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Muonium in the condensed phases of Ar, Kr and Xe

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Abstract

The first observations of $\text{Mu} (\mu^+e^-)$ in the solid and liquid phases of Ar, Kr and Xe are reported. The fraction of muons injected into the liquid phases of Ar, Kr and Xe, which form observable Mu , are $97 \pm 29\%$, $57 \pm 10\%$, $43 \pm 9\%$, respectively. The corresponding fractions in the solid phases are $91 \pm 9\%$, $99 \pm 10\%$, and $79 \pm 25\%$.

The spin relaxation rate of Mu in solid Xe is a factor of 10 larger than in the liquid. The effect is attributed to the nuclear magnetic dipole moments of ^{129}Xe and ^{131}Xe which are less effective in relaxing Mu in liquid Xe where motional narrowing is enhanced. The hyperfine splittings of Mu in solid Kr and solid Ar have been measured to an accuracy of 0.1% and are found to be in agreement with the vacuum hyperfine splitting.

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I. INTRODUCTION

When energetic (~ 4 MeV) positive muons are injected into condensed matter, they rapidly lose energy through ionization processes until they reach the range of 2-3 keV.¹ Between this energy and ~ 200 eV the muon has a velocity which is comparable with that of the valence electrons in most atoms. As a result the cross section for electron capture by a muon is large and the muon begins to capture and lose electrons in rapid succession. Below 200 eV a Mu atom (μ^+e^-) is relatively stable and will thermalize through various processes such as electron excitation, phonon creation, and ionization. Muons which slow down as bare muons create free electrons in the region where they thermalize. According to the spur model,² some muonium may result from thermalized muons scavenging these free electrons.

Mu precession has previously been observed in many insulators such as SiO_2 , solid CO_2 , and ice.^{1,3} It has also been observed in some powdered insulators such as SiO_2 , Al_2O_3 , CaO and MgO .^{4,5} In quartz, ice, and solid CO_2 the measured hyperfine frequency is consistent with the vacuum hyperfine frequency which indicates that the Mu is only weakly perturbed by the lattice.¹ This is in contrast to what is observed in semi-conductors such as Ge and Si where electron screening results in reduction of the hyperfine splitting by a factor of about two.^{6,7} Electron spin resonance (ESR) measurements of hydrogen atoms frozen in solid Xe, Kr and Ar at 4.2°K indicate that shifts in the hyperfine frequency are typically negative and less than 1%.⁸

There have been several studies of chemical reactions of hydrogen atoms with impurities such as C_6H_{10} and C_2H_6 deposited in solid Xe.^{9,10} Since Mu can be considered a light isotope of hydrogen (the muon has a

mass about 1/9 that of a proton), the observation of μ in solid Xe, Kr, and Ar makes possible the study of large isotope effects on these reactions. The muon spin rotation technique, already successful in gas and liquid phase chemistry,¹¹ has a few important advantages over conventional ESR techniques. For example, only one μ atom is present in the sample at a time so there can be no confusion as to what is reacting. In many ESR experiments the hydrogen atoms are produced by photolysis of some hydrogen compound present in macroscopic quantities. This can lead to ambiguities since a large number of radicals are produced, complicating the interpretation.

II. EXPERIMENTAL SECTION

The M13 π - μ channel at TRIUMF¹² was used in this experiment to obtain a high luminosity (10,000/sec/cm²) beam of positive spin polarized surface muons of momentum 29 MeV/c.¹³ A standard transverse field μ^+ spin rotation technique¹ was employed using the Lawrence Berkeley Laboratory surface muon apparatus (see Fig. 1). Muons entering the target region were collimated to a 3/4 in. diameter spot before detection by a thin (0.010 in.) scintillator. This detector discriminates very well against beam positrons since the energy loss for a 29 MeV/c muon is about 5 times the energy loss for a positron of similar momentum. The range of a 29 MeV/c surface muon is only 140 mg/cm² of carbon so that all muons which enter the target stop. The apparatus has four telescopes of large solid angle to detect positrons from muon decay. A transparent rear port allowed visual access to the target region which was essential when condensing the gases. Helmholtz coils were used to apply a magnetic field transverse to the muon polarization.

