INFLUENCE OF ABSORBED HYDROGEN ON THE ANGULAR DISTRIBUTION OF PHOTONS FROM TWO-QUANTUM ANNIHILATION IN TITANIUM

J. Wesołowski, B. Rozenfeld and M. Szuszkiewicz

Institute of Experimental Physics, Bolesław Bierut University, Wrocław*

(Received May 30, 1963)

The angular distribution of gamma rays from two-quantum electron-positron annihilation in titanium and in H-Ti system of 1.91 $\frac{H}{Ti}$ concentration has been measured. Curves illustrating the two distributions and normalized to the same height in the centre have been compared.

The curve corresponding to the H-Ti system seems to be broader then the one obtained for pure titanium. The experimental curves have been compared with theoretical Fermi momentum distributions calculated for 2, 3, 4, 5 and 6 free electrons per titanium atom. Usual assumption has been made that positrons are thermalized prior to annihliation and that they annihilate only with electrons of an ideal Fermi gas. From the comparison conclusion may be drawn that the number of free electrons per one titanium atom is not less than 3 in pure titanium and about 6 in the investigated H-Ti system.

The results suggest that the positive ionization of the absorbed hydrogen is almost complete.

1. *Introduction*

Although numerous investigations on physical and chemical properties of exothermic hydrogen-metal systems have been carried out, there is still considerable discrepancy between opinions regarding the nature of the bonds and the state of hydrogen atoms in these systems. It is generally assumed that hydrogen adsorbed in the exothermically occluding transition metals is very largely ionized and that the electrons supplied by the hydrogen atoms enter the unfilled shells of the metal atoms.

It seems that a comparison of free electrons density in pure and in hydrogenated metal may throw light on the structure of the hydrogen-metal system and on the degree of positive hydrogen ionization. As is known, the investigations of the angular distribution of gamma rays from two-quantum electron-positron annihilation may yield informations about the number of free electrons per one atom of the studied material. So far only one attempt, with negative result, was made in order to observe the influence of occluded hydrogen

* Address: Katedra Fizyki Doświadczalnej Uniwersytetu B. Bieruta, Wrocław, ul. Cybulskiego 36, Polska

upon the gamma-pair distribution. Lang and DeBenedetti (1957) investigated the angular correlation of two annihilation gamma rays from pure and palladium and found no gross effect.

In this paper the preliminary measurements of the angular distribution of gammas from two-quantum annihilation in pure and in hydrogenated titanium are described.

2. *Angular distribution of two-photon annihilation radiation*

The angle between two photons resulting from the two-quantum annihilation of the electron-positron pair depends on the momentum of the annihilating particle system at the time of annihilation. The relation between the projection p_z of this momentum (z-component) on a plane and the angular deviation Θ from collinearity of the projections of the quanta directions is as follows (Fig. 1):

$$\sin\frac{\Theta}{2} \approx \frac{p_z}{2mc}.$$

For small angles applied in the present measurements

$$\Theta \approx \frac{p_z}{mc}.$$

Theoretical considerations as well as conclusions drawn from the results of experimental investigations indicate that in condensed mediums, in metals for example, positrons are thermalized in a time of the order of 10^{-12} sec, by two order of magnitude smaller then the shortest observed mean lifetime against annihilation (DeBenedetti *et al.* 1952;

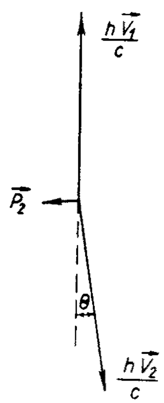

Fig. 1

Garwin 1953; Bell *et al.* 1953; Lee-Whiting 1955; Gerholm 1956). As the Coulomb repulsion prevents the low-energy positrons from appreciable interacting with the inner electrons they eventually annihilate predominantly with the conduction electrons. Consequently, the angular distribution of the two quanta should correspond to the momentum distribution of free electrons.

ACTA PHYSICA POLONICA Vol. XXIV (1963) Fasc. 6

ACTA PHYSICA POLONICA Vol. XXIV (1963) Fasc. 6

Investigations carried out on a number of metals showed a fairly good agreement between the observed angular correlations and the z-component distribution calculated for electrons of an ideal Fermi gas having the same density as free electrons in the investigated metals (Green *et al.* 1955; Stewart 1955; Lang *et al.* 1955).

Thus, the comparison of the observed angular distribution, especially in its parabolic part, with the theoretical Fermi distribution of the z-momentum component yields information about the number n of free electrons per one atom.

The Fermi z-momentum distribution is

$$N(p_z)\,dp_z = \text{const}\,(p_F^2 - p_z^2)\,dp_z,$$

where

$$p_F^2 = h^2 \left(\frac{3n_0}{8\pi}\right)^{2/3}$$

n_0 being the electron density $n_0 = \dfrac{ndL}{A}$

L, d, A denote Avogadros' number, density and atomic mass of the metal, respectively.

