Poster

HOW WELL ALIGNED ARE THE MAGNETIC ANISOTROPY AXES IN CRYSTALS OF MN_{12} MOLECULAR NANOMAGNETS? AN ANGLE-DEPENDENT AC SUSCEPTIBILITY STUDY

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Molecular clusters like Mn_{12} and Fe_8 are seen as the lowest limit of miniaturization of magnetic nanoparticles. Indeed, these clusters have a large ground-state spin ($S = 10$ in Mn_{12}) and they show magnetic hysteresis below liquid Helium temperatures, much as larger nanoparticles do [1]. The hysteresis phenomenon is a property of the isolated molecule (it is preserved in solution). It is caused by the large magnetic anisotropy energy barriers that separate spin-up and spin-down states of the whole molecule. In contrast to other nanostructured magnetic materials, single molecule magnets (SMM) are monodisperse and tend to form crystals in which the magnetic anisotropy axes are very well aligned. They offer then the possibility of investigating the Physics of *individual* molecules with conventional experimental techniques. These studies reveal the existence of quantum phenomena, like quantum tunnelling [2], which are typical of the atomic world.

Despite the intense research activity devoted to investigate these phenomena, the origin of quantum tunnelling in Mn_{12} and other SMM remains obscure. In recent years, it has been suggested that the *off-diagonal* energy terms that induce quantum spin tunnelling are caused by small distortions of the molecular core from its perfect structure [3]. In some compounds, like Mn_{12} acetate, the origin of these distortions has been associated with an intrinsic disorder in the position of solvent molecules [4]. The disorder would then tend to tilt the anisotropy axes of the molecules from their perfect alignment.

In the present contribution, we report a careful study of the degree of alignment of the magnetic anisotropy axes in crystals of Mn_{12} SMM. For this, we have made use of the extreme sensitivity that the linear susceptibility has on the angle that the applied ac magnetic field makes with the anisotropy axis. We have measured the frequency-dependent linear susceptibility (see Fig. 1) of high-quality single crystals of two Mn_{12} derivatives: Mn_{12} acetate and Mn_{12}-tBu. In the latter, the position of solvent molecules is extremely well ordered [5], which therefore makes for an interesting comparison with the acetate derivative. By adequately selecting the temperature, we have also been able to separately estimate the degree of orientation for the so-called "fast-relaxing" Mn_{12} species, a fraction of the SMM in the crystal that show faster spin tunnelling than the standard clusters.

Our results (see Figs 1 and 2) show: a) that the tunnelling rate is faster for the better ordered Mn12-tBu compound, and b) that the angular variation of is very similar in the two compounds investigated. The susceptibility data are in fact compatible with a perfect alignment of the anisotropy axes of the two molecular species within each crystal, with a maximum possible misalignment smaller than 3 degrees. These results therefore question the importance of solvent disorder in determining the tunnelling rates of Mn_{12} molecular nanomagnets.

References:

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Figures:

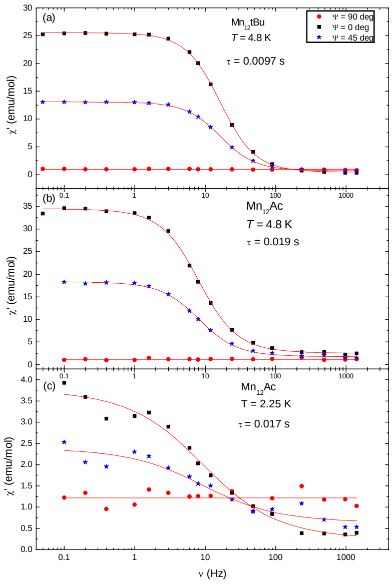

Figure 1: Frequency-dependent ac susceptibility of a single crystals of Mn_{12}-tBu (a) and Mn_{12} acetate (b and c). Solid lines are fits to a Cole-Cole function from which the tunneling time τ and the susceptibility drop Δχ between the low and high frequency limits are estimated.

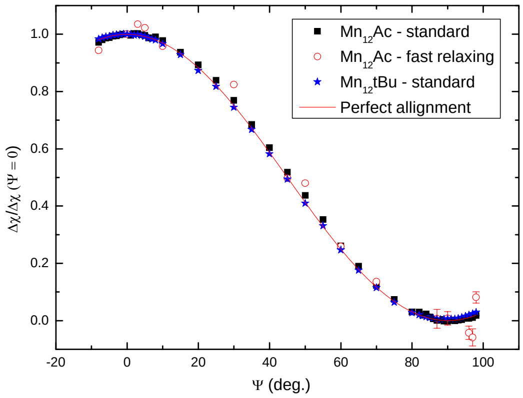

Figure 2: Angular dependence of Δχ (normalized by its value at Ψ = 0) for the slow and fast relaxing species in the two Mn_{12} derivatives investigated. Solid lines represent the predictions for different degrees of misalignment in the relative orientations of the magnetic anisotropy axes.