

## Kinetics of muonium formation in liquid helium

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The spatial distribution of electron–muon pairs in superfluid helium (He-II) is determined using a new algorithm for reconstructing the muonium ( $Mu$ ) formation probability. It is shown that because a gap is present in the excitation spectrum of He-II the thermalization time of muons and secondary electrons increases with decreasing temperature. As a result, the average distance in the electron–muon pairs increases and, correspondingly, the muonium formation rate decreases. © 1999 American Institute of Physics. [S0021-3640(99)01903-9]

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1. It is known that a muonium atom  $Mu = \mu^+ + e^-$  forms in liquid helium when a muon and an electron of the track recombine.<sup>1,2</sup> A positively charged particle in helium forms a small “chunk” with an associated mass  $M_+ \approx 50$  helium atoms, and the electron is localized in a cavity whose hydrodynamic mass is  $M_- \approx 200$  helium atoms (see the review Ref. 3 and also Ref. 4). On account of the large masses the time of approach of the charges and the formation of  $Mu$  in liquid helium are much longer than in other substances. In the simplest model<sup>5,6</sup> the interaction of a muon and a track electron is assumed to be the Coulomb interaction, and the viscous regime of approach,  $dv/dt=0$ , with the total relative mobility of the two charges  $b = b_+ + b_-$ , is studied. In a constant electric field, viscous motion

$$dr/dt = -b \cdot \nabla \phi \quad (1)$$

(here  $\phi$  is the interaction potential) is established in a time  $\tau_v = Mb/e$ , called the velocity relaxation time. For a Coulomb field the motion can be taken as viscous if the relative change in the field over the time  $\tau_v$ , viz.,  $\Delta E/E = 2v\tau_v/r = 2b^2M/r^3$ , is small. At low temperatures an electric field  $E_c = 50\text{--}100$  V/cm disrupts the formation of muonium.<sup>7</sup> This corresponds to distances  $r > (e/E_c)^{1/2}$  and for 10% accuracy gives an upper limit on the mobility  $b \approx 5\text{--}15$  cm<sup>2</sup>/V·s. Since the  $Mu$  formation time is several orders of magnitude longer than  $\tau_v$ , Eq. (1) can be used to describe the approach of the charges at temperatures to 0.7–0.8 K. These approximate considerations are confirmed by comparing the results of a direct calculation of the closing time of the charges on the basis of both Eq. (1) and the complete equations of motion: The difference does not exceed several percent at 0.7 K, and it is even smaller as temperature increases.

For a Gaussian spatial distribution of the charge the polarization function is close to  $\exp(-kt)$ .<sup>6</sup> This often justifies describing the formation of muonium by a first-order equation characteristic for chemical reactions.<sup>8</sup> In superfluid helium the conventional approach is unsatisfactory for the following reason. If in the equation (1) of viscous approach of the charges toward each other the new independent variable  $t^* = bt$  is introduced instead of the time  $t$ , then the mobility vanishes from the equation, and correspondingly the polarization function should be universal for all temperatures.

However, in the experiment of Ref. 1 it is shown that at least two exponentials, the ratio of whose amplitudes is temperature-dependent, is required in order to obtain the best fit of the muonium polarization function  $P(t)$ . Actually, the form of the function  $P(t^*)$  depends on the temperature and therefore it is not universal.

Let  $n(t)$  be the muonium formation probability as a function of time. Since half the polarization (in the  $S$  state) is not observed during the formation of muonium, in a zero magnetic field the muon polarization can be written as a sum of two integrals

$$P_{Mu}(t) = \frac{1}{2} \int_0^t n(t') dt' + \int_t^\infty n(t') dt', \quad (2)$$

which have a clear physical meaning. The first integral refers to muonium atoms in the triplet state ( $\uparrow\uparrow$ ) which have formed by the time  $t$ , while the second integral corresponds to free muons which have not yet recombined into muonium atoms.

