Theory of muon spin relaxation in condensed phases of hydrogen isotopes

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A theory of muon spin relaxation in a $(H_2\mu)^+$ $[(D_2\mu)^+]$ molecular ion forming in a crystalline phase of a hydrogen isotope has been devised. It has been shown that the $(H_2\mu)^+$ molecular ion formed does not manage to thermalize during the lifetime of the muon, but it does manage to undergo transitions to lower energy levels of the vibration–rotation spectrum during a time $\tau \ll \tau_\mu$. The spin relaxation of μ^+ is determined by the interaction of the electric dipole moment of the ion with the lattice and the spin–rotation interaction in the ion. In this sense the relaxation mechanism resembles the "muonium" mechanism, and the role of the hyperfine interaction is performed by V_{LS}. The conclusions of the study completely explain the existing experimental data, primarily the absence of a pronounced isotope effect. The qualitative results for calculating the thermalization rate of the $(H_2\mu)^+$ ion are not sensitive to the choice of the model of the interaction of the ion with lattice molecules. © *1994 American Institute of Physics.*

1. INTRODUCTION

The study of cryosystems by the muon spin relaxation (μSR) method was launched comparatively recently by the work of a group from the Leningrad Institute of Nuclear Physics and the I. V. Kurchatov Institute of Atomic Energy.[1,2] The spin relaxation of μ^+ in liquid and crystalline phases of hydrogen was investigated. Fairly curious and unexpected results were obtained already in the first experiments: a) only a diamagnetic component of the polarization was observed, apparently attesting to the complete absence of an interaction of μ^+ with the track; b) the relaxation rates in solid ortho- and para-hydrogen differed only slightly and greatly exceeded the values obtained under the hypothesis that μ^+ is stopped in interstices of the crystal lattice; c) spin relaxation was practically absent in the liquid phase.

These results found reasonable theoretical explanations under the assumption that a positive muon forms an $(H_2\mu)^+$ molecular ion in condensed phases of hydrogen.[3] The $(H_2\mu)^+$ ion is known to have the form of an equilateral triangle with a side $R_0 \approx 0.85$ Å. According to the theory in Ref. 3, muon spin relaxation is seen to be the result of "dephasing" of the precession frequencies in a randomly oriented "frozen" $(H_2\mu)^+$ ion in a crystal. The theory explained the spin relaxation mechanism in para-hydrogen, but the calculated relaxation rate greatly exceeded the experimentally observed value. Further experiments showed that the relaxation rate is strongly dependent on the temperature: it increased with decreasing temperature and exhibited a tendency to peak at $T \sim 4$–5 K.[4–6] To explain these results it was postulated in Refs. 7 and 8 that the $(H_2\mu)^+$ ion undergoes rotational diffusion by a hopping mechanism in a crystal. Then the relaxation rate should be weakly dependent on the composition of the mixture of ortho- and para-hydrogen and should have the form $\Lambda = a\omega^2_{\mu_p}/\lambda$, where $\lambda(T)$ is the rotational diffusion rate, $\omega_{\mu_p} = 2\mu_p\mu_\mu/R_0^3 \approx 2\times 10^6\ \mathrm{s}^{-1}$, μ_p and μ_μ are, respectively, the magnetic moments of a proton (or a deuteron in D_2), and the constant $a \sim 1$. As we see, the relaxation rate should depend strongly on both the temperature and the isotopic composition of the target.

A series of new experiments was subsequently carried out. In Ref. 9 it was shown that $\sim 15\%$ of the muons form an Mu atom, which rapidly ($\Lambda \sim 10^9\ \mathrm{s}^{-1}$) relaxes in ortho-hydrogen. Experiments with H_2, D_2, and HD targets were performed in Refs. 10 and 11. It was found that the behavior of the polarization precessing at the muon frequency is nearly identical in all the targets and that the relaxation rates differ only slightly. For example, at $T_1 \sim 4$ K, $\Lambda_{H_2} \approx \Lambda_{HD} \approx 1.2 \times 10^5\ \mathrm{s}^{-1}$, and $\Lambda_{D_2} \approx 0.95 \times 10^5\ \mathrm{s}^{-1}$ (H_2 and D_2 are statistical mixtures). The relaxation rate of the longitudinal component decreased as the applied magnetic field was increased. The formation of the $(H_2\mu)^+$ ion in condensed H_2, HD, and D_2 phases is probably not questionable, but the theory in Refs. 7 and 8, which predicts a strong isotope effect, does not adequately describe the behavior of the $(H_2\mu)^+$ ion in crystalline hydrogen. As was noted in Ref. 12, the latter experiments indicate that in a cryocrystal the $(H_2\mu)^+$ ion does not manage to thermalize during the lifetime of the muon $\tau_\mu = 2.2\times 10^{-6}$ s. The more recent experimental data[10,11] unequivocally indicate that muon spin relaxation in crystalline hydrogen cannot be attributed to a direct magnetic dipole-dipole interaction.

In the present work we calculated the thermalization rate of the $(H_2\mu)^+$ ion in crystalline hydrogen and devised a theory for the spin relaxation of μ^+ in an unthermalized ion. The preliminary results of the calculation were presented in Ref. 13.

2. CALCULATION OF THE THERMALIZATION RATE OF THE $(H_2\mu)^+$ ION IN A CRYSTALLINE PHASE OF HYDROGEN

1. It is well known that in "ordinary" crystals the transfer of excitation energy $\varepsilon \sim 1$ eV is generally mediated by multiphonon processes and has characteristic times $\tau(T) \sim 10^{-10}$–10^{-11} s. In molecular crystals (all cryocrystals are such) the times may be significantly longer, primarily due to the weak binding of the molecules in the crystal. When an $(H_2\mu)^+$ molecular ion forms in the ground state, a considerable amount of energy $\varepsilon \sim 4.5$ eV should be released (see, for example, Ref. 14). Therefore, the $(H_2\mu)^+$ ion clearly exists in the initial time period in a highly excited vibration–rotation state. In the ensuing time period energy is transferred to the crystal upon transitions to lower vibration–rotation energy levels. Of course, it is not possible to quantitatively consider a cascade, but, as we shall see below, the qualitative picture is fairly clear.

To describe a cascade, it is necessary to know the spectrum of vibration–rotation energy levels of the ion. This spectrum was calculated for the free ion in Refs. 13 and 15. The radiation lifetimes of the excited states of the $(H_2\mu)^+$ ion were also calculated. It was found that the radiation lifetimes are one to two orders of magnitude greater than the lifetime of the muon τ_μ.

