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THE USE OF POSITIVE MUONS IN METAL PHYSICS

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ABSTRACT

Positive muons are implanted in metals at different temperatures, ranging from 0.05 to 300 K. Their spin precession and depolarization is followed over a time interval of 10 μ s, either in applied fields or in internal fields in ferromagnets. The information obtained resembles that of proton NMR (the μ^+ is chemically equivalent, but has only 1/9 of the proton mass), with the important difference that the muons can be studied in metals in extremely low concentration.

The experiments described here deal with a) the determination of interstitial sites for the muons; b) the mechanism of diffusion at low temperatures (tunnelling effects), comparing pure metals and slightly disordered structures induced by doping with small amounts of impurities; c) trap-limited diffusion at intermediate temperatures, and d) spin-density investigations in magnetically ordered materials.

It is shown that the muon localization is very sensitive to the presence of impurities, even at the 10 ppm level, but the underlying mechanisms may be different in the low-T and intermediate-T regions. It is also shown that frequency shifts can be measured with high precision, which allows a careful study of, for instance, temperature and pressure effects on the local spin density at the muonic sites.

1. INTRODUCTION

During the last few years, the study of muon interactions in metals has developed into a new kind of spectroscopy which may serve as a complement to some of the older methods by which light impurity atoms in metals are studied. One main reason for the present interest is the high sensitivity of the method -- every muon introduced in the sample produces its own signal, which means that one can work at concentrations where the particles are truly isolated from each other; another reason is the time-scale for observation of the muon interactions, which allows the experimenters to deduce jump rates for diffusion or fluctuation times of local environments in ranges which are typical for several processes of interest in the host metals. Furthermore, the muon methods are of very high precision as far as local magnetic fields are concerned.

The positive muons, which are the subject of the present review, have charge $+e$, spin $= \frac{1}{2} \hbar$, and mass $0.11 m_p$. From a chemical point of view, they are therefore expected to behave like protons. In insulating and semi-conducting environments they do indeed tend to bind an electron and form muonium, which is the analogue of the hydrogen atom. In metals, with their high electron densities, bound states become very short-lived and the situation is better described as an impurity charge screened by conduction electrons. This is the same situation as for hydrogen impurities in metals, and results from positive muon experiments are therefore believed to be of relevance also for the interpretation of hydrogen-metal interactions at low concentrations. In some respects, the lower mass of muons ($\sim 1/9$ of the hydrogen mass) leads, however, to differences in their behaviour, notably in the diffusion rates.

The technique is usually referred to as a μ SR (Muon Spin Rotation). The method of observation will be described very briefly (see Fig. 1).

The muons are produced in the decay of π mesons; to produce the π 's with good intensity one requires a proton accelerator with an extracted beam energy around 500 MeV and an intensity of at least some μA (such accelerators exist in Europe at CERN and SIN, Switzerland, at Dubna and Gatchina, USSR, and other facilities for muon production are considered at IKO, Amsterdam, and Rutherford Lab., UK). The muons come out, with proper momentum selection, with a high degree of polarization (70-80%) and can be made to stop in metallic targets of a thickness of about 1 cm; in some special arrangements (so-called Arizona beams), even 0.01 cm. Each incoming muon starts an electronic clock.

The muons lose energy and finally arrive at thermal equilibrium in the metallic environment. This process takes a time of the order of 10^{-10} s, and it is found experimentally that the loss of polarization in this initial stage is negligible. There is a "spur" of radiation damage left behind each muon; but in metals, in contrast to the case of insulators, the muons continue to move relatively far away from the last knocked-out atom and stop in undamaged regions of the lattice¹⁾. The number of spurs per cm^3 in a normal experiment is so small that interaction with another muon's spur can be safely neglected.

The muon decays with a half-life of 2.2 μs into a positron and two neutrinos. The positrons can easily be detected since most of them have energies in the range 20-50 MeV. They give a Stop signal to the electronic clock, and all the stopped muon events in a typical experiment (10^6 - 10^7 muons) are sorted in a time histogram. The number of events recorded in a certain time interval measures the direction and magnitude of the spin polarization at the instant when these muons decayed, since the β^+ emission probability is strongly peaked in the μ^+ spin direction. If the metallic sample is placed in a *transverse magnetic field* (a field

perpendicular to the μ^+ polarization and to the plane in which the β^+ detectors are situated), the spins will precess with a Larmor frequency of 13.553 kHz/G and the time histogram may look like the example in Fig. 2, where each point has been normalized by division by the exponential decay factor.