Let us now consider a weak transverse magnetic field, such that the rotation of the free muon spin over its lifetime can be neglected. For liquid nitrogen, typically,  $H \leq 0.4$  Oe. The spins of the muonium atoms formed at the time  $t'$  start to precess with frequency  $\Omega_{Mu}$  and with a phase delay  $\Omega_{Mu}t'$ , and the polarization function will have two terms

$$P(t) = \frac{1}{2} \int_0^t n(t') \cos \Omega_{Mu}(t-t') dt' + \int_t^\infty n(t') dt', \quad (3)$$

where the first term describes the precession of the  $Mu$  spins at the Larmor frequency  $\Omega_{Mu}$  and with a delay  $t'$ , while the second term describes free muons, for which the direction of the spins remains virtually unchanged because of the weakness of the magnetic field.

The main information about  $P(t)$  is ordinarily extracted by approximating histograms of the positron counts by a model polarization function.<sup>8</sup> The true form of the function  $P(t)$  and, correspondingly,  $n(t)$  remain unknown. In the present letter we employ a new algorithm for determining the function  $n(t)$  by solving the integral equation (3) using the RECOVERY software package to reconstruct the signals.<sup>9</sup> A detailed description of the effective possibilities of this package can be found in Ref. 10.

2. In  $\mu$ SR experiments the directly measured quantity is the so-called histogram  $N(t)$ :

$$N(t) = N_0 [1 - A_0 \cdot P(t)] e^{-t/t_\mu} + B, \quad (4)$$

which actually determines the number of positrons arising in time with the decay of a positive muon. In this formula  $t_\mu = 2.197 \times 10^{-6}$  s is the muon lifetime,  $N_0$  is proportional to the intensity of the muon beam,  $A_0$  is the initial asymmetry factor, and  $B$  is the

random-coincidence background. The polarization function  $P(t)$  in Eq. (4) can be determined from the histogram  $N(t)$  only after the numerical values of the parameters  $N_0$ ,  $B$ , and  $A_0$  have been determined. At temperatures below 1.35 K the muonium formation time is much shorter than the duration of the histogram. This makes it possible to determine these parameters by a simple method. When the process leading to the formation of muonium atoms is completed, the equation for the histogram has the form

$$N_1(t) = [A + C \cos(\Omega_{Mu}t) + D \sin(\Omega_{Mu}t)]e^{-t/t_\mu} + B. \quad (5)$$

The parameters  $A$ ,  $B$ ,  $C$ , and  $D$  are determined by minimizing the quantity

$$\chi^2 = \sum_{t_i > t_0} \frac{[N(t_i) - N_1(t_i)]^2}{N(t_i)}. \quad (6)$$

This formula takes into account that the experimental data on  $N(t)$  have a Poisson distribution.

After the parameters  $A$ ,  $B$ ,  $C$ , and  $D$  are determined,  $N_0$  and the asymmetry  $A_0$  are calculated using the formulas

$$N_0 = A \exp(t_0/t_\mu), \quad A_{Mu} = \sqrt{C^2 + D^2}, \quad \text{and} \quad A_0 = \frac{A_{Mu}}{A}. \quad (7)$$

Now that all the constants in Eq. (4) are known, this formula can be inverted, and the normalized polarization

$$P(t) = \frac{\hat{P}(t)}{\hat{P}(0)}, \quad (8)$$

where the normalization constant  $\hat{P}(0)$  is an estimate of the value of the function  $\hat{P}(t)$  at time  $t=0$ , obtained from a small number of initial values using an optimal filtering program, can be calculated.<sup>11</sup> Figure 1 shows the initial histogram  $N(t)$  at  $T=0.7$  K and the function  $P(t)$ . As one can see from this figure, the polarization  $P(t)$  gradually moves away from the initial value  $P(0) \approx 1$  into a regime of uniform precession between the extreme values of the amplitudes  $+1/2$  and  $-1/2$ . This corresponds to the fact, noted at the beginning of this letter, that after muonium is formed only half the initial polarization is observed.<sup>8</sup>

It is clearly evident in Fig. 1 that the noise level of the function  $P(t)$  increases with  $t$ . We shall determine the noise level, starting from the fact that the histogram  $N(t)$  has a Poisson distribution for which  $\text{var}[N(t)] = N(t)$ . Assuming approximately that  $N(t) \approx N_0 e^{-t/t_\mu}$ , we obtain from Eq. (8)

$$\text{var}[P(t)] \approx \frac{1}{A_0^2 N(t) (\hat{P}(0))^2} \approx \frac{1}{4A_0^2 N(t)}. \quad (9)$$

The equation (9) for estimating the noise level in the function  $P(t)$  is used in the algorithm studied in the present letter.