The $(H_2\mu)^+$ ion has a dipole moment in the excited state.[13,15] The transfer of excitation energy from the ion to the crystal is mediated mainly by the interaction of the electric dipole moment of the ion with polarized molecules of the crystal.[13] The positively charged ion strongly deforms the first coordination sphere; therefore, deformation of the lattice can be realistically related to the excitation of phonons only from the second coordination sphere. The deformation of the first coordination sphere can easily be evaluated on the basis of the following arguments. As we know (see, for example, Ref. 16), the interaction of the molecules in a hydrogen lattice is adequately described by the Lennard–Jones potential

$$V_{\mathrm{LJ}}(R) = 4\varepsilon[(\sigma/R)^{12} - (\sigma/R)^6], \tag{1}$$

where $\varepsilon \simeq 5.06 \times 10^{-15}$ erg and $\sigma \simeq 2.96$ Å. Accordingly, the interaction of an ion with the molecules in the first coordination sphere may be described as

$$V_1 = \sum_a [-\alpha e^2/2R_a^4 + V_{\mathrm{LJ}}(R_a)] + \sum_{a \neq b} V_{\mathrm{LJ}}(R_{ab}). \tag{2}$$

Here the first sum describes the interaction of the ion with the molecules, the second sum describes the interaction of the molecules in the first coordination sphere, and $\alpha \simeq 5.4 a_{\mathrm{B}}^3$ is the polarizability of an H_2 molecule. In this evaluation we assume that the Lennard-Jones parameters for the interaction V_{LJ} of an ion with the molecules are the same as those for the interaction between molecules.

The following theoretical situations are possible for an $(H_2\mu)^+$ ion in a crystal: a tetrapod (four molecules in the first coordination sphere), an octapod ($N=6$), and a lattice site ($N=12$). Minimizing the interaction energy (2), we obtain the distance to the nearest neighbors R_1 and the binding energy $U = -N\varepsilon_0$. This procedure is elementary, if the second sum in Eq. (2) is neglected. For a tetrahedron and octahedron we obtain $R_1 \approx 2.5$ Å and $\varepsilon_0 \approx 0.17$ eV, and for a lattice site we obtain $R_1 \approx 2.8$ Å and $\varepsilon_0 \approx 0.11$ eV. To be specific, we assume that the $(H_2\mu)^+$ ion formed is located at a lattice site.

Despite the elementary character of the model, the evaluations are in good agreement with the exact quantum-chemical calculations of the $H_3^+(H_2)_n$ cluster;[17–19] therefore, the proposed model may be adopted for the interaction of the ion with its nearest neighbors. The constant of the hcp hydrogen lattice $R_{lH_2} \approx 3.8$ Å, and, as we see, the deformation of the first coordination sphere (the cluster) amounts to $\sim 25\%$. The deformation of the second coordination sphere is almost an order of magnitude smaller.

Thus, the Hamiltonian of the ion–cluster–crystal system has the form

$$\hat{H} = \hat{H}_{\mathrm{i}} + \hat{H}_{\mathrm{c}} + \hat{H}_{\mathrm{l}} + \hat{V}_{\mathrm{ic}} + \hat{V}_{\mathrm{cl}}, \tag{3}$$

where $\hat{H}_{\mathrm{i}}$, $\hat{H}_{\mathrm{c}}$, and $\hat{H}_{\mathrm{l}}$ are, respectively, the Hamiltonians of the ion, the cluster, and the lattice and $\hat{V}_{\mathrm{ic}}$ and $\hat{V}_{\mathrm{cl}}$ are the operators of the interactions of the dipole moment of the ion with the cluster and of the cluster with the lattice.

2. The Hamiltonian $\hat{H}_{\mathrm{i}}$, its vibration–rotation spectrum, and its states were considered in detail in Ref. 15, and we shall utilize those results.

The Hamiltonian $\hat{H}_{\mathrm{c}}$ specifies the vibration–rotation states of the molecules in the cluster and the vibrations of the molecules in the cluster. The ion at the center of the cluster strongly polarizes the molecules, inducing a dipole moment directed along the radius of the first coordination sphere $\mathbf{d} = \alpha e \mathbf{R}/R^3$. This interaction naturally alters the rotational spectrum of the molecules. However, as we shall see below, this does not have a significant influence on the rate of energy transfer and has absolutely no influence on the qualitative results. Therefore, we shall assume that the vibration–rotation spectrum and the states of the free molecules and H_2 molecules in the cluster are identical.

In performing the next calculations it will be sufficient for us to take into account only the radial vibrations of the molecules in the cluster. The remaining vibrations have a far smaller frequency. In fact, the transverse vibrations of the molecules in the cluster are determined by relatively weak interaction (1), and the frequencies of the vibrations of the cluster as a whole are small, since $M_{\mathrm{cl}} \gg M_{\mathrm{p}}$. The vibrational frequency can easily be evaluated on the basis of the interaction energy (2). We obtain

$$\omega_{\mathrm{c}}^2 = 8\alpha e^2/(M_{\mathrm{p}} R_1^6). \tag{4}$$

For a lattice site we have $\hbar\omega_{\mathrm{c}} \approx 3 \times 10^{-2}$ eV.

The Hamiltonian $\hat{H}_{\mathrm{l}}$ describes the lattice vibrations.

The form of the interaction operator $\hat{V}_{\mathrm{ic}}$ is obtained by expanding the energy of the molecules comprising the cluster in the field of the ion $V = -\alpha \mathbf{E}^2/2$ in a series in powers of the radial displacement of the molecules u_a and the dipole moment of the ion. Retaining only the terms which are linear with respect to u_a and quadratic with respect to $\mathbf{d}$, we obtain

$$V_{\mathrm{ic}} = \sum_a \frac{\alpha}{R_1^6} \left\{ 10e(\mathbf{d}\mathbf{n}_a) + \frac{3}{R_1}[7\mathbf{d}^2 - 9(\mathbf{d}\mathbf{n}_a)^2] \right\} u_a. \tag{5}$$

The summation is carried out over all the molecules in the cluster. The terms of order $\mathbf{d}^2$ in operator (5) are taken into account, if the linear terms do not make a contribution to the transition under consideration.

