Correlation of Positron Age and Pair Momentum in Radiation-Induced Voids in Molybdenum

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(Received 8 March 1976; revised manuscript received 6 June 1976)

Dual-parameter measurements of the positron annihilation energy and time spectra in neutron-induced voids in Mo show that the mean lifetime does not decrease monotonically with increasing momentum of the center of mass of the positron-electron pair. At intermediate values of the momentum, there is a minimum in the mean lifetime which deepens with increasing void size; this effect may be due to positronium formation and trapping in the voids.

Positron techniques show promise in the characterization of radiation-induced voids[1] and in investigations of their nucleation and growth processes.[2] Measurements of lifetimes and momentum spectra[3] support the concept of trapping of positrons at internal surfaces with the formation of a bound state with properties which are almost independent of void size.[4] The expectation of a simple monotonic association of increasing lifetime with decreasing momentum is not fulfilled, however, in measurements of the correlation of positron age with annihilation energy. Instead we find that intermediate values of the momentum are associated with a minimum in the mean lifetime, the depth of the minimum increasing with the void diameter. These results suggest that the long-lived trapped state may be complex despite the evidence of a simple exponential decay over several decades.

The Mo samples were cylinders, 3 mm in diameter and 2 mm in length, with a purity better than 99.99%. The void properties of these samples were characterized by Brimhall, Jimonen, and Kissinger[5] using transmission electron microscopy studies.

A ^{22}Na positron source was sandwiched between two 1.0-μm-thick nickel foils and then by the Mo samples. The momentum of the electron-positron pair was deduced from the Doppler shift of the energy of the annihilation photons, as measured with a Ge(Li) spectrometer with a resolution of 1.45 keV for the 514-keV line of ^{85}Sr. The positron age at annihilation was measured by a time spectrometer using a pair of Ne-104 plastic scintillators (2 in. $\times$ 2 in.) with a resolution of 290 ps when measured with a ^{60}Co prompt source. The dispersion of the energy spectrometer was set at 440 eV/channel and that of the time spectrometer at 81.9 ps/channel. The source strength of 3 μCi was dictated by the acceptable count rate of 10 000 counts/sec in the Ge(Li) spectrometer, resulting in very low rates of 6000 counts/sec in the start detector and 600 counts/sec in the stop detector of the time spectrometer. A total triple coincidence rate of about 3 counts/sec was obtained with a peak-to-random rate ~ 10^5. Details of the two-parameter spectrometer will be published elsewhere.[6,7]

Figure 1 shows the single-parameter time spectrum for specimen 1, containing 10^{17} voids/cm^3 with a mean diameter of 52 Å. The mean lifetime of the long-lived component is 456 ps, a typical lifetime for voids in Mo. All of our measured time spectra are well represented by two components but the shorter one is undoubtedly a composite of annihilations in dislocations, grain boundaries, and untrapped positrons. The time spectra were acquired in a system using routing and digital spectrum stabilization; the results are summarized in Table I.

Figure 2 presents two-parameter data in the form of the centroids of the time spectra plotted as a function of the relevant energy interval. Alternatively, the x axis may be considered as one

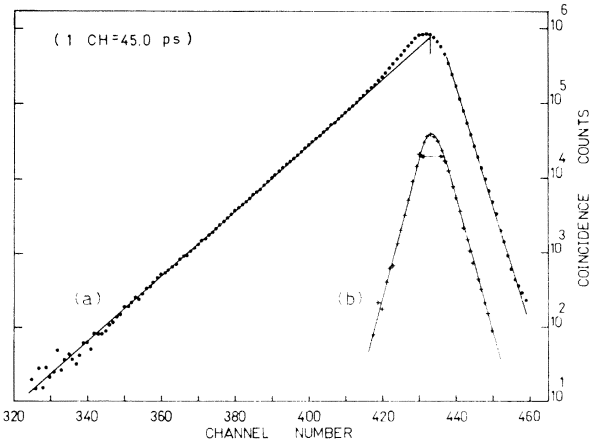

FIG. 1. (a) The annihilation-time spectrum for void specimen 1. The mean lifetime of the long-lived component is 456 ps. (b) Prompt spectrum taken with a ^{60}Co source: FWHM is 290 ps.

TABLE I. Transmission electron microscopy and positron characteristics of neutron-irradiated molybdenum specimens.

Specimen number	Irradiation temperature (°C)	Average void diameter (Å)	Void concentration (cm^{-3})	Void component Mean lifetime (ps)	Void component Intensity (%)
1	450	52	10^{17}	456 ± 3	74 ± 2
2	450	20–25	$(2-4)\times 10^{16}$	453 ± 3	47 ± 2
3	400	Not resolved	?	453 ± 3	47 ± 2
4	350	Not resolved	?	415 ± 3	46 ± 2

Cartesian component of the center-of-mass momentum. The distribution of the time centroids has the expected symmetry about zero momentum, i.e., about an energy of m_0c^2, where m_0 is the electron rest mass and c is the velocity of light. The data in Fig. 2(a) are for Mo stock which was not subjected to neutron irradiation. The departure from a horizontal straight line is clearly of statistical significance and is attributed to incomplete annealing of the dislocation structure. The feature of primary interest is the occurrence in Fig. 2(b) of deep minima in the mean lifetime at about 3 keV from m_0c^2. Such minima were observed in all the specimens listed in Table I, becoming less clearly defined with a reduction in the irradiation temperature. The minima do not appear in the unirradiated Mo and no structure of any kind was found in annealed Ni or Cd measured at room temperature.