We note that coefficients in expression (4) can also be found by the conventional method, adopted in  $\mu\text{SR}$ , using a parametric approximation of the function  $P(t)$  (helium requires at least two exponentials). However, this method can be successfully used only

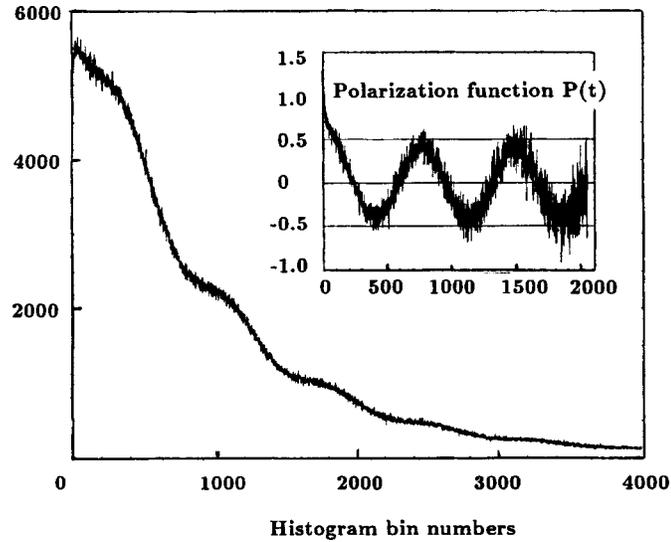


FIG. 1. Histogram  $N(t)$  and precession function  $P(t)$  of muonium in a magnetic field  $H=0.4$  G and liquid helium temperature  $T=0.7$  K. The channel width is 2.5 ns.

if reliable physical information about the form of the function  $n(t)$  and, correspondingly,  $P(t)$  is available. However, a nonparametric approach is better when the aim is to analyze these functions.

The equation (3) can be written in the equivalent form

$$P_1(t) = \int_0^t n(t') \left[ \frac{\cos \Omega_{Mu}(t-t')}{2} - 1 \right] dt', \quad (10)$$

where  $P_1(t) = P(t) - 1$ . The equation (10) is a convolution-type Volterra integral equation of the first kind for determining the nonnegative function  $n(t)$  from the input data  $P_1(t)$  with the kernel  $K(t-t')$  determined by the expression in the brackets in Eq. (10).

Introducing the modified kernel  $K_1(t-t')$

$$K_1(t-t') = \begin{cases} K(t-t'), & t' \leq t, \\ 0, & t' > t, \end{cases} \quad (11)$$

reduces Eq. (10) to a Fredholm integral equation with fixed integration limits. This equation can be solved using the DCONV2 program from the RECOVERY package.

The analysis of the errors and examples of the reconstruction of model functions which were performed in Ref. 12 showed that the method gives an average error of less than 2% in the function  $n(t)$  with a total data sample  $\sim 6 \times 10^6$  events in the histogram.

3. The results of reconstructing the function  $n(t)$  from histograms of the precession of muonium in a field  $H=0.4$  Oe are presented in Fig. 2. As the temperature decreases, the recombination rate on the initial segment  $t < 0.3 \mu s$  increases, and at long times ( $t > 0.5 \mu s$ ) the lower temperature, the lower the rate is. At  $T=0.7$  K virtually the entire recombination process is completed in the initial interval. As the temperature decreases

