Depolarization of positive muons in solid hydrogen

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The precession spectra have been investigated for positive muons in liquid and solid hydrogen with different concentrations of ortho-molecules in the temperature range 1.5–20 K. The depolarization of muon spin in solid parahydrogen has been observed. The results obtained provide a basis for studying charged-particle kinetics. A jump is observed in the depolarization rate when a mixture containing more than 30% of ortho-molecules undergoes a transition from the liquid to the solid state. In contrast, this rate varies smoothly through this phase transition in the case of parahydrogen containing less than 2% of ortho-molecules. All samples showed a reduction in the depolarization rate at temperatures below 4 K. The results obtained can be interpreted qualitatively in terms of a theoretical model in which it is assumed that the muon forms the diamagnetic ion $(H_2\mu)^+$ in hydrogen.

INTRODUCTION

Quantum crystals of hydrogen have been attracting increasing attention.[1] General theoretical considerations[2] show that such crystals constitute the most suitable media for observing nonclassical charged-particle kinetics. However, attempts to measure the mobility of charges in solid hydrogen[3] have not been successful. Estimates[4] show that the diffusion coefficient of positively charged particles in solid hydrogen near the melting point T_m is less than 10^{-9} cm^2 s^{-1}, which is at the limit of the sensitivity range of the electrode method commonly used in such measurements. As far as we know, muon spin relaxation (μSR) has not been used to investigate charged-particle kinetics in hydrogen (in this case, positive muons). The damping of the initial muon polarization by the magnetic interaction between the muon and its environment can be used to investigate μ^+ dynamics under different conditions. The depolarization of muons in parahydrogen was observed in Ref. 5. This effect provides us with a new basis for studying impurity-particle kinetics in perfect parahydrogen crystals, which could not be done by magnetic methods (NMR and ESR) because of the absence of electronic and nuclear magnetic moments. If the thermalization of the muon results in the formation of the neutral muonium atom (Mu), its kinetics should be similar to the kinetics of atomic hydrogen. It does not seem possible to predict from first principles the probability of formation of muonium in hydrogen. It has been suggested[6] that about 60% of the muons form paramagnetic muonium ions in gaseous hydrogen at room temperature. However, experiments with a liquid-hydrogen chamber[7] have shown that the angular asymmetry of positrons from muon decays is high ($A = 0.31$), which argues against appreciable amounts of muonium in condensed hydrogen.

In contrast to neutral muonium, the kinetics of the positive muon is largely determined by the interaction of its charge with the medium. The electric field of the muon polarizes the ambient hydrogen molecules, producing a polarization pressure[3)] which, in liquid hydrogen, gives rise to the so-called ice sphere around the charged particle.[8] Measurements of the mobility of charged particles in liquid hydrogen[3] confirm this picture. In solid hydrogen, the excess pressure deforms the lattice near the charge, which sharply reduces the particle mobility. It was this effect, and the presence of defects in the specimen, that have probably prevented the authors of Ref. 3 from measuring the mobility of charges in crystalline hydrogen. In view of this, there is particular interest in μSR studies of hydrogen when it crystallizes and melts.

The present paper reports a study of the depolarization of positive muons in hydrogen in a wide range of temperature and of the density of ortho-molecules.

EXPERIMENTAL METHOD

Muon spin precession in a transverse magnetic field was recorded in the standard "Muonium" apparatus[9] of the synchrocyclotron at the Leningrad Institute of Nuclear Physics (LIYaF). The magnetic field was produced by Helmholtz coils and amounted to $B_\perp = 286$ Oe. This field was stationary and uniform within the sample to within 10^{-3}. To reduce the divergence of the beam and the background due to the outer flanges of the cryostat, we used a collimator, 38 mm in diameter, and a circular start-counter phosphor, 45 mm in diameter and 1.7 mm thick. A copper filter was placed between the first counter and the collimator. Its thickness was chosen so as to ensure the maximum number of muons stopped in the sample.

