Magnetic Quenching of Positronium Decay in Water

G. FABRI, G. POLETTI, AND G. RANDONE
Laboratori Centro Informazioni Studi Esperienze, Milano, Italy
(Received 20 December 1963; revised manuscript received 19 February 1964)

Experiments on magnetic quenching of positronium have been carried out in water and the results obtained by two different techniques are compared. The results were not found to agree with theoretical predictions, the starting assumption being that positronium annihilates by "pick-off" as a free atom. Some considerations are made about the anomalous features of magnetic quenching of positronium in water.

1. INTRODUCTION

RECENT literature[1–4] has pointed out the striking behavior of the electron-positron bound system both in water and in ice.

In particular, magnetic quenching data in water at 20°C do not support the idea that the lifetime τ_2 is associated with the annihilation of the positronium free atom. These results are different from the ones found with other materials, by means of both decay measurements,[5] and triple coincidence measurements.[6]

Consequently we thought it useful to reconsider the magnetic quenching in water in more detail, because this phenomenon seems to be *a priori* strictly connected with the characteristics of the electron-positron system in the considered material.

2. PREVIOUS RESULTS

In a previous paper[4] it was shown that the functional dependence of the annihilation probability through the long lifetime on the applied magnetic field does not agree, in water at 20°C, with results found for both Lucite and Teflon.[5]

The experimental data concerning the magnetic quenching experiments, taken from the quoted paper,[4] are reported in Fig. 1. We recall that the technique employed was the one proposed by Bisi *et al.*[5] $R(H)$ is defined as

$$R(H)=\left[\frac{\int_{t_1}^{\infty} I_{2\gamma}(t)dt}{\int_{0}^{\infty} I_{2\gamma}(t)dt}\right]_H \Big/ \left[\frac{\int_{t_1}^{\infty} I_{2\gamma}(t)dt}{\int_{0}^{\infty} I_{2\gamma}(t)dt}\right], \quad (1)$$

$I_{2\gamma}(t)$ being the intensity below the delay curve and t_1 having been selected so that all the decay events involving the short lifetime were excluded from the integrals.

Figure 1 also gives for the sake of comparison two theoretical $R(H)$ curves calculated on the hypothesis of the "pick-off" conversion mechanism of the positronium atom. While the meaning of the solid line will be discussed later, the dashed curve was calculated according to the approximation used by Bisi *et al.* Following this last reference,[5] the functional dependence of $R(H)$ on the magnetic field is given by

$$R(H)=\tfrac{2}{3}+\tfrac{1}{3}\alpha \exp[-(y^2/(1+y^2))(\lambda_1-\lambda_3)t_1], \quad (2)$$

where

$$y=[(1+x^2)^{1/2}-1]/x,\ x=(4M_z/\hbar\omega_0)t\ M_z=(e\hbar/2mc)H_z,$$

is the only nonvanishing matrix element of the perturbation Hamiltonian, $\hbar\omega_0$ is the ground-state fine structure splitting constant of positronium, λ_1 and λ_3 are positronium decay constants *in vacuo*, and α is a rather complicated function calculated by Bisi *et al.* It never differs appreciably from the unity and will not be considered in the following.

Using Eq. (2) and assuming $\lambda_1=8.0\times10^9$ sec^{-1} and $\lambda_3=7.14\times10^6$ sec^{-1}, Bisi *et al.*[5] obtained two $\hbar\omega_0$ values for Teflon and Lucite. These values are in good agreement with the theoretical estimate 8.34×10^{-4} eV, which means that the long-lifetime component of positronium decay has to be attributed to the free-atom annihilation. Analysis of Fig. 1 demonstrates that agreement between Eq. (2) and experimental results cannot be obtained by

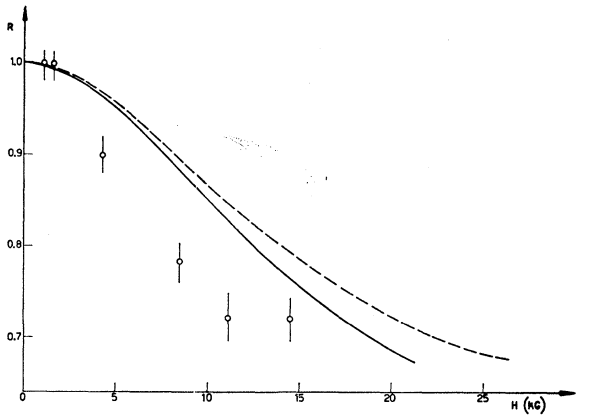

FIG. 1. Magnetic quenching of two-photon long-lived component in water. Dashed curve is calculated from Eq. (2); solid curve is calculated from Eq. (1).

