μSR Studies of Magnetism in κ-(BEDT-TTF)$_2$X

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The magnetism in the κ-(BEDT-TTF)$_2$X family of charge-transfer salts ranges from quantum spin liquid (QSL) states to weakly canted antiferromagnetic (AF) order. We study the local magnetic properties using implanted muon spectroscopy in conjunction with detailed density functional theory (DFT) calculations. From the DFT we establish molecular spin distributions, muon stopping sites and dipolar field parameters, allowing us to make a quantitative interpretation of the experimental results. We focus on X=Cu[N(CN)$_2$]Cl, which has an AF ordering transition with T_N in the region of 27 K. The DFT-calculated spin density distribution for the BEDT-TTF dimer is used to simulate the 3.7 T ^{1}H NMR spectrum of the Cu[N(CN)$_2$]Cl salt [1], finding good agreement with experiment for ferromagnetic (FM), rather than AF, interlayer ordering and an ordered moment per dimer of 0.46 μ_B. DFT is also used to explore muon stopping sites for this salt, finding one group of sites associated with muonium addition to C=C double bonds in the BEDT-TTF layer and another set of sites associated with muons stopping in the anion layer. We compute dipolar fields associated with each of the stopping sites and compare them with the precession frequencies observed in the ZF-μSR spectrum [2]. We find best match for an ordered moment of 0.52 μ_B and interlayer ordering under ZF having AF character. New measurements of TF-μSR spectra for fields up to 8 T are reported and analysed to obtain the best estimate of the magnetic order parameter under different measurement conditions, allowing us to observe the variation of transition temperature and canting of the moments with applied field. Implications are discussed for the interpretation of the μSR data of the QSL materials with X=Ag$_2$(CN)$_3$ and X=Cu$_2$(CN)$_3$.

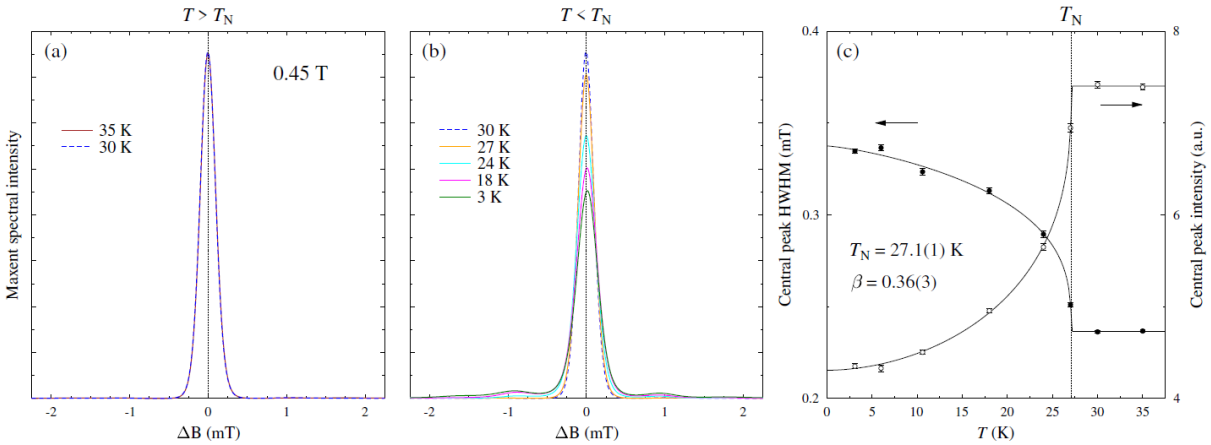

Fig. 1. TF-μSR spectra for κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl at 0.45 T.

References

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