Muon relaxation in hydrogen isotopes HD and D_2

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(Submitted 17 May 1991)
Pis'ma Zh. Eksp. Teor. Fiz. **53**, No. 12, 577–580 (25 June 1991)

Experiments on muon relaxation in hydrogen isotopes in longitudinal and transverse magnetic fields have shown that there is a significant difference in the behavior of the muon and the muonium components. The relaxation rate of the muonium component increases with the nuclear magnetic moment, whereas the depolarization of the diamagnetic component (μ^+) is virtually independent of the isotopic composition.

Several years ago damping (relaxation) was observed in the precession of the muon spin in solid parahydrogen and the temperature dependence of the relaxation rate $\Lambda(T)$ was studied.[1,2] Later these results were confirmed by the workers at TRIUMF.[3] In addition, muonium (the union of a muon and an electron) was observed in hydrogen, and comprised 10–15% of the diamagnetic component. The mechanism of muon relaxation in hydrogen was related to the formation of a $H_2\mu^+$ complex,[4] the result of which is a dipole interaction between the muon and the proton in it. The low relaxation rate and its temperature dependences are accounted for by thermally activated random rotations of the complex.[5] However, in the precession spectrum no structure characteristic of a three-spin system was observed; rather only a single relatively narrow peak was observed. If it is assumed that the $H_2\mu^+$ complex is localized in the lattice, difficulties arise in explaining the weak dependence of the relaxation rate on the concentration of orthomolecules at low temperatures.[6] The difference in the values of Λ of normal hydrogen and parahydrogen, calculated by the method of van Vleck, turns out to be four times higher than that measured at low temperatures.

Molecular hydrogen has a series of isotopes (HD, D_2, etc.) with various magnetic moments. The magnetic moment of the proton is $\mu_p = 2.793\,\mu_n$ $(S = 1/2)$ and that of the deuteron is $\mu_d = 0.857\ \mu_n\ (S = 1)$, three times smaller. This circumstance opens up the possibility of studying dipole-dipole interactions of the muon within the $H_2\mu^+$ complex (if it is formed), as well as with the surrounding magnetic moments of the hydrogen molecules.

The I. V. Kurchatov Institute of Atomic Energy and the University of British Columbia have carried out cooperative muon spin relaxation (μSR) investigations of hydrogen isotopes HD and D_2 in the TRIUMF surface muon channel. The details of the experimental method are given in Refs. 8 and 9. Here we mention only that the initial deuterium had a purity of 99.98% and the hydrogen deuteride contained 2% impurities. The gases were cooled in a special copper cell with a flow of helium vapor in an intermediate-temperature cryostat made by the firm Oxford Instruments. The pressure of the cooling helium vapor was not higher than 1 torr.

Muon transverse relaxation was studied in the liquid and solid phases of D_2 and HD. The magnetic field was 50 Oe. An analysis of the precession spectra showed that the polarization function can be approximated better by an exponential $e^{-\Lambda t}$ than by a Gaussian $\exp(-\sigma^2 t^2)$. A typical example for the relaxation at 5 K gives $\chi^2 = 1.37$ for the exponential and $\chi^2 = 2.5$ for the Gaussian. A similar dynamical situation has been noted previously.[1-3] Figure 1 shows the temperature dependence of the transverse relaxation rate $\Lambda(T)$ for D_2 and HD. In the liquid the relaxation rate is low, not exceeding 0.01 μs^{-1}. When the liquid is solidified, the value of Λ makes a large jump, which is shown on an expanded scale in Fig. 1 for deuterium. A similar, but somewhat larger jump, is observed in normal hydrogen.[2] As the temperature is lowered, the relaxation rate increases, and below 8–10 K it becomes essentially constant. The dashed lines show the values of Λ of normal hydrogen—the upper one[2-3]—and for parahydrogen—the lower one.[1] The most important feature of these curves is that *at low temperatures the relaxation rate for the hydrogen isotopes are the same within* 10%.

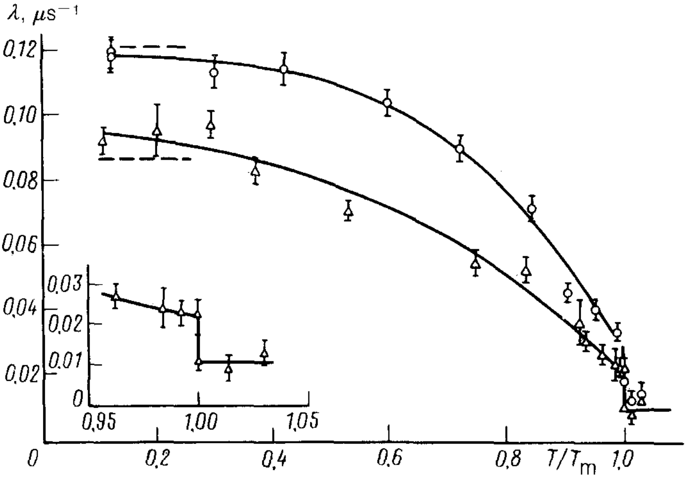

FIG. 1. Relaxation rate in HD (○) and D_2 (▽) as functions of the reduced temperature (relative to the melting temperature).