The target vessel (see Fig. 2) and gas handling apparatus were constructed totally of copper and stainless steel, except for the mylar window which allowed muons to enter. The vessel was evacuated to a pressure of 10^{-6} torr and helium leak checked before introducing the target gas. It was mounted on a rotatable liquid N₂ cryostat such that condensation of the target gas could be observed prior to facing it towards the beam. The gases were ultra high purity grade Xe and Kr and research grade Ar. The vessel was equipped with two heaters, one on the gas inlet to prevent blockages and a second, mounted between the copper vessel and the copper cold finger of the cryostat, for temperature control. The liquid Xe, solid Xe, and liquid Kr targets were cooled by maintaining a flow of cold N₂ gas in the cryostat whereas the solid Kr, liquid Ar and solid Ar were cooled with liquid N₂. A temperature controller was used to stabilize the temperature to within $\pm 1^\circ\text{K}$. The solid phases of each gas were obtained by cooling the liquid over a period of 20 min.

III. ELECTRONICS

In a μ SR experiment the measured quantity is the time interval between a muon entering the target and decaying into a positron and two neutrinos. This task was performed using the TRIUMF μ SR data acquisition system.¹⁴ The time digitization was done using a 1 GHz clock designed at TRIUMF. A pile-up gate rejected events for which two muons had entered the target within 8 μsec , reducing distortion in the time spectra considerably at the incident muon rate of 40,000 sec⁻¹ used in this experiment. Positrons from muons which were stopped in the 3/4 in. collimator or the thin scintillator (see Fig. 1) were prevented from triggering the forward, left and right positron telescopes by using counter B1 as a veto. The geometry prevented such positrons from firing the backward telescope.

IV. μSR SPECTRA

The positive muon decays into a positron and two neutrinos with a lifetime of 2.2 μsec. The positron is emitted preferentially in the direction of the muon polarization vector. The time dependence of the muon decay rate into a solid angle defined by the positron telescope allows one to measure the time evolution of the muon polarization vector (both its magnitude and direction).

If a magnetic field of strength B is applied transverse to the muon polarization direction, then the resulting μSR spectrum is of the form¹

$$N(t) = N_0 e^{-t/\tau_\mu} \left\{ 1 + A_\mu R_\mu(t) \cos(\omega_\mu t + \phi_\mu) + \frac{A_{\mu\mu}}{2} R_{\mu\mu}(t) \left[(1 + \delta) \cos((\bar{\omega} - \Omega)t + \phi_{\mu\mu}) + (1 - \delta) \cos((\bar{\omega} + \Omega)t + \phi_{\mu\mu}) \right] \right\} + BG, \quad (1)$$

where N_0 is the normalization, τ_μ the muon lifetime, A_μ the initial free muon asymmetry, ω_μ the free muon precession frequency ($2\pi \cdot 13.55 \cdot B$ kHz·gauss⁻¹), ϕ_μ the corresponding phase, $A_{\mu\mu}$ the initial Mu asymmetry, $\bar{\omega}$ the average Mu precession frequency ($2\pi \cdot 1.394 \cdot B$ MHz·gauss⁻¹), $\phi_{\mu\mu}$ the corresponding phase and BG the time-independent background. Ω and δ are functions of the Mu hyperfine splitting and the applied magnetic field.¹ $R_\mu(t)$ and $R_{\mu\mu}(t)$ characterize the spin relaxation of a free muon and a muonium atom, respectively, with $R_\mu(0) = R_{\mu\mu}(0) = 1$.

V. DATA ANALYSIS AND RESULTS

Spectra were taken at ~65 G in order to determine the fraction of muons thermalizing in a free muon state. The data were packed with 100 nsec/bin and fitted to a six parameter function given by Eq. (1) with $A_{\mu\mu} = 0$ and $R_\mu(t) = e^{-\beta^2 t^2}$. The six free parameters were N_0 , A_μ , β , ϕ_μ , ω_μ , and BG. The free muon relaxation rate was very small so that the

parameter of interest, A_μ , did not depend on whether $R_\mu(t)$ was Gaussian ($e^{-\beta^2 t^2}$) or exponential ($e^{-\lambda t}$). Raw time spectra in Cu and Ar are shown in Fig. 3. The free muon fraction in a sample was determined from

$$F_\mu = \frac{A_\mu(\text{sample}) - A_\mu(\text{Fe}_2\text{O}_3)}{A_\mu(\text{Cu}) - A_\mu(\text{Fe}_2\text{O}_3)}, \quad (2)$$

where the $A_\mu(\text{Fe}_2\text{O}_3)$ is the initial free muon asymmetry measured with Fe_2O_3 contained in a vessel similar to that used for the condensed gases. Since Fe_2O_3 has very large local fields¹⁵ it produces no μ^+ precession at 13.6 kHz·gauss⁻¹; thus any observed precession at that frequency is due to muons stopping outside the 1 in. diameter target region. It is assumed that all the muons, in Cu, precess at 13.6 kHz·gauss⁻¹.

