Theory of the muon technique for the study of solid and liquid hydrogen

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A theory of the behavior of the muon-spin polarization in solid and liquid hydrogen in a strong external magnetic field is developed. It is shown that the experimental results can be explained only if the muon forms a diamagnetic complex $(H_2\mu)^+$. It is shown that the spin interactions inside the complex lead to effective depolarization of the muon spin, and the rates of depolarization are close for para and ortho hydrogen. For a "frozen" $(H_2\mu)^+$ ion, account is taken of the influence exerted on the depolarization rate by rotational diffusion along crystallographically equivalent orientations in the hcp lattice of hydrogen. The observed polarization rate Λ decreases with increase of the rotational diffusion rate. An ice sphere is produced in liquid hydrogen around the $(H_2\mu)^+$ ion. The Brownian rotation of this sphere should lead to a noticeable decrease of the rate of depolarization on going through the melting point, as is indeed observed in experiment.

INTRODUCTION

The muon technique was recently used for the first time to study solid and liquid hydrogen.[1-3] The principles of the theory that explains the observed facts were developed in Ref. 4. We present below a complete theory of the behavior of μ^+ in liquid hydrogen.

We emphasize first of all that it is often impossible to predict, from first principles, the behavior of a muon in a particular substance, nor the chemical complexes into which it will be bound. Moreover, as shown by a number of experiments, the character of the chemical reactions of a muon is substantially altered in phase transitions even in chemical objects such as inert gases.[5] Therefore, on the one hand, the theory must be based on experiment, and on the other, it is necessary to analyze several possible variants. Experiment has shown that solid and liquid hydrogen contain only a diamagnetic muon fraction. According to the available data,[5] on the contrary, in hydrogen gas 60% of the muons produce muonium. As already noted, such changes in the behavior of the muon are quite frequently observed in phase transitions.

In solid hydrogen, the depolarization rates are of the order of $10^5\ s^{-1}$ and differ little in para- and orthohydrogen. In the liquid phase, the damping rate is decreased by about one-half ($6\cdot 10^{-4}\ s^{-1}$). The fact that the polarization of the diamagnetic fraction attenuates quite rapidly in parahydrogen indicates unequivocally that in solid and liquid hydrogen the muon is bound in some diamagnetic chemical compound. Indeed, the nuclear spin of the paramolecule is zero, and if the muon were free and in an interstice, its spin would hardly be depolarized.

The simplest diamagnetic complexes of a muon in hydrogen are the molecule MuH and the ion $(H_2\mu)^+$. Owing to the mass difference between the proton and the muon, the vibrational structure of the MuH molecule is $\omega_{\mathrm{MuH}} \approx 2.2\omega_{H_2}$. Recognizing that $\hbar\omega_{H_2} = 0.52$ eV, we see that the energy of the zero-point oscillations of the MuH molecule is approximately 0.31 eV higher than in the H_2 molecule. Thus, the exchange reaction $\mathrm{Mu} + H_2 \rightarrow \mathrm{MuH} + H$ or $\mu^+ + H_2 \rightarrow \mathrm{MuH} + H^+$ is endothermic and is possible so long as the muon (muonium) is not thermalized ("hot" channel). It can therefore be assumed that these reactions are relatively unlikely. We nevertheless consider this variant, too.

On the contrary, approximately 4.5 eV is released in the reaction $H_2 + \mu^2 \rightarrow (H_2\mu)^+$, this reaction can be freely produced in the condensed phase after thermalization, and this channel is the most probable. The behavior of muon polarization in an $(H_2\mu)^+$ "frozen" in a crystal was analyzed in Ref. 4.

