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Frequency Domain Magnetic Resonance Spectroscopy on Molecular Magnets¹

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We have advanced a novel technique of frequency domain magnetic resonance spectroscopy to investigate molecular magnets. It allows the extremely accurate determination of zero-field splitting spin Hamiltonian parameters without application of an external field. Extensive resonance lineshape studies give quantitative information on distributions within the sample. The resonance lineshapes in magnetized single-crystalline samples of Mn_{12}Ac in the blocked regime are very sensitive to the orientation of the sample with respect to the radiation propagation direction (Faraday and Voigt geometry). The asymmetric and double peak resonance lines found, could be explained by the magneto-optical properties of the material and could be simulated quantitatively. Circularly polarized radiation allows the selective excitation of $\Delta m = +1$ and $\Delta m = -1$ transitions. Finally we were able to spectroscopically observe the relaxation of the magnetization as well as quantum tunnelling of the magnetization directly. These relaxation studies using spectroscopic techniques can act as a much more local probe than magnetization measurements.

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