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Prospects for the Future of μSR

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Prospects for the future of μSR

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For 35 years, the μ^+ has been used as a probe of the intrinsic properties of materials and as a light isotope of hydrogen, whose behaviour in many media is of considerable interest. This meeting reviews mainly the latter use, which has achieved much in the last decade in fields ranging from chemistry to quantum diffusion. The next decade offers unprecedented opportunities for μSR, but its unavoidable integration with other accelerator-based programmes renders it vulnerable to 'politics'. I will explore three questions, often in parallel: (1) 'So what?' (What has μSR revealed so far of fundamental or technological importance and how should we aim to exploit its potential?) (2) 'What next?' (What new μSR capabilities are likely in the next decade if we extrapolate progress in muon beams, μSR techniques and experimental facilities?) (3) 'How?' (Given these priorities, what coherent plan might the μSR community realistically implement by cooperation and organized effort?)

1. Introduction

Having just listened to two days' worth of fascinating reviews and exciting reports on important new data, I feel a bit like the man who, having sneaked into the Sultan's harem disguised as a eunuch, belatedly begins to entertain doubts about his own adequacy to the task. However, as impossible as it may be to satisfactorily recapitulate and extrapolate from this meeting, at least it will be fun trying...

I will begin with a brief historical perspective. Before the mid-1950s the experiments we do today in μSR would have fallen into the category of pure fantasy, since they violate the 'known laws of physics' of the time. Only with the confirmation of parity non-conservation in the weak decays $\pi^+ \to \mu^+ + \nu_\mu$ and $\mu^+ \to e^+ + \nu_e + \bar{\nu}_\mu$ in 1957 did μSR enter the realm of legitimate science fiction, which extrapolates from the possible without regard for the limitations of known technology. For nearly two decades after 1957, these properties of the $\pi \to \mu \to e$ decay chain were exploited primarily for the sake of a famous series of heroic experimental tests of QED and elementary particle physics. Most of the experimental techniques we now call 'μSR' were invented to enable those fundamental physics experiments and many of today's areas of application of μSR began as peripheral problems in their implementation (e.g. mechanisms for depolarization of muons in condensed matter).

In the 1970s the meson factories increased the intensity of available muon beams by factors of 100 to 1000, triggering a veritable explosion of new techniques and applications of μSR, in recognition of which most meson factories invested in new muon beamlines and μSR facilities. Since the early 1980s these techniques have

Table 1. *Performance of muon beams for μSR*

Requirements:
high polarization (100% for 'surface' μ^+)
high luminosity
(1) high flux ($\gtrsim 2 \times 10^4$ s^{-1} on sample)
(2) small spot size (sample sizes $\lesssim 1$ cm^2)
short stopping range (140 mg cm^{-2} for 28 MeV/c surface μ^+)
low contamination of π, e, etc. ($\Rightarrow \boldsymbol{E} \times \boldsymbol{B}$ velocity selector)
transverse polarization capability ($\Rightarrow$ *big* $\boldsymbol{E} \times \boldsymbol{B}$ velocity selector)
$\Rightarrow$ QUALITY FACTOR

$$Q\ [\mathrm{s}^{-1}\ \mathrm{g}^{-1}] \equiv \frac{(\text{polarization})^2 \times (\text{flux})}{(1 + \text{contamination}) \times (\text{range}) \times (\text{spotsize})}$$

History of improvements:

before meson factories	$Q \lesssim 10^3$
backward muon beams at meson factories	$Q \lesssim 6 \times 10^6$
first surface μ^+ beams at meson factories	$Q \sim 2 \times 10^6$
today's best surface μ^+ beams	$Q \sim 3 \times 10^7$
moderated μ^+ beam at PSI (1995?)	$Q > 10^9$?
dissociated muonium μ^+ beam at BOOM (1995?)	$Q > 10^9$?

been adopted by the materials science community and μSR has become a 'state of the art' magnetic resonance tool, devoted primarily to disciplines that are rarely associated with subatomic physics.