The equation for the spin density matrix is

$$i\hbar\frac{\partial}{\partial t}\rho(s,t)=[H(s),\rho(s,t)], \tag{1}$$

where the spin Hamiltonian is

$$H(s)=\mathrm{Sp}_q V(q,\ s)\rho^{(0)}(q,\ t). \tag{1a}$$

The muon spin polarization is equal to

$$\begin{aligned} P_i(t) &= \mathrm{Sp}_s\,\sigma_{\mu_i}\rho(s,t) \\ &= \mathrm{Sp}_s\,\sigma_{\mu_i}\exp(-iH(s)t/\hbar)\,\rho(s,0)\exp(iH(s)t/\hbar). \end{aligned} \tag{2}$$

1. FROZEN $(H_2\mu)^+$ ION

We consider in this section the behavior of muon polarization in an $(H_2\mu)^+$ ion frozen in a lattice, making extensive use of the results of Ref. 4. Note that the observed diamagnetic fraction cannot be attributed to formation of a MuH molecule. Indeed, in the rotational ground state of the molecule the problem reduces, after averaging over the coordinate variables, to motion of two unbound spins in an external field. In parahydrogen, where the magnetic moment of the molecules of the medium is zero, we would observe in a perpendicular field, in both the solid and liquid medium, a practically undamped precession at the free-muon frequency. The damping rate would then hardly change in the phase transition. Thus, the hypothesis that a MuH molecule is formed must be rejected, since experiment contradicts totally both conclusions.

Neglecting the small change in shape because of the difference between the muon and proton masses, we assume that the $(H_2\mu)^+$ ion, just as the H_3^+ ion, is an equilateral

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triangle with side $R \approx 0.85$ Å. The proton binding energy in the H_3^+ ion is 4.7 eV. This effect lowers it by 0.2–0.3 eV, which is unimportant to us. The time of formation of the $(H_2\mu)^+$ ion in the ground state is limited by the rate of thermalization of the vibrational and rotational degrees of freedom. As indicated, a typical time is $t \lesssim 10^{-10} - 10^{-12}$ s, therefore the initial spin states of μ^+ and H_2 remain unchanged by the instant of completion of the elementary act. Simple estimates[4] show that in liquid hydrogen, owing to the polarization pressure, there can be produced around the ion an icy sphere, just as in helium.[6,7] The sphere radius is estimated at $r \sim 4$–7 Å. In the solid phase the polarization of the medium leads to a noticeable distortion of the lattice around the ion.

Generally speaking, the location of the $(H_2\mu)^+$ ion in the lattice cannot be unambiguously identified. Simple estimates show, however, that the potential energy of the charge-dipole interaction for the ion is a maximum in a tetrapore. Indeed, the polarizability of molecular hydrogen is $\langle\alpha\rangle = 5.4a_0^3$ ($a_0 = 0.53$ Å is the Bohr radius),[8] and we obtain for the ion $U \approx 0.5$ eV at the lattice site, $U_0 \approx 0.8$–0.9 eV in the octapore, and $U_T \approx 0.9$ eV in the tetrapore. It can be seen that the interaction energy of the $(H_2\mu)^+$ ion with the lattice is higher by one or two orders than the binding energy (30–40 K) of the hydrogen molecule in the lattice. However, the interaction between the charged and the induced dipoles does not freeze the rotation of the ion. Obviously, the potential energy of the electrostatic interaction between the ion and the lattice depends strongly on the ion orientation in accordance with the crystal symmetry. This "corrugation" of the potential relief determines the ion-rotation freezing energy. For the symmetry group D_{3h} the size of the corrugation is determined by the interaction of the octupole moment of the $(H_2\mu)^+$ ion with the quadrupole and with the induced dipole moments of the H_2 molecules. Estimates show that the height of the potential barrier (corrugation) for the ion of the center of an undistorted tetrapore is $\Delta U_T \sim 100$ K and that $\Delta U \sim 10$ K at the lattice site and in the octapore. It follows from the same estimates that the energywise favorable orientation of the ion plane is perpendicular to the threefold symmetry axis.

We assume hereafter that the localized $(H_2\mu)^+$ ion is frozen in the hydrogen lattice and that its coordinate density matrix is a function of the positions of the nuclei.

We emphasize that since the $(H_2\mu)^+$ ion interacts intensely with the lattice, an adequate description of its behavior requires the use of the density-matrix formalism. Note that for a frozen ion the energy levels with total proton spin $I = 0$ and 1 are degenerate in the coordinate variables. It is also obvious that the effective spin Hamiltonian for the frozen ion is entirely different than for a free one. Therefore, in contrast to the free ion, the spin interactions can lead to $(I = 1) \rightarrow (I = 0)$ transitions.

