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Weird Muonium Diffusion in Solid Xenon

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Abstract

Muon and muonium spin rotation and relaxation parameters were studied in liquid and solid xenon. The small diamagnetic fraction ($\sim 10\%$) observed in condensed xenon is believed to be $Xe\mu^+$. The muonium hyperfine frequency was measured for the first time in liquid Xe and was found to be in agreement with the vacuum value. A nonmonotonic temperature dependence of the muonium relaxation rate probably indicates that muonium diffusion in solid Xe is of quantum nature.

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The rare gases Ar, Kr and Xe at low temperatures form relatively simple liquids and solids, as the principal binding forces between atoms are the weak van der Waals forces. In both liquid and solid the local order is determined by the close packing of identical spheres. In the solid phase these substances have stable *fcc* configurations.

It is known that large fractions of muons form muonium atoms ($Mu \equiv \mu^+e^-$) in rare gas solids.[1] The Mu hyperfine frequency was measured in solid Kr and solid Ar and found to be in good agreement with the vacuum value. This allows study of muonium dynamics in the simplest solids. Recent measurements of muonium diffusion in alkali halides,[2,3] compound semiconductor[4] and solid nitrogen[5] have revealed unambigious evidence for the quantum nature of muonium kinetics. In comparing experimental results with the well developed theory,[6] Xe and Kr are very convenient, as one can change the nuclear hyperfine interactions of muonium by changing the isotopic content of the samples; this allows one to adjust the Mu relaxation rate into the "μ^+SR window" at all temperatures. Measurement of the nuclear hyperfine parameters of Mu atom in rare gas solids is also of intrinsic interest. Unfortunately, we were unable to determine the muonium hyperfine constant in solid xenon, so we cannot be sure of the muonium state in the solid phase.

Finally, in Ref. [1] it was found that small fractions of muons form diamagnetic states in condensed Ar, Kr and Xe. It is known that protons form bound states with Kr and Xe atoms.[7,8] Recent μLCR measurements in solid nitrogen[9] unambigiously showed that the diamagnetic fraction in N_2 corresponds to the stable $N_2\mu^+$ ion. It is essential to know the nature of the muon's diamagnetic compound in rare gas solids in order to correctly interpret the μ^+SR results.

This article presents the first detailed measurements of the temperature dependence of muon and muonium relaxation rates in liquid and solid xenon.

The experiment was performed at the JINR phasotron in Dubna. Spin-polarized muons were stopped in a sample placed in a magnetic field perpendicular to the initial polarization of the beam. The fractional inhomogeneity and time instability of the magnetic field were no worse than 10^{-3}. The registration of experimental spectra, sample preparation and cryogenic techniques using a He-flow cryostat are described in Ref. [10]. The Xe sample was cylindrical, 80 mm in diameter and 24 mm thick, with the cylinder axis along the muon beam. The fraction of muons stopping in the cryostat walls, scintillators, *etc.* (as opposed to those stopped in the sample) was estimated to be 4%. The sample temperature was measured with an accuracy better than ± 0.1 K by means of a semiconductor thermometer frozen into the sample.

Gaseous Xe with close to the natural isotopic abundances was used in this experiment. The fractions of magnetic isotopes were: ^{129}Xe: 20.0% (natural abundance 26.4%) and ^{131}Xe: 24.4% (natural abundance 21.2%). The concentrations of non-Xe impurities did not exceed 15 ppm (O_2: < 5 ppm; Ar and Kr: < 10 ppm).