The momentum distribution, which is parabolic in shape, may be visualized geometrically and compared with the graphical representation of the angular distribution of the photons.

3. *Experimental apparatus*

The angular distribution of gamma-ray pairs arising from electron-positron annihilation in titanium was measured by means of the coincidence arrangement shown schematically in Fig. 2. The source of annihilation radiation was placed at the middle of the horizontal line joining the two scintillation counters L_1 and L_2. The distance between the source and either detectors was $\approx$ 190 cm. Each counter consisted of a $40 \times 40 \times 40$ mm^3 NaJ(Tl)

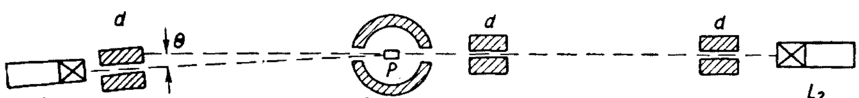

Fig. 2

scintillator mounted on FEU-19 photomultiplier tube. It was experimentally verified that the scintillation detectors showed good linear characteristic in the range of the measured photon energies. The detectors were shielded by blocks of lead d of $10 \times 10 \times 1.5$ cm^3 provided with rectangular collimating slits. Additional lead shield S was placed around the source. Collimator-detector system L_2 remained fixed, while the system L_1 could be rotated about a vertical axis passing though the sample.

The pulses from each detector were passed separately through a preamplifier and a linear amplifier to an one-channel pulse-height analyzer set to select pulses of amplitudes corresponding to the energy range of about half-width of the 0.51 MeV line. Coincidences between selected pulses from the two detectors were counted by a coincidence unit of $\approx$ 1 microsecond resolving time. The application of identical amplitude analyezrs considrrably reduced the background.

The specimens consisted of six titanium discs of 6×0.65 mm^2 between which five positron sources were sandwiched. The positron emitters were prepared by evaporating an aqueous solution of NaCl contaigning ^{22}Na on a thin ($\approx$ 1 mg/cm^2) aluminium foil. Small thickness of the foil and the very thin salt residue secured low self-absorption of the emitter.

From the detector L_2 the radiator is seen at an angle of $\approx$ 2 milliradians in the horizontal direction and of $\approx$ 40 milliradians in the vertical direction. From the specimen the detectors were seen at an angle of about 2 milliradians in the horizontal direction and at about 40 milliradians in the vertical direction. In addition, the apparatus was equipped with a special unit which continually controlled the correct functioning of the arrangement, in particular the position of the gates of the amplitude analyzers.

The rate of double coincidences was measured as function of the angular deviation θ from collinearity of the two annihilation quanta. The coincidence rate was $\approx$ 16 cpm at $\theta = 0$ and $\approx$ 0.25 cpm at $\theta = 15$ milliradians. The individual points were measured with an accuracy of $\approx 2\%$. Since the ratio of true to accidental coincidences was found to be negligible, no correction hase been made for accidental background. No correction for angular resolution of the apparatus hase also been made.

Titanium discs destinated for the investigation of H-Ti system were kindly loaded with hydrogen in the Institute I of Anorganic Chemistry of the Technical University of Wrocław. The hydrogen concentration determined by two independent methods was $1.91\ \frac{H}{Ti}$. Angular correlations for pure and loaded titanium were measured with geometrically identical samples and in identical geometry of the arrangement.

ACTA PHYSICA POLONICA Vol. XXIV (1963) Fasc. 6

4. *Results and conclusions*

Fig. 3 illustrates the results of the measurements of the angular correlation of photons from two-quantum annihilation in pure and in loaded titanium samples. The curves shown in this figure are normalized to the same hight at $\theta = 0$. The angular deviation θ expressed in radians is equal to the z-component of the pair momentum measured in units mc.

It is seen that the half-width of the curve corresponding to the H-Ti system is distincly broader than the half-with of the pure titanium curve. In Fig. 4 the experimental curve for untreated titanium sample is compared with theoretical angular correlation curves calculated on the assumptions that respectively $n = 2$, 3 or 4 electrons per titanium atom contribute to the ideal Fermi electron gas.

The analysis of the experimental curve in the region corresponding to Fermi level (maximum electron momentum) is not possible because of the large-angle tails due to such effects as the excluded volume and annihilation with bound electrons. Only central parts of the distribution, corresponding to lower and medium values of momentum, can be used for deviations not exceeding $\approx$ 4 milliradians the experimental curve follows the theoretical one calculated for $n = 3$. For larger angles the curve becomes broader and at about the half-width intersects the theoretical curve corresponding to $n = 4$. From the course of the experimental curve it may be concluded that the number of free electrons per one titanium atom is comprised between three and four.