The spin Hamiltonian of the molecular $(H_2\mu)^+$ ion in an external magnetic field **B**, obtained by averaging over the coordinate density matrix, is

$$\hat{H} = -\sum_{i=1}^{3} 2\mu_i(\mathbf{s}_i\mathbf{B}) + \sum_{i<k} 4\mu_i\mu_k r_{ik}^{-3}\{\mathbf{s}_i\mathbf{s}_k - 3(\mathbf{s}_i\mathbf{n}_{ik})(\mathbf{s}_k\mathbf{n}_{ik})\}; \quad (3)$$

where μ_i and $\mathbf{s}_i$ are respectively the magnetic moments and spin operators of the muon and of the protons, r_{ik} is the distance between the nuclei, and $\mathbf{n}_{ik} = \mathbf{r}_{ik}/r_{ik}$. The problem of three interacting spins $s_i = 1/2$ in a strong external field was considered in Refs. 9 and 10 to determine the NMRD spectrum and line shape of compounds containing three protons. We need a more complete analysis, hwoever, primarily because to determine the muon polarization we must know the spin density matrix $\rho(t)$. An important role in the calculation of the latter is obviously played by the initial conditions. Finally, considerable interest attaches to cases when the external magnetic field is comparable with the internal fields or is altogther nonexistent.

The spin density matrix of the $(H_2\mu)^+$ ion at the initial instant of time can be written in the form

$$\rho_s(0) = \rho_\mu(0)\rho_{H_2}(0), \quad (4)$$

where

$$\rho_\mu(0) = [1 + \delta_\mu \mathbf{P}(0)]/2 \quad (5)$$

is the density matrix of the polarized muon, and $\rho_{H_2}(0)$ is the density matrix of the hydrogen molecule H_2.

Let the target be an arbitrary mixture of para- and orthohydrogen with concentrations c_0 and $c_1 = 1 - c_0$. The states of the triplet are practically equiprobable, and therefore

$$\rho_{H_2}(0) = c_0\hat{1} + (1 - 4c_0)\hat{\mathbf{I}}^2/6, \quad (6)$$

where **I** is the total spin of the protons.

Substituting Eqs. (4)–(6) in the definition (2) of the polarization, we get

$$P_i(t) = S_{ik}(t)P_k(0), \quad (7)$$

where for the three 1/2 spins

$$S_{ik}(t) = {}^1/_{16}\,\mathrm{Sp}[\exp(i\hbar^{-1}H_s t)\sigma_{\mu_i}\exp(-i\hbar^{-1}H_s t)\sigma_{\mu_k}\rho_{H_2}(0)]. \quad (8)$$

It is convenient to calculate the tensor $S_{ik}(t)$ in the basis of the eigenfunctions $|\alpha\rangle$ of the spin Hamiltonian H_s:

$$S_{ik}(t) = {}^1/_{16}\sum_{\alpha,\alpha'}\langle\alpha|\sigma_{\mu_i}|\alpha'\rangle\langle\alpha'|\sigma_{\mu_k}\rho_{H_2}(0)|\alpha\rangle(i\omega_{\alpha\alpha'}t), \quad (9)$$

where $\hbar\omega'_{\alpha\alpha} = \varepsilon_\alpha - \varepsilon'_\alpha$, and ε_α are the eigenvalues of H_s.

It is known that in a strong magnetic field one need retain in the Hamiltonian (3), in first-order perturbation theory, only its secular part (see Refs. 11–13):

$$H' = -B\sum_i 2\mu_i s_{iz} + \sum_{i<k} a_{ik}(\mathbf{s}_i\mathbf{s}_k - 3s_{iz}s_{kz}), \quad (10)$$

where

$$a_{ik} = 2\mu_i\mu_k(3\cos^2\theta_{ik} - 1)r_{ik}^{-3}, \qquad \theta_{ik} = (\widehat{\mathbf{B}, \mathbf{n}_{ik}}).$$

The interactions that alter the spin projections on the external magnetic field are neglected here, since all the s_i, and hence the projections of the total spin, are good quantum numbers in a strong field. This is true if the external field is much stronger than the field produced by the protons at the muon, $B \gg B_0 = \mu_p/R^3$, where R is the molecule dimension. We choose for the estimates the mean value[14] $R = 0.85$ Å, and then $B_0 \approx 25$ G.