However, it is unwise to lose sight of the origins of μSR, particularly in view of the dependence of μSR on the continued operation of large, expensive accelerators that were (with the exception of the ISIS facility at the Rutherford-Appleton Lab in Britain) built by and for subatomic physicists and require their continued enthusiasm to maintain operation. As subatomic physics falls on harder times, it is sensible and necessary to actively work toward an alliance between subatomic physics and materials science, much like that which allowed the present blossoming of synchrotron radiation facilities. If μSR is to secure its place in the repertoire of standard materials science experimental techniques, we must meet the daunting challenge of creating enough high-quality muon beams to satisfy the demands of a growing user community – and refining the techniques and facilities to the point where they are accessible to all.

2. Progress in muon beams

Table 1 summarizes the desirable features of muon beams for μSR and shows the improvements in the overall figure of merit Q over the last few decades. The development of the 'surface muon' beam (Bowen 1985) in the 1970s was crucial to the exploitation of μSR in materials science because it made experiments feasible on targets of $\lesssim 0.1$ cm^3 (by now $\lesssim 1$ mm^3) – compatible with typically available samples of new materials – rather than the $\gtrsim 1$ cm^3 samples required for the older high-momentum, low luminosity 'backward' muon beams (still required for negative muons, a fact which impedes similar widespread exploitation of μ^-SR as a materials science probe).

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(*a*) *CW against pulsed accelerators*

To date, CW accelerators like those at TRIUMF in Canada and PSI in Switzerland, which deliver a constant primary beam intensity (apart from RF microstructure on a scale of ns), have enjoyed both the highest average intensities and the most diverse μSR programs, because they are able to produce the largest number of muons for time-integral experiments and also the most delicate time-differential (TD)-μSR experiments, which involve measuring the time interval between each muon's arrival and decay with the highest possible precision.

However, the requirement that each muon decay before another be allowed into the target limits useable muon fluxes to less than about $10^5\ s^{-1}$, often more than an order of magnitude less than the beamlines can produce. This pile-up limitation can be alleviated by up to a factor of 4 by installing a 'kicker' in the muon beam that deflects the beam after one muon has arrived in the target, waits 10–20 μs and then accepts another. Such a device has been utilized only once to date (Hutson 1986), but could be developed to multiplex a surface muon beam to several different experiments, enhancing the throughput of a given high-intensity muon channel by more than an order of magnitude.

Pulsed accelerators, such as the BOOM facility at KEK in Japan and the ISIS facility at the Rutherford Appleton Laboratory in England, overcome the pile-up limit by accepting all the muons at once; thus thousands of TD-μSR events may be accumulated simultaneously. This situation not only evades the rate limitation but also is ideally suited for experiments involving irradiation of the muons with RF, microwave or laser power. As intensities of pulsed machines increase, the prospect of 'frame-by-frame' μSR on a millisecond time scale becomes tantalizingly attractive. At ISIS, rates are currently limited mainly by the thickness of the muon production target, which is kept minimal lest it produce a perceptible loss of intensity at the neutron spallation source downstream.

However, there are two difficulties with pulsed primary beams: first, the pulses are always of finite length, typically *ca.* 50 ns, and even if the primary beam pulses were infinitely sharp the muons produced from them would be smeared out by the 26.03 ns decay lifetime of the pions from which they arise; thus pulsed beams of this sort cannot be used for conventional TD-μSR experiments requiring high time resolution, because all the muons do not arrive simultaneously. Second, because all the muons arrive at once (often accompanied by many beam positrons) and begin decaying simultaneously, the counting rate in the positron detectors varies from many kilohertz to zero in the space of a few microseconds. Since any detector's efficiency has some rate dependence, this introduces distortions in the time spectra that can only be reduced by segmenting the counters and/or using analogue counting techniques (Yamazaki 1982; Kuno 1986). While improvements of these methods may be expected, the most delicate TD-μSR experiments may remain the province of CW accelerators.

