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A new calculation technique of muonium formation rate

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Abstract

In condensed matter the formation of a muonium atom from a positive muon and an election is described usually with a first order kinetic equation which assumes that the process is random and that the charge distribution is uniform. According to this model the muon polarization function as a function of time should reduce to an exponential law. Experiments in superfluid helium demonstrates that this is incorrect.

Our proposed technique allows to reconstruct the muonium formation rate function from the μSR histogram in low transverse magnetic field without presupposing a particular theoretical form, i.e. with no parametrization. The technique is based on solving the integral equation of the first kind for the muon polarization function using the maximum likelihood method. The obtained results are of fundamental importance for the analysis of the charge kinetics in superfluid helium.

Keywords: Muonium formation; Histograms; Recovery; Maximum likelihood; Nonparametric algorithm; Superfluid helium

1. Introduction

There are basically two processes leading to creation of muonium atom

$$\mu^+ + e^- \to \text{Mu}. \tag{1}$$

The first one is “hot” muonium formation during the ionization of atoms and charge exchange process by fast moving muon. The second one takes place in condensed matter on the “cold” stage after muon thermalization as the result of recombination of charged particles at the end of the trace [1,2]. This process is usually described by the first order kinetic equation for the muonium formation rate function $n(t)$

$$\frac{dn}{dt} = -kn. \tag{2}$$

Here k is the reaction constant [3].

The resulting muonium spin state is evenly divided between the S-state and the triplet state. The polarization of those muons in the S-state is almost invisible in conventional μSR method [4]. The muon polarization can be described in zero magnetic field as

$$P(t) = 1 - \frac{1}{2}\int_0^t n(t')\,dt'. \tag{3}$$

The interaction between a muon and an electron in liquid helium can be described as a Coulomb attraction of two charges moving in the viscous regime with mutual mobility b. Let the radial density distribution function between muon and electron be a Gaussian law

$$W(r) = \frac{1}{\Delta^3\pi^{3/2}}\exp\left[-(r/\Delta)^2\right]. \tag{4}$$

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In this case the polarization function $P(t)$ should be described by the formula

$$P(t) = 1 + \frac{2}{\sqrt{\pi}} x \cdot \exp(-x^2) - \operatorname{erf}(x), \tag{5}$$

where parameter x is determined by the expression

$$x = (3bet)^{1/3}/\Delta.$$

Here Δ is the scale of the Gaussian law Eq. (4), b – mutual mobility, e – electric charge of muon and electron, and t is time.

The behavior of Eq. (5) is close to exponential $\exp(-kt)$. This explains why the muonium formation process is often described as a chemical reaction.

However in some cases (specifically in superfluid helium), the approximation (5) is inadequate [5]. The standard description of the complicated muonium formation process as a superposition of fast and slow subprocesses is known to be a very crude and inadequate model.

In superfluid helium a positive charge forms a 'snow ball' with mass $M_+ \simeq 40$–50 He atoms and the electron is localized in a cavity with hydrodynamic mass $M_- \simeq 200$ He atoms (see the review of Shikin [6]). This is the physical reason why Mu formation in superfluid helium is a rather long process compared to other substances [7].

In zero magnetic field Eq. (3) can be presented as a sum of two components

$$P(t) = \frac{1}{2}\int_0^t n(t')\,dt' + \int_t^\infty n(t')\,dt', \tag{6}$$

which have an obvious physical sense. The first integral in this formula describes of Mu atoms in triplet state (↑↑), which were formed before time t, and the second one corresponds to muons in 'snow balls', which have not yet combined with electrons to form Mu atoms.

We will now look at the muon polarization in weak transverse magnetic field (wTF), where the field is sufficiently small that the precession of the spin of the muons in 'snow balls' is negligibly small during the muon lifetime of 2.197 μs. This is possible because the gyromagnetic ratio of muonium in the triplet state $\gamma_{\mathrm{Mu}} = 8.77 \cdot 10^6\ (\mathrm{s} \cdot \mathrm{Oe})^{-1}$ is approximately 103 times larger than γ_μ for the bare muon. Typical values of magnetic field were about $H \leqslant 0.4$ Oe in the experiment [7] for liquid helium. The spins of those Mu atoms which were formed at time t' will precess with the Larmor frequency

$$\omega_{\mathrm{Mu}} = \gamma_{\mathrm{Mu}} H$$

and have a phase delay $\omega_{\mathrm{Mu}} t'$.