The hydrogen was held in the low-temperature chamber of the cryostat and took the form of a vessel, 90 mm in diameter, made from annealed stainless steel foil, 45 μm thick. Hydrogen was crystallized by bringing the liquid into contact with a cooled copper plate. This plate was in thermal contact with a thermally-insulated vessel (thermostat) containing liquid hydrogen. The sample temperature in the region of T_m could be very accurately varied, and stabilized to within 0.01 K, by pumping off hydrogen vapor from the thermostat. The crystal was cooled further by liquid helium,

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first poured into the thermostat and then onto the surface of the solid hydrogen. The temperature of the upper and lower parts of the crystal was measured to within 10 mK by two carbon thermometers located outside the muon beam. Below 4.2 K and above 10 K, the temperature was stabilized by holding the vapor pressure in the cryostat constant by means of a membrane pressure controller. The crystal temperature drift was less than 2 mK per hour under these conditions. Heaters were used to accelerate the rise in the sample temperature. Temperature equalization over the crystal height took 30–40 min. The long time taken for the temperature to settle was due to the low heat capacity and low thermal conductivity of hydrogen. At superfluid-helium temperatures ($T < 2.17$ K), the time for the temperature to settle was less than 5 min, and there was no temperature gradient. For temperatures in the range $4\ \mathrm{K} < T < 10\ \mathrm{K}$, the sample was adiabatically heated, and the temperature gradient was reduced to 0.01 K/cm.

In a prolonged experiment, the composition of hydrogen changed as a result of the ortho-para transformation. Chromatographic analysis showed that the reduction in the concentration of ortho-hydrogen in this cryostat could be described by the well-known law (to within 1%)

$$x(t) = x_0 (1 + k x_0 t)^{-1}, \qquad (1)$$

where x_0 is the initial ortho-hydrogen concentration, and $k = 0.012\ \mathrm{h}^{-1}$ for the liquid and $k = 0.017\ \mathrm{h}^{-1}$ for the solid.[10] An experiment with a given sample usually took about 24 h. The concentrations quoted below are averages over this time interval.

When the experimental spectra are analyzed by the least-squares method (LSM), it is important to take into account the fact that some of the muons stop in the cryostat walls. Accordingly, the number $N(t)$ of precessing-muon decays recorded by the positron telescope was described by

$$N(t)/N_0 = \exp(-t/\tau_\mu)$$
$$\times \{1 + A[\alpha P(t) + \beta \exp(-t/\tau_c)] \cos(\omega t + \varphi)\} + F, \qquad (2)$$

where N_0 is the normalizing constant $\tau_\mu = 2.197\ \mu\mathrm{s}$ is the muon lifetime, Λ is the initial precession asymmetry, α, β are fractions of muons stopping in hydrogen and in the warm walls of the cryostat, respectively, $P(t)$ is the time dependence of the muon polarization in hydrogen, $\tau_c = 6.3\ \mu\mathrm{s}$ is the muon depolarization time in the empty cryostat (determined in a calibration experiment), ω, φ are the precession frequency and phase of the muon spin, and F is the background. The number of events in the spectrum was usually 10^6–1.5×10^6, which meant that the average uncertainty in the depolarization rate was 5%. The coefficients α and β, obtained in the course of analysis, were found to be 0.8 and 0.2, respectively. It was shown earlier[5] that the stainless steel foil was antiferromagnetic below 20 K and that the asymmetry parameter in this foil was negligible. The absence of muon precession in cold cryostat walls ensured that muons stopping in these walls contributed only to the background F. The ratio of the "effective thickness" of all the cryostat walls to the hydrogen specimen "thickness" was less than 0.17.

The spectra were analyzed both for the exponential polarization decay $P(t) = \exp(-\Lambda t)$ and for the Gaussian decay $P(t) = \exp(-\sigma^2 t^2)$. The χ^2 value for the latter was systematically higher, so that we report only the depolarization rates Λ.

EXPERIMENTAL RESULTS

The rate of depolarization of muons in parahydrogen containing less than 2% of ortho-molecules (see the figure given in Ref. 5) in the liquid state is low and is independent of temperature to within experimental error. The depolarization rate rises in the solid state, reaching a maximum at $T \simeq 5$ K. Further cooling of the parahydrogen results in an appreciable reduction in Λ. Below 2 K, the derivative $\partial\Lambda/\partial T$ is very high and amounts to $0.02\ \mu\mathrm{s}^{-1}\ \mathrm{K}^{-1}$.