(*b*) *Future muon beams*

(i) *Muon production at high energy*

Several proposals are now being considered for 'kaon factories', accelerators with intensities comparable to those of the 'meson factories' (greater than 100 μA) but higher energies (greater than about 20 Gev), which will efficiently produce beams of kaons and other particles from the 'second generation' in large numbers

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for the first time. Such machines are also expected to produce larger numbers of pions, in rough proportion to the total power in the beam, so that muon beams with intensities more than an order of magnitude higher than today's best are on the horizon. Unfortunately, kaon factories are also more expensive than the meson factories, with the result that it is generally expected that at most one will be built in the forseeable future.

(ii) *Higher stopping luminosity*

Perhaps the greatest contributing factor in the evolution of μSR from an esoteric and exotic technique into a practical tool for materials science research has been the steady increase of muon beam stopping luminosity (measured in $s^{-1}g^{-1}$) – the most variable component of the 'quality factor' Q in table 1 and more important than sheer flux (in s^{-1}) or even luminosity (in $s^{-1}cm^{-2}$). When samples of *ca.* 100 g were needed to stop enough muons for a μSR experiment, fewer experiments were done and none of those were done on rare new materials, which are simply not available in such quantities. Even when targets two orders of magnitude smaller were feasible, measurements of rare or expensive materials required heroic acquisition efforts. Only with the full development of the 4 MeV 'surface muon beam' in the early 1980s did 'solid state physics sized' samples become accessible to μSR, with dramatic results. Today's state of the art allows use of samples 1–2 mm in diameter massing as little as 10 mg using special μSR techniques, but most samples are still much larger.

(iii) *Phase space compression*

Since surface muons (which have the highest naturally occurring Q) have a sharp momentum only at the upper limit (for pions decaying at the very surface of the production target) and are distributed as $p^{7/2}$ below 29.789 MeV/c, their stopping luminosity, while superior, is limited. Several approaches to phase space compression are being explored and the next few years may see dramatic improvements of Q. At PSI in Switzerland a slow μ^+ beam is being developed using simple moderation techniques with the very high flux available there; at the BOOM pulsed muon facility of KEK in Japan an alternative approach is being used: thermal muonium (Mu) atoms emitted from a heated-foil muon production target will be ionized by an intense laser pulse, liberating positive muons essentially at rest in vacuum. Either of these beams may be capable of producing *ca.* 10^4 μ^+/s at energies of *ca.* 1–10 eV, implying an increase of over 10^4 over present intensities and placing surface science within reach of μSR for the first time. The reacceleration of such muons would also make extremely low-emittance beams possible, leading to μ^+ implantation with few-micron precision. We may expect both facilties to be available for general use within 1–2 years.

(iv) *Dreams: muon farms and storage rings*

Further in the future we may hope to achieve complete control of the energy, phase space and time structure of low-energy muon beams, perhaps using muon storage rings less than 1 m in diameter to decelerate, cool and compress the muon beam. This would permit delivery of *ca.* 10^6 μ^+ in pulses of *ca.* 1 ns to each of 5–10 μSR experimental areas, all with stopping luminosities *ca.* 10 times higher than those available today and each capable of utilizing the full intensity in time-differential (TD)-μSR, which can accept only *ca.* 5×10^4 μ^+/s at CW

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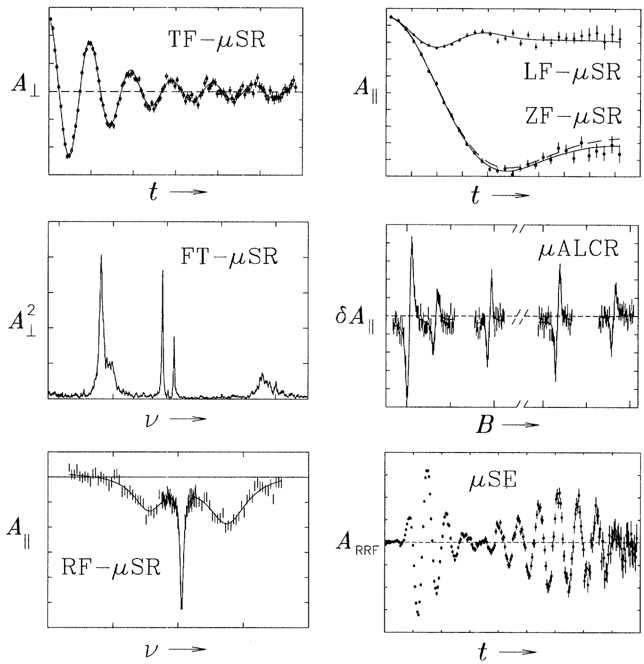