In order to determine the fraction of muons thermalizing in a diamagnetic state, several time spectra were measured in liquid and solid Xe in a transverse magnetic field of 100 Oe and fitted to

$$N(t) = N_0 \exp(-t/\tau_\mu) \{1 + A \exp(-\sigma^2 t^2) \cos(\omega_\mu t + \phi)\} + B. \qquad (1)$$

The diamagnetic fraction was determined as $F_d = A/A_0$, where $A_0 = 0.1615(13)$ is the muon asymmetry in a copper sample. It was found that the diamagnetic fraction is small and about the same in both liquid and solid xenon ($F_d = 8(1)\%$ at $T = 163$ K and $F_d = 10(1)\%$ at 70 K). In Ref.[1] the diamagnetic fraction in Xe was estimated to be slightly lower, perhaps due to a smaller wall background. The relaxation rate of the diamagnetic fraction was also the same for liquid and solid Xe: $\sigma = 0.17(3)\ \mu s^{-1}$ at $T = 163$ K (liquid) *vs* $\sigma = 0.15(4)\ \mu s^{-1}$ at T=70 K (solid). An estimate of the relaxation rate for a free μ^+ localized interstitially in the xenon lattice, taking into account ^{129}Xe and ^{131}Xe isotopic abundances, yields $\sigma \approx 0.03\ \mu s^{-1}$, significantly smaller than the observed value. Thus the dipole interaction between a free μ^+ magnetic moment and the nearest Xe nuclei cannot account for the relaxation rate observed in the experiment. A possible explanation can be obtained by taking into account Xeμ^+ ion formation. Theoretical calculations and experimental determinations of the binding energy of a XeH^+ ion agree upon a value of 5.1(1) eV.[7,8] Analogous ions are formed in cryocrystals of H_2, N_2, O_2, Kr, CO_2 and CO. The usual distance from the proton to the nearest nucleus is ~ 1 Å. In Ref. [9] it was shown that in solid nitrogen the μ^+ forms an analogous ion $N_2\mu^+$ with a μ^+-N distance of 1.09(1)Å. An estimate of σ in the case of Xeμ^+ ion formation with a 1 Å muon-nuclear separation gives a value close to that measured. Measurements of the level-crossing resonance in condensed Xe enriched with isotope ^{131}Xe should allow firm conclusions about the nature of the diamagnetic fraction in Xe.

In weak transverse field (*wTF*, $H \lesssim 15$ Oe) we observed muonium precession in liquid and solid Xe at a characteristic triplet frequency $\nu_{Mu} \approx 103\,\nu_\mu$ (where ν_μ is the Larmor frequency of the bare μ^+) due to the strong hyperfine coupling of the muon and electron. Typical *wTF* spectra are shown in Fig. 1.

At intermediate fields ($H \approx 100$ Oe) ν_{Mu} splits into two triplet muonium frequencies ν_{12} and ν_{23} from which one can obtain the hyperfine frequency A using[11]

$$A = \frac{1}{2}\left[\frac{(\nu_{12}+\nu_{23}+2\nu_\mu)^2}{\nu_{23}-\nu_{12}} + \nu_{12} - \nu_{23}\right]. \qquad (2)$$

In the case of solid Xe, Mu relaxation was so fast that the low data aquisition rate at the Dubna phasotron did not allow us to measure the hyperfine frequency. In the liquid phase we measured A in relatively high field ($H \approx 100$ Oe) to resolve two triplet frequencies. This was the first measurement of the muonium hyperfine constant in condensed xenon. Fig. 2 shows the Fourier power spectrum of muonium triplet precession in liquid xenon at $T = 163$ K. Using Eq. (2) we found $A = 4225(220)$ MHz, which is consistent with the well-known vacuum value of $A_{vac} = 4463$ MHz. This means that the muonium atom in liquid xenon is not deformed significantly by interaction with the environment.

In *wTF* the time spectra were fitted to

$$N(t) = N_0\,\exp(-t/\tau_\mu)\,\{1 + A_{Mu}\,\exp(-t/T_2)\,\cos(2\pi\nu_{Mu} + \phi)\} + B \qquad (3)$$

since the diamagnetic fraction was small at all temperatures. In liquid Xe the initial amplitude of muonium precession was $A_{Mu} = 0.040(4)$, which gives a muonium fraction $F_{Mu} = 2A_{Mu}/A_0 = 0.49(5)$. In solid Xe, $F_{Mu} = 54.5(8.5)\%$. These data are consistent with those obtained in Ref. [1].

