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Measurement of positron lifetime to probe the mixed molecular states of liquid water

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Abstract

Positron lifetime spectra were measured in liquid water at temperatures between 0 °C and 50 °C. The long lifetime of *ortho*-positronium atoms (*o*-Ps) determined by electron pick-off in molecular substances decreases smoothly by 10% as the temperature is raised. This lifetime temperature dependence can be explained by combining the Ps-bubble model and the mixture state model of liquid water.

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1. Introduction

Water has been studied extensively because it is a fundamental liquid with unique properties. For example, its boiling point under atmospheric pressure, 100 °C, is unusually high for its molecular weight, 18 u. In addition, its volume decreases as it melts, unlike other liquids. Moreover, it has an anomalous density maximum at 4 °C. These characteristics have been studied in detail using various methods. Consequently, many models have been suggested. Nevertheless, no definite mechanism has been established to explain these anomalous behaviors of water, even though each model can explain some specific phenomena.

One sensitive method to investigate molecular states in solid and liquid forms is the positron annihilation lifetime (PALT) method. When positrons are injected into a liquid, some of them form positroniums with the surrounding electrons. The positronium lifetime is determined by the rate of annihilation between the positron in the positronium and surrounding

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electrons, also known as pick-off. As described later, according to the Ps-bubble model [1–3], the lifetime should be longer at higher temperatures in liquid water. However, Vértes et al. [4] showed that the positronium lifetime has a general decreasing trend for higher temperature, although it oscillates between 38 °C to 60 °C.

We measured PALT in liquid water between 0 °C and 50 °C. This Letter explains that the positron lifetime decreases smoothly as the temperature rises. This behavior is explainable by combining the Ps-bubble model and a two-state mixture model of water.

2. Positron annihilation lifetime method and the Ps-bubble model

This section explains PALT and the Ps-bubble model.

Positrons injected in material can undergo three different processes, each with a different lifetime. The shortest lifetime (τ_1) occurs by a formation and annihilation of *para*-positronium (*p*-Ps) in which the spins are anti-parallel. In that situation, the electron forming *p*-Ps with the positron is captured from the surrounding molecules. The lifetime of *p*-Ps in vacuum, 0.125 ns, is too short to be affected by surrounding matter.

The second lifetime (τ_2) is caused by positron annihilation without forming a bound state.

The third lifetime (τ_3) results from formation of an *ortho*-positronium (*o*-Ps) with parallel spins.

The *o*-Ps has an intrinsic lifetime of 142 ns in vacuum. However, in material, the lifetime is typically several nanoseconds because it annihilates with an electron in the surrounding molecules. As a result of this pick-off process, τ_3 is sensitive to the electron-state of the surrounding substance. In liquid, *o*-Ps is generally considered to push out the surrounding molecules and form a Ps-bubble. The wave function of *o*-Ps exudes from the surface of Ps-bubble, which comprises electrons from surrounding substances. The overlap of *o*-Ps wave function and an electron wave function of the surroundings determines the pick-off rate. In the Ps-bubble model, the wave function of *o*-Ps is trapped inside a spherical infinite-depth well potential whose radius is the sum of the radius of the Ps-bubble and the exuding depth. Therefore, τ_3 is a function of the Ps-bubble radius [3,5–7].

Based on the Ps-bubble model, a semi-empirical correlation between τ_3 and the Ps-bubble radius is introduced as

$$\tau_3 = \left[2\left\{1 - \frac{R}{R+\Delta R} + \frac{1}{2\pi}\sin\left(\frac{2\pi R}{R+\Delta R}\right)\right\}\right]^{-1}, \tag{1}$$

where τ_3 is measured in nanoseconds, R is the Ps-bubble radius, and ΔR is the exuding depth of the *o*-Ps wave function into the surrounding electron wave functions. In this model, R is determined by the balance between the zero-point energy of *o*-Ps and the surface tension of the surrounding substance. The balance is represented as

$$\frac{\partial}{\partial R}\left(E + 4\pi R^2\gamma\right) = 0, \tag{2}$$

where E is the zero point energy of *o*-Ps and γ is the surface tension of bulk water. E is expressed as

$$E \sim \frac{\hbar^2 k^2}{4m_e} = \frac{\hbar^2\pi^2}{4m_e(R+\Delta R)^2}, \tag{3}$$

where m_e is the electron mass and k is the momentum of *o*-Ps.

For the case of water, γ decreases as the temperature rises; thereby the bubble becomes larger. By this argument alone, τ_3 is longer for higher temperatures.

3. Experiment

3.1. Measurement of positron lifetime

We employed the positron annihilation lifetime technique using ^{22}Na as a positron source. ^{22}Na is put in the sample center. Positrons emitted into the sample are annihilated through the process mentioned in the previous section. When ^{22}Na emits a positron, it simultaneously emits a 1275 keV photon. When a positron is annihilated with an electron, two 511 keV photons are emitted. In light of those facts, we measured the time difference between the emission of 1275 keV photon and the emission of two 511 keV photons.

We used the apparatus shown in Fig. 1.

Powder of 1.6 MBq ^{22}NaCl wrapped between 8 μm thick polyimide films was used as a positron source.