Spectra were also taken at ~8 G to look for Mu precession. The data were packed with 10 nsec/bin and fitted to a function given by Eq. (1) with A_μ , β , and ϕ_μ fixed at the values obtained from the corresponding spectra at ~65 G. In addition Ω and δ were set to zero since in low fields this is a very good approximation.¹ In most cases good fits were obtained with $R_{\mu\mu}(t) = e^{-\lambda t}$. The seven free parameters were N_0 , ω_μ , $A_{\mu\mu}$, $\bar{\omega}$, $\phi_{\mu\mu}$, λ , and BG. In the cases of liquid Ar and solid Kr substantially better fits were obtained with

$$R_{\mu\mu}(t) = \alpha e^{-\lambda_1 t} + (1 - \alpha) e^{-\lambda_2 t}. \quad (3)$$

In the case of solid Kr the χ^2 improved from 164 to 126 for 145 and 143 degrees of freedom, respectively. In the case of liquid Ar the χ^2 improved from 169 to 154 for 145 and 143 degrees of freedom, respectively. The Mu fraction in a sample was determined from

$$F_{\mu\mu} = \frac{2 A_{\mu\mu}(\text{sample})}{A_\mu(\text{Cu}) - A_\mu(\text{Fe}_2\text{O}_3)}, \quad (4)$$

where the factor of 2 arises because half the Mu ensemble oscillates at an unobservable frequency of ~ 4463 MHz. Figure 4 shows a raw spectrum in solid Ar at 8 G.

The free muon and muonium fractions determined from Eqs. (2) and (4), respectively and the Mu relaxation rates are shown in Table I. The uncertainties are due mainly to systematic differences between the results obtained for each telescope. These differences arise because the target itself degrades positrons by an amount which depends on the target material and the direction of positron emission. Since the asymmetry in muon decay depends on the positron energy¹ this leads to small differences in the measured muon and muonium fractions between telescopes.

In the case of solid Xe the relaxation of Mu was so fast that a reliable measure of the Mu asymmetry and relaxation rate was not possible in low field. Therefore, the data at 70 G were used to determine the Mu fraction and relaxation rate. The spectrum was binned at 2 nsec/bin and fitted to a function given by Eq. (1) with $R_{\text{Mu}}(t) = e^{-\lambda t}$. The hyperfine frequency ω_0 was set to the vacuum value. The parameters A_{Mu} , β , N_0 , ω_{Mu} , and ϕ_{Mu} were fixed at the values obtained from the coarsely binned fit. The four free parameters were A_{Mu} , λ , ϕ_{Mu} , and the magnetic field, B .

In the case of solid Kr and solid Ar the relaxation rate of Mu was sufficiently small to make possible a measure of the hyperfine frequency. The data at 70 G were binned at 2 nsec/bin and fitted to a function given by Eq. (1) as in the case of solid Xe. In these fits the hyperfine frequency was an additional free parameter. Figure 5 shows the two frequency precession of Mu in solid argon. The difference in the two Mu frequencies gives rise to a beating in the signal which is a function of the magnetic field and the hyperfine splitting. The measured hyperfine splittings are given in Table I.

VI. DISCUSSION

A. Mu in liquid and solid Ar

The free muon component in both liquid and solid Ar is extremely small ($<2\%$). Figure 6 shows the Mu precession signals. In the case of solid argon $91 \pm 9\%$ of the injected muons form Mu. The small relaxation rate ($0.15 \pm 0.03 \mu\text{sec}^{-1}$) is expected since there are no naturally occurring isotopes of Ar with nuclear moments. In liquid Ar there is a slow relaxing component ($0.65 \pm 0.12 \mu\text{sec}^{-1}$) which accounts for only $48 \pm 6\%$ of the muon ensemble. In addition there is an indication of a very fast relaxing ($19 \pm 11 \mu\text{sec}^{-1}$) component, accounting for $49 \pm 28\%$ of the muon ensemble and observable only within the first Mu precession period [see Fig. 6(a)]. The long-lived Mu signal demonstrates that the inert liquid or its impurities are not responsible for fast Mu relaxation. It is possible that the fast relaxing component is related to the slowing-down process during which many free electrons and radicals are produced. These free electrons and radicals could cause Mu depolarization through an electron spin exchange interaction. Each exchange will reduce the Mu asymmetry by a factor of two provided the time between spin exchanges is greater than the period of the hyperfine oscillation.