[1] R. L. De Zafra and W. T. Joyner, Phys. Rev. **112**, 19 (1958).
[2] P. Colombino, S. De Benedetti, I. De Gregori, and L. Trossi, Nuovo Cimento **8**, 508 (1958).
[3] W. Brandt, S. Berko, and W. W. Walker, Phys. Rev. **120**, 1289 (1960).
[4] G. Fabri, E. Germagnoli, I. F. Quercia, and E. Turrisi, Nuovo Cimento **30**, 21 (1963).
[5] A. Bisi, A. Fiorentini, E. Gatti, and L. Zappa, Phys. Rev. **128**, 2195 (1962).
[6] V. L. Telegdi, J. C. Sens, D. D. Yovanovitch, and S. D. Warshaw, Phys. Rev. **104**, 867 (1956).

selecting the above quoted values of the pertinent parameters. Therefore, it was thought advisable to carry out further experimental research on the dependence of three quantum decay probability on the magnetic-field strength.

3. EXPERIMENTAL LAYOUT

Measurements were made by a method proposed in a previous paper[7] with some improvements in the experimental setup in view of the smallness of the expected effect.

Essentially, the method consists of measuring the difference ΔI_2 of the intensities of the 511-keV photopeaks in the annihilation spectra with and without magnetic field. The source (about 700 μCi of carrier-free $Na^{22}Cl$ dissolved in bidistilled and deionized water) was located between the pole shoes of an electromagnet. The detector [NaI(Tl) crystal optically coupled with a RCA 6342 A photomultiplier] was carefully shielded

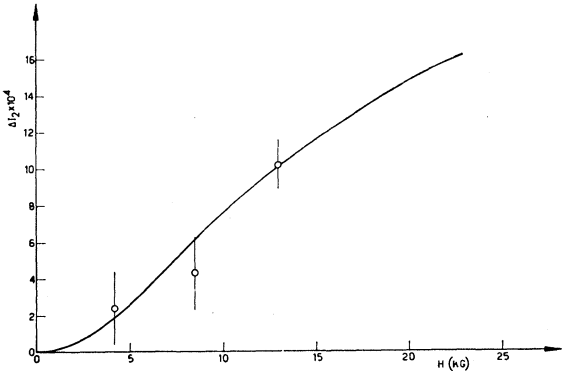

FIG. 2. Increase of two-photon counting rate in Teflon as a function of magnetic field.

from the stray flux of the magnet. The measurements were carried out by alternating short counting runs (30 sec) with the magnet both on and off. With the magnet on, the pulses belonging to the 511-keV photopeak were fed into a multichannel pulse analyzer, while with the magnet off the pulses were subtracted from the spectrum previously memorized. An electronic unit allowed us to perform in sequence the described program. It seemed advisable to check the reliability of the method by performing similar measurements with Teflon.

The experimental results in the case of Teflon were compared with the theoretical curve calculated according to the equation

$$\Delta I_2=\frac{P_T\lambda_3}{3}\left[\frac{1}{\lambda_3+\gamma}-\frac{1}{y^2\lambda_1+\lambda_3+\gamma}\right]-P_T\lambda_3\left[\frac{y^2}{\lambda_1+y^2(\lambda_3+\gamma)}\right], \quad (3)$$

[7] G. Fabri, E. Germagnoli, G. Poletti, and G. Randone, Nuovo Cimento **29**, 500 (1963).

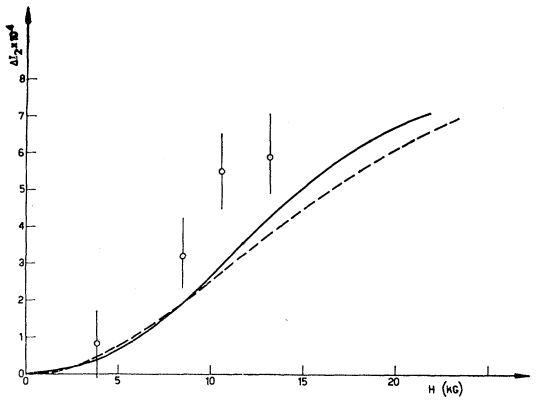

FIG. 3. Increase of two-photon counting rate in water as a function of magnetic field. Dashed curve is calculated from Eq. (3); solid curve is calculated from Eq. (6).

which is easily derived following the hypothesis of Bisi *et al.* Here P_T is the positronium formation probability and γ the triplet-singlet conversion rate through a pick-off mechanism.

It is apparent from Fig. 2 that the experimental results, though not very accurate due to the smallness of the investigated effect, are fairly well accounted for by the theoretical curve, which is practically identical with the one which can be derived for Teflon in the theory discussed below.

The same measurements were carried out in water and the results are shown in Fig. 3; in this case it has to be stressed that it is impossible to achieve agreement with the Eq. (3) by using the above mentioned values of the parameters.

4. DISCUSSION

Since Eqs. (2) and (3) do not take into account the possible influence of the magnetic field on $m=\pm1$ populations of the ortho-state, we decided to investigate how well the above mentioned procedure describes the action of the magnetic field on quenching.