Such curves reflect the precession and gradual depolarization of the muonic spins. The precision in the frequency determination is normally of the order of $5 : 10^5$ (for 10^6 events), and can be improved up to $5 : 10^6$ in specialized arrangements using the so-called stroboscopic techniques²⁾. The range of fields in which transverse-field μ SR can be conveniently measured ranges from 20 G (corresponding to two precession periods over the observation time, usually 8-10 μ s) up to 50 kG, where the time resolution sets a limit. Local fields at muon sites in ferromagnets have been found to be of the order of a few kG.

The damping of the curves in the transverse-field case is due to the dephasing of muon spins because of interactions with random local fields. For a muon fixed in the lattice, the damping is described by

$$P(t) = P_0 \exp(-\sigma^2 t^2), \quad (1)$$

where $\sigma^2 = \frac{1}{2} \overline{\gamma^2 B_{\text{dip}}^2}$, i.e. the second moment of the frequency broadening due to nearby nuclear dipoles. For a muon jumping very quickly between such sites, one would have instead

$$P(t) = P_0 \exp(-2\sigma^2 \tau_c t) = P_0 \exp(-\lambda t), \quad (2)$$

where τ_c is a correlation time related to the jump time τ of the muons.

All this is completely analogous to the concepts of NMR, which becomes clear if one makes the Fourier transforms of the muon time histograms. As a matter of fact, the terms "line-width" and "motional

narrowing" are used also in many papers on muon spin precession. The transverse-field experiments measure the " T_2 -relaxation".

Our working definition of "fixed" muons is of course related to the particular time window of the observation. We consider a muon to be immobile if the mean time of stay at its stopping site is longer than $\approx 10 \tau_\mu$ (which is about 20 μ s). We expect the jump time to decrease below this limit when we raise the temperature of a metal above a certain temperature T_f . The "text book" behaviour of damping versus T looks therefore as shown in Fig. 3. Putting in typical fields from nuclear dipoles, we find that the transition region between strong and weak damping corresponds to $\tau_c = 10^{-7}$ - 10^{-5} s. If the muon jumps between inequivalent sites in a metal with ordered electronic moments (ferromagnetic or antiferromagnetic), the local fields can be 1000 times higher and the sensitive region for the determination of τ_c will fall in the range 10^{-12} - 10^{-9} s.

In the intermediate region (around T_f) a formula of the type

$$P(t) = P_0 \exp \{-2\sigma^2\tau_c^2[\exp(-t/\tau_c) - 1 + t/\tau_c]\} \quad (3)$$

is used to fit the data. In this case it is useful to describe the damping by specifying the time required for the polarization to decay to $1/e$ of its initial value. The inverse of this time is called Λ , a parameter to be used later.

Information on the local fields at the muon sites and their diffusion can thus be obtained from transverse-field experiments. We should also note that if a material is ferromagnetic and the muons stop at well-defined sites, it is possible (and often preferable) to observe their precession in zero external field, with well-demagnetized samples. With local fields of random direction but unique strength, the spins will

still precess in phase and produce a maximum of intensity, after each full period, in the detectors, which are then placed in the forward or backward directions. However, 1/3 of the amplitude is lost, corresponding to precession around field components directed along the initial polarization direction. Figure 4 shows an example of a zero-field experiment in iron. These curves have a counterpart in perturbed angular correlation (PAC) studies in random ferromagnets³⁾.

A *zero field relaxation experiment* in a non-magnetic material with detection in the forward-backward geometry measures the ratio

$$\frac{N_f - N_b}{N_f + N_b} = AP_z(t) , \quad (4)$$

where the z-axis is now taken along the beam (i.e. the initial muon polarization); $AP_z(0)$ is about 0.25. The time histograms will now have shapes of the type shown in Fig. 5a depending on the value of the product $\tau\Delta$ ($= \tau_c\sigma$ in our notation)⁴⁾. The curve for $\tau\Delta = \infty$ corresponds to the static nuclear dipole field perturbation limit. After a sufficiently long time, the polarization is restored to 1/3 of its initial value, since precession due to the z-components of B_{dip} should not change P_z on the average. The zero-field longitudinal method can help to distinguish between effects of static field broadening and dynamic spin fluctuations, especially in systems with slow fluctuations such as spin-glasses. An additional possibility is to apply *longitudinal fields* to decouple the nuclear dipole field and thus separate a possible T_1 relaxation of electronic origin in magnetic systems from other effects.