The operator V_{cl} is isolated when the interaction energy V_{LJ}, as defined in Eq. (1), is expanded in powers of the lattice deformation and the radial displacements of the molecules in the cluster. Retaining only the linear terms, we obtain

$$V_{\rm cl}=A\sum_{a,b}(\mathbf{u}_{ab}\mathbf{n}_a)u_a, \tag{6}$$

where the summation is carried out over all the molecules in the cluster and the nearest neighbors to each of these molecules in the second coordination sphere, $\mathbf{u}_{ab}$ denotes the displacement of the lattice molecules (the deformation), and $A=(24\varepsilon/R_1)(\sigma/\mathrm{R}_1)^6(26(\sigma/\mathrm{R}_1)^6-7)$. For the parameters of a hydrogen lattice we obtain $A\approx-0.2\times10^{18}$ erg/cm^2.

3. Each transition between the states of the ion is associated with a change in the state of the entire "ion–cluster–lattice" system. The states of the ion, along with the quantum numbers of the vibration–rotation spectrum, are also determined by the total spin of the protons and the projection of the muon spin of interest to us. Clearly, the interaction of the vibrations and rotations of the ion is significant for large values of the orbital angular momentum L, so that it is difficult to specify "good" quantum numbers. However, as we shall show below, the most significant role is played by the states with $L\leqslant2$, for which separation of the vibrational and rotational states is correct.[15] Therefore, we define the states of the ion as

$$|\Psi_{\rm ion}\rangle=\sum_k a_k|N;L,M,k\rangle|I,M_I\rangle|m_s\rangle, \tag{7}$$

where N is the set of three vibrational numbers, M and k are the projections of L onto the laboratory and local coordinate systems, respectively, I is the total spin of the protons, and m_s is the projection of the muon spin. As is well known (see, for example, Ref. 20) only states which are antisymmetric with respect to the replacement $k\rightarrow-k$ are possible for a proton spin $I=1$ (the ortho-ion), and only states which are symmetric with respect to this replacement are possible for $I=0$ (the para-ion). The energy of the ion is determined only by the quantum numbers N and k.

We define the states of the cluster in the following manner:

$$|\Psi_{\rm c}\rangle=|v_{\rm c}\rangle\prod_a|v_a;K_a,M\rangle. \tag{8}$$

Here $v_{\rm c}$ is the quantum number of the radial vibrations; v_a and K_a are, respectively, the vibrational quantum number and the orbital angular momentum of molecule a in the cluster. We assume that in the initial state $v_{\rm c}=0$ and $v=0$ and that $K=0$ for para-hydrogen and $K=1$ for ortho-hydrogen.

The state of the lattice is specified by the corresponding number of phonons $|n_{\rm ph}\rangle$.

The probability of a transition between different states of the ion is determined in second-order perturbation theory for the continuous spectrum by Fermi's "golden rule" (see, for example, [20]):

$$dw_{if}=\frac{2\pi}{\hbar}\left|\sum_n\frac{\langle f|V_{\rm cl}|n\rangle\langle n|V_{\rm ic}|i\rangle}{E_i-E_n}\right|^2\delta(E_i-E_f)d\nu_f. \tag{9}$$

Here in the intermediate state $|n\rangle$ the state of the ion changes, and vibrations of the cluster are excited (in addition, rotations and vibrations of the molecules in the cluster may be excited). In the final state $|f\rangle$ a phonon with a definite energy is excited (and the vibrational state of the cluster may change). Clearly, interactions (5) and (6) do not alter the spin states of the ion and the molecules, but they determine the mechanism for transferring the excitation energy of the vibration–rotation states of the ion to the crystal.

Let ε_i and ε_f be the energies of the initial and final states of the ion. The excitation energy of an individual molecule in the cluster is

$$\Delta\varepsilon_{\rm M}=B_{\rm r}2(2K+3)+\hbar\omega_{\rm M}v, \tag{10}$$

where the rotational constant for the hydrogen molecule $B_{\rm r}\approx7.35\times10^{-3}$ eV and $\hbar\omega_{\rm M}\approx0.52$ eV. Also, $K=1$ for ortho-hydrogen, and $K=0$ for para-hydrogen. If vibrations are excited, $v=1$; otherwise, $v=0$.

The interaction results in excitation of a phonon with an energy

$$\Delta_0=\varepsilon_i-\varepsilon_f-\Delta\varepsilon_{\rm M}-\hbar\omega_{\rm c}v_{\rm c}, \tag{11}$$

where $v_{\rm c}=2$ or 0, if vibrations of the cluster are excited in the final state or not, respectively. It is easily seen that the energetic denominator in Eq. (9) then equals

$$\Delta=\Delta_0-\hbar\omega_{\rm c}(v_{\rm c}-1). \tag{12}$$

As the results of the calculation of the vibration–rotation spectrum of the $(\mathrm{H}_2\mu)^+$ ion show, the energies Δ_0 and Δ do not vanish for any two given states. However, for nearly every excited state there is a possibility of a transition for which the energy transferred to the lattice vibrations Δ_0 is small compared with the difference $\varepsilon_i-\varepsilon_f(\Delta_0\sim10^{-2}$ eV).[13,15] These transitions will clearly be most probable, and disposal of the energy will occur specifically along these channels. Since a reliable calculation of the vibration–rotation spectrum and determination of the states are possible only for the lower portion of the spectrum ($\varepsilon_f\leqslant1$ eV),[13,15] we shall henceforth restrict ourselves to only the most probable transitions for the first excited vibrational energy levels of the $(\mathrm{H}_2\mu)^+$ ion. Clearly, the transition rates in the upper part of the spectrum will clearly be of at least the same order of magnitude.

4. The transformation of Eq. (9) is carried on in the appendix. The total probability of a transition between two vibration–rotation states of the ion per unit time is given by Eq. (13):

$$W_{if}=\frac{N_2}{n}D_0\frac{\Delta_0v'_{\rm c}}{\Delta^2}\frac{|\langle v_iK_i\|\alpha\|v_fK_f\rangle|^2}{\alpha^2}\times\sum_{M_i,M_f}\frac{|\langle N_i,L_iM_ik_i|\mathbf{d}|N_f,L_fM_fk_f\rangle|^2}{(2L_i+1)d_1^2}. \tag{13}$$

Here

$$D_0=A^2C_1^2\left\langle\frac{1}{c^3}\right\rangle\frac{2\hbar e^2a_1^2}{\pi(4M_{\rm p})^4\omega_{\rm c}^2\omega_1}, \tag{14}$$

$C_1=10\alpha e/R_1^6$; $2M_{\rm p}$ is the mass of the H_2 molecule; $(\langle c^3\rangle)^{1/3}\approx2.2\times10^5$ cm/s is the mean velocity of sound in a hydrogen crystal; $\langle v_i,K_i\|\alpha\|v_f,K_f\rangle$ is the reduced matrix element of the polarizability of the H_2 molecule; $v'_{\rm c}$ = 1,2 for

TABLE I. Transitions with variation of the vibrational quantum number N.