Fig. 3 shows the temperature dependence of the muonium relaxation rate T_2^{-1}. In liquid Xe the average muonium relaxation rate is $(T_2^{-1})_{liq} \approx 3\ \mu s^{-1}$, which is slightly higher than in Ref. [1]. Solidification of the xenon gives a sharp rise of T_2^{-1}. In the temperature range 120-155 K muonium precession damps so fast that we failed to measure the relaxation rate. In the narrow temperature range 90-120 K Mu relaxation exhibits nonmonotonic temperature behaviour with minimum rate at $T \approx 110$ K. Lowering the temperature further leads to very fast relaxation which we again were unable to measure.

The sharp rise of T_2^{-1} in solid Xe can, of course, be attributed to interactions of the muonium electron with the nuclear magnetic moments of isotopes ^{129}Xe and ^{131}Xe. In liquid Xe motional narrowing probably quenches this relaxation. A decrease of T_2^{-1} with decreasing temperature for $110 < T < 120$ K can be understood in terms of quantum diffusion of Mu in solid Xe. One can explain the nonmonotonic temperature dependence of the Mu relaxation rate near 110 K by taking into account the two-phonon interaction:[6]

$$\tau^{-1} = \frac{4Z}{3}\,\frac{\tilde{\Delta}_0^2\Omega(T)}{\Omega^2(T)+\xi^2(r)} \qquad (4)$$

where τ^{-1} is the hop rate of Mu, Z is the number of equivalent potential wells in the first coordination sphere around the muonium site, Δ_0 is the tunneling amplitude, $\Omega(T)$ is the two-phonon width and $\xi(r)$ is the energy level shift between the nearest equilibrium states, caused by interaction of Mu with lattice defects. Similar behaviour of the muonium T_2^{-1} has been seen in solid nitrogen[5] with a T_2^{-1} minimum around $T \approx 20$ K. In the theoretical framework of Ref. [6] the large value of T_{min} (the temperature of the T_2^{-1} minimum) in solid Xe can be attributed to an extremely weak two-phonon interaction. On the other hand, very strong temperature dependence around T_{min} suggests a strong one-phonon interaction. The sharp temperature dependence of T_2^{-1} in the vicinity of the melting temperature can also be explained in terms of a strong one-phonon interaction with $\tau^{-1} \propto \exp(-E/T)$. Unfortunately, we were able to measure T_2^{-1} only in a very narrow temperature range near the melting point in solid Xe, which does not allow us to determine one-phonon constants as was done in Ref. [5] for solid nitrogen.

It seems very surprising that solid Xe, whose nuclear magnetic moments average less than twice as high as those in solid N_2 and whose fraction of magnetic isotopes is two times lower, exhibits a muonium relaxation rate much higher than that in nitrogen. It is known from ESR measurements that hydrogen atoms in solid Xe are trapped in substitutional sites at $T = 4.2$ K.[12] Comparison of the calculated spectral patterns with the experimental ESR spectra gives local fields up to ~ 25 G. This unusually large local field is consistent with the unusually large Mu relaxation rate in solid Xe. It should be noted that in gaseous xenon the muonium signal also exhibits extremely high relaxation,[13] which so far has no explanation.

The high value of T_2^{-1} in solid Xe may be due to interaction of muonium with free electrons from the μ^+ radiation spur, for instance *via* spin exchange. The negative charge mobility in solid Xe is known to be extremely high[14] (5-6 orders of magnitude higher than, for example, in solid O_2, N_2, CO, *etc.*). However in this case the nonmonotonic $T_2(T)$ dependence is more difficult to explain.

Finally, the large muonium relaxation rate in solid Xe may be attributable to formation of electronically excited molecules XeMu of Xe_2Mu. It is known that anomalous hydrogen-xenon dimers and trimers are comparatively long-lived ($\tau \sim$ 10-100 ns) in rare gas matrices. [See for example Refs. [15] and [16].]

Measurements of muonium relaxation rates in condensed xenon with different magnetic isotope concentrations (especially in samples without ^{129}Xe or ^{131}Xe isotopes) will probably allow us to draw a firm conclusion about the cause of Mu relaxation.