We introduce for the $(H_2\mu)^+$ ion a convenient set of spin functions, the product of the muon spin function $|\sigma_\mu\rangle$ by the singlet and triplet functions $|I, I_z\rangle$ of the protons, where I is the total spin of the protons. We use for the basis function the designations

$$|1\rangle=|+\rangle|1,\ +1\rangle,\quad |2\rangle=|+\rangle|1,\ 0\rangle,$$
$$|3\rangle=|+\rangle|1,\ -1\rangle,\quad |4\rangle=|+\rangle|0,\ 0\rangle. \tag{11}$$

Accordingly, the state vectors $|5\rangle - |8\rangle$ contain $|\sigma_\mu\rangle = |-\rangle$ in place of $|\sigma_\mu\rangle = |+\rangle$.

In our basis, the states with the maximum total spin projections $|1\rangle$ and $|7\rangle$ are eigenstates of the Hamiltonian (10) with energies

$$\varepsilon_{1,7}=[\mp(\omega_\mu+2\omega_p)-(a_{12}+a_{13}+a_{23})]/2, \tag{12}$$

where $\omega_{\mu,p} = 2\mu_{\mu,p}B$ (we put $\hbar = 1$). States having identical projections of the total spin of the system are intermixed. Therefore the states $|2\rangle$, $|4\rangle$, $|5\rangle$ and $|3\rangle$, $|6\rangle$, and $|8\rangle$, with respective total spin projections 1/2 and $-1/2$, are intermixed. Thus, the Hamiltonian matrix $\langle\alpha|H'|\beta\rangle$, where $|\alpha\rangle$ and $|\beta\rangle$ are the state vectors (11), breaks up into two diagonal elements and two 3×3 matrices. We write down the matrix for three states with total spin projection 1/2

$$\begin{pmatrix} -\omega_\mu/2 & -a_-/2 & a_+/2\sqrt{2} \\ -a_-/2 & -\omega_\mu/2 & -a_-/2\sqrt{2} \\ a_+/2\sqrt{2} & -a_-/2\sqrt{2} & (\omega_\mu+2\omega_p+a_+-a_{12})/2 \end{pmatrix}, \tag{13}$$

where $a_\pm = a_{13} \pm a_{23}$. In the matrix for the other triplet of bound states it is necessary to reverse the signs of ω_μ and ω_p.

In a strong external magnetic field we have $\omega_p \gg |a_{ik}|$, therefore the spectrum and the eigenfunctions of the matrix (13) can be obained by using the method of approximate diagonalization of a Hermitian matrix,[15,16] used in perturbation theory for close energy levels. In fact $|H'_{55} - H'_{22}|$, $|H'_{55} - H'_{44}| \sim \omega_\mu \gg |H'_{25}|$, $|H'_{45}|$, as against the difference $|H'_{22} - H'_{44}| = |a_{12}| \sim |H_{24}| = |a_-|/2$. We can therefore assume in first-order approximation [accurate to $(a_\pm/\omega_\mu)^2$] that only the states $|2\rangle$ and $|4\rangle$ are intermixed, while the state $|5\rangle$ and the matrix element H'_{55} are respectively an eigenstate and an energy level of the system. The matrix (13) can thus be approximately diagonalized with the aid of the unitary-transformation matrix

$$S(\vartheta)=\begin{pmatrix} \cos\vartheta & -\sin\vartheta & 0 \\ \sin\vartheta & \cos\vartheta & 0 \\ 0 & 0 & 1 \end{pmatrix}, \tag{14}$$

where $\tan 2\vartheta = 2H'_{24}/(H'_{22} - H'_{44}) = -a_-/a_{12}$.

Similar operations for states with total spin projection $-1/2$ yield the energy levels

$$\varepsilon_{2,4}=[-\omega_\mu+a_{12}\pm(a_{12}{}^2+a_-{}^2)^{1/2}]/2,$$
$$\varepsilon_{3,5}=[\mp(\omega_\mu-2\omega_p)+a_+-a_{12}]/2, \tag{15}$$
$$\varepsilon_{6,8}=[\omega_\mu+a_{12}\pm(a_{12}{}^2+a_-{}^2)^{1/2}]/2,$$

while the energy levels ε_1 and ε_7 are given by Eqs. (12).