The polarization function will have two components

$$P(t) = \frac{1}{2}\int_0^t n(t')\cos\bigl[\omega_{\mathrm{Mu}}(t - t')\bigr]\,dt' + \int_t^\infty n(t')\,dt', \tag{7}$$

where the first one describes the spin precession of Mu atoms with Larmor frequency ω_{Mu} with time delay t', and the second one describes the polarization of muons in 'snow balls'. We can ignore their precession in such low magnetic field.

Typically measurements are fit using some analytic model for $P(t)$ using a least squares method (LSM) [8]. The number of fit parameters is of the order of 10. The success of this standard *parametric* procedure is mainly determined by whether or not correct the theoretical function (usually analytic) was chosen for $P(t)$ or $n(t)$, respectively. This is the main drawback of the standard procedure.

In this paper we propose a new *nonparametric* algorithm for recovering the muonium formation rate $n(t)$ from the experimental μSR data by using the program package RECOVERY for restoration of signals from noisy data which is based on the maximum likelihood method.

2. Description of the method

The following is only a description of the parts of the RECOVERY algorithm relating to the special features of the μSR experiments. The detailed description of the algorithm and its applications can be found in papers [9,10].

In μSR experiments, scintillating counters detect the muon's decay positron, the direction of which is correlated with the muon spin direction at the time of its decay. The number of decay positron is histogrammed as a function of the time taken to decay

$$N(t) = N_0\bigl[1 \pm A_0 \cdot P(t)\bigr]e^{-t/t_\mu} + B, \tag{8}$$

where $t_\mu = 2.197 \cdot 10^{-6}$ s is the muon life time, N_0 is proportional to the intensity of muon beam, A_0

is the asymmetry coefficient, and B is the random background level. It is worth noting that the sign before the asymmetry coefficient A_0 may be plus or minus for two opposing scintillating counters which are along (+) or opposite (−) of the initial muon spin direction [4,8].

The polarization function $P(t)$ in Eq. (8) can only be uniquely determined from the histogram $N(t)$ if all parameters N_0, B and A_0 are known. We chose to pre-determine the parameters N_0, B and A_0 using the crude standard μSR analysis method (using Eq. (9) below) and then use those values to extract the generalized $P(t)$ from the experimental $N(t)$.

For the first step of determining N_0, B and A_0, we use the fact that in temperature range 0.5–1.5 K all muons recombine during time t_0 and this time is less than the total histogram duration $t_1 \approx 10$ μs and for $t \geqslant t_0$, when all atoms of muonium have been formed, the equation for histogram can be written as

$$N_1(t) = \left[A + C\cos(\omega_{\mathrm{Mu}}t) + D\sin(\omega_{\mathrm{Mu}}t)\right]e^{-t/t_\mu} + B. \tag{9}$$

Now we can easy find all of four parameters A, B, C, and D by minimization of the value

$$\chi^2 = \sum_{t_i > t_0} \frac{[N(t_i) - N_1(t_i)]^2}{N(t_i)}. \tag{10}$$

It is correct because the experimental data $N(t)$ have a Poissonian distribution. This procedure can also be used to determine the value of the magnetic field projection on the axis of muon spin precession.

The system of equation originating from the minimization of Eq. (10) has a symmetrical and positive-definite matrix. We solve this system by the Cholesky decomposition method (square root of matrix) with the subroutines CHOLSL and CHOLDC from the 2nd edition of the "Numerical Recipes" book by Press et al. [12]. These programs were modified for double precision (real $*$ 8) accuracy. The use of the Cholesky decomposition method for a symmetrical and positive-definite matrix provides more stable results than the Gaussian elimination method which does not use the symmetry properties of the matrix.