Figure 1. Brewer's list of approved acronyms for μSR experimental techniques. TF-μSR, transverse field muon spin rotation (includes by extension muonium spin rotation); LF-μSR, longitudinal field muon spin relaxation; ZF-μSR, zero field muon spin relaxation (technically not true 'relaxation' in most cases, but impossible to classify in the usual scheme of 'T_1' against 'T_2'); FT-μSR, Fourier transform muon spin rotation (important in the spectroscopy of muonium and radicals); μALCR, muon avoided level crossing resonance (essential for studying hyperfine couplings between other nuclei and the electron which couples to the μ^+); RF-μSR, radio frequency muon spin resonance (including by extension irradiation with the entire electromagnetic frequency spectrum); μSE, muon spin echo* (using RF resonance). Several other important techniques are LF muonium decoupling (an old technique now enjoying a resurgence, particularly in applications to Mu in semiconductors, as reported by several groups at this Meeting) and stroboscopic μSR, a time-integral TF-μSRtechnique used at PSI to take full advantage of the higher flux of muons available there. *The μSE signal is shown in a rotating reference frame at a frequency just below the muon's resonant frequency in the applied field (too high to be displayed directly).

facilities. Such a 'muon farm' would thus produce μSR results at 100 times the rate of today's typical μSR facility. The effects of such a dramatic improvement would be unpredictable, of course, but they might be expected to fall into three categories: (*a*) far more delicate and detailed measurements would be feasible; (*b*) more powerful, specialized techniques and apparati would be developed; and (*c*) μSR would become more readily accessible to a larger number and variety of scientists. The implications are obvious.

3. Progress in μSR techniques

Improved muon beams have facilitated an even more dramatic blossoming of μSR technology as this youngest branch of the magnetic resonance tree struggles to catch up with older established branches like NMR and EPR. I have no room here to document these developments, but for reference I will provide the graphical definitions of some popular μSR acronyms provided in figure 1.

4. µSR applications

The uses of µSR in materials science fall roughly into two categories or 'themes'. While this meeting is concerned mainly with the second, it is meet to mention the first, since it occupies roughly half the attention of the µSR community.

(a) *The muon as a magnetic probe*

In solids, the μ^+ generally occupies an interstitial lattice site (whereas the μ^- will go into a tight orbit about a host nucleus) where it will precess in the local magnetic field at that site. In magnetic media, this field (and its dependence on temperature, pressure, external electric and magnetic fields, etc.) is often the subject of investigation; thus the $\mu^\pm$ serves as a simple and highly reliable magnetometer. The dynamics of disordered magnetic systems like spin glasses and frustrated antiferromagnets have been widely studied using µSR, which has some advantages over more conventional methods such as neutron scattering in such cases. Recent experiments on 'heavy fermion' systems have also shown that µSR has unmatched sensitivity to very weak moments.

A similar application is found in superconductors, where the distribution of local fields found in a 'vortex lattice' in the mixed state of type-II superconductors produces a characteristic µSR frequency spectrum that can be analysed to obtain the magnetic penetration depth λ, whose magnitude reveals the ratio of n_s (the superconducting carrier density) to m^* (the effective mass of the charge carriers). The temperature dependence of n_s in turn reveals the nature of the pairing state, which has been the central mystery for the high-T_c cuprate superconductors as well as various organic superconductors. Thus µSR has made important contributions to these fields, especially in the early stages when only crude samples were available.

