

Ortho-positronium in liquid alkanes

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Abstract. Positron lifetime spectra were measured for a series of odd-numbered liquid *n*-alkanes (C_7H_{16} , C_9H_{20} , $C_{13}H_{28}$ and $C_{19}H_{40}$) in a broad range of temperature. According to the “bubble model” the *ortho*-positronium (*o*-Ps) lifetime is determined by surface tension. Bubble radius was calculated using the Tao-Eldrup model and compared to the radius expected from the surface tension of liquid. The radii are not identical, their difference rises with temperature. If one assumes the difference of radii as constant, the *o*-Ps lifetime calculated from the Tao-Eldrup model rises faster than the experimental one. The distinct rise of the difference of radii has no physical explanation in the accepted *o*-Ps annihilation model. This effect can be the result of radiation chemistry processes not taken into account so far in the bubble model.

Key words: positron annihilation • liquid alkanes • bubble model • surface tension

Introduction

Normal alkanes, C_nH_{2n+2} , are the simplest organic structures in the form of linear hydrocarbon chains. They are important components of petroleum and its derivative products, they are also important elements of many structures, like liquid crystals, surfactants and lipides. For this reason alkanes are intensively investigated using many experimental techniques, including also positron annihilation spectroscopy. Positronium properties in free (electron-less) volumes in liquid phase of alkanes depend on physicochemical properties of the medium, like surface tension, viscosity, dielectric constant, etc. [7].

There are very few data about temperature dependence of *o*-Ps lifetime in organic liquids in a broad range of temperature, e.g. the data for five selected temperatures of water in the papers by Duplâtre *et al.* [8]; as to alkanes the very scant data can be found in an old paper by Kajcsos *et al.* [17] only.

We measured the positron lifetime spectra for selected liquid alkanes in the range of temperatures covering 100–150 K, in order to check the validity and accuracy of existing positron annihilation models, in particular concerning the relation between *o*-Ps lifetime and surface tension. Surface tension of short-chain normal *n*-alkanes ($n \leq 26$) at various temperatures is well known, and can be found in the handbooks of physics [15, 16]. In pure fluids it decreases with the rise of temperature and tends to zero when the critical point is reached [30]. However, one has to remember

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that the textbook data were obtained for macroscopic samples and there is to check whether or not they can be applied to nanometer scale objects, like cavities accommodating Ps in liquids.

Experimental

The alkane samples were from Sigma Aldrich (*n*-tridecane C₁₃H₂₈, *n*-nonadecane C₁₉H₄₀ – their purity was 99%) and Fluka (*n*-heptane C₇H₁₆, *n*-nonane C₉H₂₀ – purity better than 99.5%). Liquid sample was placed in a chamber with a positron source. The sample was degassed by a freeze – thaw technique in order to avoid *ortho-para* conversion of positronium in dissolved paramagnetic molecular oxygen. The chamber volume was evacuated to about 0.5 Pa and closed, the pressure above the sample corresponded thus to the pressure of alkane saturated vapour. The temperature of the samples inside the chamber was controlled with an accuracy of ± 0.1 K using Shimaden FP21 unit.

The lifetime spectra were registered using a conventional fast-slow coincidence spectrometer with a time resolution of 0.260 ns. Two scintillation heads with BaF₂ crystals were placed at an angle of 90°. The positron source (0.5 MBq of ²²Na) was enclosed in an 8 μ m thick Kapton envelope, fixed in the centre of the chamber. The number of counts collected per one spectrum (during 1 h) was about 8×10^5 coincidences. The series of spectra was registered starting from temperature below the melting point and then temperature was gradually increased. After completing the series, a control run was registered at lowering the temperature. The results of both runs were identical regardless of the direction of temperature change.

