of surface probes, especially when defects are associated with the surface region. An interesting possibility is in the study of two-dimensional melting where defects are predicted to play an important role.

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Quantum Diffusion of Positive Muons in Iron

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Muon spin precession in iron has been observed from 300 K down to 3 K. The relaxation rate λ shows a pronounced temperature dependence with minima at 15 and 90 K. A fit with an Arrhenius function yields $E_a = 0.3$ meV below 10 K, 14 meV between 35 and 90 K, and 86 meV between 150 and 300 K. The data indicate that coherent tunneling becomes important below 35 K. The temperature region between 35 and 90 K is dominated by incoherent tunneling while above 150 K the over-barrier hopping process sets in.

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In many respects, the positive muon can be considered as a light isotope of hydrogen; in particular, its diffusion behavior should follow that of hydrogen, the main difference arising from a much smaller mass ($m_\mu = \frac{1}{9}m_p$). This difference is rather unimportant at high temperatures where only moderate isotope dependences are expected but becomes crucial at low temperatures when tunneling plays the dominant role in the diffusion process. In the present Letter, detailed experimental data are presented which provide evidence for coherent as well as incoherent tunneling at low temperatures. The transition between the two mechanisms is around 35 K.

Muon diffusion can be measured by the motional-narrowing effect in a muon-spin-rotation (MSR) experiment. In spite of several publications on muon diffusion in iron,[1-5] the experimental situation is unsatisfactory, in particular in the low-temperature region where the occurrence of the most interesting effects is expected. Because of the fast relaxation of the MSR signal no results exist below 7 K and the data between 7 and 50 K have rather poor accuracy. By using a fairly clean and perfect crystal we were able to extend the experimental data range down to 3 K and to improve the accuracy for the points at higher temperatures.

The MSR experiment was performed at the superconducting muon channel at Schweizerisches Institut für Nuklearforschung (SIN) in zero magnetic field with a coincidence counting rate of about 3 kHz. The iron target consisted of seven pieces of single-crystal iron plates, each with an area of 1 cm^2 and 1.8 mm thickness. The plates were assembled to a compact target with the surface [(100) plane] perpendicular to the muon beam. The main impurities in the sample were 6.5 ppm (wt.) Si, 8.5 ppm (wt.) Al and less than 6 ppm (wt.) Ni, all other contaminations being <1 ppm (wt.). The targets of the present experiments were prepared under the same conditions as those used in Ref. 4.

If the muon is slowed down in the target, it will finally come to rest at a tetrahedral or octahedral interstitial site in the bcc lattice of iron. Seeger[6] and Nishida *et al.*[2] have pointed out that the sign of the experimental hyperfine field favors the assumption of tetrahedral occupancy. Therefore, in the following only the tetrahedral positions will be considered.

In zero external field, the local magnetic field at the muon site is given by $B_\mu = B_L + B_{hf} + B_{dip}$, where B_L, B_{hf}, and B_{dip} are the Lorentz, hyperfine, and dipolar field, respectively. The dipolar field at the tetrahedral interstitial sites has different values depending on the relative orientation of the tetragonal axis with respect to the magnetization direction. The calculated values for parallel and perpendicular orientations are -0.52 and $+0.26$ T, respectively. The weighted average of the dipolar fields is zero. In spite of two different dipolar fields at the tetrahedral sites, only one precession signal is observed in the MSR experiment. This result is interpreted as evidence for diffusional averaging of the muon over the dipolar fields.

The measured frequency ν_μ (Fig. 1) shows a smooth decrease between 15 and 300 K, the relative temperature dependence being somewhat larger than that of the macroscopic magnetization. Below 15 K the frequency increases faster than expected from an extrapolation from higher temperatures. At present, no explanation can be given for this deviation.

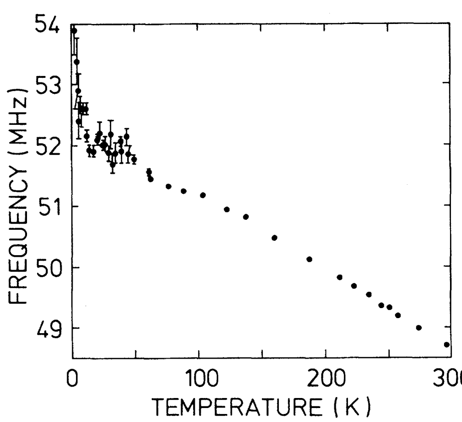

FIG. 1. Temperature dependence of the muon-spin-rotation frequency in zero external field.