With values for the parameters A, B, C and D we compute $\widehat{N}_0$ and asymmetry coefficient $\hat{A}_0$ using the formulas

$$\widehat{N}_0 = A\exp(t_0/t_\mu), \qquad \hat{A}_0 = \frac{\sqrt{C^2 + D^2}}{A}. \tag{11}$$

Now that all the constants in Eq. (8) are known, this formula can be inverted, and the normalized polarization

$$P(t) = \frac{\widehat{P}(t)}{\widehat{P}(0)},$$
$$\widehat{P}(t) = \frac{1}{\hat{A}_0}\left[1 - \frac{N(t) - B}{\widehat{N}_0}e^{t/t_\mu}\right], \tag{12}$$

where the normalization constant $\widehat{P}(0)$ is an estimate of the value of the function $\widehat{P}(t)$ at time $t = 0$, obtained from a small number of initial values using an optimal filtering program [13], can be calculated. Fig. 1 shows the histogram $N(t)$ and the polarization function $P(t)$ at $T = 0.7$ K. As one can see from this figure, the polarization $P(t)$ gradually moves away from the initial value $P(0) \simeq 1$ into a regime of uniform precession between the extreme values of the amplitudes $+1/2$ and $-1/2$. This corresponds to the fact, noted above, that after muonium is formed only half the initial polarization is observed [8].

It is obvious in Fig. 1 that the noise level in function $P(t)$ increases with t. This level can be easy seen from the fact that $N(t)$ has a Poissonian distribution which results in

$$\mathrm{var}\left[N(t)\right] = N(t).$$

If we approximate

$$N(t) \approx N_0 e^{-t/t_\mu},$$

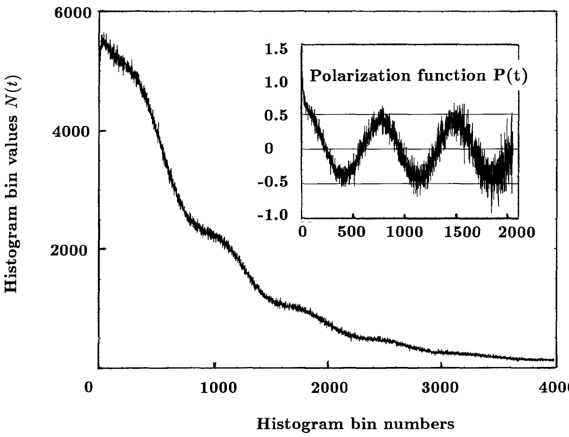

Fig. 1. Input histogram $N(t)$ and polarization function $P(t)$ for the first 2048 data points (in insert). Magnetic field $H = 0.4$ Oe, helium temperature $T = 0.7$ K, total histogram bin number $N = 3983$, one bin time $\Delta t = 2.5$ ns.

then from Eqs. (12) we obtain

$$\mathrm{var}\big[P(t)\big] \approx \frac{1}{\hat{A}_0^2 N(t)(\widehat{P}(0))^2}. \tag{13}$$

This formula for estimation of noise level in polarization function $P(t)$ is used in the algorithm presented in this paper.

After these appropriate preliminary calculations let us go to the main problem of this paper: how to determine the muonium formation rate function $n(t)$ in a model independent way, that is without assuming any particular theoretical form with fit parameters for the polarization function $P(t)$.

Eq. (7) can be rewritten as

$$P_1(t) = \int_0^t n(t')\left[\frac{\cos\omega_{\mathrm{Mu}}(t-t')}{2} - 1\right]dt', \tag{14}$$

where

$$P_1(t) = P(t) - 1. \tag{15}$$

Eq. (14) is the Volterra convolution integral equation of the first kind with the kernel

$$K(t-t') = \frac{\cos\omega_{\mathrm{Mu}}(t-t')}{2} - 1 \tag{16}$$

which is used to recover the nonnegative function $n(t)$ from the input data $P_1(t)$.

To solve this integral equation we use the program Dconv2 from the program package RECOVERY. The program Dconv2 was originally designed for the solution of the Fredholm integral equation with fixed integration limits

$$P_1(t) = \int_0^T n(t')K_1(t-t')\,dt'. \tag{17}$$

To use the program Dconv2 for the Volterra integral equation, we modified the kernel as follows:

$$K_1(t-t') = \begin{cases} K(t-t'), & t' \leqslant t, \\ 0, & t' > t. \end{cases} \tag{18}$$

The data arrays in the upgraded program have been increased to handle 2048 input points for use with μSR histograms.