The spectra were processed by LT v.9 program [18] fitting to the data a sum of three exponential components convoluted with the instrumental resolution curve. The shortest lived component ($\tau_1 \approx 0.125$ ns) belonged to *para*-positronium (*p*-Ps); the longest one ($\tau_3 \approx 3 \div 4$ ns) to *o*-Ps and an intermediate one ($\tau_2 \approx 0.300$ ns) to the annihilation of free positrons. To reduce the number of fitting parameters the ratio of *p*-Ps to *o*-Ps intensities was fixed as 1:3. The correction for annihilation in the Kapton envelope was assumed to be 10%, with the lifetime 0.374 ns.

The models of pick-off process

Positronium quenching rate by pick-off process [11] depends on the probability P to find positronium outside of free volume, in which it is trapped [9, 14]. The pick-off decay constant λ_{po} is thus [2, 13]

$$(1) \quad \lambda_{po} = P \times \lambda_b$$

where λ_b is the pick-off rate in bulk medium (commonly accepted as spin averaged constant of spontaneous positronium decay, $\lambda_b = 2 \text{ ns}^{-1}$). In this approach λ_b does not depend on electron density of the medium.

In the case of free volumes appearing in condensed media, the spacings of energy levels of a light particle in the well are much larger than the thermal energy kT ,

thus, if positronium is in an equilibrium with the surrounding medium, only the lowest energy level in the well is occupied. For that state in the spherical well (1s state), the wave function outside the well is:

$$(2) \quad \Psi_{out}(r) = B \cdot \frac{\exp(-\kappa r)}{r}$$

where $\kappa = \sqrt{[4m_e(V_0 - E) / \hbar^2]}$, V_0 is the depth of potential well, E is the energy of the state above the bottom of well, m_e is the electron rest mass. Reciprocal of κ corresponds to the average penetration range of positronium wave-function into the bulk (for $V_0 - E = 1$ eV the value of $1/\kappa$ is about 0.14 nm).

The wave-function of the state 1s inside the well is in the form:

$$(3) \quad \Psi_{in}(r) = A \cdot \frac{\sin(-kr)}{r}, \quad \text{where } k = \sqrt{\frac{4m_e E}{\hbar^2}}$$

The wave-functions inside and outside the well have to be sewn together at the well wall. A simplification of calculations was proposed by Tao [28]. Instead of a finite potential well he introduced an infinitely deep one, thus the wave-function was zero at outside. To reconstruct the penetration of wave-function to the bulk, the radius of potential well was broadened by Δ , leaving unchanged the radius of sphere R with zero electron density. The value of Δ parameter is chosen to get the same probability P of finding particle outside the void as for real finite depth well:

$$(4) \quad \int_R^{\infty} |\Psi_{out}(r)|^2 r^2 dr = \int_R^{R+\Delta} |\Psi_{\infty}(r)|^2 r^2 dr$$

where Ψ_{∞} is the Ps wave-function inside the infinite well.

The value of P for infinitely deep and broadened potential well was calculated by Eldrup [9] obtaining the equation for pick-off decay constant of *o*-Ps :

$$(5) \quad \lambda_{po} = \frac{1}{\tau_3} = 2\text{ns}^{-1}P = 2\text{ns}^{-1} \left[1 - \frac{R}{R+\Delta} + \frac{1}{2\pi} \sin\left(\frac{2\pi R}{R+\Delta}\right) \right]$$

The value of Δ is usually assumed as 0.166 nm [21]. It is comparable to the penetration depth of positronium wave-function $1/\kappa$ in the case of finite depth well.

