Annihilation Gamma-Ray Spectra from Positron-Molecule Interactions

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We report measurements of annihilation gamma-ray spectra from the isolated two-body interaction of room-temperature positrons with a range of chemical species. The Doppler broadening of these spectra provides information about the wave functions of the annihilating particles. For a variety of hydrocarbons, we observe linewidths of approximately 2.2 keV (FWHM), while for perfluorocarbons the widths are approximately 2.8 keV, as compared with 1.7 keV for molecular hydrogen. The implications of these results for a number of physical problems are discussed.

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An important tool for studying the interaction of positrons with matter is the measurement of the momentum distribution of the annihilating positron-electron pairs. This can be accomplished either by studying the angular correlation of the annihilation radiation or by high-resolution gamma-ray spectroscopy. The former technique has been used extensively to study positron annihilation in solids, liquids, and dense gases [1]. High-resolution gamma-ray spectroscopy has been used to study positron annihilation in low-density hydrogen gas [2]. It has also been used to measure the spectra of annihilation gamma rays of astrophysical origin [3].

In this Letter, we report the first study of the linewidths of gamma rays produced when positrons annihilate with electrons in a variety of molecular species. Studies of the annihilation rates of low-energy positrons interacting with molecular gases indicate that this interaction can lead to the formation of long-lived positron-molecule resonances [4–7]. The nature of such resonances is an important but unsolved problem, and the measurements presented here help to identify the electronic and positronic states involved in these resonances. As we discuss below, these measurements are also relevant to gamma-ray astronomy [8,9] and to positron ionization mass spectrometry [10,11].

The experiments were performed in a modified three-stage Penning trap, shown schematically in Fig. 1. Positrons were trapped and cooled by collisions with a nitrogen buffer gas. Details of the operation of the trap are described in Ref. [12]. Differential pumping maintains a nitrogen pressure of 2×10^{-6} torr in the third stage of the trap, yielding a positron annihilation time of 20 s or longer. Once trapped, the positrons cool to room temperature with a characteristic e-folding time of approximately 0.8 s. Since positronium formation is energetically forbidden for cold positrons, and they do not diffuse to the confining electrodes on the time scale of the experiment, direct annihilation with nitrogen and residual impurities is the only annihilation mechanism. A cold surface (typically at $-5\,^{\circ}$C) was used to reduce the pressure of residual impurities.

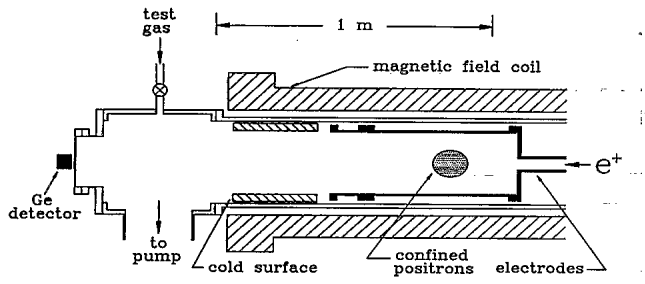

FIG. 1. Schematic diagram of the experimental apparatus.

The substance under test was introduced into the trap as a gas or vapor. The pressure of the test gas ($<1\times10^{-5}$ torr) was adjusted so that the resulting positron annihilation time was much shorter than the annihilation time on the nitrogen buffer gas.

The gamma-ray spectra were measured using a high-purity germanium detector located approximately 130 cm from the confinement region (see Fig. 1). The detector was calibrated using ^{137}Cs and ^{57}Co test sources, with gamma-ray lines at 662 and 122 keV, respectively. As shown by the inset to Fig. 2, the calibration peaks can be fitted with a combination of a Gaussian function and a small step function, which models the effects of multiple scattering, convolved with the same Gaussian. The detector resolution at the 511-keV annihilation energy was typically 1.75 keV (FWHM), estimated by interpolation between the calibration lines.

Spectra were accumulated using a multichannel analyzer (channel width, 0.25 keV) over repeated cycles consisting of a 6-s positron filling, a 1-s cooling, and a 4-s measurement phase. This sequence ensured that the measured spectra were predominantly from the annihilation of room-temperature positrons.