The program package RECOVERY, which is based on the maximum likelihood method (MLM), was chosen because this method attains the maximum possible resolution enhancement for a given signal to noise ratio in the input experimental data [9]. According to the MLM, the likelihood function

$$L = \mathcal{P}(F|G_0), \tag{19}$$

should be first defined, where $\mathcal{P}(F|G_0)$ is the conditional probability of observing a set of experimental data points

$$\{F_i\}, \quad i = 1, 2, \ldots, n,$$

which coincides with the real data set, providing that the solution is G_0. In our case G_0 is the muonium formation rate $n(t)$.

Consider the set of unknown values of the function $G_0(y_j)$, $j = 1, 2, \ldots, m$ as a vector in an m-dimensional space of solutions. Each point in this space corresponds to one possible solution, and the next step is to search for the likelihood function maximum

$$\max \mathcal{P}(F|G_0),$$

on a set of solutions limited by some necessary restrictions. For many problems, including the muonium formation rate problem, an important condition is that the solution is not negative.

An explicit form of the likelihood function for the case of polynomial statistics is given in review paper [14]. Poissonian statistics are a special case of polynomial statistics. If the data are described by Gaussian statistics, then the logarithmic likelihood function is the square of deviation between the experimental data $\{F\}$ and their approximation $\{\widehat{F}_0\}$, i.e.

$$\log L = \mathrm{const} - \tfrac{1}{2}\|F - \widehat{F}_0\|.$$

Here the two vertical lines denote the square norm and

$$\widehat{F}_0 = K\widehat{G}_0$$

is the integral transform (14) or (17) of the trial solution $\widehat{G}_0$.

The search for the likelihood function maximum is performed iteratively by the steepest ascent method which is a sign-inverted variant of steepest descent method. All explicit formulae of iterative algorithms for both polynomial and Gaussian input data statistics can be found in reference [10] and a full listing of the RECOVERY code in Fortran 77 is available from the CPC Program Library providing that person requesting the program sign the standard CPC non-profit use license. The more sophisticated conjugate gradient method is used in this paper for maximization of the likelihood function.

The number of detected positron in one time bin in our measurements was about 10^3–10^4 and it is well known that for such large numbers of events Poissonian statistics are reduced to Gaussian. It follows from the Eq. (13) that the variance of input data $P(t)$ is inversely proportional to the histogram values $N(t)$ and the main subroutine MLG8 should be used together with the program Dconv2. This subroutine is designed for input data having the Gaussian statistics with non-constant variance (see part 2 of paper [10]).

To demonstrate that our RECOVERY procedure is unbiased with respect to the analytical form of formation rate function $n(t)$ we choose two trial functions with strongly differing shapes

$$n_1(t) = \frac{C_1}{1+(t/t_0)^\alpha} \quad \text{and}$$
$$n_2(t) = C_2\left(A_1 e^{-t/t_1} + A_2 e^{-t/t_2}\right) \tag{20}$$

using the values

$$t_0 = 200, \quad \alpha = 5, \quad A_1 = 20,$$
$$t_1 = 50, \quad A_2 = 1, \quad t_2 = 500.$$

Time in the above formulae is measured in time bin units, C_1 and C_2 are the normalizing constants.

The integral (14) and function $\widehat{N}(t)$ from Eq. (8) were computed for each of the above trial functions. The parameters in Eq. (8) $A_0 = 0.2$, $B = 0$, and $N_0 = 4 \cdot 10^4$ for $n_1(t)$ and $N_0 = 1.7 \cdot 10^4$ for $n_2(t)$ were chosen for Monte Carlo simulations. The Poissonian deviates were applied to both trial functions in Eq. (8): $\widehat{N}_1(t)$ and $\widehat{N}_2(t)$ and in result we have obtained two simulated histograms $N_1(t)$ and $N_2(t)$ for 2048 data points. The results of estimation of the trial functions from these quasi-experimental' data by our recovering procedure are shown in Fig. 2 together with the exact solutions. They are in good agreement. We emphasize that the discrepancy between the estimate and exact solution in Fig. 2 does include two sources of errors: the systematic and the statistical ones. The total error in Fig. 2 is about 2%.