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Figure Captions

1. wTF-μ^+SR spectra for Mu in condensed Xe.
2. Fourier transform of Mu precession signals in liquid Xe.
3. Temperature dependence of the muonium relaxation rate T_2^{-1} in condensed xenon.

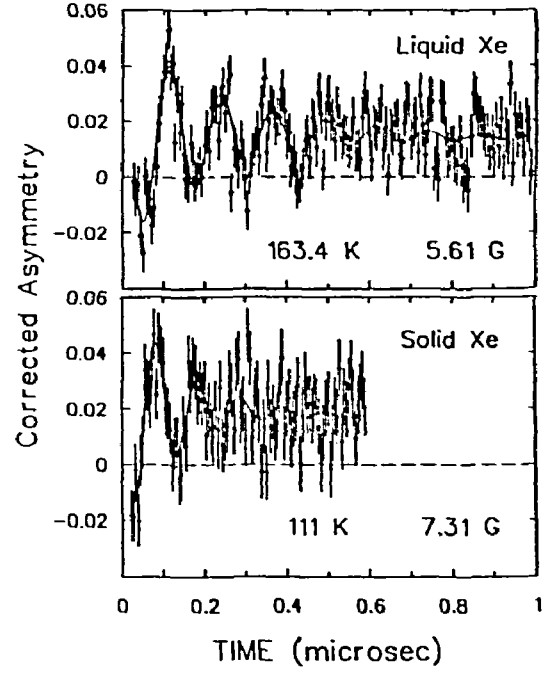

Fig. 1

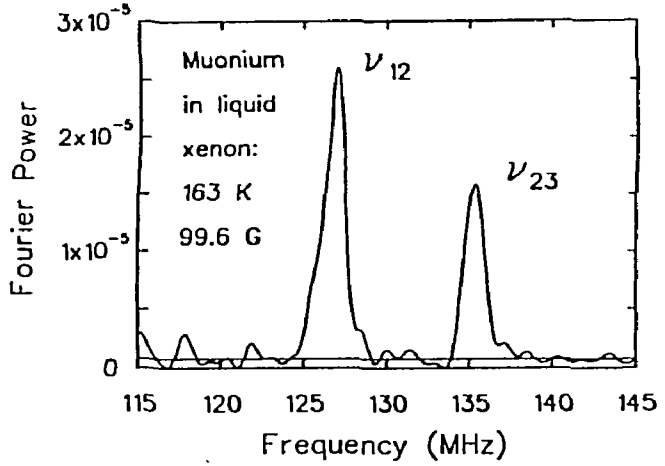
3x10⁻⁵
2x10⁻⁵
1x10⁻⁵
0
Fourier Power
Muonium
in liquid
xenon:
163 K
99.6 G
ν12
ν23
115
120
125
130
135
140
145
Frequency (MHz)

Fig. 2

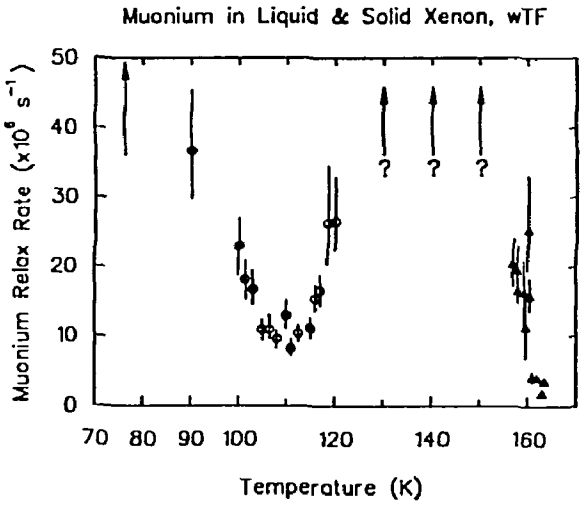
Muonium in Liquid & Solid Xenon, wTF
Muonium Relax Rate (x10⁶ s⁻¹)
50
40
30
20
10
0
?
?
?
70
80
90
100
120
140
160
Temperature (K)

Fig. 3