Bubble model and free volumes in liquids

Positronium behaviour in liquids is described by the model proposed by Ferrel [10] and developed by Buchikhin [3, 4] and Roellig [23]. The radii of natural free volumes in liquids are of the order of 0.1 nm, density fluctuations [12] cannot increase these radii to more than 0.2 nm [29]. The observed *o*-Ps lifetimes are much longer than expected for the radii mentioned above; it means that positronium forces itself the free volume in liquid. The pressure exerted by zero-point oscillations of positronium produces a bubble around the Ps, whose dimensions are determined by the minimum of total energy, composed of positronium energy $E(R)$, surface tension energy $E_{\sigma}(R)$ and external pressure $E_p(R)$ [22]:

$$(6) \quad d/dR \cdot [E(R) + 4\pi R^2\sigma + 4\pi R^3\pi / 3] = 0$$

where $E(R)$ is the energy of the particle confined in the bubble, σ is the surface tension, p is the external pressure. Surface tension of alkanes is of the order $\sigma \approx 10^{-3}$ eV/Å², and atmospheric pressure $p \approx 10^{-6}$ eV/Å³. The pressure over the sample in our experiment was always less than the atmospheric one, thus the term $4\pi R^3 p/3$ is by 3, or more, orders of magnitude smaller than that related to the surface tension and will be neglected in further discussion.

According to the Tao-Eldrup model, the radius of empty volume is R , while the radius of potential well is $R_0 = R + \Delta$. The question arises, which of these radii determines the energy $E(R)$ of positronium in the well. Transition from finite to infinite depth of the well -increases the energy, while broadening of the well -diminishes. One can estimate the Ps energy in two variants of potential well using an example taken from the literature. Mogensen and Jacobsen [20] have estimated the depth of potential well in liquid benzene as 0.961 eV (the value of about 1 eV is typical of many organic media), the Ps energy in the well as 0.575 eV, and the radius of the well 4.1 Å. In the Tao-Eldrup model we receive the Ps energy 0.576 eV assuming the well radius $R + \Delta$. It is almost exactly the same energy as obtained in the realistic model using the finite well depth. So, in further calculations we put into the equation for Ps energy the radius equal to $R + \Delta$.

$$(7) \quad E(R) = \frac{\hbar^2}{2m_{\text{Ps}}} \cdot \frac{\pi^2}{(R + \Delta)^2} \\ = 1.904 \cdot 10^{-2} \cdot \frac{\pi^2}{(R + \Delta)^2} \text{ [eV} \cdot \text{nm}^2 \text{]}$$

where $m_{\text{Ps}} = 2m_e$ - positronium mass.

The free volume radius (or rather bubble radius) appears also in the expression for surface tension energy, and we do not know whether that radius is identical to that in Eq. (5). Real potential well is not strictly rectangular, thus R in Eqs. (5) and (7) is rather an "equivalent" radius. In the term describing the surface tension the bubble radius is also not too precisely defined; the bubble limit should be located somewhere between the outer filled orbital of atoms on the bubble surface and the atom centers. So, let us assume an arbitrary value of radius acting in surface tension by introducing a correction term a , which is to be determined. Finally, we receive:

$$(8) \quad \frac{d}{dR} \left[1.904 \cdot 10^{-2} \cdot \frac{\pi^2}{(R + \Delta)^2} + 4\pi(R + \Delta + a)^2 \sigma \right] = 0 \\ -1.904 \cdot 10^{-2} \cdot \frac{2\pi^2}{(R + \Delta)^3} + 8\pi(R + \Delta + a)\sigma = 0$$

Using the R value from the Tao-Eldrup model and macroscopic value of surface tension we can estimate the constant a for each experimental point

$$(9) \quad a = \frac{1.904 \cdot 10^{-2} \pi}{4(R + \Delta)^3 \sigma} - (R + \Delta) \text{ [nm]}$$

Experimental results and discussion

Mean lifetime of *o*-Ps equal to about 3.1 ns is typical of alkanes just above their melting points, and this

