

DIRECT TWO QUANTUM JUMPS IN HIGH FREQUENCY ELECTRON PARAMAGNETIC RESONANCE DESCRIBED BY A 3-LEVEL DENSITY MATRIX FORMALISM.

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High frequency EPR spectroscopy is a powerful tool for revealing the magnetic resonance properties of Nickel(II) complexes with an $S=1$ (triplet) ground state. For $\text{Ni}(\text{ethylenediamine})_3(\text{NO}_3)_2$ we observed first-order spectra at 95 GHz. A single crystal of this compound shows two normal $M_S = 0 \leftarrow M_S = 1$ and the $M_S = -1 \leftarrow M_S = 0$ transitions with an effective zero-field splitting depending on the orientation in the magnetic field. Moreover, a direct transition that corresponds to $M_S = 1 \leftarrow M_S = -1$ is observed. For this transition, the intensity is strongly orientation dependent. At the magic angle all three transitions fall on top of each other for this axially symmetric crystal. All three transitions show the same dependence on microwave power. The existence of a two quantum jump ($\Delta M_S = 2$ transition) and its dependence of the microwave power cannot be understood and explained in a (conventionally used) 2-level picture of each transition in a triplet system.

In the current work we present the results of a 3-level density matrix calculation. The calculation starts from the general equation of motion of the density matrix operator in the Heisenberg picture^a. The resulting set of differential equations is solved numerically. The solutions reproduce very well the observed linewidths and intensities. The two quantum jump follows intrinsically from the model.

^aN. Dam, Ethylene Hot-band Spectroscopy and Relaxation phenomena, Thesis 1988, University of Nijmegen.