The depolarization rate in the solid state increases with increasing ortho-hydrogen concentration. A radical change in the behavior of $\Lambda(T)$ is observed near T_m. Figure 1 shows the temperature dependence of the depolarization rate for a sample containing approximately 55% of ortho-molecules. A jump occurs in Λ when crystallization takes place. It is clear from the figure that this jump appears in a very narrow temperature range which, at any rate, is less than 0.05 K. The depolarization rate increases with decreasing temperature down to 2.5–2 K, and thereafter decreases systematically. A special experiment showed that the reduction of Λ was not related to the penetration of the sample by superfluid helium.[11] We note that the depolarization rate in the liquid is low and practically independent of temperature. Five samples with ortho-molecule concentrations up to 66% were studied. The depolarization rate in the temperature range 14–20 K was found to be 0.04–0.05 $\mu\mathrm{s}^{-1}$ and was independent of composition. It is still not entirely clear whether this result actually represents the depolarization of muons in the liquid phase and is not a consequence of instrumental effects.

Figure 2 shows the muon depolarization rate as a function of the concentration of the ortho-molecules and gives the maximum values of Λ^2 for different specimens. In the

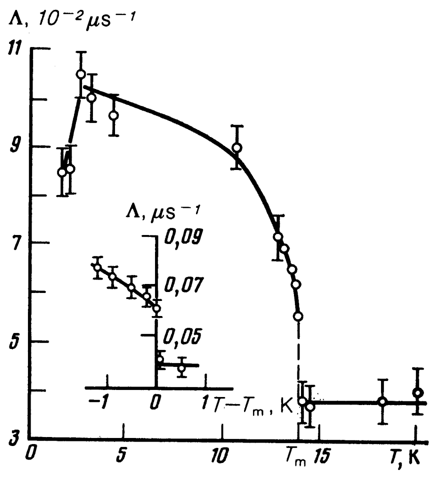

FIG. 1. Temperature dependence of the rate of depolarization of muons in hydrogen containing 55% of ortho-hydrogen. The solid lines constitute conjectures. Inset shows the region near T_m.

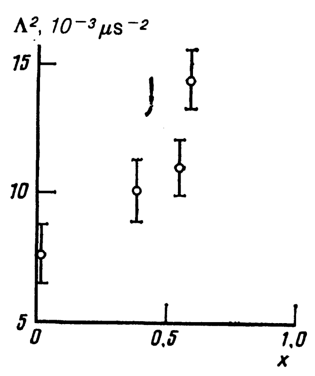

FIG. 2. Maximum depolarization rate as a function of the concentration of the ortho-molecules.

case of para-ortho mixtures of hydrogen, the decay of polarization due to the dipole-dipole interaction between the muon and the nuclear magnetic moments of the ortho-molecules must be taken into account. For a stationary muon and high ortho-hydrogen concentrations, the polarization is given by the Gaussian law[12]

$$P(t)=\exp(-\sigma^2t^2),$$
$$\sigma^2={}^2/_{15}J(J+1)(\hbar\gamma_p\gamma_\mu)^2x\sum_k r_k^{-6}, \tag{3}$$

where $J=1$ is the nuclear spin of the ortho-molecules, γ_p, γ_μ are the gyromagnetic ratios of the proton and muon, and x is the ortho-molecule concentration. For mixtures containing small amounts of ortho-hydrogen, dipole-dipole relaxation is described by the exponential law[13]

$$P(t)=\exp(-\Lambda t), \tag{4}$$
$$\Lambda\approx4.5\hbar\gamma_p\gamma_\mu\rho^{-1}x\langle|m|\rangle, \tag{5}$$

where ρ is the volume per hydrogen molecule and $\langle|m|\rangle$ is the average spin component. For high ortho-hydrogen concentrations, the calculated and measured depolarization rates can be made to agree by assuming that the muon is located in an interstitial position in the lattice. However, it is clear from Fig. 2 that the experiment with practically pure parahydrogen ($x<2\%$) indicates that the observed depolarization cannot be due to the magnetic interaction (5) between the muon and the impurity orthohydrogen molecules. Fourier analysis of the muon precession spectra in condensed hydrogen showed no evidence of the presence of the muonium component (to within 2% of the amplitude of the spectrum at the muonium frequency). This is in agreement with Ref. 7.

