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RESONANCE ANNIHILATION OF POSITRONS IN CHLORINE AND ARGON*

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The recent discovery of the "shoulder"¹⁻³ in the lifetime spectra of positrons in argon has raised a possibility of the existence of resonance in positron annihilation in matter. A further study of lifetime spectra of positrons in chlorine, Cl₂-Ar, and Cl₂-N₂ mixtures has been made. The measurements were carried out by using two time-to-amplitude converters, of which one has a resolution $(W_{1/2})$ of 0.5 nsec and a useful range of 25 nsec, $\frac{4}{4}$ and another a resolution of 1.2 nsec and a useful range of 360 nsec.⁵ These instruments of higher resolution have revealed fine structure in the positron lifetime spectrum. The purpose of this note is to report some preliminary results which indicate the existence of resonance annihilation.

No third component is observed in positron annihilation lifetime spectra in pure chlorine, but a shoulder appears in the second component (Fig. 1). This shoulder also appears in Cl_2 -Ar mixtures but not in Cl_2 -N₂ mixtures. Plots of $\lambda_2(t)$ against t for the lifetime spectra² have been analyzed.⁶ It was found that after the initial rise² $\lambda_2(t)$ oscillates instead of leveling off. The oscillations in the $\lambda_2(t)$ plots for different pressures of argon and chlorine follow a certain systematic pattern. There are at least four peaks to be traced in $\lambda_2(t)$ plots for Cl_2 -Ar mixtures. Two of them can also be observed in $\lambda_2(t)$ plots for pure chlorine and another two for pure argon. Apparently, these peaks are due to the combined effects of the resonance annihilation of free positrons in chlorine or argon at certain energy levels and the non-negligible slowing down time of positrons in chlorine or argon. After some assumptions are made for the initial energy distribution of positrons, the resonance energies are esti-



FIG. 1. Lifetime spectra of positron annihilation in chlorine.

mated to be about 0.9 and 0.7 eV for argon, and 0.5 and 0.3 eV for chlorine⁶ using the cross section observed by Teutsch and Hughes.⁷ The average value of $\lambda_2(t)/p$ at the tail part (*t* large) in chlorine is estimated to be about 0.3 nsec⁻¹ atm⁻¹, which is about 1000 times greater than the Dirac free-electron annihilation rate. The value of $\lambda_2(t)/p$ for t = 0 is estimated to be less than 0.15 nsec⁻¹ atm⁻¹.

The third component only appears in Cl_2 -Ar or Cl_2 -N₂ mixtures with low chlorine content. In Cl_2 -Ar mixtures it starts to appear at a chlorine pressure less than 0.05 atm and in Cl_2 -N₂ mixtures at less than 0.5 atm. The changes of the intensities of three components for positron annihilation in Cl_2 -Ar mixtures at different pressures measured by the wide-range timeto-amplitude converter are shown in Fig. 2. The changes of the intensities for positron annihilation in Cl_2 -N₂ mixtures follow the same pattern but at a higher chlorine pressure.

An entirely new phenomenon is that a new shoulder with a shoulder broadness and chlorine pressure product of the order of 10^{-2} nsec atm appears behind the prompt peak on the top of the larger shoulder when the third component starts to appear for positron annihilation in Cl_2 -Ar mixtures. This shoulder, as well as the first shoulder, is not observed in Cl₂-N₂ mixtures. It was found that the sum of the intensities of the new shoulder and the third component remains practically unchanged even though the intensity of the third component changes considerably. Therefore, the new shoulder and the third component must arise from a single origin, the annihilation of positronium. This suggests the existence of a strong annihilation resonance at a certain energy. As shown before,⁸ the reaction

$$Ps + Cl_2 \rightarrow PsCl + Cl$$

requires a threshold energy of approximately 0.5 eV, and since the annihilation lifetime of PsCl has been calculated to be very short,^{9,10} it is not surprising to find out that a strong resonance annihilation occurs at a positronium energy just above this threshold.

In Cl_2-N_2 mixtures, because of the more efficient thermalization power of nitrogen, pos-



FIG. 2. The intensities (I) of first, second, and third components of positron annihilation spectra in Cl_2 -Ar mixtures.

itronium escapes the resonance region much faster than in a Cl_2 -Ar mixture of the same composition. Therefore, it is expected that the third component appears at higher Cl_2 partial pressure in N_2 and no second shoulder can be detected.

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Force Office of Scientific Research Grant No. 62-398. †Present address: New England Institute for Medical

- Research, Ridgefield, Connecticut. ¹S. J. Tao, J. Bell, and J. H. Green, Proc. Phys.
- Soc. (London) <u>83</u>, 453 (1964).
- ²D. A. L. Paul, Proc. Phys. Soc. (London) <u>84</u>, 563 (1964).
- ³W. R. Falk and G. Jones, Can. J. Phys. <u>42</u>, 1751 (1964).
- ⁴J. Bell, S. J. Tao, and J. H. Green, to be published.
- $^5 J.$ Bell and S. J. Tao, to be published.
- ⁶S. J. Tao, to be published.
- ⁷W. B. **T**eutsch and V. W. Hughes, Phys. Rev. <u>103</u>, 1266 (1956).
- ⁸S. J. Tao, thesis, University of New South Wales, 1964 (unpublished).
- ⁹L. Simons, Phys. Rev. <u>90</u>, 165 (1953).
- ¹⁰R. A. Ferrell, Phys. Rev. <u>103</u>, 1266 (1956).