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Electric Field Dependence of Muonium Atom Formation in Solid Nitrogen

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Abstract

Muon spin precession signals arising from both muonium (μ^+e^- or Mu) and diamagnetic muon species have been studied in condensed molecular nitrogen in the temperature range 10-78 K. Muonium is formed both in "prompt" epithermal processes and in "delayed" convergence of the thermalized μ^+ with an electron from the muon's ionization track. The latter process is strongly correlated with changes in the electron's mobility in solid N_2; it is inhibited by an electric field $E \sim 3.5$ kV/cm applied in the direction of the muon's motion and enhanced by a comparable electric field in the opposite direction, indicating that the μ^+ stops on average about 60 nm "downstream" of the e^- liberated in its last ionization of the medium.

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Muonium (Mu) is a light hydrogen-like atom with a positive muon (μ^+) as its nucleus [1, 2], formed by stopping high energy ($\sim$ 4 MeV) μ^+ in matter. Although the chemical fate of the μ^+ has long been known [3] to depend critically upon the composition of the stopping medium, our understanding of Mu formation in condensed media is still incomplete and fraught with controversy. Meanwhile, experiments using the techniques of μ^+ and muonium spin rotation/relaxation/resonance (μSR) [1] are providing valuable information on a variety of solid state physics phenomena in systems of considerable interest (magnets, conventional and high-T_c superconductors, metals, semiconductors, insulators *etc.*) where it is generally assumed that the muon is gently inserted into the lattice and subsequently interacts with a host environment that is undisturbed except for the irreducible effects of the muon's presence. In reality, the incoming μ^+ leaves behind an ionization track whose radiolysis products could subsequently interact with the muon and mask the effects one wishes to study. It has been customary to assert [4] that in solids the μ^+ thermalizes (*i.e.* its translational degrees of freedom reach thermal equilibrium with its surroundings) far enough from its own last ionization products that such complications are unimportant. It is extremely important to test this assumption in order to ensure the validity of interpretation of many previous and future experimental results; unfortunately, the spatial distribution of positive muons with respect to their final ionization products has not been experimentally accessible until now.

In this paper we present the first unambiguous observation of Mu formation *via* convergence of the μ^+ with its own radiolysis electron in a *solid*, namely solid molecular nitrogen (s-N_2). Measurements of muon spin precession in external electric fields applied parallel and antiparallel to the initial muon momentum reveal a strong anisotropy in the spatial distribution of such muon-electron pairs: the μ^+ consistently thermalizes "downstream" from the end of its ionization track — which also demonstrates conclusively that the effects of multiple scattering do *not* completely randomize the direction of motion of the muon even at the very end of its range, contrary to conventional wisdom. These results show that "muon radiolysis" does play an important role in some solids; in s-N_2 such effects are strongly correlated with *electron mobility*, allowing us to predict which other solids are likely to exhibit similar phenomena.

In the process of slowing down from about 10 keV in *low pressure gases*, the μ^+ undergoes numerous "charge exchange" collisions in which it picks up an electron directly from the atoms or molecules of the medium to form Mu and loses its electron to become a μ^+ again. The charge state of the muon upon thermalization depends upon the relative affinities of the μ^+ and the host species for electrons, the energy dependence of the various charge exchange processes and the rate at which the muon slows down through the critical energy region [5]. Thermal muonium atoms produced in this way are herein designated "prompt Mu." Any free electrons liberated earlier in the passage of the muon (while it still has sufficient energy to ionize the medium) are expected either to recombine with their parent positive ions or to become attached to other atoms or molecules, which remain distant from the thermalized muon. Muonium formation in low-pressure gases is therefore usually assumed to be "all prompt" as defined above.

In *liquids* (including, by extension, high-pressure gases [6]) and in *solids*, this picture must be adapted to take collective phenomena into account; the image of isolated "collisions" is no longer generally valid and it may be misleading to think of a se-

quence of "charge exchange cycles" in the same way as for gases. Nevertheless, the outcome (differentiation of the muons into several charge states upon thermalization) will be similar and one expects a "prompt Mu" fraction in condensed media as well. Meanwhile, ions (including free electrons) generated in the muon's ionization track may sometimes be mobile enough to reach the thermalized muon and form muonium and/or interact magnetically with the spin of a Mu atom formed earlier; such ions may be created *via* direct ionization of the medium by the high energy incoming muon, *via* secondary ionization by scattered electrons or *via* $\mu^+ \rightleftharpoons$Mu charge exchange cycles at lower energy, where electron capture by the muon leaves behind a positive ion and electron loss by muonium deposits the electron again further downstream. This complicated scenario is often referred to as the "spur model" [7-10]. In the past, the strongest argument against this model was the absence of any effect of external electric field on Mu formation [11], contrasting with the strong electric field effects observed for positronium formation [12].

