in light of our own experience with the James and Coolidge wave functions.

ACKNOWLEDGMENTS

We wish to thank Professor Jan Korringa and Professor A. Yoshimori for their many detailed suggestions. We have discussed our results and our interpretations with many people and we wish to thank them all, particularly R. McWeeny, K. Ohno, and A. Fröman. The calculations were carried out on the ALWAC-III computer of the Uppsala Quantum Chemistry Group with the indispensable assistance of K. Appel and F. Bystrand. We are grateful to Professor P. O. Löwdin for the use of the facilities of the Uppsala Quantum Chemistry Group where most of this work was carried out.

PHYSICAL REVIEW VOLUME 130, NUMBER 1 1 APRIL 1963

Positronium Decay in Teflon. Influence of Lattice Transitions

G. Fabri, E. Germagnoli, and G. Randone

Laboratori Centro Informazioni Studi Esperienze, Milano, Italy

(Received 26 November 1962)

The decay of positronium in Teflon has been investigated at temperatures between −200 and +280°C and in greater detail in the region of lattice transitions around room temperature. It is found that the long lifetime τ_2 increases sharply in the interval between 20 and 50°C, while the probability of formation of positronium shows a drop around 20°C. Evidence of the reversibility of the lattice transitions results from the decay features of positronium.

THE decay of positrons in Teflon has been investigated by several authors[1] with the help of different techniques and in a wide range of temperature, up to 300°C. Teflon is considered to be a suitable material for checking the validity of the theory of pickoff annihilation of positrons bound in orthopositronium, because both the τ_2 lifetime and the probability of positronium formation were found to be smooth functions of temperature. This seems to indicate that the well-known temperature effects can be rather accurately described in terms of the "excluded volume" effect.[2]

However, remarkable differences in the values of the τ_2 lifetime around room temperature were found with different samples and are reported in the literature. The occurrence of lattice transitions involving changes of density (at 19 and 30°C) might be thought to be responsible for these differences, though a search for changes in the τ_2 decay due to the lattice transitions was reported to be negative.[3]

In the measurements described here, differently processed specimens from a number of suppliers were used, the τ_2 lifetime at 20°C proving different from specimen to specimen, in the range between 1.8×10^{-9} sec and 3.3×10^{-9} sec. Moreover, it was noticed that τ_2 increases remarkably after short thermal treatments such as the ones usually performed to obtain commercial Teflon in the shape of rods or plates. Systematic measurements of positronium decay were consequently carried out with a method already described,[4] the specimens being in the shape of cylinders, 10 mm long and 8 mm in diameter, obtained with pure powder[5] compacted at room temperature. Pure powder was chosen for the final measurements because it exhibits the shortest τ_2 lifetime at 20°C, according to preliminary results.

Figure 1 shows the τ_2 lifetime vs temperature for such specimens, and the percentage of positrons decaying through τ_2 is given as a function of temperature in

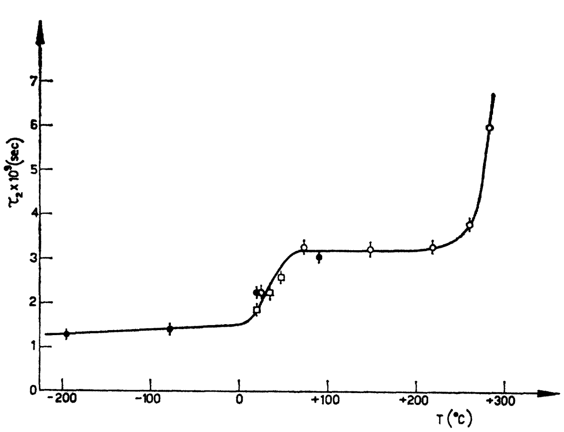

FIG. 1. The long lifetime (τ_2) of positronium vs temperature. The results obtained with three specimens are plotted.

[1] R. E. Bell and R. L. Graham, Phys. Rev. **90**, 644 (1953); R. L. de Zafra and W. T. Joyner, *ibid.* **112**, 19 (1958); A. Bisi, A. Fasana, E. Gatti, and L. Zappa, Nuovo Cimento **22**, 266 (1961).

[2] W. Brandt, S. Berko, and W. W. Walker, Phys. Rev. **120**, 4 (1960).

[3] G. P. Furukawa, R. E. McCoskey, and G. J. King, J. Res. Natl. Bur. Std. **49**, 273 (1952).