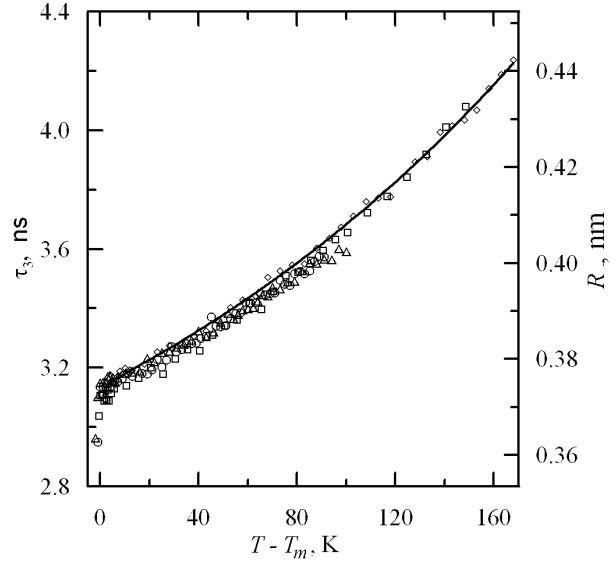


Fig. 1. Ortho-positronium lifetime as a function of temperature above the melting point T_m for *n*-heptane (squares, $T_m = 182.5$ K), *n*-nonane (diamonds, $T_m = 221$ K), *n*-tridecane (circles, $T_m = 268$ K) and *n*-nonadecane (triangles, $T_m = 305$ K).

value is almost independent of the chain length. With the rise of temperature we observe the lengthening of that lifetime (Fig. 1). The data for two short chain alkanes, C_7H_{16} (*n*-heptane) and C_9H_{20} (*n*-nonane), show that 150 K above the melting point T_m the lifetime τ_3 increases by ≈ 1 ns. That rise of lifetime for both the above-mentioned alkanes is roughly described by the same exponential function:

$$(10) \quad \tau_3 = 2.209 + 0.934 \cdot \exp[0.0046(T - T_m)] \text{ [ns]}$$

(for longer chains the form of $\tau_3(T)$ is very similar, as can be seen in Fig. 1). Increase of τ_3 in the investigated range corresponds to an increase of free volume from 0.21 nm³ to 0.33 nm³.

An attempt to estimate the possible difference of radii calculated from surface tension and from the lifetime τ_3 in the Tao-Eldrup model was undertaken, i.e. the parameter a was calculated from Eq. (9). The results are shown in Fig. 2. In the case of $C_{13}H_{28}$ (*n*-tridecane) and $C_{19}H_{40}$ (*n*-nonadecane), the a parameter in the range (0–30) K above the melting point is close to zero, it means that near the melting point the bubble radius estimated from surface tension R_σ is very close to $R + \Delta$, i.e. it exceeds the radius of electron-less void by about 0.16 nm. This is comparable to the Van der Waals radii of hydrogen and carbon atoms. However, a rises evidently with temperature, reaching finally ≈ 0.15 nm. The difference of radii R and R_σ should be constant, depending on the sizes of atoms on the bubble surface, i.e. dependent on the assumption how far from the centre of atoms on the bubble surface we place the "equivalent" edge of electron density distribution, and how far the centre of molecular force producing the surface tension is located. Intuitively, the difference of radii R and $R_\sigma = (R + \Delta + a)$ should be less than Van der Waals radius R_{vdw} of involved atoms. Due to short range of molecular forces, the use of Van der Waals radius in this discussion seems to be more suitable than that of Wigner-Seitz one, proposed by Byakov and Stepanov

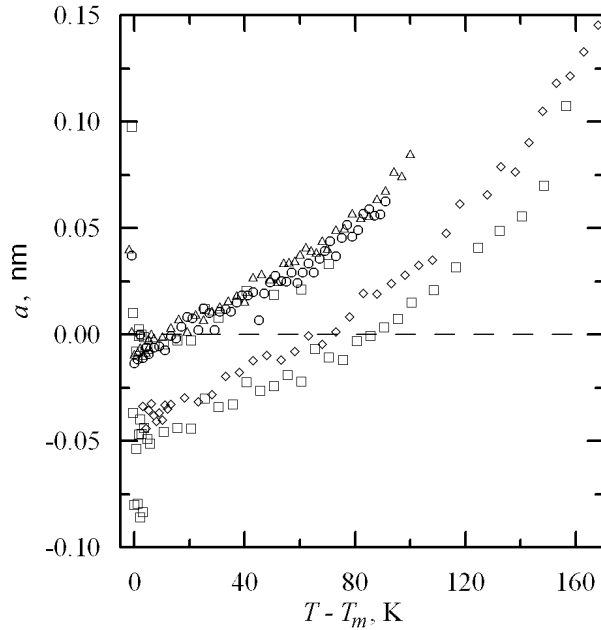


Fig. 2. Divergence of radii of electron-less void and of positronium bubble as a function of temperature above the melting point. The meaning of symbols as in Fig. 1.