2. LATTICE SITES OF POSITIVE MUONS IN METALS

Most metals crystallize in one of the following simple structures: b.c.c. (body-centred cubic); f.c.c. (face-centred cubic); or h.c.p.

(hexagonal close-packed). The two cubic structures are shown in Fig. 6. In both types there exist interstitial sites around which the nearest neighbouring metal atoms have octahedral (O) or tetrahedral (T) symmetry. These sites are also indicated in the figures. One of the principal problems is to decide which of the two types is the most stable site for the positive muon. It is expected that a muon and a hydrogen (or deuterium) atom should take up the same type of site, but the information on hydrogen (from channelling, neutron diffraction, etc.) is still fairly incomplete, and muon data would give a valuable confirmation of or complement to these data.

The magnetic interaction of the muon moment with nearby dipole moments provides a way of determining the muonic sites. Several metals are composed of isotopes with large nuclear dipole moments (Cu and Al among the f.c.c. metals; V, Nb, Ta among the b.c.c. metals). These moments produce fields of the order of a few gauss at the muonic sites, with a spread characterized by the parameter $\langle B_{\text{dip}}^2 \rangle$. Calculating the dipole sums with the applied field B_0 (the z-axis) in the principal directions $[100]$, $[110]$, and $[111]$, one finds distinctly different values for σ for the two types of interstitial positions, as illustrated by a few examples in Table 1. These are the so-called van Vleck values of σ . For the b.c.c. lattice there are also sites which are magnetically inequivalent although they are electrostatically identical. This fact shows up for certain directions of the applied field.

In principle, therefore, a measurement of σ for a single crystal sample should be enough for a site determination.

The first experiments of this type (on muons in Cu single crystals) gave very small differences in σ for the three directions $[100]$, $[110]$, and $[111]$. It was later pointed out by Hartmann⁵⁾ that such experiments

must be performed in sufficiently strong applied fields in order to get the answer expected from the simple theory of dipolar sums. The reason is that the expectation value of the dipolar field is changed by the electric quadrupole interaction experienced by the neighbouring nuclei when the muonic charge is introduced in an interstice. If the muon can be considered as a point charge, it produces a radially directed field gradient EFG (Fig. 7), and the nuclei are subjected to a combined electric and magnetic interaction. The symmetry axis of the EFG forms, in general, a certain angle β with the direction of the applied field (z-axis) for each neighbouring nucleus.

The Hamiltonian for the combined interaction

$$H = H_{\text{magn}} + H_{\text{el}} \quad (5)$$

acting on the neighbouring nuclei was set up and diagonalized by Hartmann, and the expectation values $\langle I_z \rangle$ and $\langle I_x \rangle$ were calculated.

Each nucleus will contribute a dipolar field component (θ, ϕ are the polar angles of nucleus i as seen from the muon)

$$\Delta B_z^{(i)} = \gamma \hbar \langle I_z \rangle_i^{\text{stat}} \frac{3 \cos^2 \theta - 1}{r^3} + \gamma \hbar \langle I_x \rangle_i^{\text{stat}} \frac{3 \sin \theta \cos \theta}{r^3} \quad (6)$$

to the local field at the muon site. There are also oscillating components of $\langle I_z \rangle$ and $\langle I_x \rangle$ giving fluctuating fields, but these can be neglected as long as their periods are low compared to the Larmor frequency of the muon. The lattice sums were carried out over about 70 nuclei around the interstitial, but the major contribution to the sums comes from the nearest neighbours. The EFG was assumed to decrease with the distance from the muon as $1/r^3$.

The resultant values of $\langle \Delta B_z^2 \rangle$ depend strongly on the parameter $y = \omega_B / \omega_E$, where ω_E is the quadrupole interaction frequency. At large- y

values, where the magnetic interaction dominates, the $\langle \Delta B_z^2 \rangle$ curves approach the van Vleck values. For most nuclei the ratio of the magnetic moment has such a value that this "strong field limit" is approached in a magnetic field of a few thousand gauss, but for strongly deformed nuclei, such as Ta, it would be difficult to reach this limit.