A typical annihilation spectrum is presented in Fig. 2. This includes contributions from the background radiation spectrum, which consists of a continuum with a superimposed annihilation peak. For each data run, the background spectrum was measured separately with the positron beam switched off. The scaled background data were fitted with a Gaussian plus a second-order polynomial (see Fig. 2) and subtracted from the data spectrum. The total number of counts in the corrected peak was in the range 5000–25 000. In Fig. 3, we show the corrected

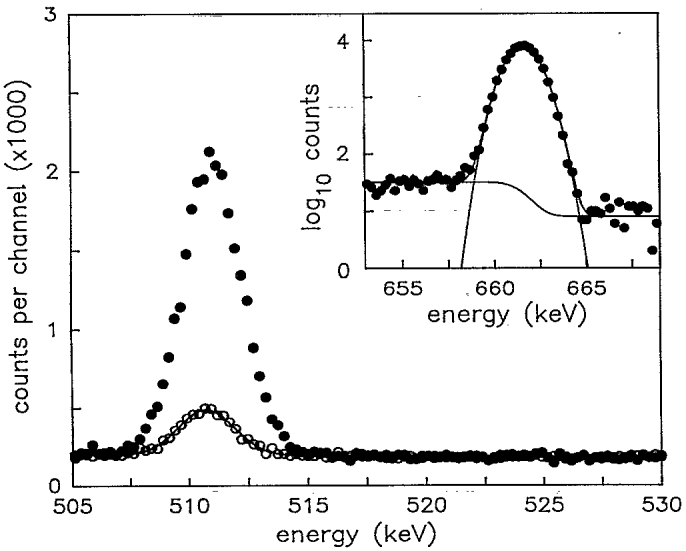

FIG. 2. Annihilation spectrum of positrons in *n*-hexane. The data were accumulated in 4150 cycles, each consisting of a 6-s filling, 1-s cooling, and 4-s measurement phase. Raw data (●) and scaled background (○) with a fitted Gaussian plus second-order polynomial. Inset: The 662-keV calibration line from ^{137}Cs with fitting function consisting of a Gaussian and a Gaussian-convolved step function.

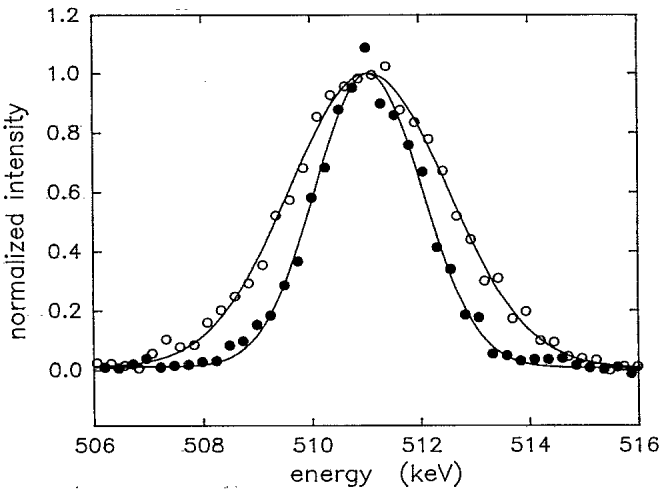

FIG. 3. Background-corrected annihilation spectra, with fitted Gaussians, for hydrogen (●) and sulfur hexafluoride (○). The data are normalized to the peak values of the fitted curves.

spectra for hydrogen and sulfur hexafluoride, representing the extremes of the range of linewidths measured.

The data can be fitted with a Gaussian plus a small convolved step, as for the calibration lines. Since the corrected spectrum is the convolution of the detector response with the annihilation line shape, deconvolution of the data would yield a nearly Gaussian line of width ΔE given by $\Delta E^2 = \Delta E_t^2 - \Delta E_d^2$, where ΔE_t is the measured total linewidth and ΔE_d is the detector resolution. Using this method, we obtained the annihilation linewidths for a number of chemical species. These results are summarized in Table I. The uncertainties reflect the scatter in the data from different runs, where available. The linewidths for toluene and *n*-dodecane have been increased by a measured correction of 0.17 keV to account for a non-Gaussian detector response during these particular runs.

Table I also includes data for annihilation on the nitrogen buffer gas and the residual impurities present in the third stage of the trap, for which the combined linewidth is 2.36 keV. The annihilation time of the confined positrons was 20 s or longer before the introduction of the test gas and was reduced to approximately 3.5–4 s by the presence of the test gas. Thus the contribution from the nitrogen and impurities is expected to have only a small effect on our measurements for the hydrocarbons. However, the linewidth for hydrogen may be slightly smaller than reported here, and the linewidths for the perfluorocarbons and SF_6 may be slightly larger than those quoted.

The measured linewidths of the annihilation spectra show distinct trends for different chemical families. The linewidth for H_2 is considerably narrower than that of any other molecule studied. Our measurement agrees with the only previous H_2 measurement in dilute gas [2] and with theory [14]. The spectra for the hydrocarbons all have linewidths in the range 2.15–2.3 keV, while linewidths for perfluorocarbons are all significantly higher, falling in the range 2.75–2.85 keV [15]. This suggests that, within a chemical family, the annihilations involve electrons in equivalent states. In view of the simi-

TABLE I. Annihilation linewidth, ΔE (FWHM), and the normalized annihilation rate, Z_{eff}, for a variety of molecular gases, including hydrocarbons and perfluorocarbons.