Whereas reactions of the muon with its own radiolysis products must be exothermic (since said products have usually thermalized), charge-exchange processes are presumed to occur epithermally even in condensed phases; therefore the prompt Mu fraction has sometimes been referred to as "hot" muonium [13, 14, 2, 15, 16]. Although this terminology accurately evokes the epithermal formation of prompt Mu, it now also carries a burden of contentious connotations which we prefer to avoid. Because of the delay between muon thermalization and the arrival of a free electron or mobile negative ion, muonium atoms formed from muon-radiolysis electrons are herein designated "delayed Mu," although obviously the formation time can in some cases be too short for practical experimental discrimination between "prompt" and "delayed" Mu.

Muonium formation has recently been observed in superfluid liquid helium (ℓ-^{4}He) [17], in liquid and solid N_2 [18] and in liquid and solid Ne [19], where the ionization energies of isolated atoms or molecules (25, 15.6 and 22 eV, respectively) are much higher than that of Mu (13.5 eV) and so Mu formation must be either epithermal or associated with radiolysis. In normal ℓ-^{4}He there is no Mu formation; moreover, the amplitude of Mu precession in superfluid ℓ-^{4}He depends strongly on magnetic field strength as well as on the magnitude of an applied electric field [17]. Those results show that *all* Mu formation in that system is due to "delayed" convergence of the thermalized μ^+ with a radiolysis electron.

During the slowing down process there may also be a finite probability for reactions in which the epithermal μ^+ or Mu atom (usually denoted μ^* or Mu*) combines chemically with a host atom or molecule to incorporate the muon into a molecular species. Such "hot atom reactions" [13, 14, 2, 15] are usually assumed to be too endothermic to occur after thermalization; they can therefore be clearly distinguished *theoretically* from "delayed" radiolysis reactions. However, since both usually occur before the beginning of direct observation of the muon polarization by μSR, the two types of reactions are difficult to distinguish *experimentally*.

In gaseous, liquid or solid molecular nitrogen (as in noble gases or in *any* non-reactive molecular environment), the most likely diamagnetic species incorporating muons are simple molecular ions (in this case $N_2\mu^+$) formed by addition of the μ^+ to a host molecule; $N_2\mu^+$ has been shown experimentally [22] to be the dominant diamagnetic muon species (herein generally designated μ_D) in s-N_2. Since this ion can form by addition of a thermal μ^+ to a host molecule, all muons which do *not* form prompt

Mu are presumed to thermalize in this form initially. The μ^+ is therefore effectively immobilized at $t = 0$. However, the positive charge of the molecular ion still serves to attract negative ions or free electrons from muon radiolysis, promoting formation of a delayed Mu fraction by the exothermic process $N_2\mu^+ + e^- \rightarrow N_2 + \mathrm{Mu}$, where e^- could also be any mobile negative ion. In s-N_2 the mobility of free electrons is such that it is possible to discriminate experimentally between the prompt and delayed Mu fractions, as shown below.

When a beam of spin polarized positive muons is stopped in a sample, the Mu and μ_D fractions can be distinguished easily by their precession frequencies in a weak transverse magnetic field (wTF). In wTF-μSR, μ_D precesses at the Larmor frequency of the free muon, $\omega_\mu = \gamma_\mu H$ (where H is the applied magnetic field and $\gamma_\mu = 2\pi \times 0.01355$ MHz/G), whereas triplet Mu precesses at a characteristic frequency $\omega_{\mathrm{Mu}} \approx 103\,\omega_\mu$ in the same H. Because half the muon polarization in Mu oscillates at a frequency which is normally too high to observe experimentally [1], the "asymmetry" (muon decay anisotropy) of the Mu "signal" (A_{Mu}) is half that of μ_D (A_D) for the same formation probability. Both are compared with the full asymmetry A_o obtained with a metallic sample to obtain the Mu and μ_D polarization fractions, $P_{\mathrm{Mu}} = 2A_{\mathrm{Mu}}/A_o$ and $P_D = A_D/A_o$ respectively. When some fast depolarization takes place before the signals are observed experimentally, it is helpful also to define a "lost [polarization] fraction" $P_\ell = 1 - P_{\mathrm{Mu}} - P_D$. One cannot tell directly whether P_ℓ (if any) should be considered part of the Mu or μ_D fractions.