Under the working conditions of the cryostat, a small amount of atmospheric oxygen ($<10^{-4}$) may have entered the hydrogen sample. In principle, paramagnetic impurities may have been responsible for the depolarization effect. However, estimates showed that, in the solid phase, the contribution of oxygen was unimportant. The point is that the solubility of oxygen in condensed hydrogen is less than 10^{-8}, and it forms a suspension with particle sizes of 0.1–10 μm (Ref. 10). It follows from (5) that this type of impurity in solid hydrogen can produce a depolarization rate Λ of less than 100 s^{-1}.

It is readily shown that the probability that a muon will be captured by impurities is negligible. If, for the purposes of an estimate, we take the maximum diffusion coefficient for charged particles in solid hydrogen to be approximately 10^{-9} cm^2 s^{-1} (Ref. 14), we see that the muon traverses a distance of the order of 10^{-7} cm in a time corresponding to its lifetime. A basically new mechanism of depolarization, related to the specific interaction between the muon and hydrogen, is thus seen to occur in solid hydrogen.

MUON DEPOLARIZATION MECHANISM IN SOLID HYDROGEN

The theory of the behavior of the muon in hydrogen has been developed by Belousov and Smilga.[14] The theoretical model rests on the assumption that the muon forms the ion $(H_2\mu)^+$ with hydrogen. The alternative possibility is the formation of the MuH molecule. However, this is relatively unlikely because the exchange reaction $\mu^+ + H_2 \rightarrow MuH + H^+$ can proceed only at the hot stage, when the muon has not yet been thermalized. Actually, because of the isotope effect, the lowest vibrational state of the MuH molecule occurs at approximately 0.3 eV above the corresponding state in H_2, and the exchange reaction is endothermal. Moreover, analysis shows that depolarization does not occur in the MuH molecule. The molecule is rapidly ($t\simeq10^{-10}$–10^{-12} s) thermalized, and is found in the ground state with angular momentum $K=0$. The dipole-dipole interaction between the magnetic moments of the muon and proton, averaged over the spherically symmetric wave function of the rotational state, vanishes in exactly the same way as the dipole-dipole term in the calculation of the hyperfine interaction between the proton and electron magnetic moments in the ground state of the hydrogen atom. The muon polarization will therefore persist in the MuH molecule in pure parahydrogen.

It would thus appear that the muon forms the ion $(H_2\mu)^+$ in condensed hydrogen. It is well known that the binding energy of the proton in the ion H_3^+ is 4.7 eV (Ref. 15). Apart from the isotope effect (0.2–0.4 eV), this is also the binding energy of the muon in the ion $(H_2\mu)^+$. The formation of the $(H_2\mu)^+$ ion is energetically very convenient, and this state has the highest probability. In the adiabatic approximation, the configuration of the ion is determined by the Coulomb interaction, so that it may be assumed that the $(H_2\mu)^+$ ion (like the H_3^+ ion) has the configuration of an equilateral triangle with side lengths $R=0.85$ Å (Ref. 15). Usually, the duration of a chemical reaction is determined by the elementary release of energy into the medium, and amounts to less than 10^{-10} s. It is readily shown that, if the $(H_2\mu)^+$ ion is formed in a time interval of this order, the spin states of the proton and muon remain unaltered. We shall assume that the rotational degrees of freedom of the $(H_2\mu)^+$ ion are frozen in the lattice. This is a crucial assumption. The rotational levels of the frozen ion are infinitely degenerate and, because of this, the energy barrier to proton spin transitions from the antiparallel to the parallel states is removed. Under the above assumptions, the muon-spin dynamics in ortho- and parahydrogen differs by the initial conditions and the form of the spin density matrix at $t = 0$. In a strong magnetic field, the secular part of the effective spin Hamiltonian is

$$\mathcal{H} = -2B \sum_i \mu_i s_{iz} + \sum_{i<k} a_{ik} (\mathbf{s}_i \cdot \mathbf{s}_k - 3 s_{iz} s_{kz}),$$

$$a_{ik} = 2\mu_i \mu_k (3\cos^2\theta_{ik} - 1) r_{ik}^{-3}, \qquad \cos\theta_{ik} = \mathbf{B}\cdot\mathbf{r}_{ik}/Br_{ik}, \tag{6}$$