(b) *The muon as a light isotope of hydrogen*

(i) *Muonium chemistry*

Muonium is in every sense a true light isotope of the H atom; they have almost exactly the same size, reduced mass and ionization potential and both obey the Born–Oppenheimer approximation. But what an isotopic difference – Mu is a factor of *ca.* 18 lighter than D! The lighter Mu atom exhibits dramatic quantum tunnelling effects in barrier penetration that are rarely seen so unambiguously in chemistry experiments but which provide very important tests for the few *ab initio* theories of chemical reaction kinetics (Fleming 1992). Once the differences between Mu and H chemistry are well understood (as is now the case for many types of reactions), measurements of Mu reactivity (which are often easy) can be used to reliably predict the reactivity of H under circumstances where H atoms cannot even be detected.

(ii) *Radical chemistry*

Chemical reactions often incorporate Mu into radicals (paramagnetic molecules with unpaired electrons) where a weakened hyperfine interaction persists between the electron and muon spins (and also between the electron and any other nuclei with magnetic moments). These molecules so closely resemble their analogues in which the μ^+ is replaced by a proton that they have nearly identical reaction kinetics. The muon version may thus reveal the chemical behaviour of the proton version even when information on the latter is unavailable by other methods. The very existence of some radicals (never observed by any other means) has been confirmed by μSR.

Using μALCR techniques one can measure not only the hyperfine coupling of the muon to the unpaired electron but also the couplings of that electron to the other nuclei; thus the muon can be used to study the molecular structure of regions of the molecule far from the muon's site, much as in a conventional ENDOR experiment (Percival 1987). Sometimes the detailed structure of such radicals is influenced by the mass of the adatom: Mu tends to have slightly longer bond lengths and larger zero-point motions, which influence bond angles. Combined with the fact that the nuclear hyperfine couplings can often be measured more accurately by μSR than by ENDOR, this makes μALCR a very important probe of molecular structure.

(iii) *Hydrogen in semiconductors*

Hydrogen is known to be an ubiquitous impurity in semiconductors (e.g. Si and GaAs), where it is now being incorporated intentionally for the purpose of *passivating* electrically active impurities. Consequently, a knowledge of the location, electronic structure and dynamical behaviour of H atoms in semiconductors is vital to that industry. So far, very little has been learned about isolated H atoms from any method that observes H itself, but μ^+SR spectroscopy techniques including longitudinal-field decoupling of Mu and Mu*, channelling of muons from π^+ decay or positrons from μ^+ decay, FT-μSR and μALCR have provided a wealth of such information (Patterson 1988; Kiefl 1988).

Many such results were presented at this Meeting; I despair of making a thorough summary, so will mention only the newly developed RF-μSR techniques which have revealed the temperature dependence of the fractions of muons in Mu, Mu* or ionized states like μ^+ or Mu^- at times comparable to the muon lifetime (long after any initial formation or reaction of paramagnetic states). These probabilities are strong functions of p- or n-type doping in Si, and thus reflect interactions between H-like species and impurities, the very information required for understanding industrial uses of H. This profuse new information seemed to me to dramatically increase the community's confusion level – which is, after all, the main purpose of new results! One or two years from now the resulting cognitive dissonance should have motivated a far deeper understanding of this commercially important subject.

(iv) *Quantum diffusion*

The theory of quantum tunnelling with dissipation, a currently important field of condensed matter physics, has implications for the electronic and diffusive transport properties of all types of materials (Stamp 1991; Kagan 1992). Given its light mass, its affinity for electrons and its repulsion from nuclei, the μ^+ is an ideal light interstitial particle with which to study such phenomena (Flynn 1970; Kagan 1974; Petzinger 1982). For these reasons, the quantum diffusion of μ^+ and Mu in solids has been the subject of numerous μSR experiments (Kadono 1992) and associated theoretical analyses (Kagan 1992).