The first μSR investigation of condensed nitrogen [20] revealed a pronounced maximum in the temperature dependence of P_D at the $\alpha - \beta$ transition temperature ($T_{\alpha\beta}$) below which the N_2 molecules freeze into fixed orientations along the diagonals of *fcc* unit cells in α-N_2; above $T_{\alpha\beta}$ the molecules are free to rotate so that β-N_2 is orientationally disordered. In that work the μ_D species was presumed to be the linear $N_2\mu^+$ ion analogous to N_2H^+ [21]. This assumption was confirmed experimentally by detection of the characteristic ^{14}N-μ^+ nuclear quadrupolar level-crossing resonance in longitudinal magnetic field [22]. Further investigations led to observation of long-lived Mu atoms in s-$^{14}N_2$ with a muonium hyperfine parameter $\nu_o = 4494(5)$ MHz [18] close to the vacuum value (4463 MHz), but the details of Mu formation in condensed N_2 remained unknown.

The present experiment was performed on the M13 beam line at TRIUMF. Ultra high purity nitrogen ($^{14}N_2$ with $\sim 10^{-5}$ impurity content) was condensed from a glass bulb into a rectangular copper cell 22 mm on a side and 6 mm thick. The front and back of the cell were sealed with clear Mylar windows 0.125 mm thick. A 10 mm square window in the external wall of the cryostat allowed us to visually inspect the growth of s-N_2 crystals, which usually took about 5 hours. Only perfectly transparent crystals without any visible defects were used in the experiment. An external electric field of up to 3.5 kV/cm was generated by means of two parallel grids of very fine wires located in front and back of the sample cell. Positive muons of 28 MeV/c momentum and 100% spin polarization were stopped in the condensed N_2 and wTF-μSR measurements were made in various magnetic fields.

The typical wTF-μSR spectra in Fig. 1 show μ_D signals in s-N_2. It was found that the diamagnetic signal itself clearly has both a slow-relaxing (S) and a fast-relaxing (F) component in liquid N_2 (ℓ-N_2) and in solid β-N_2. These signals plus twice the Mu amplitude accounted for the full asymmetry at each temperature; accordingly, we set

$P_\ell = 0$ throughout the analysis. Therefore, taking into account also the Mu component, the overall muon decay asymmetry was described by the following expression:

$$A(t) = A_{\mathrm{Mu}} \exp(-\lambda_{\mathrm{Mu}} t) \cos(\omega_{\mathrm{Mu}} t + \varphi) + [A_F \exp(-\lambda_F t) + A_S \exp(-\lambda_S t)] \cos \omega_\mu t, \tag{1}$$

where A_{Mu}, A_F and A_S are the muonium, fast-relaxing diamagnetic and slow-relaxing diamagnetic asymmetries, respectively, and λ_{Mu}, λ_F and λ_S are the corresponding depolarization rates. The S component has been unambiguously identified as the $N_2\mu^+$ molecular ion [22]; it is reasonable to assume the same identity for the F component. The decay rate λ_F of the F signal varies with temperature and is always at least two orders of magnitude higher than that of the S component; this is too fast to be due to interaction with nuclear moments. This component is probably a direct manifestation of delayed muonium formation due to mobile electrons created in the incoming muon's ionization track. No F component was seen in $\alpha - N_2$, probably because λ_D^{-1} is shorter than the experimental time resolution.