Experiments giving the damping constant σ as a function of applied magnetic fields, with the field along the main symmetry axes, are shown in Figs. 8a ⁶⁾ and 8b ⁷⁾ and compared to the theoretical predictions. It is quite evident that the data can only be fitted with the assumption of *octahedral* sites for muons in Cu and *tetrahedral* sites for muons in Mn-doped Al at 15 K. In both cases it has been necessary to scale down the theoretical curves by 15-20% in order to obtain a fit. This effect is interpreted in terms of a dilation of the lattice of about 5% around the muon.

From the same data one can find a value of ω_E , and therefore the electric field gradient at the neighbouring nuclei, by fitting the curves with an appropriate parameter y . The results are of low accuracy, mainly due to the large uncertainties in the quadrupole moments of ^{61,63}Cu and ²⁷Al,

$$q = 0.27 \pm 0.15 \text{ \AA}^{-3} \text{ Cu (octahedral } \mu^+)$$

$$q = 0.18 \text{ \AA}^{-3} \text{ Mn-doped Al (tetrahedral } \mu^+),$$

but could be of interest for comparison with electronic screening calculations of the impurity charges.

When the nuclei of the host material have no quadrupole moment ($I = \frac{1}{2}$) the effects discussed above are not relevant, and zero-field experiments should give information directly on the muonic site from measured σ -values. As has been shown by Yamazaki and collaborators ^{4,8)},

a zero-field longitudinal measurement is in fact more sensitive to the local dipole environment than a normal high-field transverse measurement because of a higher effective σ , and it can also establish clearly if the muons are static. An example is presented in Fig. 5b for the case of ZrH_2 (where the dipolar fields are dominated by the hydrogen). The value of σ deduced favours octahedral interstitials with respect to Zr sites for the muons, rather than substitutional sites.

Site determinations for the muons can also be performed in magnetic materials using the unique local fields expected for different sites, as has been discussed for muons in Co by Meier et al.^{9,10}.

3. LOCAL ELECTRONIC STRUCTURE AROUND A MUON IN A METAL

Like protons, the muons form the simplest impurity centres imaginable for scattering of itinerant electrons in a metal. Several theoretical treatments exist already¹¹⁻¹⁴), most of them based on linear response theory, but attempts are also being made to introduce the muon as a centre in cluster calculations¹⁵). As far as hyperfine interaction parameters are concerned, a muon (and a proton) are of course much easier to handle than heavier impurities because of the absence of core electrons. In all these treatments the muon itself has so far been considered as point-like.

One experimental observable which can be compared with theory has been mentioned already: the field gradient (EFG) produced by the muonic charge on neighbouring nuclei and screened by the electrons. The EFG data are, however, scarce and imprecise, and the main quantities of interest are the *Knight shift in the non-magnetic metals* and the *strong contact fields at the interstitial sites in ferromagnetic metals*. Another quantity which is obtainable indirectly is the local volume dilatation (causing decreases in σ -values and changes in activation

energies for diffusion). These lattice distortions can, in principle, be calculated by screening charge methods¹⁶⁾, to be compared with experiment.

A series of Knight shifts have been reported by Schenck et al.¹⁷⁾ and are shown in Fig. 9. The various theories referred to in this figure have not been particularly successful in predicting the Knight shifts. More recent approaches seem, however, to be on the way to improving this situation¹⁸⁾.

In ordered magnetic materials the magnetic field seen by the muons is the sum of the local dipolar field and the contact field due to the strong spin polarization of the conduction electrons (no field is applied in the experiment):

$$B_{\text{loc}} = B_{\text{dip}} + B_{\text{cont}} , \quad B_{\text{cont}} = - \frac{8}{3} \pi \mu_B \eta(0) (n_0^{\uparrow} - n_0^{\downarrow}) . \quad (7)$$

In ferromagnetic metals, both terms may be of the order of kilogauss. The dipolar term disappears for both types of interstitial sites in the f.c.c. lattice (Ni), but should take two different values for octahedral as well as tetrahedral sites of b.c.c. metals (Fe). However, if there is a fast jump process between magnetically inequivalent sites in Fe, the dipolar fields are expected to average out in this case also, whereas the contact field remains. This fast averaging seems to be working even at relatively low temperatures in Fe, as will be discussed in more detail in the next section.

