

Supplementary Material (ESI) for Chemical Communications

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Description of the ZF- μ^+ SR Technique

μ^+ SR in its various forms (ZF, LF, TF) is a powerful probe of the magnetic properties of materials.⁵ The 100% spin-polarised μ^+ beam is stopped in the sample and the time histograms of μ^+ decay positrons are recorded by forward (F) and backward (B) counters as a function of time. Since a positron is emitted preferentially towards the μ^+ spin direction, the time-dependent asymmetry $A(t) = (F - B)/(F + B)$ reflects the time evolution of the μ^+ spin polarisation. After coming to rest at interstitial sites of the solid, the μ^+ precess in the local magnetic fields $\langle B_{\mu} \rangle$ with a precession frequency, $\nu_{\mu} = (\gamma_{\mu}/2\pi)\langle B_{\mu} \rangle$, where $\gamma_{\mu}/2\pi = 13.55$ kHz/G. In the absence of an applied field, a precession signals the onset of an ordering (FM or AF) transition. Application of a longitudinal magnetic field (LF) allows the decoupling of the μ^+ spin from the static internal fields. Although for powder samples such as the present fulleride, the exact μ^+ site is not known, it is expected that the μ^+ will reside near the negatively charged C_{60}^{3-} units.