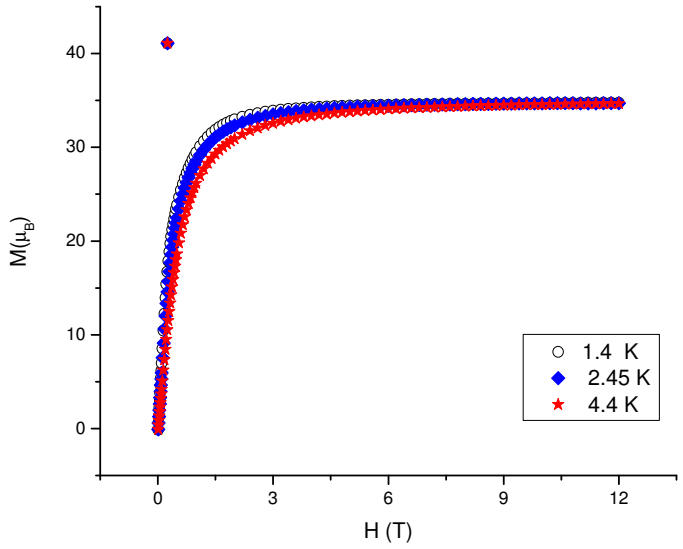

Figure S1: *Field dependence of magnetization at 1.4K (black circles), 2.45K (blue squares) and 4.4K (red stars).*