The nonzero components of the tensor $S_{ik}(t)$ are

$$S_{zz}(t)=1,$$
$$S_{+-}(t)=S_{-+}^{*}(t)={}^1/_3\{2(1-c_0)\cos\omega_1 t+(1+2c_0)\omega_2{}^{-2}$$
$$\times(a_{12}{}^2+a_-{}^2\cos\omega_2 t)\}\exp(i\omega_\mu t), \tag{16}$$

where $\omega_1 = a_+$, $\omega_2 = (a_{12}^2 + a_-^2)^{1/2}$.

The off-diagonal components of the tensor $S_{+-}(t)$ are defined to make

$$P_+(t)=S_{+-}(t)P_+(0), \tag{17}$$

where $P_+ = P_x + iP_y$.

Equations (16) determine the behavior of the muon polarization in a field $(\mathrm{H}_2\mu)^+$ that is arbitrarily oriented relative to the external magnetic field. Obviously, the observed polarization of the muon ensemble is determined by a tensor averaged over all the ion orientations. For a frozen ion all the plane orientations are equally probable. Let us average the tensor component $S_{+-}(t)$ over all the equiprobable orientation of an equilateral triangle relative to the external magnetic field **B**.

Let θ be the angle between the field **B** (the z axis) and the normal to the plane of the ion, and φ the angle between the projection of **B** on the ion plane and the axis joining the protons; we have then

$$\omega_1(\theta,\ \varphi)=\gamma\omega_{pp}[3\sin^2\theta(1+{}^1/_2\cos 2\varphi)-2],$$
$$a_-=(3\sqrt{3}/2)\gamma\omega_{pp}\sin^2\theta\sin 2\varphi, \tag{18}$$
$$\omega_2{}^2(\theta,\ \varphi)=\omega_{pp}{}^2\{1+3\sin^2\theta[{}^3/_4(1+3\gamma^2)\sin^2\theta-1]$$
$$-3[{}^3/_4(3\gamma^2-1)\sin^2\theta\cos 2\varphi+1]\sin^2\theta\cos 2\varphi\},$$

where $\omega_{pp} = 2\mu_p^2R^{-3}$, $\gamma = \mu_\mu/\mu_p = 3.18$, and R is the distance between the nuclei (the side of the triangle). The correspondingly averaged tensor components are:

$$\langle S_{+-}(t)\rangle=\frac{1}{4\pi}\int_0^{2\pi} d\varphi\int_0^{\pi} d\theta\sin\theta S_{+-}(t)$$
$$=\frac{1}{3}\{2(1-c_0)S_1(t)+(1+2c_0)[S_0+S_2(t)]\}\exp(i\omega_\mu t), \tag{19}$$

where

$$S_0=1-\langle[a_-(\theta,\ \varphi)/\omega_2(\theta,\ \varphi)]^2\rangle, \tag{20}$$
$$S_1=\langle\cos\omega_1(\theta,\ \varphi)t\rangle, \tag{21}$$
$$S_2=\langle[a_-(\theta,\ \varphi)/\omega_2(\theta/\varphi)]^2\cos\omega_2(\theta,\ \varphi)t\rangle. \tag{22}$$

It is shown in Ref. 4 that the dephasing of the muon-polarization precession frequencies in a frozen field $(\mathrm{H}_2\mu)^+$ leads to depolarization with a characteristic rate $\Lambda\sim\gamma\omega_{pp}$. It is also shown there that the functions $S_1(t)$ and $S_2(t)$ decrease to $\lesssim 1\%$ of the initial value at $t\sim 10^{-5}$ s. Thus, the calculated depolarization rate is several times larger than the experimental one.

2. ROTATIONAL DIFFUSION OF THE $(\mathrm{H}_2\mu)^+$ ION

Thus, for the frozen $(\mathrm{H}_2\mu)^+$ ion the calculated relaxation rate is several times larger than the observed one. It is natural to associate the model refinement needed for quantitative agreement with experiment with some diffusion of the muon. Diffusion always decreases the relaxation rate. The potential well for the muon in the ion, however, is approximately 4.5 eV, the distance to the nearest equilibrium positions is ~4.0 Å, and it is easily seen that hopping diffusion of the muon is excluded in practice. This excludes all the more translational diffusion of the $(\mathrm{H}_2\mu)^+$ ion as a whole. In particular, this can be directly verified by the data on the translational self-diffusion in solid hydrogen and on diffusion of orthohydrogen in parahydrogen.[17–19] It is perfectly obvious that the diffusion rate of an ion, whose effective mass is very large because of the strong polaron effect, should be smaller by several orders.

