons that form positronium. Applying such a correction to our data would give a mobility of about 7 cm^2 V^{-1} sec^{-1}. This value is about four times smaller than that of Ref. 14. We conclude that polyethylene samples may exhibit large variations in their positron mobilities, and that the experiment should be repeated using well characterized samples.

One should note that the drift velocities measured in Fig. 2(a) are more than ten times smaller than those previously measured. Consequently our improved Doppler shift technique should make it possible to measure positron mobilities in a number of interesting materials. A useful conclusion regarding the spur mechanism[17] follows from our present measurement. At the 60-kV/cm electric field corresponding to a 100% change in the Ps formation probability,[18] the positron drift velocity would be no more than 6×10^5 cm/sec. Assuming a spur radius of 100 Å allows us to conclude that the spur lifetime[17] is less than 2×10^{-12} sec.[21]

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*Present address: Grambling State University, Grambling, LA 91245.

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