ε_i, eV	ε_f, eV	$\vert N_i L_i k_i\rangle$	$\vert N_f L_f k_f\rangle$	Transition	v'	$\vert K-K'\vert$	Δ_0, eV	W, $10^6 s^{-1}$
0.398	0.164	$\vert 1;-;2,2\rangle$	$\vert 0;-;3,2\rangle$	o–o	0	2	0.11	14
0.349	0.055	$\vert 1;-;1,1\rangle$	$\vert 0;-;2,1\rangle$	o–o	0	2	0.17	19
0.634	0.029	$\vert 1;+;3,(0,2)\rangle$	$\vert 2;+;2,(0,2)\rangle$	p–p	1	2	0.05	1.8
0.367	0.327	$\vert 1;+;2,(0,2)\rangle$	$\vert 0;+;3,(0,2)\rangle$	p–p	0	2	0.04	$1.4\cdot 10^4$
0.422	0.131	$\vert 1;+;3,(1,3)\rangle$	$\vert 0;+;2,1\rangle$	p–p	0	2	0.19	0.85
0.378	0	$\vert 1;+;1.0\rangle$	$\vert 0;0.0\rangle$	p–p	0	2	0.27	45
0.578	0.174	$\vert 2;-;2,2\rangle$	$\vert 0;-;2,1\rangle$	o–o	0	2	0.40	$2.3\cdot 10^{-3}$
0.549	0.233	$\vert 2;+;2,(0,2)\rangle$	$\vert 0;+;2,1\rangle$	p–p	0	2	0.31	$7.2\cdot 10^{-3}$
0.651	0.029	$\vert 2;+;2,1\rangle$	$\vert 0;+;2,(0,2)\rangle$	p–p	1	2	0.06	0.14
0.681	0.395	$\vert 3;-;1,1\rangle$	$\vert 1;-;2,1\rangle$	o–o	0	2	0.16	19
0.706	0.471	$\vert 3;+;1,1\rangle$	$\vert 1;+;2,1\rangle$	p–p	0	2	0.13	23
0.831	0.054	$\vert 4;-;1,1\rangle$	$\vert 0;-;2.1\rangle$	o–o	1	2	0.14	1.8
0.856	0.131	$\vert 4;+;1,1\rangle$	$\vert 0;+;2,1\rangle$	p–p	1	2	0.11	2.1

$v_c=0,2$, respectively; $\langle N_i;L_i,M_i,k_i|\mathbf{d}|N_f;L_f,M_f,k_f\rangle$ is the matrix element of the dipole moment operator between the initial and final states of the $(H_2\mu)^+$ ion; N_2 is the coordination number for the interaction of the molecules in the first and second coordination spheres. Clearly, the transition between different vibration–rotation energy levels is specified by the first term in (5).

The matrix elements for the polarizability of the H_2 molecule were calculated in Ref. 21. For transitions without a change in the vibrational quantum number $\langle 0,K\|\alpha\|0,K+2\rangle\approx\alpha=(\alpha_\|+\alpha_\perp)/3$. When vibrations of a molecule are excited, the matrix elements decrease by almost an order of magnitude: $\langle 0,0\|\alpha_\|\|1,0\rangle\approx 1.15a_B^3$, $\langle 0,0\|\alpha_\perp\|1,0\rangle\approx 0.54a_B^3$, and $\langle 0,0\|\alpha\|1,0\rangle\approx 0.74a_B^3$.

Plugging in the values of the parameters and the constants, we obtain the numerical estimate $D_0\approx 7.2\times 10^{16}$ erg/cm$^3\cdot$s.

Table I presents the results of the numerical calculations of the most probable transitions with variation of the vibrational quantum numbers. Here we took into account only excited states for which it makes sense to separate the vibrational and rotational states.[13,15] The coordination number N_2 for an ion at a lattice site equals 36. As we see, the transition rates are of the order of 10^7 s^{-1} for symmetric vibrational states and 10^3 s^{-1} for an antisymmetric vibration. The transition rate can increase only as a result of an accidental resonance, as occurs for the para-transition $|1;+;2,(0,2)\rangle\rightarrow|0;+;3,(0,2)\rangle$. Transitions to the ground $|0,0,0\rangle$ state are possible only from the $|N;+;1,0\rangle$ states, but only the transition from the level with $N=1$ has an appreciable probability. Thus, we see that during a cascade with changes in the vibrational quantum numbers, the ion undergoes transitions to excited rotational states. The transitions from the $|2;L,k\rangle$ levels are hindered by two factors: 1) the smallness of the dipole moment, 2) the "unfortunate" location of the level in the spectrum of excitations of the crystal. The results in Table I graphically illustrate that the thermalization time of the $(H_2\mu)^+$ ion in a crystal is comparable to the lifetime of a muon $\tau_\mu=2.2\times 10^{-6}$ s.

Let us now consider transitions without variation of the vibrational quantum numbers. We again take into account that it makes sense to consider only the values $L\leqslant 3$. In this case vibrations of the molecules are clearly not excited. The numerical results for $N=0$ are presented in Table II. Since

TABLE II. Transitions without variation of the vibrational quantum number.