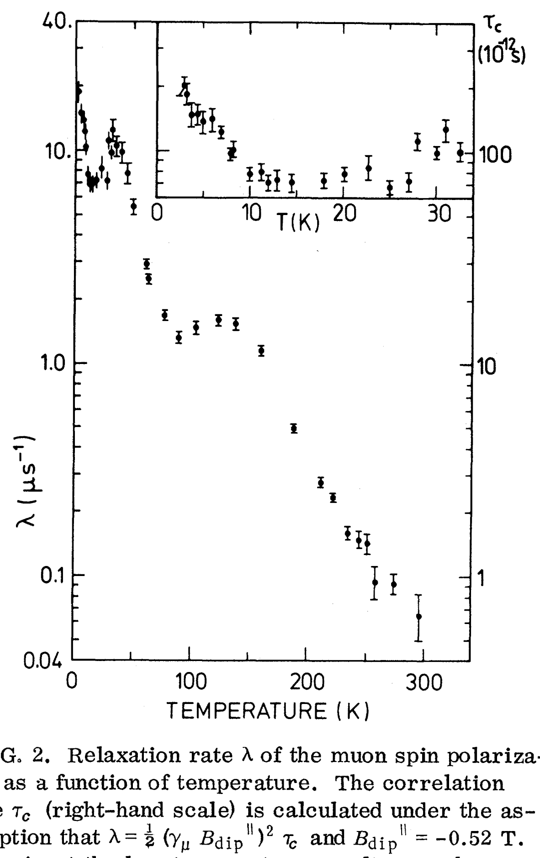

FIG. 2. Relaxation rate λ of the muon spin polarization as a function of temperature. The correlation time τ_c (right-hand scale) is calculated under the assumption that $\lambda = \frac{1}{2}(\gamma_\mu B_{dip}^{\parallel})^2 \tau_c$ and $B_{dip}^{\parallel} = -0.52$ T. In the inset the low-temperature results are shown on an extended temperature scale. There additional data points are included.

The relaxation rate λ as a function of temperature (Fig. 2) exhibits a pronounced structure with minima at 15 and 90 K and three regions of strong decrease of λ with temperature. In the earlier experiments on muon diffusion in iron,[1-5] the structure of λ was not recognized with this clarity and therefore the deduced activation energies and the interpretation of the data differ considerably from the present results. The absolute values of λ in the present experiment are smaller than in most of the earlier publications,[1-3] but similar low relaxation rates were reported in Ref. 4 and 5. Small relaxation rates are obtained only if clean and perfect crystals are used.

The relaxation rate λ is connected with the correlation time τ_c by the relation (motional-narrowing limit)

$$\lambda = \gamma_\mu^2 \langle B_{\rm dip}^2 \rangle \tau_c , \qquad (1)$$

with

$$\gamma_\mu = 8.52 \times 10^8 \ {\rm s}^{-1}\,{\rm T}^{-1}, \qquad (2)$$

$$\langle B_{\rm dip}^2 \rangle = \tfrac{1}{3} B_{\rm dip}^{\parallel 2} + \tfrac{2}{3} B_{\rm dip}^{\perp 2}. \qquad (3)$$

The scale on the right-hand side of Fig. 2 is calculated on the basis of this formula.

The diffusion coefficient for muons jumping between tetrahedral interstitial sites, $D = a^2/72\tau_c$ (a being the lattice constant) is plotted in Fig. 3 versus reciprocal temperature. For comparison the hydrogen diffusion data are included (solid line), thereby taking the average value[7] of the widely scattered experimental data.

It is obvious from Fig. 2 and 3 that the temperature dependence of the relaxation rate (or equivalent of the diffusion coefficient) cannot be fitted by a single Arrhenius law. However, there are three regions with a strong decrease of λ, to which such a fit may be applied separately. The values $D_0 = (4.7 \pm 0.9) \times 10^{-4}$ cm^2/s and $E_a = 86 \pm 3$ meV obtained in region III agree remarkably well with those of hydrogen diffusion above 300 K [D_0(H) = 7.5×10^{-4} cm^2/s and E_a(H) = 105 meV[7]]. The close correspondence of the diffusion data (see also Fig. 3) suggests a common diffusion mechanism for the two particles. Since this mechanism apparently governs the high-temperature region, it may be identified with the over-barrier hopping process.