where μ_i, s_i are the magnetic moments and spins of the muon and proton, the z axis lies along the magnetic field $\mathbf{B}$, and $r_{ik} = R = 0.85$ Å is the side length of the triangle $(H_2\mu)^+$. The condition for the validity of (6) is obvious: the field B must be much greater than the local field due to the proton at the muon, i.e., $B \gg \mu_p/R^3 \simeq 25$ G. The Hamiltonian (6) has eight levels that depend on the orientation of the plane of the ion in the magnetic field:

$$\varepsilon_{1,7} = -{}^1/_2 [a_{12} + a_+ \pm (\omega_\mu + 2\omega_p)],$$

$$\varepsilon_{3,5} = -{}^1/_2 [a_{12} - a_+ \pm (\omega_\mu - 2\omega_p)],$$

$$\varepsilon_{2,4} = {}^1/_2 [-\omega_\mu + a_{12} \pm (a_{12}^2 + a_-^2)^{1/2}],$$

$$\varepsilon_{6,8} = {}^1/_2 [\omega_\mu + a_{12} \pm (a_{12}^2 + a_-^2)^{1/2}], \tag{7}$$

where $\omega_{\mu,p} = \gamma_{\mu,p} B$, $a_\pm = a_{13} \pm a_{23}$, and a_{ik} are the coefficients in the Hamiltonian (6).

In a strong magnetic field, the polarization component parallel to $\mathbf{B}$ is conserved, and the transverse polarization component is given by

$$P_\perp(t) = P_\perp(0) \{{}^2/_3 x s_1(t) + (1 - {}^2/_3 x)[0{,}22 + s_2(t)]\} \cos\omega_\mu t, \tag{8}$$

where

$$s_1(t) = \frac{1}{2} \int_0^1 \cos[(2-3z)\omega_{\mu p} t] J_0\left(\frac{3}{2}\omega_{\mu p} t z\right)(1-z)^{-1/2}\, dz,$$

$$s_2(t) = 0{,}39 \int_0^1 J_0\left(\frac{3^{3/2}}{2}\omega_{\mu p} t z\right)(1-z)^{-1/2}\, dz,$$

$J_0(x)$ is the Bessel function of order zero, and $\omega_{\mu p} = 2\mu_p \mu_p/\hbar R^3 \simeq 2\times 10^6\ \mathrm{s}^{-1}$. We emphasize that the observed depolarization arises as a result of averaging over different orientations of the ion $(H_2\mu)^+$.

DISCUSSION OF RESULTS

Figure 3 shows the muon spin precession in a parahydrogen crystal at $T = 4.2$ K. The time spectrum is described by the interpolation formula (2) with damping rate $\Lambda = 0.09 \pm 0.01\ \mu\mathrm{s}^{-1}$ and χ^2 is then 1.02. The solid line shows the polarization function (8), calculated for the model of a stationary $(H_2\mu)^+$ ion. As can be seen, the calculated depolarization rate is much greater than the measured value of Λ. This is so because the low energy of interaction between the $(H_2\mu)^+$ ion and its environment does not ensure complete fixing of the triangle of positive charges in the lattice, as assumed in Ref. 14. It is well known that the mutual displacement of the spins produces an averaging of the local fields and a reduction in the depolarization rate.[16] The translational motion of the μ^+ from one H_2 molecule to another is unlikely because of its high binding energy in the $(H_2\mu)^+$ ion ($E \simeq 4$ eV). The rotational motion of the ion is much more realistic. In the hcp hydrogen lattice (symmetry group D_{3h}), the ion lies on a threefold axis and its charge polarizes the ambient molecules, whose electric field produces a corrugated potential distribution for different orientations of the ion. The potential distribution is difficult to calculate from first principles, but estimates show that the anisotropic part of the interaction between the $(H_2\mu)^+$ ion and the induced dipole moments of the H_2 molecules gives rise to potential barriers of the order of 10–100 K. The role of the dipole–quadrupole interaction in the freezing of the $(H_2\mu)^+$ ion was noted in Ref. 17.