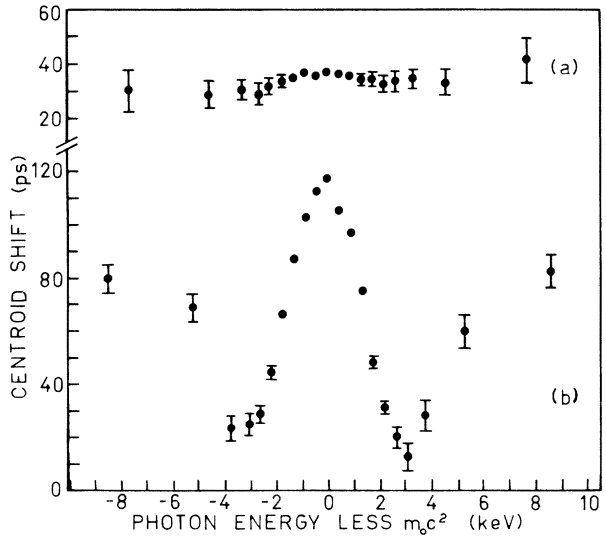

FIG. 2. The position of the centroids of the time spectra as a function of the annihilation energy. The zero of time is not true zero but an arbitrary reference channel. (a) Unirradiated Mo specimen; (b) Mo specimen of Fig. 1(b).

It is not clear whether the occurrence of the minima seen in Fig. 2(b) is a consequence of trapping in large vacancy aggregates (microscopic or submicroscopic voids) or if they merely become apparent with large voids because of the reduction in competitive trapping by dislocations. As shown by Brimhall, Simonen, and Kissinger,[5] the increase in void diameter is accompanied by a large decrease in dislocation density.

Although we do not resolve three separate components in the time spectrum, it is useful to assume their existence for the following discussion. τ_1 is the mean lifetime of positrons annihilating in a free state, τ_2 for those in traps such as dislocations or grain boundaries, and τ_3 for those in large vacancy aggregates. There is abundant evidence[1-3] that $\tau_1<\tau_2<\tau_3$ and that the difference is substantial in each case for Mo specimens. The momentum spectrum for each of these states, as measured by angular correlation, can be represented by a single Gaussian function to reasonable accuracy.[8] When these functions are convoluted with the response function of the Ge(Li) detector, the Gaussian approximation must be improved further (aside from a slight asymmetry which is not relevant). The widths of the Gaussians are in the reverse order to their mean lifetimes, i.e., $w_1>w_2>w_3$, and again the differences are substantial.[8] The centroid shift from true zero for the composite time spectrum associated with momentum p is simply $\Delta\tau = I_1(p)\tau_1 + I_2(p)\tau_2 + I_3(p)\tau_3$, where $I_i(p)$ is the intensity of the ith component of the momentum spectrum. It is readily shown that this is a decreasing function of $|p|$ because of the Gaussian weighting functions; hence we have the "expectation" referred to in the first paragraph.

Since the data show unexpected structure only when large vacancy clusters are present, we assume that the state formed by trapping positrons in these defects must be the source of that struc-

ture. One possibility is that the trapped state is not pure but is a mixture in which some of the positrons are trapped as positronium atoms while other are "free." There is fragmentary experimental evidence[9-11] that the trapped state in voids has some positroniumlike properties and recent calculations[12] also indicate that such a state is favored energetically. Contrary evidence comes from the lack of sensitivity to strong magnetic fields which should cause ortho-para conversion.[1] It is also argued that, since positronium cannot form in the bulk metal and there is no other medium present, there is no "source" of this state. However, this argument does not apply to external metallic surfaces, a fact which has been demonstrated convincingly in the study of excited states of positronium.[13] Internal surfaces in voids are widely accepted as good approximations to external surfaces.[4]

We have found in two-parameter measurements in a polymer[6] that a plot of the time centroid versus energy has a deep minimum at m_0c^2. This occurs because of an intense component from self-annihilation of parapositronium, combining low momentum with a short mean lifetime. It is possible that the structure in Fig. 2 could arise from the superposition of spectra from trapped positronium and trapped positrons, provided that the central dip in the former spectrum does not have the same width as the peak in the latter. The postulate of a complex trapped state may seem incompatible with the excellence of the fit of the long-lived component in Fig. 1 with a single exponential but it is not possible to exclude an additional weak component of comparable lifetime from analysis of such data. It has been suggested by R. N. West, and independently by D. N. Lowy, that each trapped positron may spend part of its time "free" and part in positronium with frequent conversions occurring on reflection at the void surfaces. An attractive feature of this model is that it would not require that the long-lived state be anything other than a single exponential.

The extreme sensititivy of time spectrometers to magnetic fields makes it impractical to test the positronium hypothesis by the usual technique of ortho-to-para conversion in strong fields.[1] Furthermore we could not be sure that such a test would give an unequivocal answer since it depends on the momentum distributions from ortho- and para-annihilations being appreciably different. The usual approximation that self-annihilation dominates for para-positronium while pickoff dominates for ortho-positronium is possibly invalid for positronium in voids.

The work is supported by the National Research Council of Canada. We would like to thank H. E. Kissinger of Battelle Memorial Institute for providing the void specimens and to thank R. N. West and J. H. Evans for helpful discussions.

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