Pressure Dependence of Positron Annihilation Rates in the Alkali Metals

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The annihilation rate of positrons in the alkali metals has been measured at room temperature for pressures up to 55 kbar. The observed departure from simple predictions of the free-electron gas model are explainable on the basis of changes in the fraction of core annihilations. This interpretation is not, however, consistent with the changes in the Doppler-broadened line shape.

Theories of annihilation rates in metals are tested commonly by comparing theoretical and experimental values for Al and the alkali metals, all of these having angular-correlation curves like those expected for a free-electron gas. Each metal is characterized by a value r_s which is the radius of the sphere containing one valence electron. The measured rates, when plotted against r_s, fall on a smooth curve which closely resembles the predictions of a free-electron model, modified by enhancement factors which are applied to electron density at the positron site.[1]

A much simpler test of theory should result from subjecting one of the alkali metals such as sodium to a very high pressure so that the value of r_s could be altered over a wide range without any appreciable change in the ion core. The very high compressibility of the alkali metals makes it possible to effect useful density changes with simple pressure systems. We can, for example, increase the density of potassium by more than 40% by subjecting it to a pressure of 50 kbar.[2] In addition, the alkali metals have unusually long positron lifetimes which makes precise measurements possible with a system of moderate time resolution.

Our pressure system uses the opposed-anvil technique, originated by Bridgman, in the form developed by Vaisnys and Montgomery.[3] We use Carboloy 883 anvils and a 6S Isomica sealing ring. The opposing faces of the anvils have a diameter of 0.300 in. and apply pressure on a flat annular sealing ring with a thickness of 0.031 in. The sample consists of a pair of disks of 0.13 in. diameter and 0.012 in. thickness. These are spaced by a source-sandwich arrangement consisting of about 5 μCi of ^{22}Na between two aluminum foils of thickness 0.0005 in. The cylindrical pellet, consisting of source and sample disks, fits tightly in the aperture of the annular sealing ring. All fabrication and assembly is done in a dry–inert-atmosphere glove box.

The timing system has no unusual features except that the large scintillators, $1\frac{3}{4}$-in.-diam cylinders of NE-102 with a length of 2 in., limit the resolving time to about 490 psec. True coincidences were obtained at a rate of about 1000/hr in a typical run, and 10 h of counting were required to obtain the desired precision using slope determinations. It would be preferable to use centroid-shift analyses, but pressure systems of the opposed-anvil type do not lend themselves to pressure cycling, a basic requirement if one were to employ the centroid-shift method with confidence.

The data in Fig. 1 show that λ, the annihilation rate, is not a universal function of r_s but that

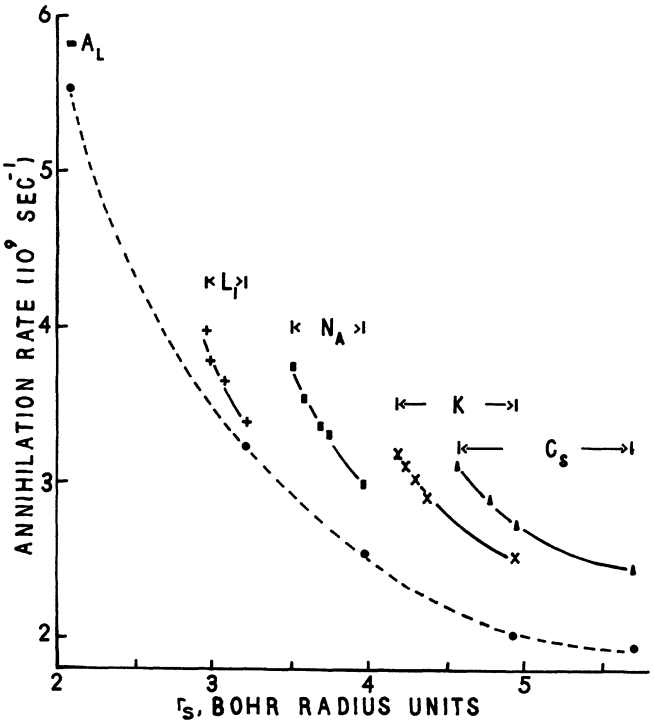

FIG. 1. Annihilation rates at various pressures up to 55 kbar. The data points along the solid curves are measured total annihilation rates. The dashed curve represents the rate attributed to valence electrons alone on the basis of angular-correlation data. The standard deviation of all points is less than 2%.

TABLE I. Percentage of annihilations assigned tentatively to core electrons in the alkali metals at different pressures.

Metal \ Pressure (kbar)	0	20	50
Li	5	7	8.5
Na	15	16.5	20
K	20	22.5	25
Cs	20	25	29.5

there is a different relationship between these parameters for each of the metals studied. The rates at normal pressures agree closely with published figures.[4] The four solid curves are simply visual fits to the sets of data points for the four alkali metals. In each case the data extrapolate towards very high annihilation rates at high pressure (small r_s), and this is most marked in Na and K, the two metals which are the closest approximation to a free-electron gas. There is incentive here to extend the measurements to higher pressures, but this will require extensive modifications to our high-pressure apparatus. The aluminum result is for a single-crystal sample of 99.9999% purity which was annealed and etched prior to measurement with a system of higher resolution (330 psec, fullwidth at half-maximum).

There are at least two reasons why one should not make a simple comparison of the observed annihilation rates with theory. Annihilations with core electrons are expected to contribute significantly to the total, and this component should have a different pressure dependence from the valence-electron component. Even if it were possible to extract the core contribution satisfactorily, we would still have to modify the horizontal scale to allow for the excluded volume effect, i.e., the valence electrons are not distributed over the same volume as the positrons because the latter are repelled by the ion cores.

