MUON LEVEL-CROSSING RESONANCE IN MAGNETICALLY ORDERED MATERIALS

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The positive muon is widely used as a microscopic probe of internal fields at interstitial sites in magnetically ordered materials. Recently, we have demonstrated that the hyperfine fields on the neighboring host nuclear spins can be measured using a novel muon level-crossing resonance technique, thus providing a more detailed picture of the electronic and magnetic environment around the muon. In this paper I will describe the fundamentals of muon level-crossing resonance as applied to magnetically ordered materials, and report an example in MnF₂.

1. Introduction

The technique of positive muon spin rotation has recently gained considerable recognition for its usefulness as a probe of magnetism. For example, the recent discovery of magnetic ordering in YBa₂Cu₃Oₓ (x < 6.4) [1,2] and the subsequent determination of the phase diagrams in YBa₂Cu₃Oₓ [3] and La₁₋ₓSrₓCuO₄ [4–6] as functions of x were made using positive muons to detect the spontaneous internal magnetic fields in the ordered state. In such studies the positive muon serves as a point-like probe of internal fields at the interstitial site where the muon localizes. This technique has several advantages over more conventional magnetic resonance techniques in that:

1. The muon is a spin 1/2 particle and thus is a pure magnetic probe since there is no muon quadrupolar interaction.

2. The muon is an implanted probe. For this reason and the nature of muon decay it is virtually guaranteed to give a signal in any sample, whether it be a single crystal, polycrystal or glass, and under almost any experimental condition (temperature, pressure, or magnetic field, including zero-field).

3. By virtue of parity non-conservation of weak interactions the muons are created with 100% spin polarization from pion decay (π⁺ → μ⁺ + νμ) and also the time evolution of the muon polarization is observable through muon decay (μ⁺ → e⁺ + νe + ¯νμ) (the positron is emitted preferentially along the polarization direction). Consequently the technique obviates the need for radio frequency (RF) fields which is particularly useful in conductive samples.

The μSR technique opens a time window, corresponding to fluctuation rates in the internal field in the range 10⁻⁶–10⁻¹² s⁻¹ [7], which is unique. However,
conventional μSR provides rather limited information on static magnetic environment of the host compared to neutron scattering, NMR or NQR.

It would therefore be desirable to somehow use the muon to probe internal fields of the neighboring host nuclear spins, following the example of nuclear double resonance techniques. Muon-nuclear double resonance techniques are now possible using RF fields [8] but the method is technically very difficult because the muon lifetime is short compared to the time it takes to precess a nuclear spin with reasonable RF fields. In addition, this sacrifices one of the main advantages of μSR. In special cases (for example MnSi), it has been possible to measure nuclear spin relaxation rates using conventional μSR techniques [9]. Abragam was the first to suggest how the muon could be used to measure energy splittings in the surrounding host medium by the use of level-crossing resonance [10]. This is a technique for which the muon is ideally suited. Experiments on muons in copper [11], muonium substituted free radicals [12–15], muonium defect centers in semiconductors [16,17], and most recently the antiferromagnet MnF₂ [18] have demonstrated clearly that muon level-crossing resonance is a powerful probe of the hyperfine or quadrupolar interaction of neighboring nuclear spins. A typical μLCR spectra for a muonium substituted free radical in the liquid phase is shown in fig. 1. The information gained from such spectra exceeds that of ESR and ENDOR since both the magnitude and relative signs of the hyperfine parameters are obtained in lowest order.

![Fig. 1. The μLCR spectrum for the 13C enriched 13C₆H₆Mu free radical taken from ref. [15]. Three additional proton resonances were also observed but are off scale. The numbers in brackets refer to the ring positions ending where the muonium adds.](image-url)