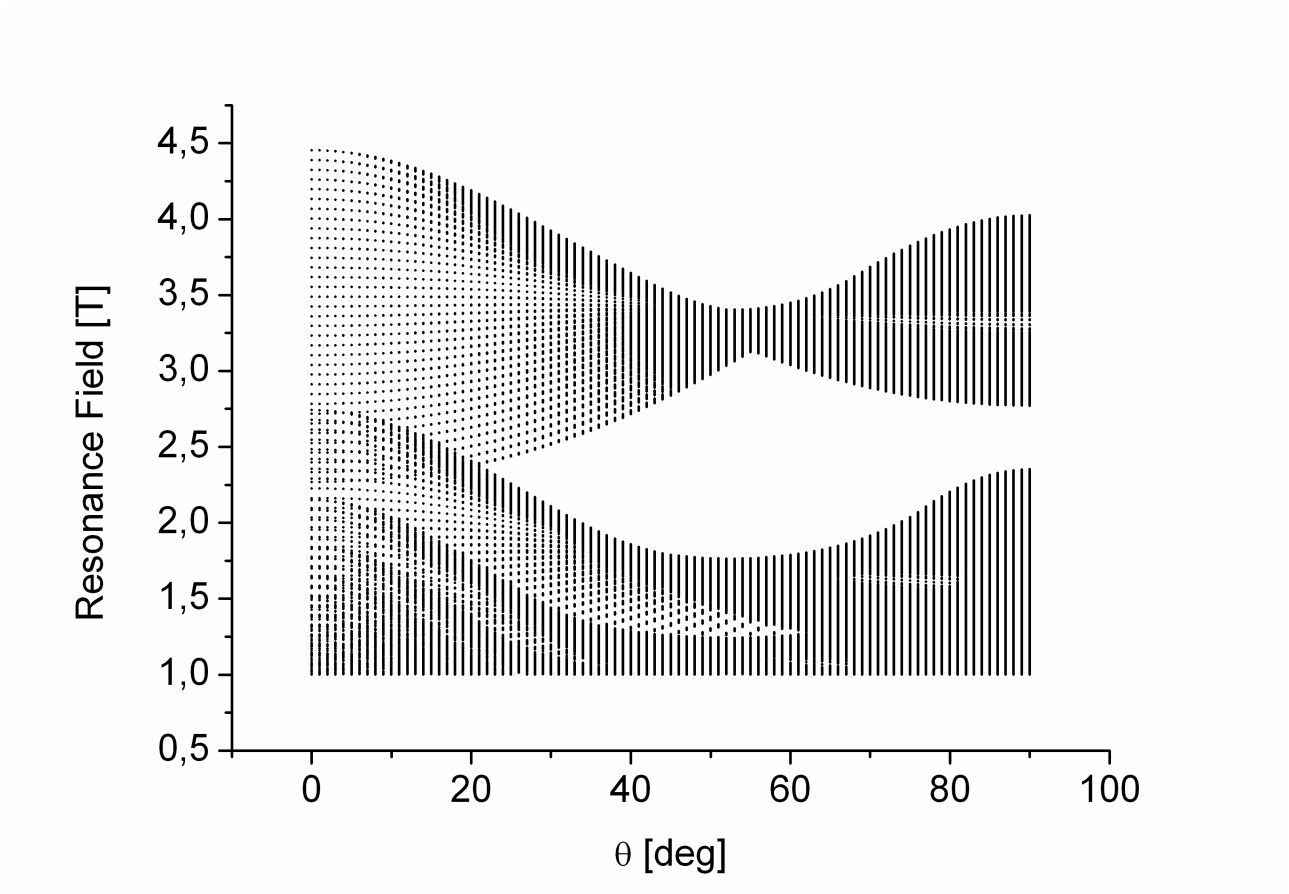

Figure S2: Angular dependence of resonance fields for S=35/2, D=-0.03 cm^{-1}, E=0.0015 cm^{-1}. Three bands can be distinguished, corresponding to the principal transitions, the $B_0/2$ ones and the $B_0/3$ ones.

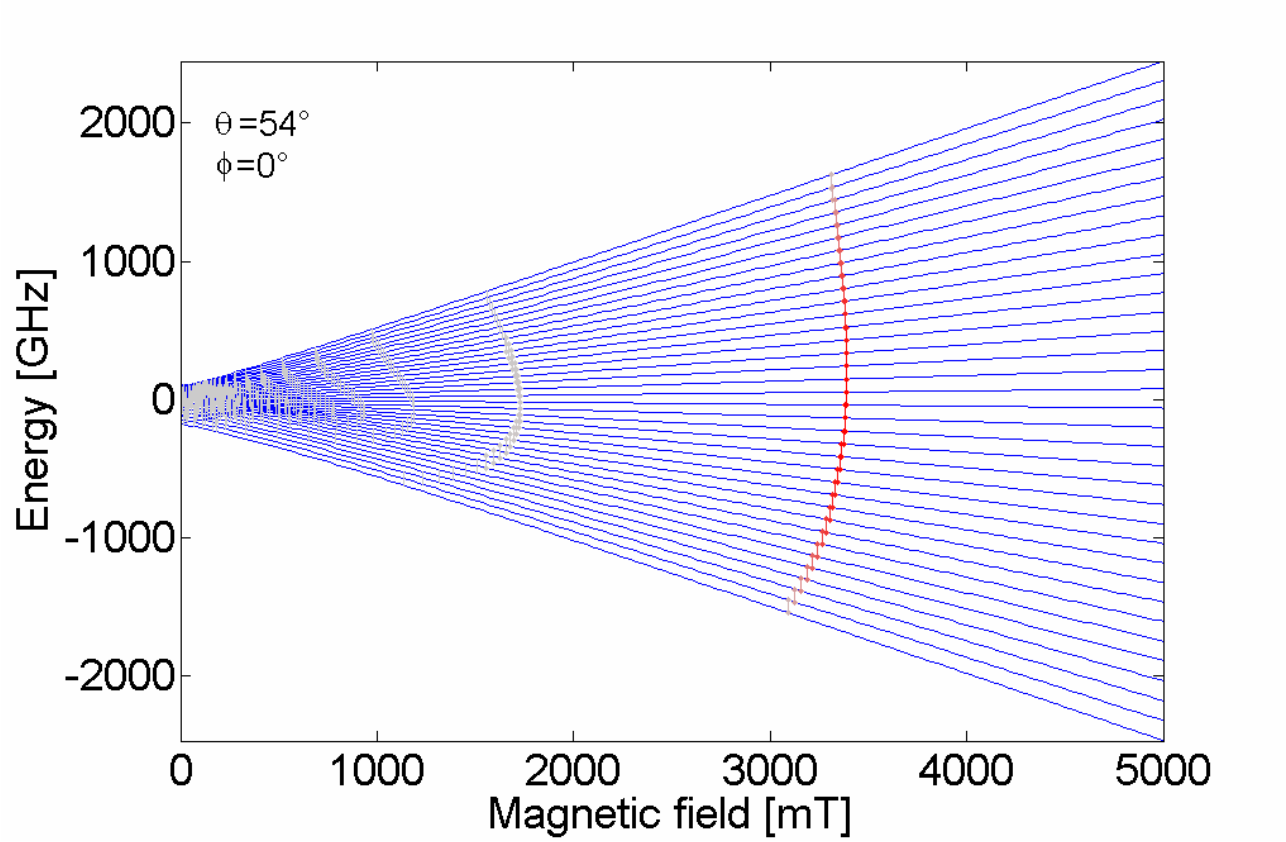

Figure S3: *Energy levels of the S = 35/2 state for θ =54°and ϕ = 0°. The red arrows indicate allowed transitions at W-band for the S = 35/2, the grey arrows the forbidden ones.*