The statistical accuracy of the recovering procedure was also quantitatively evaluated by a Monte Carlo procedure which was applied M times, $10 < M < 20$ (see [11])

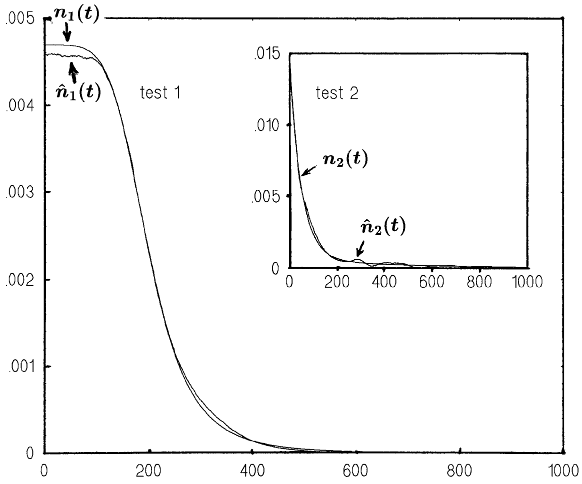

Fig. 2. Trial functions $n_1(t)$ and $n_2(t)$ together with their respective extracted values $\hat{n}_1(t)$ and $\hat{n}_2(t)$ from simulated data.

$$N_j(t_i) = \text{Entier}\left[\widehat{N}(t_i) + \text{NRAN}(i) \cdot \sqrt{\widehat{N}(t_i)}\right],$$
$$j = 1, 2, \ldots, M, \quad i = 1, 2, \ldots, N_{\text{data}}. \tag{21}$$

Here Entier[$\cdot$] denotes the greatest integer function, $\widehat{N}(t_i)$ are the histogram function values as computed by the formula (8) for $t \leqslant t_0$ (t_0 is the muonium formation time) and by the formula (9) for $t > t_0$, and NRAN(i) is a random number generator with Gaussian distribution. The statistical accuracy estimation is

$$\text{var}\left[n(t)\right] = \frac{1}{M-1} \sum_{j=1}^{M} \left[n_j(t) - n(t)\right]^2, \tag{22}$$

where $n(t)$ is the solution of the integral equation Eq. (14) for the real histogram $N(t)$, and $n_j(t)$ is the solution of the integral equation for Monte Carlo simulated input data $N_j(t)$.

The muonium formation rate function $n(t)$ in liquid helium at temperature $T = 0.7$ K is shown in Fig. 3 together with the estimation of its statistical accuracy (at level $\pm 1\sigma$).

It is seen from this figure that mean accuracy is less than 10% for total histogram statistics

$$\sum_{i=1}^{4000} N(t_i) \sim 6 \cdot 10^6.$$

The function $n(t)$ decreases to zero with increasing time faster than an exponential – in another words the

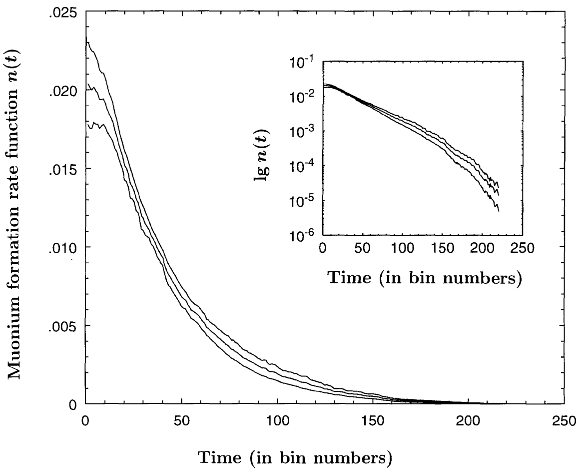

Fig. 3. Muonium formation rate function $n(t)$ in liquid He and its accuracy at the level $\pm\sigma$ (main plot). The same functions is shown in insert in logarithmic scale on Y axis. Time on X axis is measured in the histogram bin numbers, one bin time $\Delta t = 2.5$ ns. Temperature $T = 0.7$ K.

curve $n(t)$ is upwards convex in Y logarithmic scale plot.