We have attempted to account for the core contribution by the technique of Terrell, Weisberg, and Berko[5] using the more recent data on angular correlation from the University of North Carolina group.[6] This provides the estimates of core fraction listed in column 2 of Table I. The data in the other columns are based on the additional assumption that the valence annihilation rates for all the alkali metals should depend smoothly on r_s as indicated by the dashed curve in Fig. 1.

In order to observe the predicted change in the core fraction more directly, we have attempted

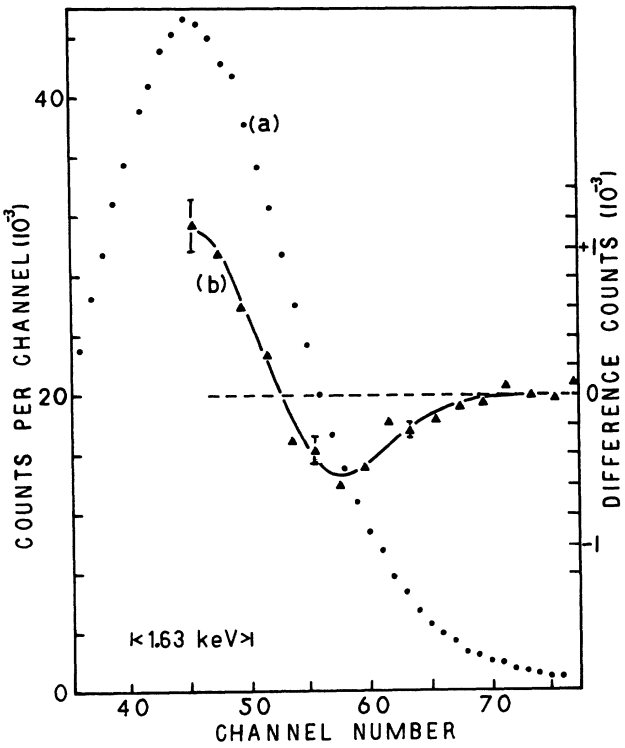

FIG. 2. (a) Annihilation line shape in K at normal pressure using a Ge(Li) spectrometer (left-hand scale). (b) Difference curve for the annihilation lines in K at 0 and 55 kbar normalized to equal areas (right-hand scale).

to measure the change in momentum distribution with pressure for both Na and K. This was done by using a Ge(Li) spectrometer which had a resolution of 2.0 keV on the 514-keV line of ^{85}Sr. Digital stabilization of gain and the zero of the analog-to-digital converter were used so that a reliable comparison of line shapes could be made by studying their difference curves. Figure 2(a) shows the shape of the annihilation line in K at normal pressure, and Fig. 2(b) shows the difference between it and a similar curve obtained at a pressure of 55 kbar and normalized to the same area.

The most striking feature of these data is that the difference curve comes to zero at much smaller values of the momenta than do the individual spectra. The same observation is made with data on K at 25 kbar and for Na at 50 and 25 kbar. Although the statistics are not satisfactory on any single set of data, the general reproducibility of the effect is convincing. In no case do we see any evidence of enhancement of the high-momentum components which would be

expected to occur if the core fraction increased with pressure. The only effect apparent is that the central parabola becomes broader with pressure, as one would expect on the basis of the increased mean density of valence electrons. The statistical reliability of the data does not justify using them as a test of free-electron theory in this connection.

We are planning to check the entirely unexpected line-shape results using a Ge(Li) spectrometer of much improved resolution and efficiency. The implication of the present results, namely that the core fraction is independent of pressure, is so difficult to accept that we have refrained from comparing our measured annihilation rates with theory.

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Resolved "Fine Structure" in the Magnetic Resonance of a Localized Moment in a Metal*

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The first observation of the resolved fine-structure splitting of a localized moment in a metal is reported in the EPR spectrum of single-crystal dilute Au:Gd alloys. A theory of line narrowing of fine-structure lines from spin-spin couplings and localized-conduction electron exchange is developed. The narrowing distorts the observed resonance line shape, and is essential to the extraction of the fine-structure splitting parameter: $b_4 = -20 \pm 1$ G.

We report the first observation of a resolved crystal-field splitting in the electron paramagnetic resonance (EPR) of a localized moment in a metal. Dilute Au:Gd single crystals were used for our measurements. For low temperatures and specific angles, the spectrum exhibits two resolved resonance lines. At other angles, where only one line is resolved, a strong angular variation in linewidth is observed. We attribute the former behavior to resolved, the latter to unresolved, fine structure. Dynamic effects, originating with a Gd spin-spin interaction arising from the Ruderman-Kittel-Kasuya-Yosida indirect exchange, and the Gd conduction-electron relaxation strongly affect the form of the spectrum. The distortion of the line shape (substantial narrowing) can be described by a population-weighted difference in stochastic transition matrices, arising from these interactions, appropriate to fine-structure resonance splittings. The concentration, temperature, and angular variation of the spectrum enable us to extract the functional behavior, and explicit values, for the transition rates, as well as the value for the fine-structure splitting parameter. We fit the spectrum by a fourth-order crystal-field parameter $b_4 = -20 \pm 1$ G, and by an isotropic g factor $g = 2.05 \pm 0.01$.

Electron-spin-resonance measurements were performed on single-crystal Au:Gd samples, grown in the form of cylinders, with nominal Gd concentrations of 1000, 300, and 150 ppm. Complementary measurements were performed on powder samples with nominal Gd concentrations of 1000, 500, 150, 75, and 35 ppm. The measurements were conducted at 3 and 0.8 cm wavelength, and as a function of angle and temperature in the liquid-He range. The magnetic field was rotated in a plane perpendicular to the cylinder axis. The spectra at 3 cm exhibit the following features:

(1) At 4.2°K a "single" line is observed for