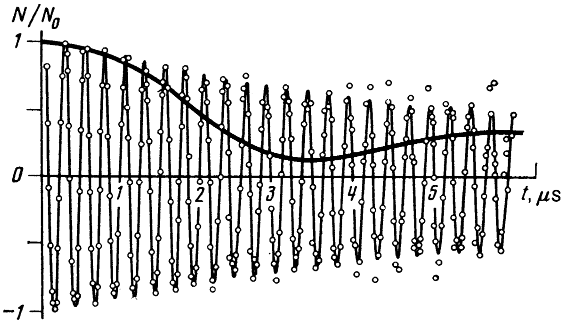

FIG. 3. Muon spin precession at $T = 4.2$ K in a field $H = 286$ Oe. Solid line shows the polarization function calculated from (8).

Random rotations of the ion are conveniently characterized by a rotational diffusion frequency $\Omega = \tau_c^{-1}$, where τ_c is the expectation time of the rotation of the ion from one equilibrium position to another. For a diffusing ion in a strong field, the transverse polarization can be qualitatively described by the simple exponential formula $P(t) = P_\perp(0)\exp(-\Lambda t)$, and the rate of depolarization decreases with increasing rotational frequency:

$$\Lambda = 2\omega_{\mu p}^2 \tau_c. \tag{9}$$

When $\tau_c = 10^{-8}$ s, the rate of depolarization agrees with the experimental value of Λ at $T = 4$ K.

Rotational diffusion of the ion can explain the temperature dependence of the depolarization in solid hydrogen and its low value in the liquid. It is well known that processes involving tunneling through potential barriers can be described by the Arrhenius law $\Lambda \sim \exp(E/kt)$. It would appear that thermal activation of rotational diffusion occurs in hydrogen at temperatures above 4 K. In particular, $E \simeq 10$ K for parahydrogen (Fig. 2), and this confirms that the anisotropic part of the interaction with the lattice is small.

The weak depolarization in the liquid phase is due to Brownian rotation of the ice ball.[8] A similar effect is known in NMR as dynamic line narrowing. The radius of the ice ball at $T = 15$ K is $R \simeq 10$ Å. Random rotation of the ball through $\varphi = \pi/3$ occurs in a time $\tau_c = 0.5(\varphi R)^2 \mathscr{D}^{-1} \leqslant 10^{-9}$ s, where $\mathscr{D} = 4kT/6\pi\eta R$, and η is the viscosity of hydrogen. For such rapid rotations, the local magnetic fields due to the protons are effectively averaged out and, in accordance with (9), the depolarization rate is low ($\Lambda \lesssim 0.01$

μs^{-1}). As already noted, the observed depolarization rate Λ in the liquid may possibly be due to instrumental effects, and the retardation of Brownian rotation was not seen in the experiment. Because of this, the depolarization rate Λ in parahydrogen varies smoothly across the melting point T_m.

In mixtures containing ortho-hydrogen concentrations $x > 30\%$, a small but well-defined jump in $\Lambda(T)$ is observed at $T = T_m$. This jump is naturally interpreted in terms of the dipole-dipole relaxation due to the interaction between the magnetic moment of the ion and the magnetic moments of the orthomolecules in the environment. Estimates of the change in Λ in the course of melting, based on (3), are in good agreement with experimental data. It is important to note that, if the normal hydrogen crystals are imperfect, especially in the case of rapid crystallization, this may additionally increase the jump due to freezing of the rotational diffusion of the $(H_2\mu)^+$ ion by defects. This explains the early experiments[18] with normal hydrogen, crystallized by liquid helium, in which the jump in the depolarization rate was higher by a factor of two.

All the samples investigated showed a sharp reduction in the rate of depolarization at low temperatures $T < 2$ K. It is possible that this was due to quantum phenomena, namely, sub-barrier rotations of the ion. Theory shows[2] that low-temperature diffusion processes are described by high powers of temperature, which may explain the large value of the derivative $\partial\Lambda/\partial T$ at low temperatures.

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[3)] The pressure is given by $p = \alpha e^2/2\Omega r^4$, where $\alpha = 0.8 \times 10^{-24}$ cm^3 is the polarizability of molecular hydrogen, Ω is the volume per molecule, and r the distance from the muon.

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