Resonant Microwave Power Absorption and Relaxation of the Energy Levels of the Molecular Nanomagnet Fe_8 Using High Frequency Superconducting Quantum Interference Device-Detected Electron Paramagnetic Resonance (SQUID-EPR)

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Energy levels and saturation of the molecular nanomagnet Fe_8 were investigated using a novel new approach to broadband high frequency electron paramagnetic resonance (HFEPR). This approach is based on dc detection of the suppression of the magnetic moment under resonant microwave irradiation. The change in magnetic moment is obtained by a superconducting quantum interference device (SQUID) magnetometer retrofitted with high frequency sources and waveguides. The technique provides quantitative determination of the EPR as a function of microwave power (up to 400 mW), frequency (up to 141 GHz), magnetic field (0 – 7 Tesla) and temperature (1.8 - 300 K). For both low spin S=_ $CuSO_4 \cdot 5 H_2O$ and high spin S=10 Fe_8 we suppressed up to 80% of the magnetization at liquid helium temperatures. The degree of suppression returns discrete values for the temperature and field dependence of the longitudinal relaxation time, T_1, over a wide range for S=1/2 systems. For the high spin S=10 Fe_8 system we found an anomalous large suppression of the magnetization. This indicates that resonantly pumping low lying energy levels effectively excites higher energy levels and demagnetizes the sample. Possible mechanisms such as resonant heating, cross-relaxation, and phonon blockades will be discussed.