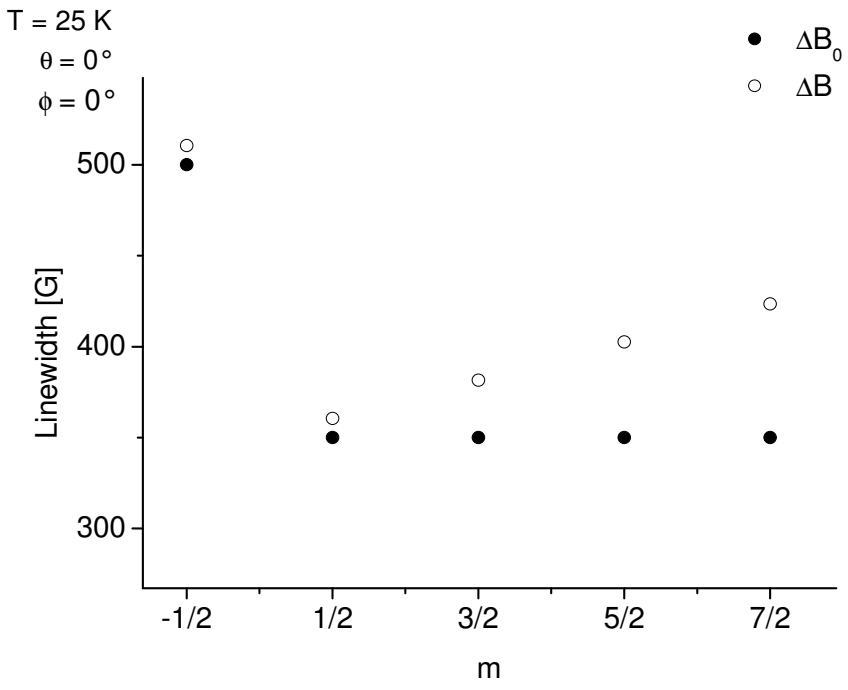

Figure S4: *ΔB_0 and ΔB linewidth (see text) assumed for the observed single transitions of the S=33/2 multiplet as a function of m, at T=25 K with $\theta = \phi = 0°$.*

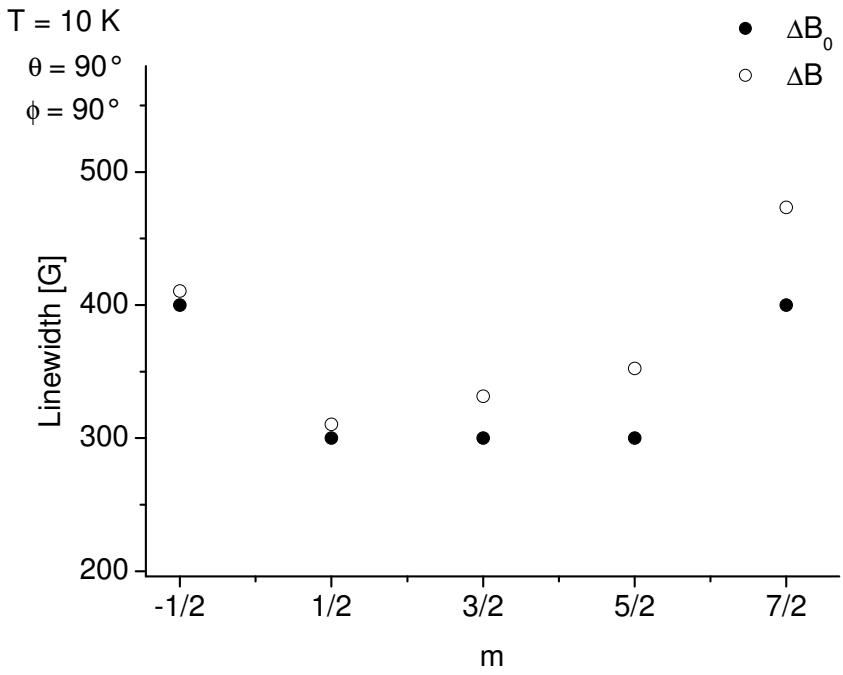

Figure S5: *ΔB_0 linewidth assumed for the observed single transitions og the S=33/2 multiplet as a function of m, at T=10 K with $\theta = \phi = 90°$.*