Nonethless, it is precisely the diffusion mechanism that provides the necessary decrease of the relaxation rate for the $(H_2\mu)^+$ ion. The hcp lattice for the $(H_2\mu)^+$ triangle has three crystallographically equivalent orientations, separated by a potential barrier determined by the corrugation of the potential relief of the ion + lattice system. As noted above, the height of the barrier is ~ 10–100 K, depending on the position of the localized ion.

Consider the simplest variant of rotational diffusion of the $(H_2\mu)^+$ ion, when the equivalent equilibrium positions are specified by rotation of the triangle through an angle $\alpha \pm 2\pi/3$ about the C_3 axis.[1]

We introduce a coordinate frame xyz rigidly connected with the lattice and with $z \| C_3$. In our case the normal to the ion plane coincides then with the z axis and the ion orientation is determined by the angle between the axis joining the protons and the x axis. We denote the muon polarization for the orientations α, $\alpha + 2\pi/3$ and $\alpha + 4\pi/3$ by $P_0(t)$, $P_1(t)$, $P_2(t)$, respectively.

The magnetic moments have in an external field a preferred direction. The dipole-dipole interaction between the magnetic moments of the protons and of the muon depend therefore on the ion orientation in the magnetic field. The transition frequencies that determine the behavior of the muon polarization in different equilibrium are correspondingly dependent on the orientation. Let us consider the case of a strong magnetic field.

In each of the three equilibrium positions, the muon polarization is determined by the secular Hamiltonian (10). The polarization component parallel to the field is conserved: $P_\|(t) = P_\|(0)$. The precession of the perpendicular component is determined by one complex function $S_{+-}(t)$ [Eq. (16)] that depends on the ion orientation in the field **B**. The polarization of a muon jumping from one equilibrium position to another is, naturally, conserved: $P_{i+1}(t) = P_i(t)$. The longitudinal polarization component is thus preserved in the course of diffusion, just as for the frozen ion. The transverse component attenuates, since the orientation of the ion relative to the field changes during the jumps, and the time dependence of the function $S_{+-}(t)$ is determined by the equilibrium position of the $(H_2\mu)^+$ ion.

Thus, in each equilibrium position the complex transverse polarization $P_i(t) = (P_{xi} + iP_{yi})$ is determined by a corresponding function $S_i(t) \equiv S_{+-i}(t)$, where $i = 0, 1, 2$. We denote by $\lambda/2$ the probability of a transition from a position "i" to a neighboring position "k," so that the muon polarization $P_i(t)$ is given by the integral equation[2] (Ref. 20)

$$P_i(t) = e^{-\lambda t} S_i(t) P_i(0) + \frac{\lambda}{2} \int_0^t e^{-\lambda(t-\tau)} S_i(t-\tau) \sum_{k \neq i} P_k(\tau)\, d\tau. \tag{23}$$

The system of integral equations is reduced by a Laplace transformation to an inhomogeneous system of three algebraic equations. All the equilibrium positions for the muon are equally probable, therefore $P_0(0) = P_1(0) = P_2(0) = P(0)/3$, where $P(0)$ is the total muon polarization at the initial instant of time. The solution of the system of equations for the Laplace transform of the total polarization $P(p) = \Sigma P_i(p)$ is

$$P(p) = \frac{1}{3} \frac{S_0+S_1+S_2+\lambda(S_0S_1+S_0S_2+S_1S_2)+3(\lambda/2)^2 S_0S_1S_2}{1-(\lambda/2)^2(S_0S_1+S_0S_2+S_1S_2)-2(\lambda/2)^3 S_0S_1S_2} P(0), \tag{24}$$

where the argument of the functions $S_i \equiv S_i(p+\lambda)$ is left out for brevity.