Figure 2(a) shows the temperature dependences of A_{Mu} and A_S in s-N_2. Most prominent is the minimum in the Mu asymmetry and corresponding maximum in the S asymmetry near the $\alpha - \beta$ transition. This anti-correlation suggests competition between Mu formation and incorporation of stopping muons into molecular ions. Muonium has an ionization potential of 13.5 eV, whereas $N_2\mu^+$ has a binding energy of about 5 eV in the undisturbed s-N_2 lattice; the polarization of neighbouring nitrogen molecules by the charge of the $N_2\mu^+$ ion will contribute a characteristic interaction energy of ~ 1 eV [22], so the ion cannot form spontaneously from thermal Mu and a neutral N_2 molecule. The maximum in A_S is therefore not directly due to an increased rate of $N_2\mu^+$ formation, but rather indirectly due to the decreased probability of Mu formation.

"Prompt" (epithermal) Mu formation, like any "hot-atom" reactions of Mu^* with N_2 to form $N_2\mu^+$, is unlikely to exhibit any dramatic temperature dependence. More probably, a large fraction of the muons thermalize in the charged state and form molecular ions, only to capture a radiolysis electron and form "delayed" Mu. In s-N_2 positive charges have been found to be immobile [23], so the e^- must move to the μ^+. In this case one would expect the Mu formation probability to depend critically upon the mobility of electrons in the lattice.

Figure 2(b) presents the temperature dependence of the electron mobility b_e in s-N_2. From 63 K down to 50 K, b_e is fairly constant, $b_e \approx 1.7 \times 10^{-3}$ cm^2 s^{-1} V^{-1}; it then decreases to about 0.7×10^{-3} cm^2 s^{-1} V^{-1} at $T_{\alpha-\beta}$. The muonium asymmetry has the same temperature dependence; moreover, it changes in the same proportions. Below $T_{\alpha-\beta}$ the electron mobility appears to increase sharply to about 2.0×10^{-3} cm^2 s^{-1} V^{-1}, although the mobility data do not reveal the temperature dependence. Our data show a sharp increase in the muonium asymmetry, accompanied by a corresponding (approximately twice as large) drop in the diamagnetic asymmetry just below the transition. Electron drift mobility measurements usually have comparatively low precision; nevertheless the temperature dependence of the Mu asymmetry convincingly follows that of $b_e(T)$. We conclude that Mu formation in s-N_2 is at least partially due to convergence of the μ^+ and a radiolysis electron.

The most obvious test of this conclusion is to apply an external electric field E to the sample and see if it affects the Mu and μ_D signals. Figure 3 displays the results of

just such a test in α-N_2. Both A_{Mu} and A_S do indeed change with E, confirming that delayed Mu formation takes place in s-N_2. The E-dependences shown suggest that the spatial distribution of the radiolysis electrons relative to the thermalized muons is highly anisotropic: $N_2\mu^+$ ions are formed downstream (*i.e.* in the direction of the initial muon momentum) from the last radiolysis electrons of the muon's ionization track. A positive sign for E corresponds to an electric field applied "forward" (along the initial μ^+ momentum direction) which thus pulls the μ^+ and e^- apart, giving rise to an increased long-lived diamagnetic component. Negative E is "backward" (opposite to the initial μ^+ momentum) and thus pushes the μ^+ and e^- together to form more Mu atoms. It should be noted that the electric field dependences of A_{Mu} and A_S also (like their temperature dependences) suggest the presence of competing channels which determine the final states of positive muons. The scale of the A_{Mu} decrease is about half that of A_S increase, as expected [1] from the 50% loss in polarization in the former.

No nonlinear "hot electron" effects have been observed in s-N_2 up to applied electric fields of $E \simeq 10^2$ kV/cm [23]. Therefore the leveling off of A_{Mu} and A_S for $E > +3$ kV/cm probably reflects compensation by the external electric field of the Coloumb attraction between μ^+ and e^-, thus providing an estimate of $R_s \sim 6 \times 10^{-6}$ cm for the $\mu^+ - e^-$ distance in s-N_2 from the relation $E = e/\epsilon R_s^2$, where $\epsilon = 1.45$ is the dielectric constant of s-N_2. Even at high temperatures, this value of R_s is an order of magnitude less than the Onsager length $R_c = e^2/\epsilon k_B T$, which is usually considered to determine the escape probability for an ion pair: $W \sim \exp(-R_c/R_s)$ [24]. Since a substantial P_D is observed in s-N_2 at *all* temperatures, treating the μ^+ and e^- as an isolated pair is clearly an oversimplification. Rather, the muon has to compete with positive ions from its own track for neutralization.