Electron spin resonance experiments on H atoms in a solid Ar lattice at 4.2°K indicate at least two trapping sites.⁸ The major component has a hyperfine splitting shift of -0.46% relative to the vacuum value. At temperatures above 39°K the H atoms become mobile. In the present experiment we have measured a mean hyperfine splitting of Mu in solid argon at 77°K to be 4463.8 ± 6.0 MHz which gives a shift of $+0.01 \pm 0.13\%$. It is likely the value we have measured is due to Mu which is diffusing in the bulk, in which case an averaging of the matrix effects which cause

hyperfine shifts (Van der Vals and Pauli exclusion forces)¹⁶ might occur. Real isotope effects between Mu and H atoms will be studied by repeating the present experiment at lower temperatures where the Mu becomes trapped.

B. Mu in liquid and solid Xe

As in the case of Ar the free muon fraction in liquid and solid Xe is small ($3.3 \pm 0.8\%$ and $5 \pm 3\%$, respectively). In liquid Xe only a single Mu component was observed which accounts for $43 \pm 9\%$ of the muon ensemble. It is possible there is also a fast relaxing Mu component, as in the case of liquid Ar, but it is not resolvable. In solid Xe the Mu component accounts for $79 \pm 25\%$ of the muon ensemble.

The Mu relaxation rate increases sharply from $2.1 \pm 0.2 \text{ usec}^{-1}$ in the liquid to $19.0 \pm 2.5 \text{ usec}^{-1}$ in the solid. Figure 7 shows the Mu precession signals in the two phases. The nuclear dipole moments of ^{129}Xe and ^{131}Xe , which comprise 26.44% and 21.18% of the naturally occurring Xe, give rise to random local magnetic fields which can cause Mu spin relaxation. Motional narrowing, an effect greatly enhanced in the liquid, may be responsible for the discontinuity in relaxation rates between phases. The relaxation rate in liquid Xe may be due to reactive impurities present at a level of several ppm.

The ESR spectrum for H atoms trapped at interstitial sites in a Xe lattice at 4.2°K are multicomponent in nature because of hyperfine splittings induced by the lattice.⁸ The spectrum has an overall spread of 98.2 G indicating mean local fields of ~ 25 G at interstitial positions. The μSR spectrum for Mu in solid Xe at 150°K has only one component with a relaxation rate of $19.0 \pm 2.5 \text{ usec}^{-1}$. This corresponds to mean local fields of ~ 1 G. Thus it is likely that the Mu is diffusing fairly fast

at 150°K or possibly trapped in defects where the local fields are much smaller. It should be noted that H atoms are mobile in solid Xe at temperatures above 20°K .^{9,10}

C. Mu in liquid and solid Kr

Again the free muon fractions in solid and liquid Kr are small ($1.4 \pm 1.8\%$ and $6.5 \pm 0.1\%$, respectively). As in the case of liquid Xe the Mu component accounts for only $57 \pm 10\%$ of the muon ensemble. As mentioned in Sec. V, a considerably better fit to the solid Kr data was obtained using a two-component Mu relaxation function [see Fig. 8(b) and Table I]. The sum of these components accounts for $99 \pm 10\%$ of the muon ensemble. One possible interpretation is that the slowly relaxing component ($0.89 \pm 0.04 \text{ usec}^{-1}$) is due to Mu trapped in crystal defects where the effect of nuclear moments (^{83}Kr accounts for 11.48% of the natural Kr) is reduced. The fast relaxing component could be due to Mu trapped or diffusing slowly interstitially.

In the present experiment the hyperfine frequency of Mu in solid Kr has been measured to be 4462.9 ± 3.7 MHz, corresponding to a shift of $-0.01 \pm 0.08\%$ relative to the vacuum value. ESR data on H atoms in solid Kr at 4.2°K indicate a primary component with a hyperfine splitting shift of -0.59% . As in the case of solid Ar this is believed not to be a real isotope effect but rather due to the temperature difference.