The time dependence of the polarization $P(t)$ is determined by the inverse transform

$$P(t) = \frac{1}{2\pi i} \int_{-i\infty-0}^{i\infty-0} P(p)\, e^{pt}\, dp. \tag{25}$$

We put $p + \lambda = x - i\omega_\mu$, and then the Laplace transform of the function takes the form

$$S_i(x) = f_i(x)/\Pi_i(x), \tag{26}$$

where

$$f_i(x) = x^4 + {}^1/_3[(1+2c_0)\omega_{1i}^2 + a_{12i}^2 + 2\omega_{2i}^2 - 2c_0 a_{-i}^2] \times x^2 + {}^1/_3(1+2c_0)\, a_{12i}^2 \omega_{1i}^2, \tag{27}$$

$$\Pi_i(x) = x(x^2+\omega_{1i}^2)(x^2+\omega_{2i}^2). \tag{28}$$

Substituting (26) in (24), we get

$$P(x) = [f_0\Pi_1\Pi_2 + f_1\Pi_0\Pi_2 + f_2\Pi_0\Pi_1 + \lambda(f_0f_1\Pi_2 + f_0f_2\Pi_1 + f_1f_2\Pi_0) + {}^3/_4\lambda^2 f_0f_1f_2] \times [\Pi_0\Pi_1\Pi_2 - (\lambda/2)^2(f_0f_1\Pi_2 + f_0f_2\Pi_1 + f_1f_2\Pi_0) - 2(\lambda/2)^3 f_0f_1f_2]^{-1} \cdot {}^1/_3 P(0). \tag{29}$$

The denominator of (29) contains a polynomial of order 15. The residues in the integral (25) must accordingly be taken at 15 points. The main contribution to the integral of (25), however, is made by poles with small real parts that determine the low depolarization velocity. It is known that poles with small real parts can exist if $\lambda \gg \omega_{1,2}$. We separate from the argument of the functions (27) and (28) the large real part λ: $x = y + \lambda$, $y = p - i\omega_\mu$. We have $|y| \ll \lambda$ for the sought solution.

It follows from the structure of the denominator of (19) that there exists a single small root proportional to $1/\lambda$. Retaining in the denominator of (29) only terms linear in y and leading in λ, and equating the denominator to zero, we obtain the desired pole with small real part:

$$y_1 = \Lambda = -(\Delta_0 - \delta_0 + \Delta_1 - \delta_1 + \Delta_2 - \delta_2)/3\lambda, \tag{30}$$

where

$$\delta_i = {}^1/_3(1+2c_0)\omega_{1i}^2 + a_{12i}^2 + {}^2/_3(1-c_0) a_{-i}^2, \tag{31}$$

$$\Delta_i = \omega_{1i}^2 + \omega_{2i}^2. \tag{32}$$

The remaining 14 poles have large real parts $\sim\lambda$ and the corresponding residues are of the order of Λ/λ. We obtain as a result, accurate to terms $\sim 1/\lambda$, a simple time dependence of the muon transverse polarization:

$$P_\perp(t) = P_\perp(0) \exp(i\omega_\mu - \Lambda)t. \tag{33}$$

Note that the relaxation rate depends on the orientation of the C_3 axis relative to the external field.

Let the direction of the field **B** be specified in the *xyz* frame by the angles θ and φ. The components of the unit vectors joining the nuclei along the field direction are then

$$n_{13}={}^1/_2\sin\theta[3^{1/2}\cos(\varphi-\alpha)\pm\sin(\varphi-\alpha)]. \tag{34}$$

We express the functions (31) and (32) in terms of unit vectors:

$$\Delta_i-\delta_i=9\gamma^2\omega_{pp}^2\{(n_{13i}^2+n_{23i}^2)-{}^4/_3(1+c_0)n_{13i}^2n_{23i}^2 -{}^8/_9(1-c_0)(n_{13i}^2+n_{23i}^2-{}^1/_3)\}. \tag{35}$$

We substitute (34) in (35) and sum over the three equilibrium positions. The summation over three-equiprobable values of the angle α is equivalent to averaging over this angle. Sinc the argument everywhere in (34) is only $(\varphi-\alpha)$, we find that in the presence of axial symmetry the result depends only on the angle θ:

$$\Lambda(\theta)=3\gamma^2\omega_{pp}^2\{[(7-4c_0)/8]\sin^4\theta-8(1-c_0)\times(3\sin^2\theta-1)/27\}\lambda^{-1}. \tag{36}$$