3. Results

Fig. 4 shows the muonium formation rate $n(t)$ in superfluid helium over the temperature range

$$0.5\ \mathrm{K} \leqslant T \leqslant 1.35\ \mathrm{K}, \tag{23}$$

as extracted by our method.

The muonium formation rate increases for time interval $t < 0.1\ \mu$s with decreasing helium temperature and the rate decreases for time $t > 0.5\ \mu$s. It is interesting to compare these results with the measurements of muonium precession amplitude published in paper [15]. Mu spins conserve their coherence in transverse magnetic field if they are formed during the time less than half of a precession period. For magnetic field $H = 0.4$ Oe

$$t_{\mathrm{Mu}} = 1/(2\omega_{\mathrm{Mu}}) \simeq (2\gamma_{\mathrm{Mu}} H)^{-1} \simeq 0.2\ \mu\mathrm{s}. \tag{24}$$

Muonium precession amplitude A_{Mu} is determined by the first integral in Eq. (7). Using the data shown in Fig. 4 one can see, that the value $n(0)$ increases in temperature range $0.5 \leqslant T \leqslant 1.35$ K with reduction in helium temperature, but the A_{Mu} has maximum at $T \simeq 0.8$ K.

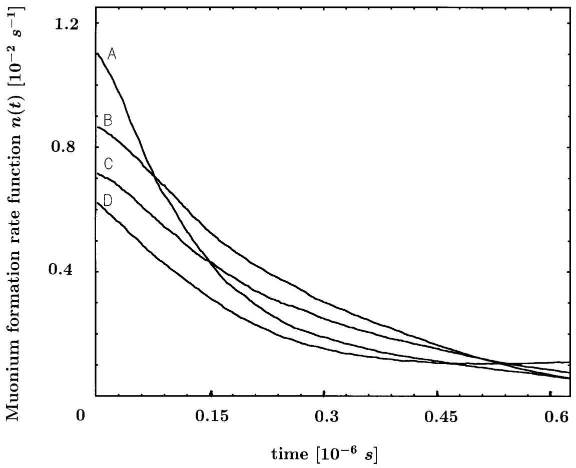

Fig. 4. Muonium formation rate functions $n(t)$ in liquid He in the temperature range 0.5 K $\leqslant T \leqslant 1.35$ K; (A) $T = 0.5$ K, (B) $T = 0.8$ K, (C) $T = 1$ K, (D) $T = 1.35$ K.

Let us consider the function $W(r)$ which is radial density distribution of muon–electron pairs. This function makes obvious physical sense for the Coulomb attraction of a muon and an electron in liquid helium in the viscous regime. This distribution function is appropriate when the ions have relaxed to their final local equilibrium and their relative velocity vector will be

$$V = -b\nabla\varphi, \tag{25}$$

where b is the mutual mobility and φ is the electric field potential.

We will ignore for simplicity the space asymmetry of distribution function $W(r)$, which was discovered in paper [16]. Then we obtain the equation

$$P(t) = 1 - 2\pi \int_0^{r(t)} W(\xi)\xi^2\, d\xi \tag{26}$$

instead of Eq. (3). Let us introduce the new variable $\tilde{t} = bt$ to eliminate the mobility b from Eq. (25). If $W(r)$ is independent of temperature, then both $P(t)$ and $n(t)$ should be universal for all temperatures. However we can see from Fig. 4 that a scaling law does not hold, because as the temperature changes, the mobility changes by a few orders of magnitude, while the function $n(t)$ values changes only in shape.