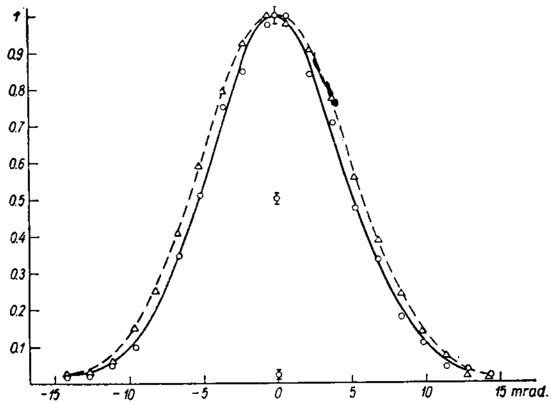

Fig. 3

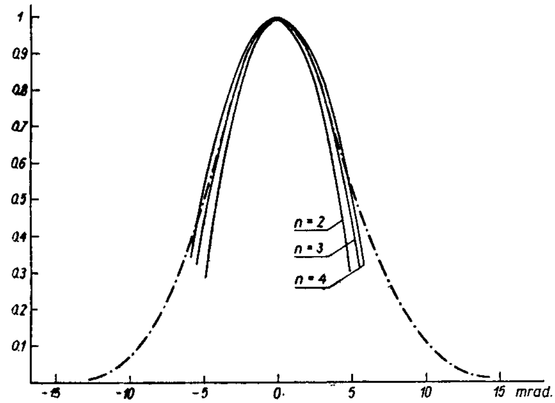

Fig. 4

Titanium is one of transition metals of the Fe group with unclosed $3d$ configuration. The free atom electron configuration of this metal consists of an "argon core" and 4 outer electrons in $3d$ and $4s$ states. Little is known about the state distribution of these outer electrons and on their contribution to the Fermi electron gas. Our results may suggest that beside $4s$ electrons considerable proportion of the $3d$ electrons also enters into the Fermi gas. It can be houped that more precise measurements carried out with better statistic would throw more light on this problem.

In Fig. 5 the experimental curve for H-Ti system is shown together with the two theoretical distribution curves calculated for $n = 5$ and $n = 6$ free electrons per titanium atom.

ACTA PHYSICA POLONICA Vol. XXIV (1963) Fasc. 6

The density change of the specimen caused by the occluded hydrogen has been taken into account.

If annihilation in H-Ti system is assumed to occur chiefly with conduction electrons, then the comparison of the curves may lead to the conclusion that a considirable portion of occluded hydrogen gives up electrons to the free electron gas.

This conclusion supports the view that hydrogen, at least partially, exists in some hydrogen-metal systems in a positively ionized state. The simplified method of analysis used in these preliminary measurements do not allow for precise determination of the fraction of ionized hydrogen atoms; the more so that the lattice expansion caused by the occluded gas may influence the energy distribution of electrons. It can only be stated that the positive ionization of hydrogen is almost complete in the investigated H-Ti system.

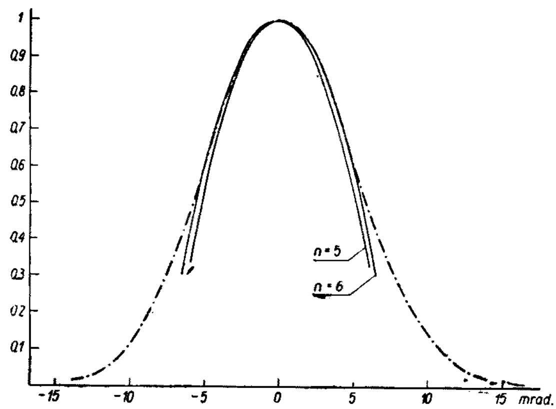

Fig. 5

We would like to express our gratitude to Dr B. Staliński who kindly charged our samples to the high hydrogen concentration.

REFERENCES

Bell, R. E. and Graham, R. L., *Phys. Rev.*, **90**, 644 (1953).
DeBenedetti, S. and Richtings, H. J., *Phys. Rev.*, **85**, 377 (1952).
Garwin, R. L., *Phys. Rev.*, **91**, 1571 (1953).
Gerholm, *Arkiv Fysik*, **10**, 523 (1956).
Green, R. E. and Stewart, A. T., *Phys. Rev.*, **98**, 486 (1955).
Lang, G., DeBenedetti, S. and Smoluchowski, R., *Phys. Rev.*, **99**, 596 (1955).
Lang, G. and DeBenedetti, S., *Phys. Rev.*, **108**, 914 (1957).
Lee-Whiting, G. E., *Phys. Rev.*, **97**, 1557 (1955).
Minton, G. H., *Phys. Rev.*, **94**, 758 (1954).
Stewart, A., *Phys. Rev.*, **99**, 594 (1955).