Experiments in both Ni and Fe therefore measure the contact field at the muon site. For the theoretical comparison the local spin polarization to be put into Eq. (7) is the pure metal value $(n_0^{\uparrow} - n_0^{\downarrow})$, multiplied by an enhancement factor $\eta(0)$ calculable from theory in the scattering models. Some attempts to calculate this quantity are reproduced in Table 2 together with the measured values¹⁸⁾.

The unperturbed interstitial spin densities in the B_{cont} calculations are taken from neutron scattering data. The agreement for $\eta = 0$ in Ni is good but may be fortuitous. The over-all situation is not at all satisfactory. The theory may indeed more easily predict second-order effects, such as changes with temperature or pressure on the contact fields, rather than their absolute values.

Several groups have measured the difference in the temperature dependences $B_c(T)$ as compared to that of the host metal magnetization $M(T)$, usually expressed by the parameter $A(T)$ in

$$\frac{B_c(T)}{B_c(0)} = A(T) \frac{M(T)}{M(0)} . \quad (8)$$

This has been done for Ni, Fe, Co, and Gd^{18,19)}. Recent measurements from CERN have given the results shown in Fig. 10. The full curve is a theoretical prediction for Ni by Katayama et al.²⁰⁾, based on muffin-tin potentials. The local spin density is a result of the hybridization of 1s electrons on the μ^+ sites (assumed to be octahedral) and d-electron states from Ni. The temperature dependence follows from the rearrangement of spin-up and spin-down electrons near the Fermi level.

The thermal expansion of the host lattice is, however, also a reason for this spin density variation. A high-pressure experiment (up to 7 kbar) at room temperature was performed²¹⁾ in order to pick out the volume dependence explicitly. This variation is shown in Fig. 11. Using these constants it was found for the case of Ni that about half of the variation in $A(T)$ could be explained by expansion, which would bring the theory into good agreement for this metal. Similar calculations for muons in Fe do not yet exist.

A number of examples of data from μ^+ interactions in magnetic materials is given in a recent review article by Meier²²⁾.

4. DIFFUSION OF MUONS IN METALS

As for hydrogen, diffusion is very fast in metals. Jump times shorter than 10^{-7} s are observed at room temperature for most metals, and for some of them even much further down in temperature. Muons in Cu, for instance, show a $\sigma(T)$ curve of the Fig. 3 type for temperatures between 5 and 250 K, and one can evaluate the rate $1/\tau_c$ as a function of temperature in the interval 100-250 K and fit it with an Arrhenius law,

$$\frac{1}{\tau_c} = \frac{1}{\tau_0} \exp (-E_a/kT) . \quad (9)$$

For this example the muon activation energy E_a comes out around 50 meV ²³⁾, whereas it is 400 meV for protons in Cu (studied in the temperature range 500-1000 K). The pre-exponential factor $1/\tau_0$ is $10^{7.5} \text{ s}^{-1}$ for muons, but 10^{12} s^{-1} for the protons, in the respective temperature ranges. The low pre-exponential factor has been interpreted by several authors as a result of strong sub-barrier diffusion for muons, which have high zero-point energies in the interstitial potential wells. For phonon-assisted tunnelling, which is an incoherent tunnelling process, one expects a relation derived by Holstein²⁴⁾ and Flynn and Stoneham²⁵⁾:

$$\frac{1}{\tau_0} = \left(\frac{\pi}{4\hbar^2 E_a kT} \right)^{\frac{1}{2}} J^2 . \quad (10)$$

This equation has later been provided with correction terms for intermediate temperatures by Teichler²⁶⁾. The transfer matrix element J comes out for muons in Cu as 32 μeV . Similar derivations of diffusion parameters for muons in other metals have appeared in the literature²⁷⁾, but as more and more experimental information has been collected it has become increasingly doubtful whether these numbers are really representative for the pure metal $\rightarrow$ muon interactions.

Starting with *the b.c.c. metals*, we can briefly recapitulate the development in Nb. Figure 12 shows the damping parameter Λ versus T for Nb of three different purities: a) contains 3700 ppm N, b) ≈ 60 ppm N, and c) 15 ± 5 ppm N; in addition, all of these samples contain some C, O, and about 100 ppm substitutional impurities, mainly Ta²⁸⁾. Curve (a) is confusingly similar to the "textbook" case of Fig. 3, while (b) and (c) show a complicated structure indicating several stages in the diffusion, detectable on the microsecond time-scale. As shown by the fits, and a detailed theoretical treatment by Kehr, Richter and Honig²⁹⁾, these experimental data can be interpreted in terms of a two-stage model of diffusion involving

- i) a jump probability $1/\tau$ for muons starting at low temperature, < 15 K;
- ii) a characteristic rate (concentration dependent) $1/\tau_1$ for muons to reach traps, probably associated with the N-atoms;
- iii) a rate $1/\tau_0$ for leaving these traps.