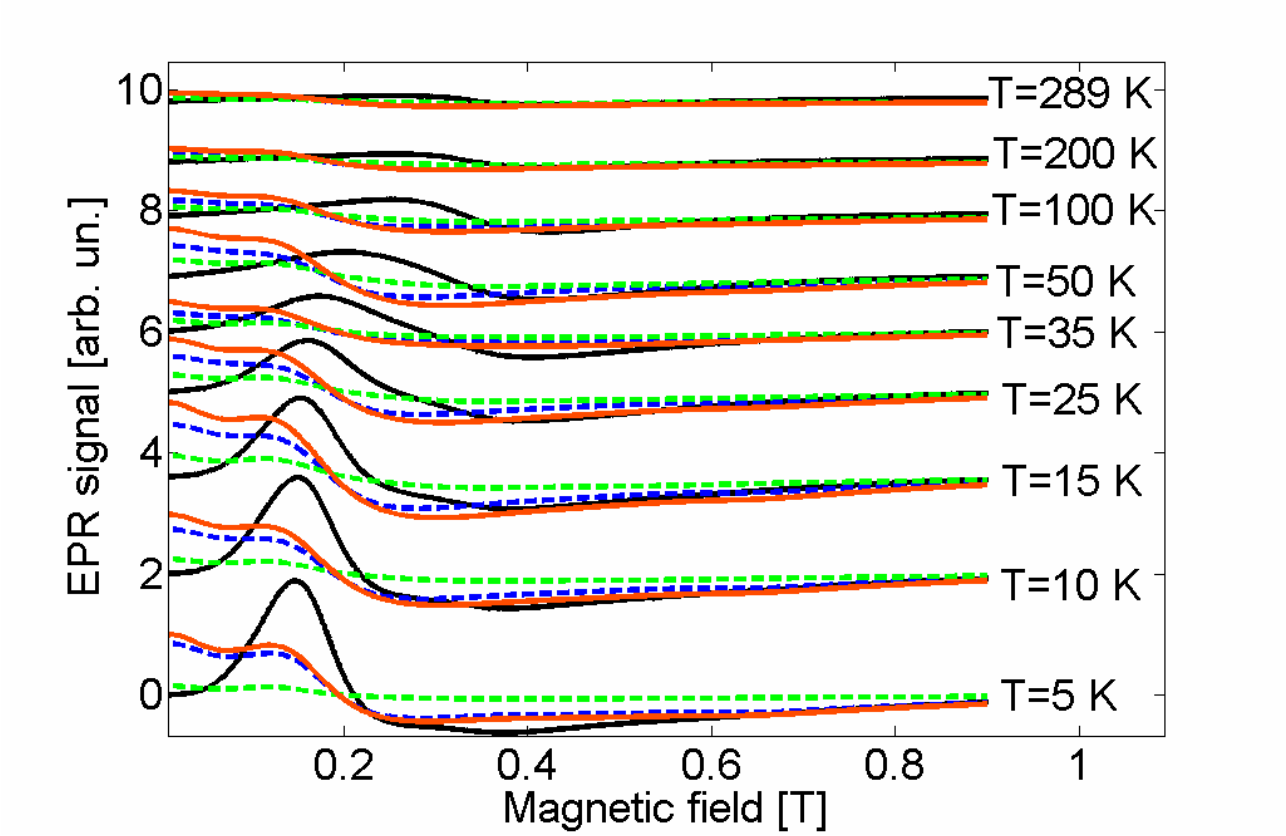

Figure S6: *X-band EMR powder spectra of* Fe_{19} *(black) recorded at different temperatures with a power of 65.57* μW *and simulations (red), sum of the S = 35/2 (dashed blue) and S = 33/2 (dashed green) contributions.*

