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MUON SPIN-ROTATION STUDY ON MAGNETITE*

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ABSTRACT

Muon spin-rotation (μ SR) results on synthetic single crystals of magnetite (Fe_3O_4) support the idea of muon bond formation in oxides. The anomaly in the temperature dependence of the μ SR signal observed in Fe_3O_4 may be attributed to the existence of molecular polarons in the Verwey transition-temperature region.

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The muon spin rotation (μ SR) technique has been used to investigate the well known ferrimagnet Fe_3O_4 (magnetite). At the Verwey temperature ($T_V=121$ K) magnetite shows a metal-insulator transition. This phase transition has a complex nature and shows electronic as well as lattice features^{1,2,3}.

This investigation serves two purposes. Our first aim is to understand muon behaviour in magnetic oxides, particularly the muon trapping site(s), the electronic structure, and the local magnetic field B_μ at the muon site. Secondly, these μ SR measurements might reveal information about solid state effects occurring in the interstitial region. Magnetite is well suited for these purposes. More detailed information on this study will appear in a forthcoming paper.

In a μ SR experiment⁴ one measures the muon spin polarization at the time of decay of the muon. Superimposed on the muon decay pattern one observes the Larmor precession signal, due to B_μ . The μ SR signal characteristics - the Larmor frequency, the asymmetry and the depolarization rate - provide information about the surroundings of the muon site.

In the magnetic oxides the muon samples its environment through both dipole-dipole interactions and covalent (electron transfer and overlap) effects⁵. μ SR studies⁵ in antiferromagnetic oxides have shown the localization of the muon. An interpretation of the muon behaviour in these oxides can be sought in terms of muon bond formation which is analogous to hydrogen bonding⁵.

Above and below T_V μ SR signals have been observed in synthetic and natural single crystals at zero applied field². At room temperature the observed μ SR frequency corresponds to an internal field of 4 kG. External transverse field measurements were performed at room temperature at the stopped muon channel of the Clinton P. Anderson Meson Physics Facility. The magnetic field, perpendicular to the initial muon spin polarization, was applied along the $\langle 111 \rangle$, $\langle 110 \rangle$ and $\langle 100 \rangle$ crystal directions. Figure 1 depicts the field dependence of the observed μ SR signals in Fe_3O_4 for the case $B_{\text{ext}} // \langle 110 \rangle$. As can be seen, fields much higher than the demagnetization field cause a splitting of the signal. This splitting appears to be orientation dependent and maximum for the $\langle 110 \rangle$ direction.

The field dependent data, neglecting the splitting, can be described by⁶

$$B_{\text{eff}} = B_{\text{ext}} + B_{\text{dem}} + B_{\text{int}} \quad (1)$$

In general the demagnetization field B_{dem} and the internal field B_{int} depend upon the angle between the external field B_{ext} and the $\langle 111 \rangle$ -axis, the easy magnetization axis. The observed demagnetization fields on the order of 1 kG for the samples investigated are in reasonable agreement with the calculated values for the sample geometry. In the fitting procedure, known values for K_1 , the anisotropy constant, and M_S , the saturation magnetization, have been used. The direction of the internal field in magnetite is found to be along the direction of the magnetic moments of the iron ions. At zero applied field B_{int} is parallel to the $\langle 111 \rangle$ direction. With this information it is possible to locate the muon stopping site. Calculations to perform this search, taking into account dipole and covalent contributions to the local field, are in progress.

The splitting occurring at higher applied fields deserves some extra discussion. This kind of effect has also been observed in the μ SR spectra of the orthoferrites⁷. Here the split signals can be described by (1), or, less likely, explained by assuming a distortion of the iron spin system in a large magnetic field. However for magnetite the upper signals are about 3 % larger than the maximum possible vectorial sum (1). This means a field enhancement effect occurs.

Such magnetic (anti-) shielding effects are known from proton NMR⁸. Assuming muon bond formation the observed splitting (see fig. 1) can be due to differences in magnetic shielding of the electron cloud around the muon which is located at different sites. Conversely, the observed splitting strongly indicates the occurrence of muon bond formation.

As reported earlier², two jumps in the μ SR frequency, of about equal magnitude but opposite sign, occur at T_V and 245 K. New measurements on synthetic crystals, yielding much stronger signals, agree with these results but there are strong indications of a splitting below T_V . Below 90 K no μ SR signals are observed, which may be due to the different magnetic surroundings of the muon sites.

The observed extra transition at 245 K can be due to either a change in the muon state or a magnetite effect. The former has also been observed in other magnetic oxides^{5,7}. However the characteristics of these changes are very different from what has been seen in magnetite.

The latter possibility can be investigated by studying titanomagnetite (Fe_2TiO_4). Because of the nearly identical structure, in which the Ti ions substitute for the iron ions at the B-sites in the spinel

structure, the same muon behaviour and similar stopping sites can be expected. Below 140 K Fe_2TiO_4 is an antiferromagnet. Preliminary transverse μSR measurements show no drastic changes in the depolarization rate in a temperature range around 245 K. Further comparative measurements in both oxides will be performed in an attempt to show that the transition at 245 K is a solid state effect, presumably a precursor of the Verwey transition.

In this context it is important to emphasize the fact that various experimental studies^{1,2} have indicated that in the temperature range from 100 K to 250 K at least two transitions occur in magnetite. These effects can be due to some kind of charge fluctuation or localization, associated with local lattice changes, for instance oxygen displacements. It appears that the existence of a coupled charge density-lattice distortion complex involving several ions, as suggested by Yamada et al.³ may serve as a basis of an interpretation for these anomalies. These 'molecular polarons' may be the reason for the observed shift in μSR frequency below 245 K. The shift can be due to oxygen displacements in the unit, causing muon displacements, which are seen as a change in the hyperfine interactions.

In conclusion, this μSR investigation on magnetite has shown that muon bond formation is an important ingredient in the interpretation of μSR studies on magnetic oxides. The possible occurrence of 'molecular polarons' may explain the temperature dependence of the μSR frequency signal observed in magnetite above the Verwey transition.

REFERENCES

1. Proc. Int. Mtg. on Magnetite and other materials,
showing a Verwey transition, Cambridge (1979),
published in Phil. Mag. B42 (1980)
2. C. Boekema, Phil. Mag. B42 (1980) 409
3. Y. Yamada et al., Solid State Comm. 32 (1979) 827
4. J. Brewer and K. Crowe, Ann. Rev. Nucl. and Part. Sci. 28 (1978) 239
5. C. Boekema et al. Hyperfine Interactions 8 (1981) 609
and references therein
6. P. F. Meier, Hyperfine Interactions 8 (1981) 591
7. E. Holzschuh et al. Hyperfine Interactions 8 (1980) 77
8. U. Haeblerlen et al. Chemical Physics 3 (1974) 248

FIGURE CAPTION

Fig. 1. Field dependence of the observed μ SR signal in magnetite at room temperature. The external field is applied parallel to the $\langle 110 \rangle$ axis. The solid line is a fit to the data, neglecting the splitting. The splitting suggests magnetic shielding effects in the muon bond.