Each of these processes dominates in a specific temperature range, and it was possible to derive activation energies for the "starting positions" and for the traps. The E_a for muons in N-associated traps turned out to be 48 meV, as compared with 167 meV for hydrogen in a similar situation studied by neutron scattering³⁰⁾.

More detailed discussions of trapping will be given in the accompanying paper by Herlach³¹⁾. Here, we will indicate the extreme sensitivity of muon diffusion to impurities by showing the results for the purest Nb obtained so far (Fig. 13), with interstitial impurities between 1-2 ppm and substitutional ones < 3 ppm. In this Nb sample, muons seem to be more or less mobile down to $T = 6$ K³²⁾.

A parallel investigation was carried out on b.c.c. Fe, where the higher sensitivity to high-frequency jumps makes it possible to follow the damping factor [in this case λ , see Eq. (2)] between $\lambda = 10$ and $\lambda = 0.01$. Arrhenius laws are observed in the temperature range 50-300 K, with somewhat different parameters depending on sample purity. Fits with small-polaron theory diffusion (Flynn-Stoneham) give tunnelling matrix elements of the order of a few meV and activation energies of the order of 100 meV ³³⁾.

It is evident that the tunnelling probability is very large in the b.c.c. metals, which may be traced back to the low barriers and short distances between tetrahedral sites in these structures. An intriguing fact is that for muons in Fe, below 40 K the λ -values are almost constant down to 10 K, which cannot be explained by small-polaron diffusion. Other mechanisms must therefore be invoked, and the possibility of coherent motion with the development of band-like states ³⁴⁾ will be discussed later.

For *f.c.c. metals*, we have already mentioned the case of Cu, which seems "normal" in the 5-250 K range. Very recent investigations down to 0.05 K, using dilution refrigerators ²³⁾, showed however an unexpected decrease of σ below 2 K (Fig. 14). This behaviour remains unexplained at the moment, but a development of the point-like muonic states towards more extended ones at low T has been suggested. This extension may still be limited by inhomogeneities due to remaining impurities in the Cu samples used.

The greatest number of investigations of impurity effects have been made on Al-metal. Here it has been known for some time that muons are mobile down to 2 K even in material of modest purity (~ 5 N). A recent investigation ²³⁾ shows that the damping parameter remains small down to

0.03 K, with a value corresponding to jump rates of the order of 10^7 s^{-1} (if a jump model should apply). The situation in Al-metal can hardly be explained in terms of a polaron theory, which predicts a T^7 behaviour at low T for the jump rate and would therefore require unrealistically large tunnelling matrix elements.

In the absence of multi-phonon processes, it remains for us to look for one-phonon processes, which could occur in the presence of strains in the lattice and have a linear T-dependence [Rollefson³⁵], or for a completely coherent process without phonon interaction. Since even the one-phonon expression leads to inconsistencies below 1 K for the purest samples, we will consider in more detail the conditions for the coherent motion.

We should keep in mind that when the muon is at equilibrium in an interstice, it produces a lattice distortion around it, which lowers the total energy. A shift of the particle to the next interstice means shifting the whole configuration one step. For the muon to tunnel to the next position, the matrix element J of the simple "undistorted" overlap is therefore reduced to $J_{\text{eff}} = J \exp(-s)$, where the low-temperature limit for $\exp(-s)$ can be evaluated using a Debye model³⁶). The quantity J_{eff} is expected to be of the order of a few μeV .

Coherent motion is equivalent to a spreading of the muon wave function over a certain region of space. It is expected that the coherent process should be very easily destroyed by perturbation of the energy levels at adjacent sites. The Anderson theory of localization says that a delocalized state can develop only if

$$J_{\text{eff}} \gtrsim W, \quad (11)$$

where W is the spread of energy levels.