In β-N_2 and in liquid N_2, we were unable to detect any variation of A_{Mu} or A_S with external electric field up to 3.5 kV/cm. However, we were able to estimate R_ℓ, the characteristic $\mu^+ - e^-$ distance in ℓ-N_2, by measuring the *magnetic* field dependence of A_{Mu} at $T = 75$ K. Assuming that the conversion from μ^+ to Mu occurs at exponentially distributed times,

$$dn_{\mathrm{Mu}}(t) = -dn_\mu(t) = \lambda n_\mu(t) dt, \tag{2}$$

where λ is the characteristic formation rate, the muonium asymmetry has been shown [7] to be

$$A_{\mathrm{Mu}} \sim \frac{\lambda}{(\lambda^2 + \omega_{\mathrm{Mu}}^2)^{1/2}}. \tag{3}$$

Expression (2) holds true for a uniform spatial distribution of the e^- with respect to the μ^+, which is obviously not the case here; nevertheless, (3) gives a reasonable estimate for the parameter λ.

Figure 4 shows the magnetic field dependence of A_{Mu} in liquid nitrogen. The line on the figure is a fit according to Eq. (3) including a prompt Mu fraction which is not dependent on magnetic field. The field dependent part of A_{Mu} is due to delayed Mu formation: different Mu atoms are formed at different times and the phase coherence among the precessing delayed Mu atoms is lost. The higher the magnetic field, the stronger the effect of dephasing. At high enough field only the prompt Mu asymmetry remains — which, according to the fit, has the value 0.042(1). The Mu formation rate was determined to be $\lambda = 2.3(1) \times 10^7$ s^{-1}. This parameter determines the average time

$\tau \equiv \lambda^{-1}$ needed for the μ^+ and e^- to meet. In low electric fields, when charged particle mobilities are independent of electric field the time of approach can be estimated as

$$\tau = \lambda^{-1} = \frac{R_\ell^3 \epsilon}{3eb}, \qquad (4)$$

where R_ℓ is the typical $\mu^+ - e^-$ distance and $b = b_{\mu^+} + b_e$ is net mobility. In ℓ-N_2 positive and negative charges turn out to be almost equally mobile, giving $b \simeq 2.5 \times 10^{-3}$ cm^2/V·s [23] at $T = 75$ K. Equation (4) gives an estimate of $R_\ell = 3.2(1) \times 10^{-6}$ cm for the $\mu^+ - e^-$ distance in liquid N_2. This distance corresponds to an electric field between μ^+ and e^- charges of $E \sim 10$ kV/cm. Such a high field is probably the reason for our failure to observe any E-dependence of A_{Mu} or A_S in liquid nitrogen.

The characteristic initial distance R_s between μ^+ and e^- in solid nitrogen is about twice as large as R_ℓ; in gaseous N_2 this parameter was found [6] to be $R_g = 11.9(6) \times 10^{-6}$ cm at a pressure of 50 atm and room temperature. It should be noted that in gaseous N_2 at pressures above 10 atm the "spur model" was concluded to be the dominant mechanism of Mu formation while at lower pressures the "hot model" was the best [6]. A possible reason why the $\mu^+ - e^-$ distance in α-N_2 is larger than in ℓ-N_2 is a lower cross section for μ^+ scattering by frozen N_2 molecules; the same argument would hold for β-N_2, where the almost freely rotating N_2 molecules present a higher μ^+ scattering cross section. Alternatively one may postulate additional low energy collective modes (librons) as an extra energy-loss channel for μ^+ thermalization in α-N_2. Either way, we expect to observe electric field dependence of A_{Mu} and A_S at high enough electric fields in both ℓ-N_2 and β-N_2.

In conclusion, we have presented the first direct evidence for muonium formation *via* convergence of the μ^+ with a radiolysis electron from the muon's ionization track in a solid. A strong anisotropy was found in the $\mu^+ - e^-$ spatial distribution in solid nitrogen. Characteristic initial $\mu^+ - e^-$ separations were determined to be on the order of several hundred angstroms in both solid and liquid nitrogen.