Molecule	Formula	ΔE (keV)	Z_{eff}
Hydrocarbons			
Benzene	C_6H_6	2.16 ± 0.10	18400[a]
Toluene	C_7H_8	2.15	155000[a]
n-Hexane	C_6H_{14}	2.29 ± 0.05	98000[a]
n-Dodecane	$C_{12}H_{26}$	2.19	1780000[b]
Perfluorocarbons			
Carbon tetrafluoride	CF_4	2.84 ± 0.05	54[a]
Perfluoro-hexane	C_6F_{14}	2.76 ± 0.05	535[a]
Perfluoro-octane	C_8F_{18}	2.81 ± 0.05	1064[a]
Other gases			
Sulfur hexafluoride	SF_6	2.92 ± 0.05	97[c]
Hydrogen	H_2	1.72 ± 0.11	15[c]
Nitrogen	N_2	2.24 ± 0.05	29[c]
Nitrogen fill gas and impurities		2.36	

[a]Reference [7].
[b]Reference [13].
[c]Reference [5].

larity among measured hydrocarbons, it appears likely that these states correspond to electrons in the C-H bond, which is the only bond common to all of the hydrocarbons studied. The compact structures of CF_4 and SF_6 suggest that the most accessible, and therefore the most likely, annihilation sites in these molecules are the fluorines. Furthermore, the similarity between the linewidths of these molecules and the other perfluoro compounds studied indicates that the fluorines are the most likely annihilation sites in all of these compounds.

Also listed in Table I are the values of the corresponding positron annihilation rates, expressed in terms of the parameter Z_{eff} [16]. The large values of Z_{eff} observed for the hydrocarbons have been interpreted as evidence for the occurrence of long-lived positron-molecule resonances [4–7]. Recent studies have shown that there are large, systematic differences between the annihilation rates of positrons in hydrocarbons and those in the analogous perfluorocarbons [7]. A remarkable feature of the hydrocarbon results shown in Table I is that, while Z_{eff} varies by 2 orders of magnitude, the measured linewidths are essentially the same. The similarity of the linewidths indicates that the electronic and positronic states in the resonances are also similar. Thus, the observed differences in Z_{eff} within a particular chemical family are likely to reflect differences in the formation or dissociation rates of the resonances.

There has been extensive work on positron annihilation in solids, liquids, and high-density gases, using angular correlation techniques to study the spectra from positrons slowing down from relativistic energies. However, these measurements often show complications due to competing annihilation channels that are avoided using the positron trapping techniques described here. For molecular solids and liquids, a component is often seen in the annihilation spectrum in a regime of energies greater than that attributed to positronium formation and condensed-matter effects [17,18]. We note that, in liquid hydrogen and liquid nitrogen [17], the widths of these components agree quite well with our results in dilute gases. Similarly, the hydrocarbon linewidths reported here agree with the widths seen in liquid hexane and in solid polyethylene, while our perfluorocarbon linewidths agree with data from Teflon [18], a solid perfluorocarbon. The fact that we are now able to study isolated positron-molecule interactions with precision should provide added insight into the annihilation processes occurring in these condensed-matter systems. For example, the question arises as to whether the long-lived resonances believed to occur in low-pressure gases may also exist in solids and liquids, where they could be true bound states.

Astrophysical measurements of gamma-ray spectra show a narrow 511-keV annihilation line ($\Delta E < 5$ keV) from a compact source located near the galactic center with a superimposed diffuse component ($\Delta E < 3$ keV) which appears to arise from annihilation on the interstellar medium [9]. At present, the energy resolution of these measurements is not sufficiently high to assign accurate values to these linewidths. However, much more accurate measurements will become available within the next decade [19,20]. At present, the minority constituents of the interstellar medium are uncertain, but are believed to include dust grains and molecular gases, possibly including large aromatic molecules [21]. The data presented here may help to identify the annihilation medium in these astrophysical processes in that we have shown that the linewidth has a significant dependence on the particular chemical species involved.

The identification of annihilation sites may also be valuable in interpreting data from positron ionization mass spectrometry (PIMS) [10]. Ionization of large molecules by positrons can produce very different fragmentation patterns from those obtained by electron impact [11]. By identifying the annihilation sites in such interactions, we may be able to resolve some of the outstanding questions in PIMS such as the differences between "pickoff" ionization, which occurs slightly above the positronium formation threshold, and annihilation following positron attachment, which occurs for positrons with energies below this threshold [11].

In summary, we have studied the spectra of annihilation gamma rays from two-body, positron-molecule interactions in the isolated environment of a Penning trap. The data can help to identify the specific annihilation sites of the positrons in these molecules. This information should be useful in the formation of theoretical models of positron-molecule resonances. It can also be expected to be relevant to the understanding of the ion spectra produced in positron ionization mass spectrometry. Finally, the experimental techniques described here should permit a systematic study of gamma-ray spectra in neutral and ionized gases directly relevant to the interpretation of astrophysical annihilation spectra.

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