ε_i, eV	ε_f, eV	$\vert L_i,k_i\rangle$	$\vert L_f,k_f\rangle$	Transition	$\vert K-K'\vert$	Δ_0, MeV	W, $10^9 s^{-1}$
0.299	0.131	$\vert +;3,(1,3)_+\rangle$	$\vert +;2,1\rangle$	p–p	2	66	1.2
0.082	-	$\vert +;3,(1,3)_-\rangle$	-	-	-	-	forbidden
0.164	0.058	$\vert -;3.2\rangle$	$\vert -;2,2\rangle$	o–o	2	51	6.5
0.164	0.055	$\vert -;3,(1,3)_+\rangle$	$\vert -;2,1\rangle$	o–o	2	53	8.3
0.057	0.055	$\vert -;3,(1,3)_-\rangle$	$\vert -;2,1\rangle$	o–o	0	2	0.08
0.294	0.131	$\vert +;3,(0,2)_+\rangle$	$\vert +;2,(0,2)_-\rangle$	p–p	2	61	0.31
0.086	0.027	$\vert +;3,(0,2)_-\rangle$	$\vert +;2,(0,2)_+\rangle$	p–p	2	21	0.17
0.058	-	$\vert -;2,2\rangle$	-	-	-	-	forbidden
0.055	0.009	$\vert -;2,1\rangle$	$\vert -;1,1\rangle$	o–o	0	46	4.2
0.145	0.038	$\vert +;2,(0,2)_-\rangle$	$\vert +;1.0\rangle$	p–p	2	5	0.67
0.027	-	$\vert +;2,(0,2)_+\rangle$	-	-	-	-	forbidden
0.013	0.039	$\vert +;2.1\rangle$	$\vert +;1,1\rangle$	p–p	2	54	8.4
0.01	-	$\vert -;1,1\rangle$	-	-	-	-	forbidden
0.036	0	$\vert +;1.0\rangle$	$\vert 0.0\rangle$	p–p	0	36	$2.8\cdot 10^2$
0.037	-	$\vert +;1,1\rangle$	-	-	-	-	forbidden

the rotational energy levels are weakly dependent on N, the results presented are valid for any $N \leqslant 4$.

Comparing the results presented in Tables I and II, we see that all the allowed transitions without a change in N have rates which are two or three orders of magnitude higher than the transitions with a change in N. Thus, during a time $\tau \leqslant 10^{-9}$ s, the ion undergoes transitions to vibration-rotation states with small values for the orbital angular momentum L. Next, from states corresponding to symmetric vibrations the ion makes transitions to rotational states with $N=0$, and from antisymmetric vibrational states it makes transitions to rotational states with $N=2$.

Thus, as is seen from the data in the tables, as a result of a cascade the ion must reach one of the long-lived excited rotational states

$$\begin{aligned} &|+;3,M,(1,3)_-\rangle=0{,}21|+;3,M,1\rangle-0{,}98|+;3,M,3\rangle \\ &|+;2,M,(0,2)_+\rangle=0{,}51|+;2,M,0\rangle+0{,}86|+;2,M,2\rangle \\ &|+;1,M,1\rangle \quad \text{para-ion,} \end{aligned} \tag{15}$$

$$|-;2,M,2\rangle, |-;1,M,1\rangle \quad \text{ortho-ion.} \tag{16}$$

The only exception is the channel leading to an $|+;1,M,0\rangle$ rotational state of the para-ion, from which the ion can rapidly thermalize. However, as is seen from Table I, this channel has only a small probability due to its small statistical weight.

Thus, it may be assumed that the main conclusion regarding the impossibility of thermalization of the $(H_2\mu)^+$ ion during the lifetime of the muon does not depend on the assumptions which were employed in the numerical calculations.

3. MECHANISM OF MUON SPIN RELAXATION IN AN UNTHERMALIZED $(H_2\mu)^+$ ION

1. Let us now consider the $(H_2\mu)^+$ ion in states with a nonzero value of L. In these states the following hyperfine interactions of the magnetic moment of the muon are nonzero: the dipole-dipole interactions with the magnetic momenta of the nuclei (V_{dd}) and with the magnetic field of the orbital motion of the charges (spin–rotation, V_{LS}). Of course, $V_{LS}=0$ for both a thermalized ion and a "frozen" ion; therefore, this interaction has not hitherto been taken into account. Attention was turned to the importance of V_{LS} in Ref. 22 during an analysis of the behavior of the spin polarization of μ^+ in noble gases. Since the $(H_2\mu)^+$ ion is an asymmetric top, in the general case the spin–rotation interaction has the form (see, for example, Ref. 20)

$$V_{LS}=\hbar A_1 L_\xi s_\xi+\hbar A_2 L_\eta s_\eta+\hbar A_3 L_\zeta s_\zeta, \tag{17}$$

where L_α and s_α are, respectively, the projections of the orbital angular momentum of the ion and of the spin of μ^+ in the local coordinate system $\xi\eta\zeta$, which is defined in the appendix.

The constants A_1, A_2, and A_3 are determined by the interaction of the magnetic moment of the muon with the magnetic field created at the muon by the orbital motion of all the charges of the ion, as is usually done for molecules (see, for example, Ref. 20). Clearly, this interaction is significantly stronger for a charged system than for neutral systems. As usual, the constants A_1, B_2, and A_3 in spin Hamiltonian (17) are obtained after averaging of the interaction over the orbital state of the system. Averaging the magnetic field with respect to the ground orbital state of the electrons and the rotational state of the nuclei in the ion, we obtain the following numerical estimates for the parameters of V_{LS}:

$$A_1=6.4A_0; \quad A_2=0.6A_0; \quad A_3=-0.3A_0, \tag{18}$$

where $A_0=4\mu_N\mu_{0\mu}/\hbar R_0^3 \approx 1.4\times 10^6 \ \text{s}^{-1}$. As we see $|A_1| \gg |A_{2,3}|$. The value of V_{LS} is nonzero for both ortho- and para-ions, while $V_{dd}\neq 0$ only for an ortho-ion with $L\neq 0$.

Since the projection of the muon spin is determined experimentally in the laboratory coordinate system, it is convenient to rewrite Eq. (17) in the form

$$V_{LS}=\hbar(A_1 L_\xi \mathbf{n}_\xi+A_2 L_\eta \mathbf{n}_\eta+A_3 L_\zeta \mathbf{n}_\zeta)\mathbf{s}. \tag{19}$$

The expression in parentheses in Eq. (19) is similar to expression (A3) for the dipole moment operator, but is slightly more complicated due to the fact that the projection of the field onto the η axis appears in (19).

In long-lived states (15) and (16) with definite L and k, interaction (19) may be represented by the effective Hamiltonian

$$\hat{V}_{LS}^{\text{eff}}=\hbar(A_x(Lk)\hat{s}_x\hat{L}_x+A_y(Lk)\hat{s}_y\hat{L}_y+A_z(Lk)\hat{s}_z\hat{L}_z), \tag{20}$$

where the constants $A_i(Lk)$ are obtained as a result of the averaging of (19) over states (15) and (16). For example, for the $|-;1,M,1\rangle$ states $A_x=A_y=A_z=A_{11}^-=(A_3-A_2)/2$, and for the $|+;1,M,1\rangle$ states $A_x=A_y=A_z=A_{11}^+=(A_3+A_1)/2$. As we see, for the states with $L=1$ the effective Hamiltonian is isotropic, but the constants for the ortho- and para-ions have different values. For the states with $L=2$ we have $A_x=A_z\neq A_y$, and the effective Hamiltonian is anisotropic.