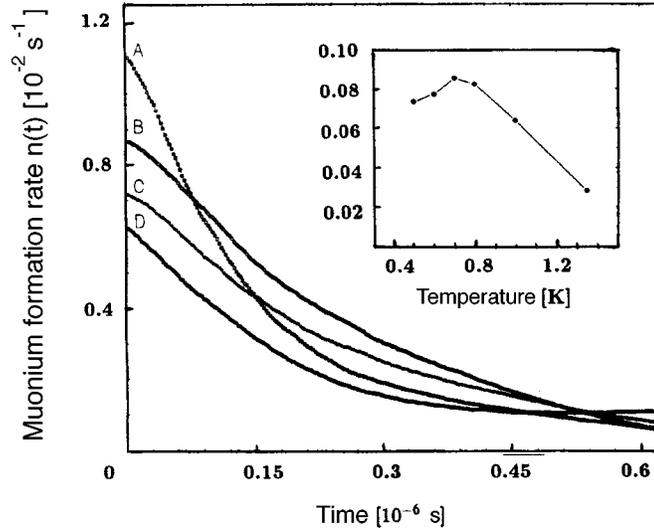


FIG. 2. Muonium formation rate  $n(t)$  at various temperatures: A:  $T=0.5$  K; B:  $T=0.8$  K; C:  $T=1$  K; D:  $T=1.35$  K. Inset: Temperature dependence of the muonium precession amplitude  $A_{Mu}$  calculated from Eqs. (7).

further, a long-time component reappears, though  $n(0)$  once again increases. Here a nonmonotonic temperature variation of the form of the function  $n(t)$  is most clearly manifested. After constructing  $n(t)$  in logarithmic coordinates, it can be verified that the recombination rate is not a simple exponential. The values of  $A_{Mu}$ , calculated using Eq. (7) and shown in the inset in Fig. 2, attest to the nonmonotonic character of the temperature dependence of  $n(t)$ . These values are virtually identical to the results obtained in Ref. 5.

Let us introduce the density function  $W(r)$  for the initial spatial distribution of muon–electron pairs. In the problem of the viscous closing of the particles on one another, the initial radial distribution of the pairs corresponds to the condition that the process of thermalization of the ionized particles has been completed and their velocity vector  $V = -b\nabla\phi$  has been established along the electric field lines. If for simplicity the spatial asymmetry of the distribution function  $W(r)$  found in Ref. 7 is neglected, then instead of Eq. (2) the polarization can be written in the form

$$P(t) = 1 - 2\pi \int_0^{r(t)} W(\xi) \xi^2 d\xi. \tag{12}$$

Since  $dP/dt = V \cdot dP/dr$ , it is easy to find from Eqs. (1) and (12) a relation between the recombination probability and the initial pair distribution density

$$n(t) = 4\pi beW(r)r^2 \cdot V. \tag{13}$$

In this formula it is assumed that for a purely Coulomb attraction the velocity is  $V = eb/r^2$  and the corresponding pair recombination time as a function of distance  $r$  is  $t = r^3/3be$ . Figure 3 shows the function  $W(r)$  for different temperatures in the range  $0.7 \text{ K} \leq T \leq 1.35 \text{ K}$ . One can see that as temperature decreases, the average interparticle

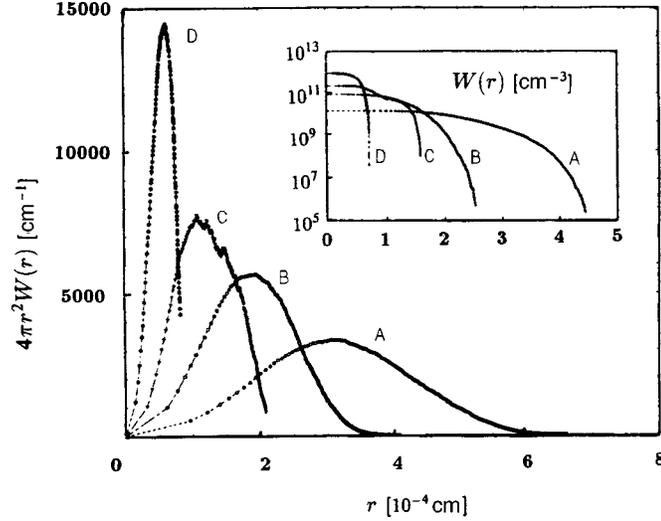


FIG. 3. Radial distribution function  $4\pi r^2 \cdot W(r)$  of muon–electron distances at various temperatures. A:  $T = 0.7$  K; B:  $T = 0.8$  K; C:  $T = 1$  K; D:  $T = 1.35$  K. Inset:  $W(r)$  on a logarithmic scale for the same temperatures as in the main plot. The distance in units of  $10^{-4}$  cm is plotted along the abscissa in both plots.