μ^+ diffusion in metals. In pure, defect-free metallic crystals, one generally observes the intrinsic interactions between the μ^+ and the lattice. (In metals with

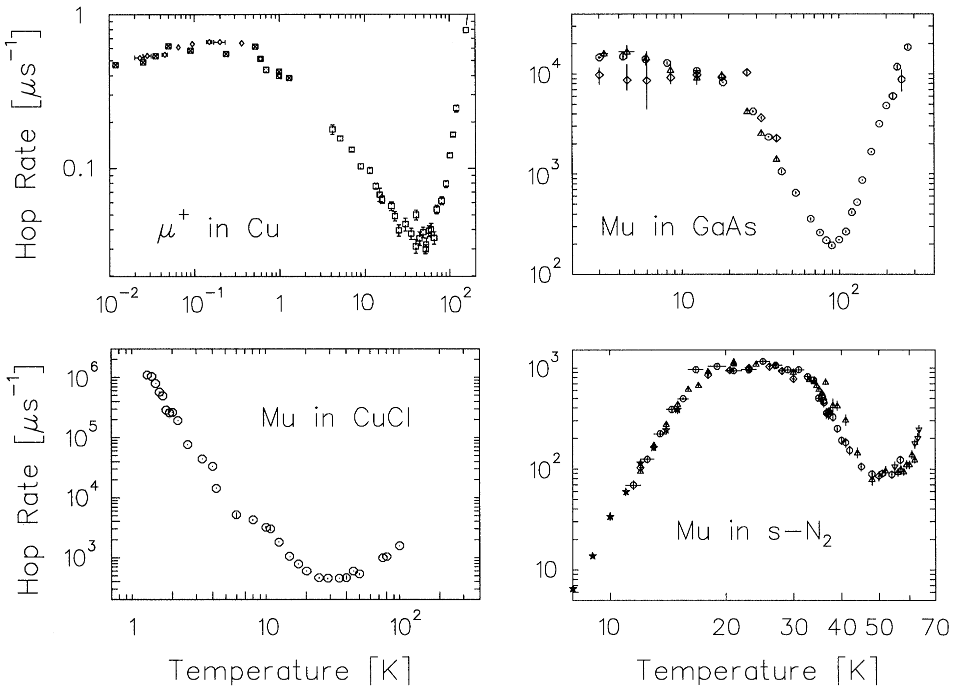

Figure 2. Temperature dependence of the μ^+ or Mu hop rate τ_c^{-1} in various pure crystalline materials. Upper left: μ^+ in copper metal (from Luke 1991). Upper right: Mu in semiconducting GaAs (from Schneider 1992). Lower left: Mu in semiconducting CuCl. Lower right: Mu in insulating solid nitrogen (from Storchak 1994). In each case there is a region where τ_c^{-1} increases with decreasing T, the signature of coherent tunnelling, and in each crystal $\tau_c^{-1}(T)$ shows a power-law behaviour in that region; note, however, the huge differences in horizontal and vertical scales!

defects or impurities, trapping is likely, leading to complicated dynamics (Petzinger 1982).) The hopping of positive muons between sites in metals has been studied using TF-, ZF- and LLF-μ^+SR experiments, revealing a consistent qualitative temperature dependence typified by the classic example shown in the upper left-hand corner of figure 2: at high temperatures ($\gtrsim 80$ K in Cu) the muon exhibits semiclassical thermally activated hopping over the energy barriers between adjacent interstitial sites; at lower temperatures ($\lesssim 20$ K for Cu) the μ^+ actually hops faster as T decreases due to the enhancement of quantum tunnelling as the disorder due to lattice vibrations is reduced. At still lower T tunnelling is inhibited by intrinsic disorder. The observed 'electron drag' effect (additional dissipation due to electronic degrees of freedom) giving a much weaker power law $\tau_c^{-1} \propto T^{-\alpha}$ for screened positive muons in metals than for Mu in insulators, is predicted by theory (Kondo 1992). Similar behaviour has been observed indirectly in normal aluminium, whereas in superconducting aluminium, which can be driven normal by an applied magnetic field, muon diffusion is considerably faster in the superconducting state than in the normal state, because the opening of a superconducting gap at the Fermi surface effectively quenches the dissipation due to screening electrons (Kondo 1992).