It can easily be shown that the velocity relaxation time

$$\tau_{\mathrm{v}} = Mb/e \tag{27}$$

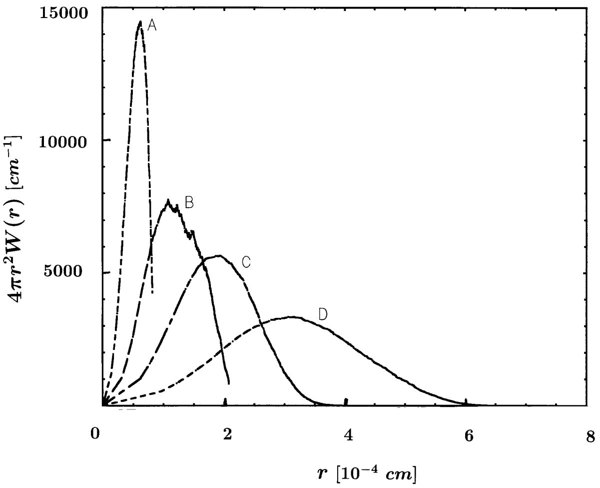

Fig. 5. Radial distribution functions $4\pi r^2 \cdot W(r)$ for helium temperature range $0.7\ \mathrm{K} \leqslant T \leqslant 1.35\ \mathrm{K}$; (A) $T = 1.35$ K, (B) $T = 1$ K, (C) $T = 0.8$ K, (D) $T = 0.7$ K.

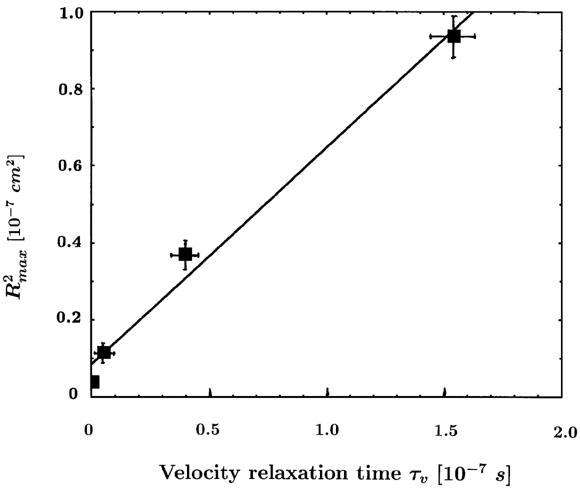

Fig. 6. The dependence $R^2_{\max}$ from the Fig. 5 versus of the velocity relaxation time τ_v at different temperatures and the straight line approximation of this dependence.

is much less than the muonium formation time above 0.7 K. Hence it follows that the lack of the scaling law is connected to the shape of distribution function $W(r)$. This function can easily be found from Eqs. (3), (25) and (26)

$$W(r) = \frac{n(t)}{4\pi eb} = \frac{n(r^3/3eb)}{4\pi eb}. \tag{28}$$

The muonium formation time for a muon and an electron spaced initially at distance r apart is

$$t = \frac{r^3}{3eb}, \tag{29}$$

which was substituted in the second equality in Eq. (28).

The distribution functions $4\pi r^2 W(r)$ are shown in Fig. 5 for helium temperature range $0.7\ \mathrm{K} \leqslant T \leqslant 1.35$ K. It is seen from this figure that both the mean and the dispersion of the distance between muon and electron pairs increase as the helium temperature is reduced. This follows by virtue of increasing mobility for particles in superfluid helium. The thermalization process in normal liquids are completed by a time of 10^{-12} to 10^{-10} s because of elastic phonon interactions. In contrast there is a gap $\delta E = 8$ K in the superfluid helium excitation spectrum [17] which results in an anomalous high mobility of impurity particles, such as muons. As the energy of particles is reduced to less than 8 K, the velocity relaxation time τ_v rises greatly.

The positions of the maximums in the Fig. 5 correspond to mean distances between muon and electron and their displacement is determined by dispersion of particles over the time τ_v.

Fig. 6 shows the squared position $R^2_{\max}$ of the maximum of the distribution function versus of the velocity relaxation time τ_v at different temperatures. It is easy to see that the dependence is close to linear

$$R^2_{\max} \propto \tau_\mathrm{v}.$$

This dependence reflects obviously the random diffusive character of the low-temperature thermalization of the charges.

4. Conclusions

The use of our non-parametric procedure based on the maximum likelihood method permits the effective solution of problems in μSR experiments. In our view it is especially helpful when analytical form of solution is unknown and it is a subject of research itself.

We have demonstrated that the direct reconstruction of the muonium formation rate function proves the lack of a scaling law for muon depolarization in superfluid helium with changing temperature, which results from the change of the radial distribution function $W(r)$ in a velocity relaxation process.

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