[4] C. Cottini, G. Fabri, E. Gatti, and E. Germagnoli, J. Phys. Chem. Solids **17**, 65 (1960).

[5] Kindly supplied by Firm Montecatini.

Fig. 2. It is apparent that τ_2 nearly doubles when the temperature crosses the interval of the triclinic $\rightarrow$ hexagonal lattice transition in the proximity of room temperature and levels off until the higher temperature transition at about 275°C is reached. Conversely, the percentage I_2 of positronium decaying through τ_2 shows a peculiar drop near 20°C and sharply increases around 275°C. It will be noticed that the quoted I_2 values are lower than the one reported in the literature (about 30%). The usual percentage (30%) of the τ_2 decay was observed with samples of commercial Teflon, for which τ_2 is 3×10^{-9} sec, the scatter of experimental values from specimen to specimen being about 15%, and neither τ_2 nor I_2 being dependent critically on temperature in the interval of the transitions.

The curves of Figs. 1 and 2 are only partially reversible, in the sense that both τ_2 and I_2 are left unchanged with respect to the values given in these figures only if the specimens have been heated at temperatures much lower than 275°C and slowly cooled down. As an example, the results obtained with one specimen undergoing a simple thermal cycle are reported in Table I.

TABLE I. Data from temperature-induced changes in the decay of positronium.

Run No.	Thermal treatment	Measuring temperature (°C)	$\tau_2\times10^9$ (sec)	I_2 (%)
1	none	−196	1.28±0.03	21±2
2	none	−78	1.43±0.03	22±2
3	none	20	2.25±0.03	19±2
4	6 h at 90°C;	90	3.07±0.03	13±2
5	slow cooling down to 20°C (0.3°C/min)	20	2.22±0.03	21±2
6	1 h at 100°C; slow cooling down to 20°C (0.06°C/min)	−196	1.27±0.03	23±2

Treatments at higher temperature, on the contrary, are responsible for large differences in the decay features of positronium. Roughly speaking, $\tau_2 \simeq 3\times10^{-9}$ sec and $I_2 \simeq 30\%$ at room temperature, provided Teflon has been heated at 275°C or at higher temperature. Some tendency towards the behavior of unheated Teflon has been observed after specimens had been annealed or

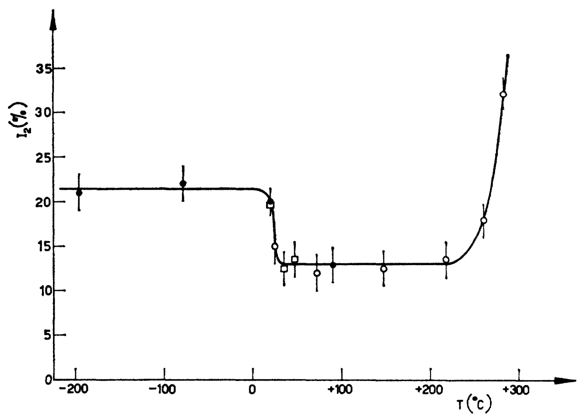

FIG. 2. The intensity of the τ_2 component vs temperature. The results obtained with three specimens are plotted.

slowly cooled down, though the extent of recovery has not been investigated in detail.

It is worthwhile to point out that a similarity exists between the present results and the ones obtained from infrared spectroscopy,[6] an absorption peak at 625 cm^{-1} having been observed at temperatures above 19°C and being absent below 19°C: the intensity of the peak was found to increase up to 90°C. Also dimensional changes due to the low-temperature transitions appear to recover[7] after reasonably short thermal treatments (typically 60 h around 30°C). This is fairly consistent with the present results, though it is not suggested that the above-described behavior of positronium in Teflon is simply explainable in terms of changes in density. Actually a comparison between the present data and the analogous ones, obtained with other materials whenever changes in density occur,[8] seems to suggest that the dependence of τ_2 on temperature observed here is far too large to be accounted by the small change in density (1.23%) that occurs in the room-temperature lattice transition of Teflon. Moreover, the observed drop of I_2 represents a rather anomalous feature.

[6] H. E. Moynihan, J. Am. Chem. Soc. **81**, 1045 (1959).
[7] F. A. Quinn, Jr., D. E. Roberts, and R. N. Work, J. Appl. Phys. **22**, 1085 (1951).
[8] See, for instance, H. S. Landes, S. Berko, and A. J. Zuchelli, Phys. Rev. **103**, 828 (1957).