Since Al-metal is a suitable test case for quantum diffusion, some investigations have been made on impurity-doped material, with concentrations of Mn, Mg, Zn, and Cu ³⁷⁾ in the range 10-1000 ppm. Mn-doping shows one of the most dramatic effects. The first experiments on $\text{AlMn}_{1300 \text{ ppm}}$ ⁷⁾ showed a peak in $\sigma(T)$ around 15 K, corresponding approximately to point-like muons at fixed positions in a pure Al lattice. The symmetry of the muonic sites was found to be tetrahedral (experiment in Fig. 8b). Later experiments have explored the temperature range down to 0.1 K for two Mn concentrations, 1300 ppm and 42 ppm ³⁴⁾, as shown in Fig. 15. The present indications are that trapping occurs in the 10-20 K range in a relatively wide sphere around each Mn impurity, as in the case of doped Nb, and that these traps are reached in the low-concentration sample after a relatively fast diffusion from initially random positions at this temperature. A trapping model for the 42 ppm alloy and a few other dilute impurity systems is the subject of one of the contributions to this conference ³⁸⁾. At the lowest temperatures, where there seems to be no other mechanism for diffusion other than the coherent one, we see a gradual increase of σ from 0.05 at the purest Al (a few ppm impurities) to ~ 0.15 at 42 ppm Mn and ~ 0.22 at 1300 ppm Mn. Furthermore, for lower temperatures there is a tendency towards octahedral site occupation.

There are therefore strong indications that the low-temperature phenomena are connected with the break-down of coherent propagation (band-state formation) when going from pure to impure samples. An order of magnitude estimate of the disorder parameter W can be made, assuming that the energy spread is due to elastic energy differences among adjacent muon sites induced by the impurities.

The elastic energy decreases approximately as r^{-3} when going outward, according to a formula given by Leibfried ³⁹⁾,

$$E_{\text{int}} = - \frac{1}{r^3} \frac{15}{18\pi} d \left(\frac{\overline{C_{11}} + 2\overline{C_{12}}}{3\overline{C_{11}}} \right)^2 \Delta V^{\mu} \Delta V^{\text{Mn}} \left| \frac{3}{5} - \sum_j \rho_j^4 \right|, \quad (12)$$

where $d = C_{11} - C_{12} - C_{44}$ is the elastic anisotropy of the lattice; $\overline{C_{12}} = C_{12} + 1/5 d$ and $\overline{C_{11}} = C_{12} + 2C_{44} + 2/5 d$ are averaged elastic constants; ΔV^{μ} and ΔV^{Mn} are volume expansions induced by the defects; and ρ_j are the direction cosines with respect to the cubic axes.

Introducing empirical values for the constants, it is found that the energy change between adjacent sites at a distance Δr can be written

$$\Delta E_{\text{int}} = \frac{dE_{\text{int}}}{dr} \Delta r = 3 \times 0.39 \Delta r / r^4 \text{ eV}. \quad (13)$$

For the 1300 ppm AlMn sample referred to in Fig. 8b, one can estimate a value of $\Delta E_{\text{int}} \approx 70 \mu\text{eV}$, and for 42 ppm around $1 \mu\text{eV}$. The corresponding quantity for 6N-purity Al should be of the order of $0.01 \mu\text{eV}$.

It seems reasonable that the Anderson localization condition is fulfilled for AlMn_{0.001} but not for 6N Al, since

$$\Delta E(\text{Al}) < J_{\text{eff}} < \Delta E(\text{Al Mn}_{0.001}) \quad (14)$$

if J_{eff} is of the order of $1 \mu\text{eV}$. The case of the 42 ppm Mn alloys is an intermediate one where muon delocalization may occur in certain regions of the crystal, leading to the observed values of σ . It should be pointed out that already a spreading out of the muon wave function over a few sites gives an appreciable reduction of σ . Similar considerations might apply to the low-T situation for muons in Cu: here the strain effects would be even more critical since the high-T data indicate a J_{eff} of the order of $0.01 \mu\text{eV}$.

It seems that the positive muon is a particle which can be put into environments where it is unstable against quantum mechanical delocalization. Its behaviour is therefore of interest in the wider perspective

of delocalization in disordered systems in general. As we all know, these topics are being studied intensely at present as far as electron transport is concerned. Also numerical models for disorder, giving detailed localization conditions, begin to appear in the literature.