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FIGURE CAPTIONS

1. Diamagnetic precession signals in s-N_2 in a transverse magnetic field of 65 G at several temperatures. The muonium signal is eliminated by adjusting the channel width to a large integer multiple of the Mu precession period. Note the two-component (S and F) relaxation at high temperatures. Typical muonium precession signals are shown in [18].
2. (a) Temperature dependences of the Mu (circles, in a wTF of 5.2 G) and slow-relaxing diamagnetic (stars, in a TF of 65 G) asymmetries in solid nitrogen. The minimum of A_{Mu} and maximum of A_S take place at the temperature of the $\alpha - \beta$ transition in s-N_2. (b) Temperature dependence of the electron mobility in solid nitrogen, from Ref. [23]. Different symbols refer to different specimens.
3. Electric field dependence of Mu (circles) and slow-relaxing diamagnetic (stars) asymmetries in α-N_2 at $T = 20$ K. The change in A_S is about twice that in A_{Mu}.
4. Mu asymmetry *vs.* transverse magnetic field in liquid nitrogen at $T = 75$ K. The line is a fit according to Eq.(3) taking into account an additional magnetic field independent "prompt" Mu fraction.

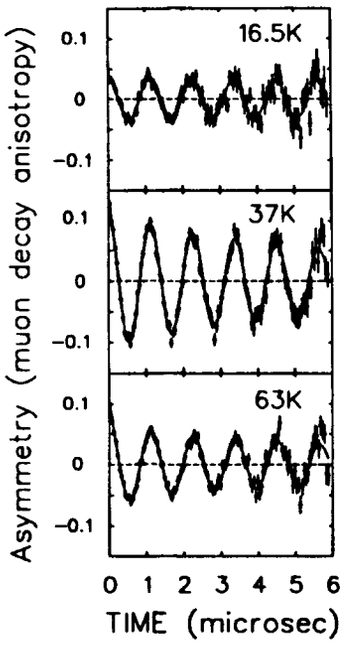

Fig. 1

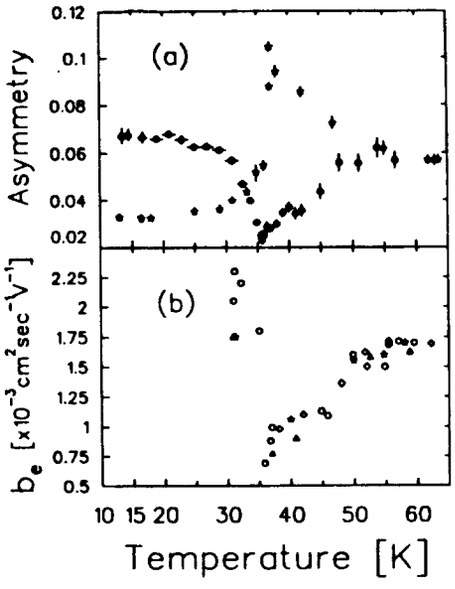

Fig. 2

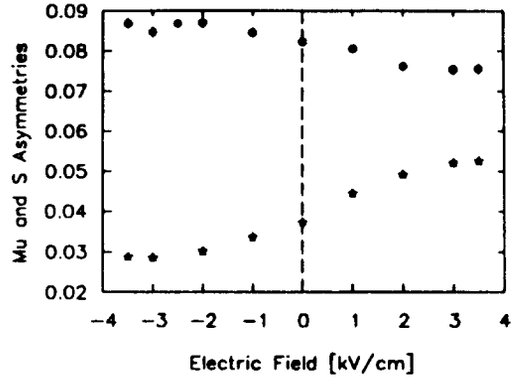

Fig. 3

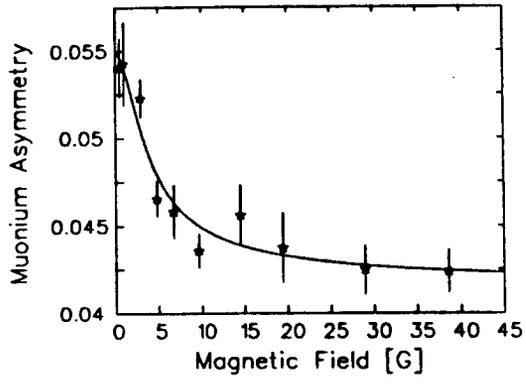

Fig. 4