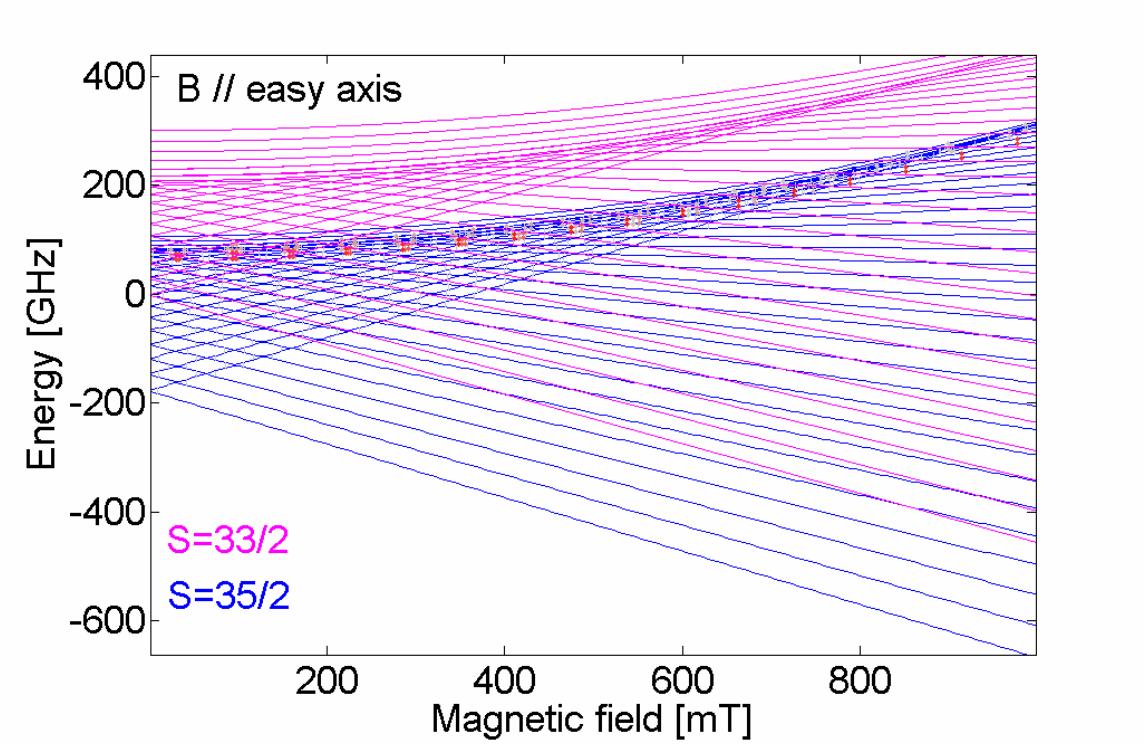

Figure S7: *Energy levels of the S = 35/2 (blue) and S = 33/2 (magenta) for $\theta = 0°$ and $\phi = 90°$. The red arrows indicate allowed transitions at X-band for the S = 35/2, the grey arrows the forbidden ones.*