It can be shown that only V_{LS}(20) should henceforth be taken into account. In fact, in states (16) the magnitude of the dipole-dipole interaction decreases significantly as a result of the averaging over the rotational states, and it turns out that for the ortho-ion $V_{dd}^{\text{eff}} < V_{LS}^{\text{eff}}$. The operator V_{dd} couples the spin of the muon $\hat{\mathbf{s}}$, the total spin of the protons $\hat{\mathbf{I}}$, and the orbital angular momentum $\hat{\mathbf{L}}$. The interaction V_{dd} is effectively averaged over the equally probable states with different projections of the total spin of the protons I, and then it may be neglected in the total spin Hamiltonian.

2. As was shown in the first section, second-order perturbation theory gives definite long-lived excited states of the $(H_2\mu)^+$ ion, in which the new interaction V_{LS} now turns out to be significant. Consideration of this interaction leads to the appearance of muon spin relaxation as a result of an interaction with the lattice in third-order perturbation theory. We note that the spin relaxation effect is simply absent in lower orders. In complete analogy to the calculation of the thermalization rate, we use Fermi's "golden rule" in third-order perturbation theory for the continuous spectrum (see, for example, Ref. 20):

$$dw_{if}=\frac{2\pi}{\hbar}\left|\sum_{n,n'}\frac{\langle f|V_{cl}|n'\rangle\langle n'|V_{ic}|n\rangle\langle n|V_{LS}|i\rangle}{(E_i-E_n)(E_i-E_{n'})}\right|^2$$

$$\times \delta(E_i - E_f) d\nu_f, \tag{21}$$

where the vibration–rotation state of the ion is now fixed and is specified by one of Eqs. (15) or (16). The largest probability is assigned to the processes under which vibrations of the cluster are virtually excited, but other states are not excited. In the final state only the spin state of the muon is different, and the energy surplus (or deficit) is transferred to phonons: $\Delta_0 \sim A_{Lk}$. Since $A_{Lk} \ll T$ over the entire permissible temperature range, it may be supposed that the phonons have a Bose distribution function $n_{\rm ph}(\Delta_0) \approx {\rm T}/\Delta_0 \gg 1$, which results in a linear temperature dependence. Accordingly, a contribution to the mean value with respect to the vibration–rotation state of the ion is made only by the second term in $V_{\rm ic}(5)$, which is quadratic with respect to the dipole moment of the ion $\mathbf{d}$.

Inasmuch as $V_{\rm LS}(20)$ couples the orbital angular moment of the ion and the muon spin and maintains their total momentum $\mathbf{J} = \mathbf{L} + \mathbf{s}$, the muon spin can change its projection only if there is a simultaneous change in the projection of the orbital angular momentum M. This greatly complicates the formal writing of the expressions. However, since all the states with different M are equally probable, the muon spin "flip" rate $\nu = w_{if}$ can be expressed by a fairly general formula. After some very cumbersome mathematical operations, for the states of an ion with a definite value of L we obtain

$$\nu_L = \frac{N_2}{n} \Gamma T \sum_M \left(\frac{A_{Lk}}{\Delta_{\rm M}} \right)^2 \frac{(L-M)(L+M+1)}{2L+1}. \tag{22}$$

Here

$$\Gamma = \frac{80 A^2 C_2^2 \hbar^2 (e a_{\rm B} R_0)^4}{3\pi (4\hbar\omega_{\rm c})^4 M_{\rm p}^3} \left\langle \frac{1}{c^3} \right\rangle, \tag{23}$$

where $C_2 = 3\alpha/R_1^7$ and $\Delta_{\rm M} = \varepsilon_i - \varepsilon_f$ is the difference between the energy levels of the ion in the initial and final states. The mathematical operations omitted here are similar to the operations performed in the appendix during the derivation of Eq. (13).

Of course, the muon spin "flip" rates are dependent on the rotational state of the ion and on the strength of the external magnetic field. If $V_{\rm LS}$ is large compared with the energy of the magnetic moment of the muon in the external field $\hbar A_{Lk} \gg 2\mu_{0\mu} B$, it may be assumed that $\Delta_{\rm M} \approx \hbar A_{Lk}$ and that the constants ν_L do not depend on the field B and are identical for the ortho- and para-ions. Also, as can easily be seen, for example, $\nu_2/\nu_1 = 3$. In a strong field $\nu_L \propto B^{-2}$, and the spin "flip" rates for the para- and ortho-ions have different values. Plugging in the values of the parameters, we obtain an estimate for the flip rate ν_1 in a weak field

$$\nu_1 \simeq 1.5 \times 10^6 T \ {\rm s}^{-1},$$

where T is measured in degrees Kelvin. Accordingly, at $T > 5$ K we have $\nu_1 \sim 10^7 \ {\rm s}^{-1}$. Thus, in the temperature range $T \gg 1$ K we obtain $\nu \sim A_{Lk}$, and determination of the spin relaxation rate of a muon in hydrogen requires solving the relaxation equation for the spin density matrix of the $(H_2\mu)^+$ subsystem.

3. Under the conditions considered the relaxation processes are described well by the Wangsness–Bloch equation for a spin density matrix (see, for example, Refs. 12, 23, and 24):

$$\frac{\partial \rho}{\partial t} + \frac{i}{\hbar} [\hat{H}_{0\rm s}, \rho] = \frac{1}{2} \nu (\sigma_m \rho \sigma_m - 3\rho), \tag{24}$$

where ρ is the spin density matrix of the $(H_2\mu)^+$ ion, ν is the muon spin "flip" rate (frequency), σ_m denotes the muon spin operators, and $\hat{H}_{0\rm s}$ is the spin Hamiltonian of the ion. Clearly, $\hat{H}_{0\rm s}$ depends both on the spin variables and on the orbital state of the ion. We need to consider only the long-lived excited states. Since system of equations (24) for the components of the density matrix is very cumbersome, we take into account only the principal interactions, discarding the interactions which do not directly "affect" the muon spin. Then in the states with $L = 1$ we have

$$\hat{H}_{0\rm s} = -\hbar\omega \hat{s}_z + \hbar A_{Lk} (\mathbf{sL}), \tag{25}$$

where $\hbar\omega = 2\mu_{0\mu} B$. Accordingly, in Eq. (24) the flip rate ν_L is specified by Eq. (22). In states with $L \geq 2$ the anisotropic interaction should be written down.