Mu diffusion in semiconductors and insulators. In ionic crystals containing highly negative ions such as F^- or O^{2-}, the μ^+ tends to form relatively strong hydrogen bonds, suppressing μ^+ diffusion until quite high temperatures ($\gtrsim 100$–

200 K). However, if muonium is formed, the Mu atom is more or less decoupled from local electric fields and usually diffuses freely through the lattice.

The T-dependence of τ_c^{-1} for Mu in semiconducting GaAs (Schneider 1992) is startlingly similar to that seen for the μ^+ in Cu, as shown in figure 2. Similar results are also seen in CuCl, one of the most ionic of the diamond-lattice semiconductors, and in ionic insulators such as KCl (Kadono 1990), except that the power-law behaviour $\tau_c^{-1}(T) \propto T^{-\alpha}$ has a quite different exponent in the region where τ_c^{-1} decreases with T, namely $\alpha \approx 3$ rather than $\alpha < 1$. This is because the quantum tunnelling of Mu (which is not coupled to charge carriers) is governed by Mu-phonon interactions, whereas that of the μ^+ is dominated by interactions with conduction electrons.

In insulating *cryocrystals* of solid nitrogen (s-N_2), the T-dependence of the Mu hop rate is somewhat different, as shown in the lower right-hand corner of figure 2. Such Van der Waals crystals have very low Debye temperatures and weak couplings between the neutral Mu atom and the host lattice; they are therefore interesting systems in which to test theoretical predictions of the temperature dependence of Mu quantum diffusion (Storchak 1994).

(c) *Summary and speculations*

My view of science is a process of 'desperately seeking simplicity', in which we begin with a conventional understanding of a subject which motivates some crude measurements using standard methods on some apparently peripheral topic, which often yields both a simple (expected) result (destined for obscurity) and a confusing extra bit that forces us into cognitive dissonance. This being an unpleasant state, we are motivated to perform higher precision measurements using the same standard methods, which almost always yield incomprehensible, seemingly chaotic results, further deepening the dissonance, which we then spread around to as many others as possible to share the frustration. This eventually spurs thoughtful reinterpretation of the data as well as the development of new measuring techniques. Together, these efforts reveal the order in complexity, a rich phenomenology and possibly some new physics – which leads to a 'feeding frenzy' as everyone tries to cash in on the latest excitement. When this finally settles down, of course, we are back to a new (we hope improved) conventional understanding, and we start over again.

This cycle reached the 'incomprehensible chaos' stage in the study of Mu/H in semiconductors sometime in the last several years, and is now in the process of digestion. Watch for a major breakthrough in understanding of metastability in these systems in the near future!

However, the H atom community and the Mu community need to come together before this can have practical impact; so far the former study mainly high H concentrations while the latter are concerned only with the isolated atom, as might be expected – we take little interest in that which we cannot see for ourselves!

Another issue that needs to be addressed by the μSR community is the difference between thermal incorporation of H and implantation of Mu: how important is radiolysis in the formation of Mu states in solids generally, and in semiconductors in particular? Here the chemists and phyicists have much to teach one another and a strong cooperative effort is needed.

The study of μ^+/Mu quantum diffusion has gone through at least one full cycle of the above-mentioned process in its 20-year history; it seems that a comprehensive theory is essentially complete and we need only verify its predictions in a few more cases. This is an exciting time to work in this area, but I predict that we are in for another session of confusion when we begin to seriously study inhomogeneous diffusion in imperfect crystals – potentially the most relevant to understanding transport in real materials.

On the whole, an exotic and rather expensive suite of experimental techniques called μSR have certainly begun to have a significant impact upon materials science. Whether this should be regarded as 'forced' productivity driven by the implacable persistence and ingenuity of a community of brilliant enthusiasts or as the intrinsic power of the μSR technique inexorably asserting itself despite the ignorance and ineptitude of its promoters is for others to decide.

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