distance and its variance increase. In an ordinary condensate the thermalization of particles due to elastic collisions (phonons) is completed in a time  $10^{-12} - 10^{-10}$  s. A gap of the order of  $\delta = 8$  K is present in the excitation spectrum of superfluid helium,<sup>13</sup> as a result of which the mobility of the impurity particles is anomalously high. When the kinetic energy of the charges drops below 8 K, the scattering probability drops sharply and the velocity relaxation time  $\tau_v$  increases. Evidently, as a result, the average electron–muon distance increases with decreasing temperature. Assuming that before viscous closing is established the impurity particles move with velocity close to the Landau critical velocity,  $V_L \approx 60$  m/s,<sup>13</sup> the order of magnitude of the additional separation is  $R \approx V_L \cdot \tau_v$ .

Analysis shows that the character of the separation is more complicated. Figure 4 shows the squared position  $R_{\max}^2$  of the maximum of the distribution function as a function of the relative mobility of the muon and the electron at this temperature. It is easy to see that the dependence is linear  $R_{\max}^2 \propto b$  to a high degree of accuracy. Since the mobility  $b$  is proportional to  $\tau_v$ , this dependence reflects the random diffusive character of the low-temperature thermalization of the charges. At temperatures below 0.7 K the separation of the particles become so large that some of them fall outside the Onsager radius  $r_c = e^2 / \epsilon k_B T$ , where  $\epsilon$  is the dielectric constant and  $k_B$  is Boltzmann's constant. The probability of recombination of these muon–electron pairs is small, and a free (muon) fraction appears as a result.

Although, as one can see in Fig. 2, the initial formation rate of muonium increases with decreasing temperature, the total muonium asymmetry decreases. The decrease of  $A_{Mu}$  below 0.7 K is due to the fact that its formation time is  $\tau_{Mu} \approx (\gamma_{Mu} H)^{-1} \approx 0.2 \mu\text{s}$ . In this interval  $n(0)$  increases but the integral

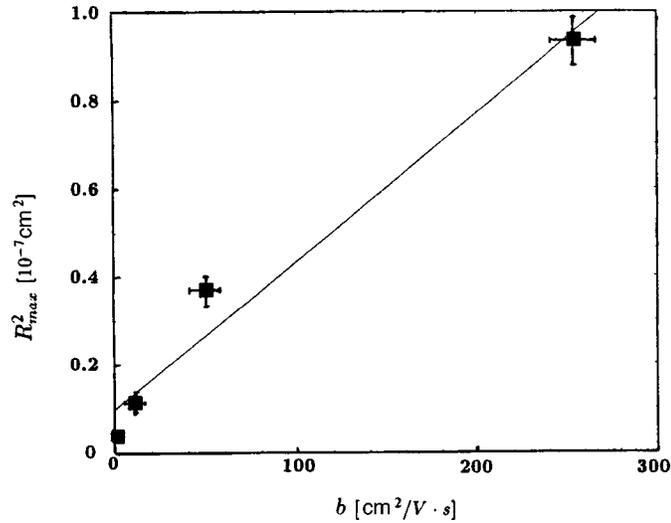


FIG. 4. Squared average separation in muon–electron pairs versus the relative mobility.

$$A_{Mu} = \int_0^{\tau_{Mu}} n(t) dt$$

decreases with decreasing temperature.<sup>5</sup>

In summary, a direct reconstruction of the muonium formation rate in superfluid helium shows that the absence of universality of the function  $P(t)$  as a function of temperature (its scaling) is due to an increase in the average interparticle distance in the muon–electron pairs in the process of velocity relaxation.

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