[6]. In the case of multiatomic elongated molecules, like our alkanes, the Wigner-Seitz radius, being of the order of 0.5 nm, is evidently too large what will imply that the centre of molecular forces is placed behind the cavity wall by 4 atomic radii! The difference $R_\sigma - R$ is not surprising, but its increase with temperature, reaching 0.16 nm, is hard to accept, when there are no changes of the medium structure.

In Fig. 2 we also see that for shorter alkanes, C_7H_{16} and C_9H_{20} , the curves $a(T - T_m)$ are distinctly shifted toward smaller a . The curves in Fig. 2 are drawn assuming the values of surface tension from the literature, e.g. [15, 16]; however, these data cover the range from 0°C to 100°C only. In that range σ is linearly dependent on temperature:

$$(11) \quad \sigma = c - d \cdot T$$

(coefficients c , d are given in the literature; parameter d decreases with increasing chain length). For C_7H_{16} and C_9H_{20} the values of σ were taken by us from an extrapolation of lines (11) toward low temperatures. The values of τ_3 lifetime for some alkanes and respective values of surface tension at a temperature of 2 K above the melting point are shown in Fig. 3. It is seen that in the range of data taken from the tables (not from an extrapolation) σ values are almost independent of chain length, but in the region $n < 13$ they rise rapidly when the chains become short. It is thus possible that the extrapolation of Eq. (11) to lower temperatures by several scores of K is not justified. If, alternatively, one determines the surface tension for $n < 13$ and for the temperature near the melting point, by extrapolating linearly the data from Fig. 3 for $15 \leq n \leq 20$, we receive for C_7H_{16} and C_9H_{20} in the point $T - T_m = 2$ K the value of a also close to zero.

There is to remember that the value of σ depends on surface curvature. In the case of concave surface (bubble) σ rises with curvature [1]. For the radii over

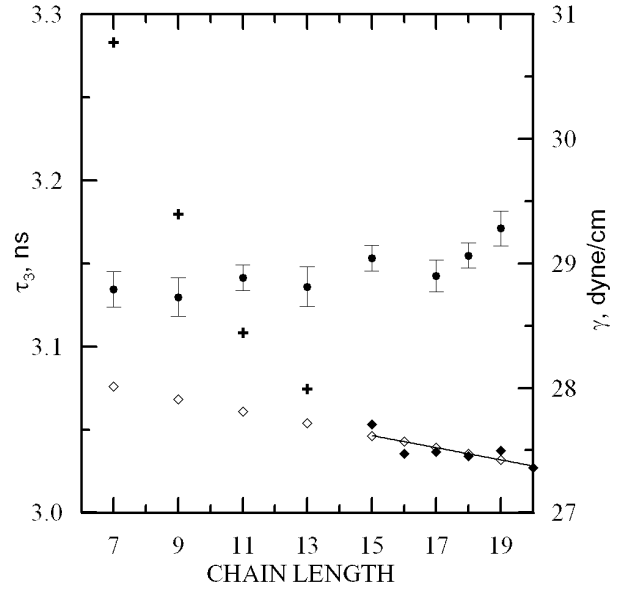


Fig. 3. *Ortho*-positronium lifetime (circles) and surface tension (diamonds) 2 K above the melting point as a function of chain length (crosses – surface tension extrapolated by Eq. (11); empty diamonds – surface tension extrapolated by equation: $\sigma = -0.49n + 28.35$ dyn/cm.