In contrast to region III, the muon data below 150 K are completely different from the expected behavior. The measured relaxation rates indicate an overall strong enhancement and a pronounced structure in the diffusion coefficient at

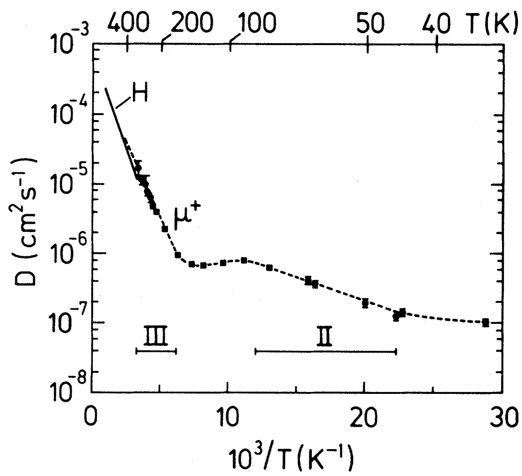

FIG. 3. Diffusion coefficient D for positive muons (μ^+) in iron vs the reciprocal temperature. D is derived from the relaxation rate λ, for diffusion over tetrahedral sites. Temperature regions II and III mentioned in the text are indicated. For comparison, the average hydrogen diffusion data (Ref. 7) are given (solid line).

low temperatures. In particular, the fact that only one frequency is observed (indicating motional averaging over dipolar fields) demonstrates that the muon is still highly mobile down to 3 K. The extrapolated correlation time τ_c below 15 K would be years, if the parameters from region III are used to calculate τ_c, while the present data require τ_c in the order of 10^{-10} s. This very strong enhancement of the muon diffusion is a clear evidence of an efficient tunneling process at low temperatures. At present the most interesting question is whether tunneling occurs via a coherent or incoherent process. In this respect it is of particular interest that the complex behavior of λ (Fig. 2) below 150 K cannot be explained by a single process but rather requires at least two different mechanism. This aspect will be stressed in the following discussion.

The data are first discussed in the framework of the incoherent small-polaron hopping model.[8-10] The basic idea of this model is that the muon is self-trapped because of lattice relaxations with the consequence that the ground-state levels of an occupied and unoccupied interstitial site are at different energies. In this situation the tunneling process requires the assistance of phonons to equalize the levels of the neighboring sites. Considering realistic self-trapping energies,[11] it is obvious that only region II (35–90 K) can be identified with the small-polaron hopping process.

The experimental activation energy in this region of 14± 1 meV is close to the expected values,[11] whereas $E_a = 0.3 \pm 0.03$ meV from region I (3–10 K) is clearly outside of realistic estimates.

The experimentally observed decrease of λ below 35 K (Fig. 2) is not understandable in the framework of the small-polaron hopping model but is expected if coherent tunneling is assumed. The formation of band states depends very critically on the periodicity of the lattice and therefore can be observed only in rather pure and perfect crystals. A rough estimate[12, 13] of whether coherent tunneling is possible in principle can be obtained from a comparison of the tunneling matrix element J with the static energy shift ΔE of adjacent sites due to impurities or defects. A calculation similar to that of Ref. 5 yields ΔE of the order of 0.1 μeV for positions midway between the impurities whereas an estimate of the tunneling matrix element from the decrease of λ in region II yields $J \approx 1$ meV. Thus, coherent tunneling should at least in principle be possible in large portions of the crystal.

Experimental evidence for coherent tunneling is provided by the decrease of λ between 35 and 20 K. This behavior, which corresponds to an increase of the diffusion coefficient with decreasing temperature, is a strong indication for coherent tunneling.

The new increase of λ with decreasing temperature below 15 K (Fig. 2) can be explained by an effect recently pointed out by Fujii[14]: In magnetic systems like Fe, the dipolar fields follow the periodicity of the lattice. This implies that each tunneling state sees a distinct but in general different average dipolar field. This results in a field distribution which causes a depolarization of the muon spin. In order to understand the structure in the temperature dependence of λ below 35 K it must be assumed that motional narrowing between tunneling states plays a role too. Again, in view of relation (1), the decrease of λ between 35 and 20 K is caused by the strong reduction of $\langle B_{\rm dip}^2 \rangle$ when tunneling states develop. However, at even lower temperatures the increase of the correlation time becomes larger than the reduction of $\langle B_{\rm dip}^2 \rangle$, resulting in an increase of λ.

In the present experiment muon diffusion in iron was measured over a large dynamical range from 3 to 300 K. The data indicate that probably all basic diffusion processes, coherent tunneling (3–35 K), incoherent tunneling (35–90 K) and over-barrier hopping (above 150 K) were observed. The diffusion parameters (J, E_a, D_0) obtained have reasonable values and are consistent with theoretical estimates of these quantities.

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