An experiment on a single crystal can determine the orientation of the $(H_2\mu)^+$ ion in the crystal lattice. Production of large hydrogen single crystals suitable for μSR investigations is rather complicated task. All the experiments performed to date were made on polycrystals. To obtain an equation that describes the behavior of the muon polarization in polycrystals, Eq. (33) must be averaged over all the orientations of the C_3 axis in the magnetic field **B**. We recognize here that $\Lambda\ll\omega_\mu$ and replace approximately averaging of the exponential by averaging of its argument:

$$\langle P_\perp(t)\rangle\approx\exp[(i\omega_\mu-\langle\Lambda\rangle)t]P_\perp(0), \tag{37}$$

$$\langle\Lambda\rangle=(23+4c_0)\gamma^2\omega_{pp}^2/45\lambda. \tag{38}$$

It can be seen from (38) that the depolarization rate $\langle\Lambda\rangle$ depends little on the ortho/para ratio in the hydrogen: in pure parahydrogen it is approximately 10% higher than in pure orthohydrogen. In the reduction of experimental results for a mixture of para- and orthohydrogen, account must also be taken of the known dipole-dipole relaxation mechanism.[21]

CONCLUSION

The results obtained within the framework of the assumed model permit interpretation of the entire experimental material presently available on the behavior of μ^+ polarization in solid and liquid hydrogen. The hypothetical hopping rotational diffusion of the frozen $(H_2\mu)^+$ ion explains the observed depolarization in pure parahydrogen. The depolarization is due to para and ortho transitions in the $(H_2\mu)^+$ ion and should therefore vanish even in strongly slowed-down rotation of the ion, for in this case the para and ortho states differ in energy by more than the dipole-dipole interaction energy.

A final check on our hypothesis that a frozen $(H_2\mu)^+$ ion is formed can be obtained by studying the muon polarization in the absence of an external magnetic field and in longitudinal fields of the order of 20–200 G. Indeed, as already noted [see Eq. (16)], the longitudinal polarization is conserved, within the secular approximation, in a strong external field. The characteristic scale of the internal field at the muon in the ion is approximately 25 G, so that the depolarization is practically zero in longitudinal fields $\gtrsim 200$ G.

Note that rotational diffusion of the ion does not lead to any qualitative changes of the behavior of the longitudinal polarization. Quantitative diffusion-induced changes in the secular part of the Hamiltonian do not influence the conservation of the longitudinal polarization. Quantitative diffusion-induced changes in the secular part of the Hamiltonian do not influence the conservation of the longitudinal polarization component in strong magnetic fields. In exactly the same manner, rotational diffusion does not influence the behavior of the muon polarization in the absence of an external magnetic field. Oscillations of the muon polarization, with frequencies on the order of 10^6 s^{-1}, should be observed. As shown in Ref. 22, six oscillation frequencies will be observed in the general case, and three in parahydrogen.

Note that, strictly speaking, the question of the influence of a very small admixture of a MuH fraction (or of a free muon) remains at present experimentally unanswered. Over long times $t\gtrsim 10^{-5}$ s, the polarization would asymptotically reach a small constant value.

The experimentally observed abrupt decrease of the depolarization on going into the liquid phase is interpreted as the result of Brownian rotation of the ice sphere produced around the ion.[4,21] The increase of the depolarization rate with decrease of temperature is apparently due to a decreased rate of rotational diffusion. The observed tendency of the depolarization rate to decrease at $T\lesssim 4$ K can be connected with quantum effects in rotational diffusion of the $(H_2\mu)^+$ ion. Answers to these questions can be provided by more accurate experiments.

[1] Of importance for the subsequent solution of the problem is only that the diffusion has the same velocity along the three positions that replace one another upon rotation.

[2] Equation (23), which is valid for the case of "incoherent" diffusion $P_i(t+\tau)=S_i(t+\tau)P_i(\tau)$, can be used for our problem, since the interactions that determine the functions S_i in various equilibrium positions of the ion are not related to one another.

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