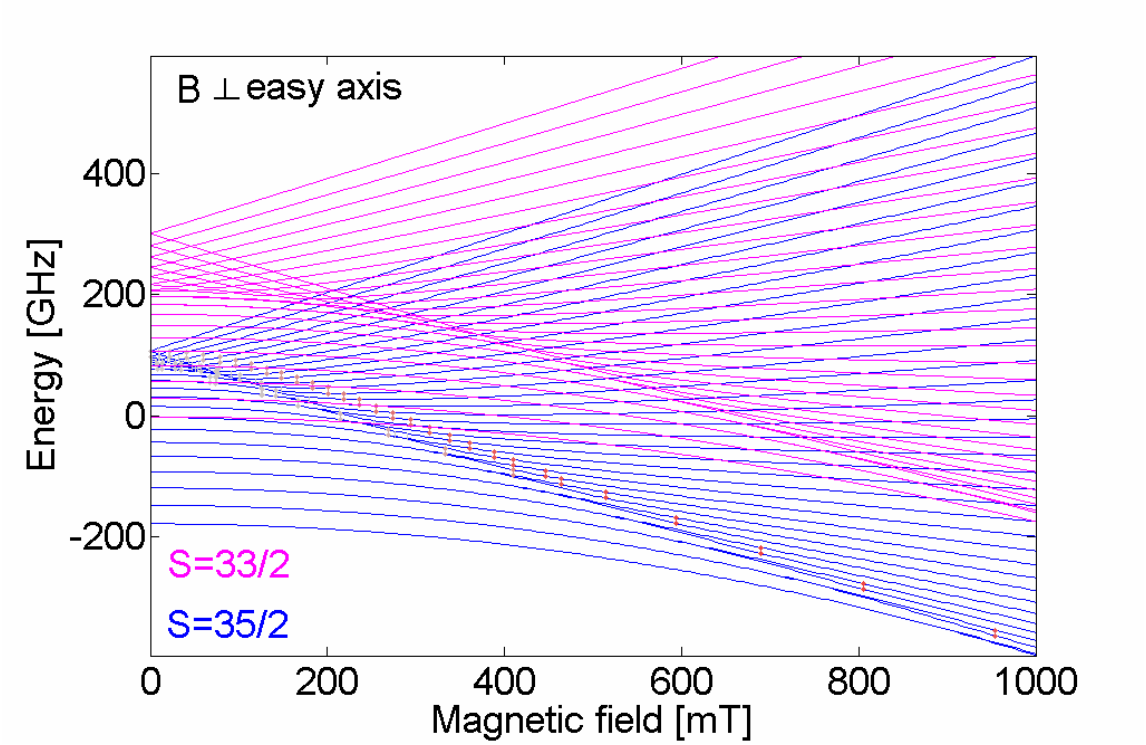

Figure S8: *Energy levels of the S = 35/2 (blue) and S = 33/2 (magenta) for $\theta = 90°$ and $\phi = 90°$. The red arrows indicate allowed transitions at X-band for the S = 35/2, the grey arrows the forbidden ones.*