The population changes of the magnetic substates—singlet (n_1), triplet with $m=0(n_3^0)$, and triplet with $m=\pm1(n_3^1)$— were calculated as functions of time for the decay of the free atom of positronium by pick-off. The following differential system was integrated:

$$\begin{aligned}
\frac{dn_1}{dt}&=-\left(\Lambda_1+\frac{3\gamma}{1+y^2}\right)n_1+\frac{\gamma}{1+y^2}n_3^0+\gamma n_3^1,\\
\frac{dn_3^0}{dt}&=-\left(\Lambda_3+\frac{\gamma}{1+y^2}\right)n_3^0+\frac{\gamma}{1+y^2}n_1,\\
\frac{dn_3^1}{dt}&=-(\lambda_3+\gamma)n_3^1+\frac{2\gamma}{1+y^2}n_1,
\end{aligned} \quad (4)$$

where

$$\Lambda_1=\left(\frac{1}{1+y^2}\right)[\lambda_1+y^2\lambda_3],$$

$$\Lambda_3=\left(\frac{1}{1+y^2}\right)[\lambda_3+y^2\lambda_1].$$

This system is derived from the paper by Telegdi *et al.*,[6] who explicitly took into account all the possible transitions between the para- and ortho-states controlled by the conversion constant γ. The only modification to the hypothesis of these is that the functional dependence of the decay constants on the magnetic field intensity was put into the rigorous form calculated by Bisi *et al.*,[5] instead of making use of the approximate treatment attempted by Telegdi *et al.*,[6] which is not valid for high magnetic fields.

By integrating the differential system (4) in the case $H=0$ it is possible to calculate γ as a function of τ_2, λ_1, and λ_3.

$$\gamma=\frac{\tau_2^{-2}-\tau^{-1}(\lambda_1+\lambda_3)+\lambda_1\lambda_3}{4\tau_2^{-1}-(\lambda_1+3\lambda_3)}.$$

The value of τ_2 is derived from the experimental delay curve at $H=0$. In the case of water ($\tau_2=1.82\ 10^{-9}$ sec) γ has been taken equal to $6.929\ 10^8$ sec^{-1}.

The numerical solutions, which were obtained with the help of a digital computer ELEA 6001, fit equations like

$$n(t)=Ae^{-\lambda_S t}+Be^{-\lambda_T{}^0 t}+Ce^{-\lambda_T{}^1 t},$$

for each of the three populations. λ_S, $\lambda_T{}^0$, $\lambda_T{}^1$ are the decay constants of the positronium atoms that are in the singlet and triplet states with $m=0$ and $m=\pm1$; of course λ and A, B, C are functions of the magnetic field.

The intensity of the two-quantum decay is, therefore,

$$I_{2\gamma}(t)=(1+y^2)^{-1}[n_1(t)+y^2n_3{}^0(t)], \qquad (5)$$

and enables one to calculate the $R(H)$ function by using Eq. (1).

The curve calculated according to (1) for water is the solid line in Fig. (1). Likewise, the positronium three-quantum decay is given by

$$\Delta I_2=\left[\int_0^\infty I_{2\gamma}(t)dt\right]_H-\left[\int_0^\infty I_{2\gamma}(t)dt\right]_{H=0}, \qquad (6)$$

and the curve calculated by Eq. (6) is the solid line in Fig. 3.

By inspection of Fig. 1 and Fig. 3, it becomes clear, that the experimental data concerning the magnetic quenching of the long lifetime in water cannot be accounted for theoretically, either if the assumptions by Bisi *et al.*[5] are maintained, or if the more general treatment sketched here is followed; really, the discrepancy between experimental and theoretical results is much larger than the difference between the two theoretical curves.

It can be pointed out that for both sets of experimental results a satisfactory agreement with the experimental data can be achieved by assuming the conversion rate between triplet and singlet states to be a function of the applied magnetic field.

This might enable one to put forward the hypothesis that a conversion mechanism may exist, which is different from the one described as pick-off and which may be competitive with it. This possibility has already been considered by other authors.[3,8]

The hypothesis that the conversion rate γ is a function of the magnetic field H is not of course the only possible means for making theory and experimental results agree. As a matter of fact it can be pointed out that the agreement can be reached by putting in Eqs. (1) and (6) a value of the fine structure splitting constant $\hbar\omega_0\simeq5\times10^{-4}$ eV.

Though we are aware that such a conclusion results from an over-simplified view of the problem, nevertheless the above-mentioned results seem to suggest clearly that the electron-positron bound system in water does not show the characteristics of the positronium free atom.

However, due to the particular structure of liquid H_2O,[9] the wave function $1S$ of the positronium free atom may be distorted to such an extent as to lose spherical symmetry and such a distortion can easily account for the previously hypothesized change of the $\hbar\omega_0$ constant, without strongly varying the ground-state energy of the system and the decay constants λ_1 and λ_3.

It would be interesting to make a more detailed experimental study of the above mentioned possibility that the ground-state wave function of positronium is distorted. To this end the quoted results could be compared with those for water near the boiling point or with those for other materials characterized by strong electric-dipole interactions.

ACKNOWLEDGMENTS

We gratefully thank Professor E. Germagnoli for his continuous assistance and helpful comments. Thanks are also due to Professor I. F. Quercia for several suggestions and to Dr. A. Ghirardi for her work on the numerical calculations.

[8] R. K. Wilson, P. O. Johnson, and R. R. Stump, Phys. Rev. **129**, 2091 (1963).

[9] G. Nemethy and H. A. Scheraga, J. Chem. Phys. **36**, 3382 (1962).