10 nm the effect is negligible, but at $R_\sigma = 1$ nm in typical organic liquids σ is about twice of that for the flat surface [1, 19]. However, this simple relation is valid for $R_\sigma > 1$ nm, near $R_\sigma = 0.5$ nm (depending on the medium) the slope $d\sigma/dR$ changes the sign, surface tension begins to decrease with decreasing radius [24]. In our experiment the radii $R + \Delta$ are about (0.5–0.6) nm, thus near the maximum of σ . There are no exact data for this range of radii, but if we double the value of σ in Eq. (9) the variation of $(R_\sigma - R)$ with temperature reduces by a factor of two, but at the expense of non-physical reduction of R_σ i.e. $(a + \Delta) < 0$. Thus, an account of the rise of surface tension at a concave surface cannot solve the problem of rising divergence of radii.

In the case of alkanes with $n = 13$ and $n = 19$ (and may be also others) the parameter a near the melting point is close to zero. If one assumes that $a = 0$ is a correct value, the radius R can be calculated from the equation:

$$(12) \quad R = \sqrt[4]{\frac{1.904 \cdot 10^{-2} \pi}{4\sigma} - \Delta}$$

and then one can calculate the mean lifetimes using the Tao-Eldrup equation. The result is shown in Fig. 4. One can see that the rise of mean *o*-Ps lifetime with temperature is slower than it would be expected from the Tao-Eldrup equation.

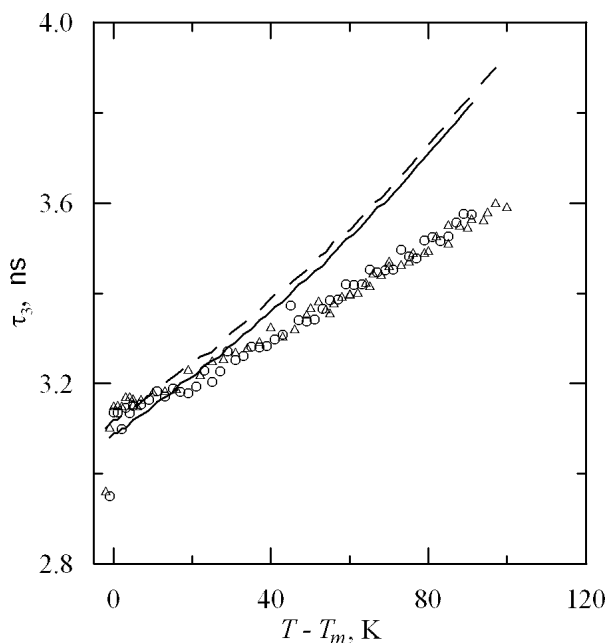
One can draw the dependence of *o*-Ps decay constant directly on surface tension. Tao [28] proposed to write this relation in the form:

$$(13) \quad \lambda_{po} = A\sigma^\beta$$

where σ is as for flat surface, σ_π . Fitting the curve (13) to the data for various organic liquids at room temperature Tao received: $A = 0.06$; $\beta = 0.50$. This equation was proposed as empirical only, however Byakov and Krištiak [5] have shown that expanding $\sin x \approx x - x^2/\pi$ in Eq. (5) and neglecting the divergence of radii ($a = 0$) one obtains:

Table 1. Pick off constant vs. surface tension

<i>n</i> -Alkane	$\lambda_p = \alpha \cdot \sigma^\beta$, (this paper)	Schrader [25]	Tao [28], normal alkanes
C ₇ H ₁₆	$\lambda_p = 0.072 \cdot \sigma^{0.44}$		
C ₉ H ₂₀	$\lambda_p = 0.08 \cdot \sigma^{0.41}$		
C ₁₃ H ₂₈	$\lambda_p = 0.08 \cdot \sigma^{0.42}$	$\lambda_p = 0.06 \cdot \sigma^{0.4}$	$\lambda_p = 0.061 \cdot \sigma^{0.5}$
C ₁₉ H ₄₀	$\lambda_p = 0.09 \cdot \sigma^{0.39}$		