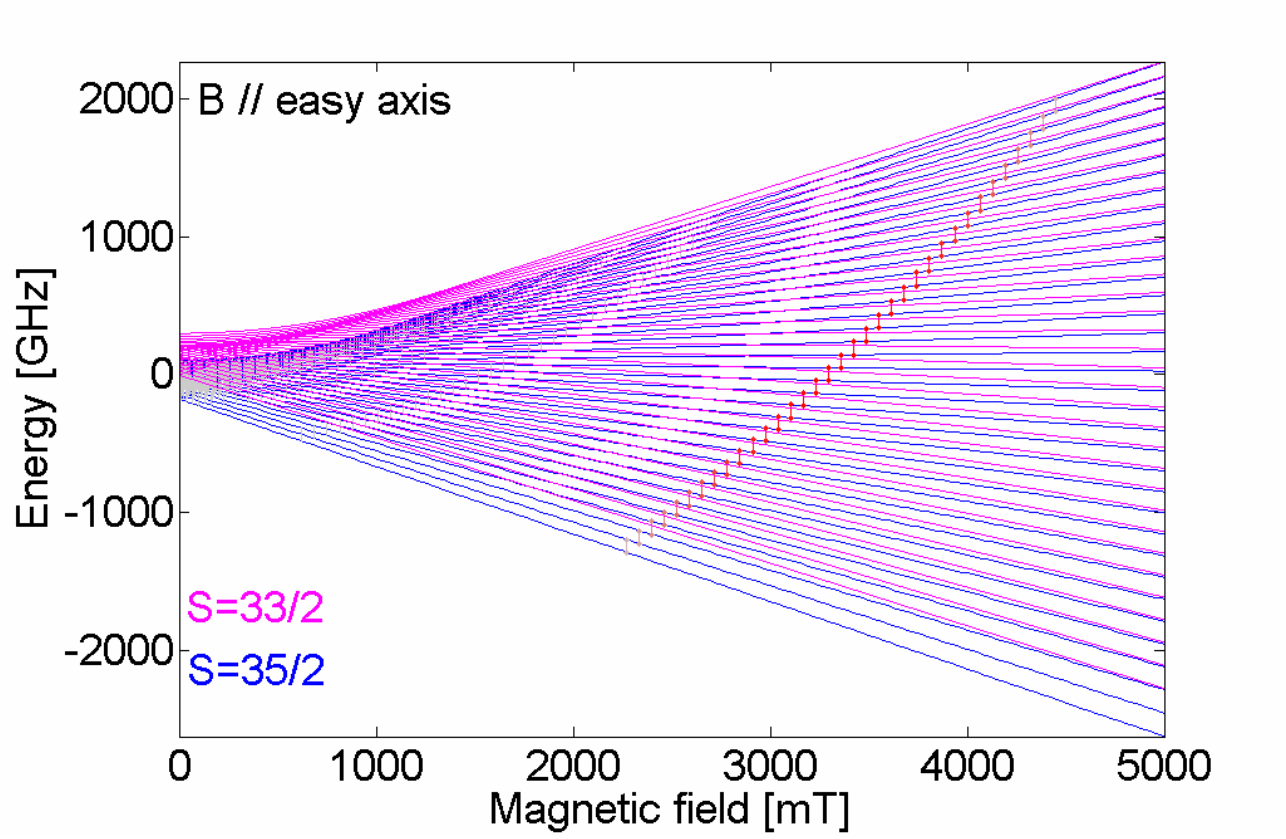

Figure S9: *Energy levels of the S = 35/2 (blue) and S = 33/2 (magenta) for* θ *=0° and* ϕ *= 90°. The red arrows indicate allowed transitions at W-band for the S = 35/2, the grey arrows the forbidden ones.*

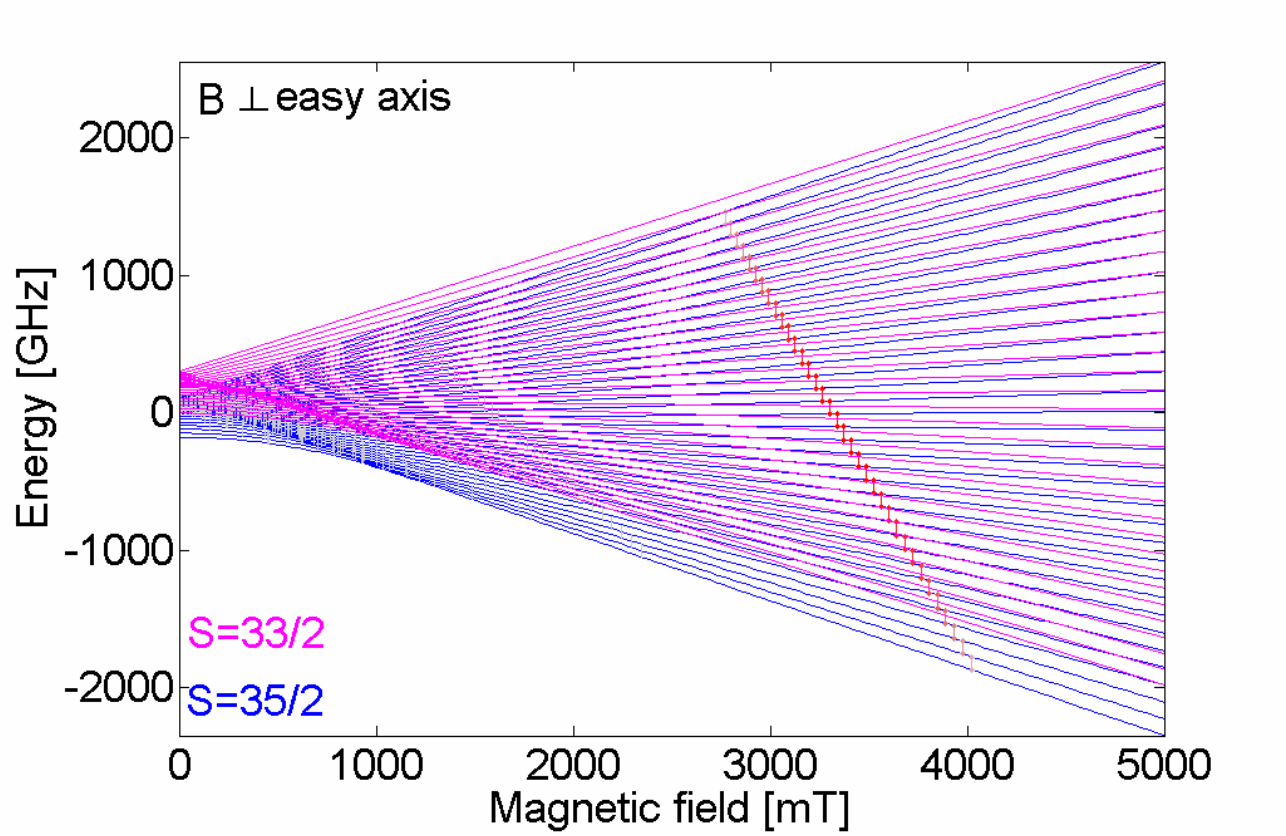

Figure S10: *Energy levels of the S = 35/2 (blue) and S = 33/2 (magenta) for* θ *=90° and* ϕ *= 90°. The red arrows indicate allowed transitions at W-band for the S = 35/2, the grey arrows the forbidden ones.*