The configuration of a muon cluster is governed by the Coulomb interaction, and, presumably, this configuration should be the same for $H_2\mu^+$, $HD\mu^+$, and $D_2\mu^+$; i.e., we can assume that the configuration is triangular in all the cases, with a distance between the muon and the nuclei of $R = 0.85$ Å. Within this theory[4] it is easy to estimate the muon relaxation rate in hydrogen and in deuterium. Since $\sigma \simeq \mu_p/R^3$, the precession should be three times as fast for a stationary $H_2\mu^+$ ion than it is for a stationary $D_2\mu^+$ ion. In the case of a rotating complex the relaxation rate decreases in proportion to the expectation value of the rotation time $(\Lambda_\perp = \sigma^2\tau)$, which, in turn increases with the mass of the complex: $\tau \simeq \sqrt{m/g}$. Assuming that the ion–molecule interaction parameter g depends only weakly on the isotopic composition of the complex, we can expect that the relaxation rate in hydrogen and deuterium will differ from each other as $\Lambda_H/\Lambda_D \simeq 9\sqrt{m_H/m_D}$, i.e., by about a factor of 6. However, in the experiments all the isotopes studied had nearly the same value of Λ. It seems unlikely that there should be a change in the static part of the magnetic interaction (σ^2) that exactly equalizes the change in the hopping frequency $(\omega = \tau^{-1})$. Nonetheless, if the complexes do rotate, then at $T \simeq 4$ K the time is $t \simeq 10^{-8}$s for HD and for D_2 this time is an order of magnitude greater.

The suppression of the relaxation in a longitudinal magnetic field for not too fast muon kinetics is described by the expression[10]

$$\Lambda(H) = \frac{\Lambda_0}{1 + (\gamma H\tau)^2}, \tag{1}$$

where $\Lambda_0 = 2\sigma^2\tau$ is the relaxation rate in the absence of the external field. For the HD complex we should expect a decrease in the relaxation by a factor of two in a longitudinal field of $\simeq 1$ kOe, and for the deuteron in a field of $\simeq 100$ Oe. The functions $P(t)$ were measured for D_2 and HD in various longitudinal fields at 7 K. Figure 2 shows the dependence of the relaxation rate on the longitudinal field strength in the approximation of $P(t)$ by a single exponential. It is easy to see that the relaxation is suppressed with increasing field, and in a field above 30 Oe the asymmetry is nearly constant. In zero field the relaxation rate is about 1.4 times greater than in a transverse field. It is well known that $\Lambda_0/\Lambda_\perp \geqslant 2$ for the usual mechanism of dipole–dipole relaxation.[10]

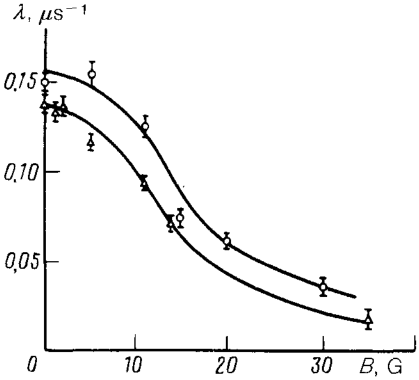

FIG. 2. Depolarization rate as a function of the longitudinal magnetic field.

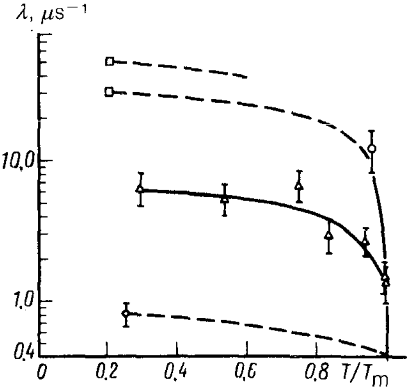

FIG. 3. Muonium relaxation in hydrogen isotopes. From top to bottom: normal H_2; HD; normal D_2, and para-H_2.

Nevertheless, we assume that the depolarization rate decreases in a longitudinal field as in the case of a diffusing muon. We can then estimate from (1) the dispersion of the magnetic field and the correlation parameter τ. For deuterium approximation (1) gives $\sigma = \sqrt{\langle H^2 \rangle} \simeq 3.5$ Oe and $\tau \simeq$ μs. For HD, as seen from Fig. 2, these parameters are nearly the same. We recall that in the $H_2\mu^+$ complex the protons create a local field of about 20 Oe at the muon.

At present, it is difficult to give an adequate explanation of the small relaxation rate or, more importantly, its weak dependence on the isotopic composition. In any case, this result brings into doubt the hypothesis of the rotating $H_2\mu^+$ complex.[4,5] The low rate of muon relaxation may be a consequence of the slowness of the formation of the bound state of the muon. A two-step process, in which the muon is not depolarized in the free state is possible; instead, the depolarization occurs after the formation of the ion, as a result of the dipole coupling with the nuclear moments, as discussed in Ref. 4. Another possible mechanism is that in which the relaxation takes place through the interaction of the muon with the orbital angular momentum of the ion in an excited state.

In the isotopes we have investigated, just as in hydrogen,[3] precession was observed at the frequency of muonium, with an asymmetry of $\simeq 15\%$ of the diamagnetic component. The temperature dependence of the attenuation rate of the precession at the frequency of muonium in a field of $\simeq 5$ G was studied. In liquid D_2 and HD the attenuation of the muonium precession is small and amounts to $\lambda_{\mathrm{Mu}} \simeq 0.2\ \mu s^{-1}$. In the crystalline phase the transverse relaxation rate increased rapidly as the temperature is lowered, and the precession in HD (and in normal H_2) becomes virtually unobservable. Figure 3 shows the results of measurements of the depolarization rate of muonium in hydrogen isotopes. The horizontal axis is normalized to the melting temperature. The squares denote the expected values of the relaxation, calculated from the magnitudes of the internal local fields.[3] Although the picture of the relaxation of muonium requires refinement, nonetheless, two important conclusions follow from the results shown in Fig. 3. First, the relaxation rate of the *Mu* spin (in contrast with that of the μ^+) is proportional to the nuclear spin of the hydrogen isotope, and second, in parahydrogen the relaxation occurs because of the presence of a small amount of orthomolecules (estimated at less than 0.5%). The latter factor may be particularly important in the study of the diffusion of muonium.

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Translated by J. R. Anderson