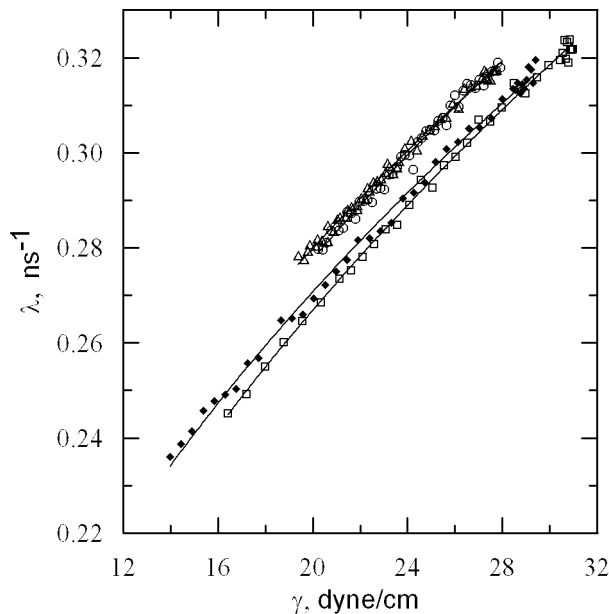
**Fig. 4.** Ortho-positronium lifetime as a function of $(T - T_m)$: experimental (points) and calculated from Tao-Eldrup model assuming the divergence of radii $a = 0$ (lines). Circles and solid line – tridecane; triangles and dashed line – nonadecane.

$$(14) \quad \lambda_{po} = 2\lambda_b \left(\frac{\Delta}{a_B} \right)^2 \cdot \left(\frac{8\sigma a_B^2}{\pi R y} \right)^{1/2} \approx 0.07 \sigma^{1/2}$$

where a_B is the Bohr radius, Ry is the Rydberg constant. Byakov and Krištiak compared their results with the same experimental data about organic liquids as in [28], which are rather scattered. Analogous data for a given alkane as a function of temperature run much more smoothly and this time we receive the values of constant A from 0.07 to 0.09 depending on the chain length, and β from 0.44 to 0.39, with a tendency to diminish with lengthening of the molecule (Table 1). This value of β is close to the estimate given by Schrader [25]. The decay rate vs. surface tension for the investigated *n*-alkanes is shown in Fig. 5.

Conclusions

The rise of *o*-Ps lifetime with temperature is slower than expected from existing models of the pick-off process. To obtain the consistency of estimates based on surface tension and experimental data one needs to assume that the bubble radius R_σ , acting in the case of surface tension is larger than R estimated from lifetime data using the Tao-Eldrup model. Moreover, that divergence of radii rises systematically with temperature. That distinct rise of divergence, reaching 0.15 nm at high temperature, is hard to explain without assumption that the *o*-Ps lifetime

**Fig. 5.** Correlation between the *o*-Ps pick-off rate and the surface tension of *n*-heptane (squares), *n*-nonane (diamonds), *n*-tridecane (circles) and *n*-nonadecane (triangles).

is influenced by other temperature dependent factors, beside surface tension, not included into consideration. One can suppose, after Byakov and Stepanov [26, 27], that the *o*-Ps lifetime, shorter than expected from Tao-Eldrup model at fixed difference of radii, is the result of positronium reactions with radiolytic products, e.g. Ps oxygenation by the radicals and ions produced during the sample irradiation. Among the products of radiolysis one can find also the paramagnetic ones, thus *ortho-para* conversion of *o*-Ps can be also the source of lifetime shortening. There is to note that *ortho-para* conversion occurs also on molecular oxygen from the air dissolved in liquid sample. We have eliminated the latter effect, able to distort the results, by thorough degassing the samples before the measurements.

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