Thus, we obtain an equation which is similar in structure to the equation for the spin density matrix of muonium (see, for example, Ref. 12). The role of the hyperfine interaction is played by the spin–rotation interaction, and the electron spin operators are replaced by the orbital angular momentum operator of the ion. In our system $L \geq 1$, but even for $L = 1$ we now have a system of $2^3 = 8$ coupled equations for different components of the density matrix. Of course, these systems of equations cannot be solved analytically; nevertheless, the qualitative picture of muon spin relaxation should be similar to that observed in an Mu atom for the longitudinal (parallel to $\mathbf{B}$) and transverse (perpendicular to $\mathbf{B}$) components of the polarization.

To be specific, we consider the states with $L = 1: |\pm; 1, M, 1\rangle$. Just as for Mu, we analyze the regions of large $(\nu_1 \gg A_{11})$ and small $(\nu_1 \ll A_{11})$ "flip" rates.

At large ν, the transverse polarization $P_\perp(t)$ precesses with the muon frequency and slowly decays with a rate $\Lambda_\perp \propto \nu^{-1}$. At small ν, $P_\perp(t)$ precesses with the muonium frequencies and decays slowly with a rate $\Lambda_\perp \propto \nu$. In both cases the precession frequency is fast compared with the decay rate.

In our case for $L = 1$ the roles of the triplet and the singlet in the Mu atom are played by the quartet $(J = 3/2)$ and the doublet $(J = 1/2)$ in the ion. Accordingly, at small ν precession should be observed with frequencies corresponding to transitions between states with different J and M_J. Since states of $(H_2\mu)^+$ ions with different L are possible for an ensemble of muons, the presence of different frequencies should be manifested as effective spin relaxation with a rate $\Lambda \sim A_{Lk}$.

To find the relaxation rates of the transverse polarization we isolate the large frequencies in the corresponding components of the density matrix. Averaging the system obtained with respect to the period of fast oscillations, we can reduce the system of eight equations to a simple system of four equations, whose characteristic equation has the form

$$[(\nu-\lambda)^2+4\Omega^2][(\nu-\lambda)^2+\Omega^2]-\nu^2(\nu-\lambda)^2=0, \quad (26)$$

where $\Omega=A_{11}^2/\sqrt{2}\omega$ when $\omega \gg A_{11}$ and $\Omega=\sqrt{2}\omega$ when $\omega \ll A_{11}$. As is easily seen, when $\nu \geqslant 3\Omega$ (at large ν) there is a single solution, which decreases with increasing ν:

$$\Lambda_\perp=5\Omega^2/2\nu. \quad (27)$$

When $\nu<3\Omega$, the character of the solution changes, and we obtain the same real part $\mathrm{Re}\,\lambda=-\nu$ (at small ν) for all four roots. Just as in the case of Mu, the spin relaxation rate should have a maximum. In our case it peaks when $\nu \sim 3\Omega$. When $T \geqslant 1$ K, we have the case of large ν, and, accordingly, $\Lambda_\perp \propto T^{-1}$. For a strong field $\Delta_M \approx \hbar\omega$ [see Eq. (27)], and we find that in this case $\Lambda_\perp$ is virtually independent of the field $\mathbf{B}$.

At large ν the longitudinal polarization $P_\parallel(t)$ decays with a rate $\Lambda_\parallel \propto \nu^{-1}$, and at small ν it oscillates with the hyperfine splitting frequencies and decays with a rate $\Lambda_\parallel \propto \nu$. Thus, at small ν effective spin relaxation should again be observed owing to the multifrequency oscillations of the polarization in the ensemble of muons. Carrying out the procedure of isolating the large oscillation frequencies precisely as for the components determining $P_\perp(t)$, we obtain a system of three coupled equations for the corresponding components of the density matrix. For the system obtained we find the small root corresponding to slow decay:

$$\Lambda_\parallel=\frac{\nu\Omega^2}{2\Omega^2+\nu^2}. \quad (28)$$

We again see that as the field increases, $\Lambda_\parallel$ decreases, and at $\omega \gg A$, we find that $\Lambda_\parallel \propto \omega^{-2}$.

Unfortunately, the most interesting region of values of the parameters is not subject to analytical description. However, the results qualitatively correspond to the observed picture. In fact, a relatively broad maximum is observed in the relaxation rate of $P_\perp(t)$ at $T \sim 4$ K. According to the numerical evaluations, $A_{Lk} \sim 10^6$ s^{-1}. In a field $B \sim 100$ G the precession frequency $\omega \sim 0.8 \times 10^7$ s^{-1}, and we have the case of a strong field, in which $\nu \propto \omega^{-2}$ [see Eq. (22)]. From Eq. (27) we find that the relaxation rate of the transverse component of the polarization $\Lambda_\perp \propto nA_{Lk}^2/N_2\Gamma T$ does not depend on the field and is determined by the interaction of the ion with the crystal. At $T \sim 4$ K we obtain the estimate $\Lambda_\perp \sim 2 \times 10^5$ s^{-1}.

For the longitudinal polarization we obtain $\Lambda_\parallel \approx \nu/2 \propto B^{-2}$ from Eq. (28). Since the spin-rotation interaction constants correspond to a field $B \sim 10$–30 G, the longitudinal component scarcely relaxes in a field $B \sim 100$ G.

Of course, the evaluations performed can claim only qualitative correspondence, but they provide the correct order of magnitude for the decay rate and are in complete agreement with the experimental results in Refs. 10 and 11. It should, however, be recalled that in ortho-hydrogen the dipole-dipole interaction of the magnetic moments of a muon and the nuclei of the molecules in the crystal makes an additional contribution to the relaxation rate.