VII. SUMMARY AND CONCLUSION

A large fraction of the muons which stop in the liquid and solid phases of Ar, Kr, and Xe form Mu, as in the gas phase of these elements ($100 \pm 10\%$ for Kr and Xe and $73 \pm 6\%$ for Ar).¹⁷ The missing fractions in the solid phases are consistent with zero whereas in the liquid phases roughly half the muon ensemble is unobservable after 100 nsec. In liquid

As there is evidence of a fast relaxing Mu component which would account for the missing fraction. In solid Kr at 90°K there is evidence that Mu exists in two different environments. Also, the Mu relaxation rate is found to be very sensitive to a change of phase in the case of Xe, where the fraction of nuclei with magnetic dipole moments is large.

More data at low temperatures are necessary in order to make firm comparisons with existing ESR data on H atoms in these solid inert elements. It should be possible to study the trapping of Mu atoms by observing the Mu relaxation rate as a function of temperature.

The presence of Mu in the solid phase of these elements is of practical importance since it makes possible the study of Mu reactions in the solid phase. A variety of H reactions with substances deposited in these inert moderators are currently being studied.^{9,10}

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Table I. μ SR results in condensed phases of Ar, Kr, and Xe.

Target Substance	Temperature K	Free Muon Fraction ^a %	Mu Fraction(s) ^b %	Missing Fraction %	Mu Relaxation Rate(s) μsec^{-1}	Mu Hyperfine Splitting ^c MHz
Liquid Ar	85	1.6 ± 1.0	(1) 48 ± 6 (2) 49 ± 28	3 ± 29	(1) 0.65 ± 0.12 (2) 19.0 ± 11.0	
Solid Ar	77	0.8 ± 0.2	91 ± 9	8 ± 9	0.15 ± 0.03	4463.8 ± 6.0
Liquid Xe	162	3.3 ± 0.8	43 ± 9	54 ± 10	2.07 ± 0.21	
Solid Xe	150	5.0 ± 3.3	79 ± 25	16 ± 28	19.0 ± 2.5	
Liquid Kr	120	6.5 ± 0.1	57 ± 10	36 ± 10	3.6 ± 0.9	
Solid Kr	90	1.4 ± 1.8	(1) 71 ± 7 (2) 29 ± 7	0 ± 10	(1) 6.68 ± 0.20 (2) 0.89 ± 0.04	4462.9 ± 3.7

^a These fractions were determined from data at ~ 70 G.

^b These fractions were determined from data at ~ 8 G except for solid Xe where the fraction was from ~ 70 G data.

^c The vacuum hyperfine splitting is 4463.302 MHz.

FIGURE CAPTIONS

1. Experimental apparatus used to measure the time interval between a muon entering the target through the defining counter (D) and the emission of a positron from its decay. The positron event is defined as R1.R2, L1.L2, B1.B2 or F1.F2. The carbon degrader between the counters of each telescope is used to discriminate against low-energy positrons (< 30 MeV).
2. The copper target vessel used to condense the various gas targets. The muons enter through the 0.005 in. mylar window. The target gas is fed in through a tube at the top.
- 3(a). Muon decay rate in Cu at -62 G as a function of time after the muon arrival.
- 3(b). Muon decay rate in solid Ar at $+67$ G as a function of time after the muon arrival.
4. Muon decay rate in solid Ar at 77°K in a magnetic field of 8.9 G as a function of time after the muon arrival.
5. Two-frequency precession in solid Ar at 77°K in a magnetic field of 66.5 G. The flat background, free muon precession, and exponential decay have been removed. The splitting of the two frequencies gives rise to a beating effect.
- 6(a). Mu precession in liquid Ar at 85°K in a magnetic field of 6.9 G.
- 6(b). Mu precession in solid Ar at 77°K in a magnetic field of 8.9 G.
- 7(a). Mu precession in liquid Xe at 162°K in a magnetic field of 10.9 G.
- 7(b). Mu precession in solid Xe at 150°K in a magnetic field of 70 G.
- 8(a). Mu precession in liquid Kr at 120°K in a magnetic field of 10.5 G.
- 8(b). Mu precession in solid Kr at 90°K in a magnetic field of 10.7 G.

Fig. 1

Fig. 2

Fig. 3(a)

Fig. 3(b)

Fig. 4

Fig. 5

Fig. 7(a)

Fig. 7(b)

Fig. 6(a)

Fig. 6(b)

Fig. 8(a)

Fig. 8(b)