Several open questions still remain in this area, such as

- 1) If coherent motion exists at low T , it will evidently be destroyed by thermal motion at some transition temperature T_{tr} and go over into the incoherent regime. A first theoretical estimate of this temperature has been given by Kehr⁴⁰⁾
- 2) The potential wells for the muons (including the lattice distortion in the self-trapped state), activation energies, and tunnelling matrix elements should in principle be calculable from an electron theory of the muon-near-neighbour system, which forms a connection to the observables discussed in Section 2. Such calculations have been performed for muons and hydrogen in Cu with encouraging results [Teichler¹⁶⁾] but not yet for other metals.
- 3) The question has been raised, whether muons implanted in metals reach the self-trapped state only after passing a barrier, which might require the presence of impurities [Browne and Stoneham⁴¹⁾].

For further reading about muons in solid-state physics and chemistry, the following books are recommended: 1) Muon Spin Rotation, Volume 6 of Hyperfine Interactions (1979) (North-Holland Publishing Co., Amsterdam, 1979), and 2) Muon Physics III, Chemistry and Solids (eds. V.W. Hughes and C.S. Wu, Academic Press, New York, 1975).

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Table 1

Theoretical damping parameters σ (μs^{-1}) for point-like muons in some cubic metals

Host metal	Site	Field direction		
		[100]	[111]	[110]
Cu (f.c.c.)	octahedral	0.307	0.067	0.164
	tetrahedral	0.077	0.319	0.279
Nb (b.c.c.)	octahedral	0.555*	0.208	0.331*
	tetrahedral	0.404*	0.326	0.347*
		* averages over two magnetically inequivalent sites		

Table 2

Hyperfine fields (in kG)

	B_c (exp) ref. 18	B_c (theory)			
		ref. 12	ref. 14	ref. 20 ^{b)}	ref. 15 ^{c)}
Ni	-0.688	-0.66 ^{a)} -6.9	-0.924	-0.72	-0.59
Co	-6.1	-1.58 -13.4			-5.7
Fe	-11.1	-1.06 -9.4			-10.1
Gd	-7.3	-2.87 -24.4			-8.7

a) For $\eta = 0$.

b) *Ab initio* muffin-tin potentials.

c) Cluster calculation.

Figure captions

- Fig. 1 : Principles of μ SR. The pion decay in flight leaves the muons in a state of high polarization.
- Fig. 2 : Example of time histogram showing muon precession in a transverse field.
- Fig. 3 : Expected temperature dependence of the μ SR damping factor (or line-width).
- Fig. 4 : Zero-field μ SR in a ferromagnet (Fe at 19 K). The damping at this temperature is fairly strong and only a few periods are seen clearly.
- Fig. 5 : a) The theoretical longitudinal relaxation factor $G_z(t)$ as expected for zero external field and the transverse relaxation factor $G_x(t)$ for strong external fields.
b) Observed functions $G_z(t)$ and $G_x(t)$ for muons in ZrH_2 (ref. 8).
- Fig. 6 : The EFG produced by a muon at an interstitial site.
- Fig. 7 : Interstitial sites available for muons in b.c.c. and f.c.c. lattices.
- Fig. 8 : Fits to experimental magnetic field dependence for
a) Cu-metal (O-sites) (ref. 6);
b) Mn-doped Al-metal at 15 K (T-sites) (ref. 7).
- Fig. 9 : Muonic Knight shifts for a number of simple metals (ref. 17).
The theory 1 refers to ref. 12, theories 2 and 3 to ref. 13, theory 4 to ref. 42 and theory 5 to ref. 43.
- Fig. 10 : Temperature dependence of the hyperfine field $B_{hf} = B_c$ as expressed through the parameter $A(T)$.

- Fig. 11 : Pressure variation of the precession frequency in Fe and Ni at room temperature.
- Fig. 12 : Temperature variation of the damping parameter Λ for muons in Nb of different purities (see text).
- Fig. 13 : The damping parameter for muons in the purest Nb available so far (ref. 32).
- Fig. 14 : Damping parameters σ observed at low temperatures for muons in Cu and Al.
- Fig. 15 : Low-temperature investigation of the effect of Mn-doping on muon mobility in Al.

a) Pions decaying at rest.

Momenta (p) and spins (S)
for muon (μ), neutrino (ν) and positron (e^+)

b) Pions decaying in flight.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

a)

b)

Fig. 5

Fig. 6

Fig. 7

Fig. 8

Fig. 9

Fig. 10

Fig. 11

Fig. 12

Fig. 13

Fig. 14

Fig. 15