4. The model considered is supported, first or all, by the weak isotopic dependence of the relaxation rate.[10,11] In fact, the principal constant A_1 in V_{LS} has nearly identical values in the $(H_2\mu)^+$, $(D_2\mu)^+$, and $(HD\mu)^+$ ions, since it is determined by the orbital motion of the muon relative to the ξ axis. The isotope effect is manifested in the flip rate ν through both the density of the crystal n and the dimensional factor Γ. Since the frequencies of the radial vibrations of the molecules in a cluster $\omega_c \propto M^{-1/2}$ (where M is the mass of the isotope), $\Gamma \propto M^{-1}$. However, the constant $C_2 \propto (\sigma/R_1)^{12}$ also varies. The polarizability of the molecules and the parameters σ are nearly identical for H_2 and D_2 (Ref. 16). However, for H_2 $R_{lH_2} \simeq 3.79$ Å, and for D_2 $R_{lD_2} \simeq 3.6$ Å; therefore, $C_2(H_2)/C_2(H_2) \approx (R_{lH_2}/R_{lD_2})^{12} \approx 1.85$. Since $n_{H_2}/n_{D_2} \approx 1.15$, we find that under the same conditions $\nu_{D_2}/\nu_{H_2} \approx 1.1$. For the relaxation rate we have $\Lambda_{D_2}/\Lambda_{H_2} \approx \nu_{H_2}/\nu_{D_2} \approx 0.9$, which is in complete agreement with experiment.[10,11]

4. CONCLUSIONS

In conclusion we stress the main results of this work.

1. The results of the experiments in Ref. 10 and 11 have been attributed to spin relaxation of the unthermalized $(H_2\mu)^+$ ion in a crystalline phase of hydrogen isotopes. The spin relaxation is determined mainly by the spin-orbit coupling of the magnetic moment of the muon in the unthermalized ion.

2. The $(H_2\mu)^+$ molecular ion formed does not manage to thermalize during the lifetime of the muon, but it does manage to undergo transitions to lower energy levels of the vibration–rotation spectrum during a time $\tau \ll \tau_\mu$.

3. The spin–rotation interaction in the ion scarcely depends on the isotopic composition of the target, but it is nonzero only for an unthermalized ion.

4. The spin relaxation of μ^+ is determined by the interaction of the electric dipole moment of the ion with the lattice and the spin–rotation interaction in the ion. In this sense the relaxation mechanism resembles the "muonium" mechanism, and the role of the hyperfine interaction is performed by V_{LS}. A rigorous analytical calculation of the relaxation rate is not possible, but the qualitative conclusions completely account for the existing experimental data.

5. The qualitative results for calculating the thermalization rate of the $(H_2\mu)^+$ ion are not sensitive to the choice of the model of the interaction of the ion with lattice molecules.

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APPENDIX

To transform Eq. (9) we assume that a linear isotropic spectrum is valid for the phonons (acoustic phonons are excited). Since $\Delta_0 \gg T$, we always have $n_{ph}(\Delta_0) \approx 1$, and there is no temperature dependence in this stage. Performing the integration over the states, the summation over the phonon polarizations, and the summation over the orientations of the unit vectors $\mathbf{n}_a$, we obtain

$$w_{if}(N_2/n)A^2C_1^2\langle c^{-3}\rangle$$
$$\times(\Delta 0 v c\prime\,|\langle vi,Ki\|\alpha\|vf,Kf\rangle|2/\pi(4M\mathrm{p})3\omega\mathrm{c}2\nu 2\Delta 2\alpha 2)|$$

$$\times\langle N_i,L_i,M_i,k_i|\mathbf{d}|N_f,L_f,M_f,k_f\rangle|^2. \tag{A1}$$

The dipole moment operator of $(H_2\mu)^+$ has nonzero projections only in the plane of the ion. We direct the ζ axis of the local coordinate system along the second-order axis and the ξ axis in the plane of the ion, and we place the origin of coordinates at the center of mass of the ion. The nonzero components of the dipole moment operator, expressed in terms of the normal coordinates q_1, q_2, and q_3, then have the form[15]

$$d_\xi=ea_2q_2;\quad d_\zeta=e(a_0R_0+a_1q_1+a_3q_3), \tag{A2}$$

where $a_0=0.243$, $a_1=-0.471$, $a_2=0.019$, and $a_3=1574$. The corresponding vibrations have the frequencies $\hbar\omega_1=0.375$ eV, $\hbar\omega_2=0.648$ eV, and $\hbar\omega_1=0.972$ eV. The normal coordinates q_1 and q_3 correspond to symmetric vibrations of the ion, and q_2 corresponds to antisymmetric vibrations. We direct the unit vectors $\mathbf{n}_\xi$ and $\mathbf{n}_\zeta$ along the corresponding axis of the local coordinate system, and we then have

$$\mathbf{d}=d_\xi\mathbf{n}_\xi+d_\zeta\mathbf{n}_\zeta=\begin{pmatrix} \frac{1}{2}(D^1_{11}+D^1_{-1-1}-D^1_{1-1}-D^1_{-11})d_\xi+\frac{1}{\sqrt{2}}(D^1_{0-1}-D^1_{01})d_\zeta \\ -\frac{i}{2}(D^1_{11}-D^1_{-1-1}+D^1_{1-1}-D^1_{-11})d_\xi+\frac{i}{\sqrt{2}}(D^1_{0-1}+D^1_{01})d_\zeta \\ -\frac{1}{\sqrt{2}}(D^1_{10}-D^1_{-10})d_\xi+D^1_{00}d_\zeta \end{pmatrix}. \tag{A3}$$

Here the $D^1_{\mu m}$ are Wigner functions. Thus, the calculation of the dipole moment matrix elements reduces to the known formula (see, for example, Ref. 20):

$$\langle N_i,L_i,k_i|D^1_{\mu m}d_\alpha|N_f,L_f,k_f\rangle$$
$$=(-1)^{M_i+k_i}\sqrt{(2L_f+1)/(2L_i+1)}\,C^{L_iM_i}_{1m,L_fM_f}C^{L_ik_i}_{1\mu,L_fk_f}$$
$$\times\langle N_i,L_i\|d_\alpha\|N_f,L_f\rangle, \tag{A4}$$

where the $C^{LM}_{1m,L'M'}$ are the Clebsch–Gordan coefficients. As we see, either transitions which alter the vibrational quantum number corresponding to the particular normal coordinate by unity or do not alter the vibrational state of the ion are possible.

The first excited vibrational level corresponds to the normal coordinate q_1.

Now it would be convenient for us to measure all the matrix elements in (A4) in units of the matrix element[15]

$$d_1=\langle 0,0\|d_\zeta\|1,0\rangle=a_1(\hbar/2M_p\omega_1)^{1/2}. \tag{A5}$$

Performing the summation over all the equally probable projections of the momentum in the initial state M_i, we obtain Eq. (13).

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