Electronic Supplementary Material (ESI) for Dalton Transactions

Electronic Supplementary Information

Determination of the relevant magnetic interactions in low-dimensional molecular materials: the fundamental role of single crystal high frequency EPR.

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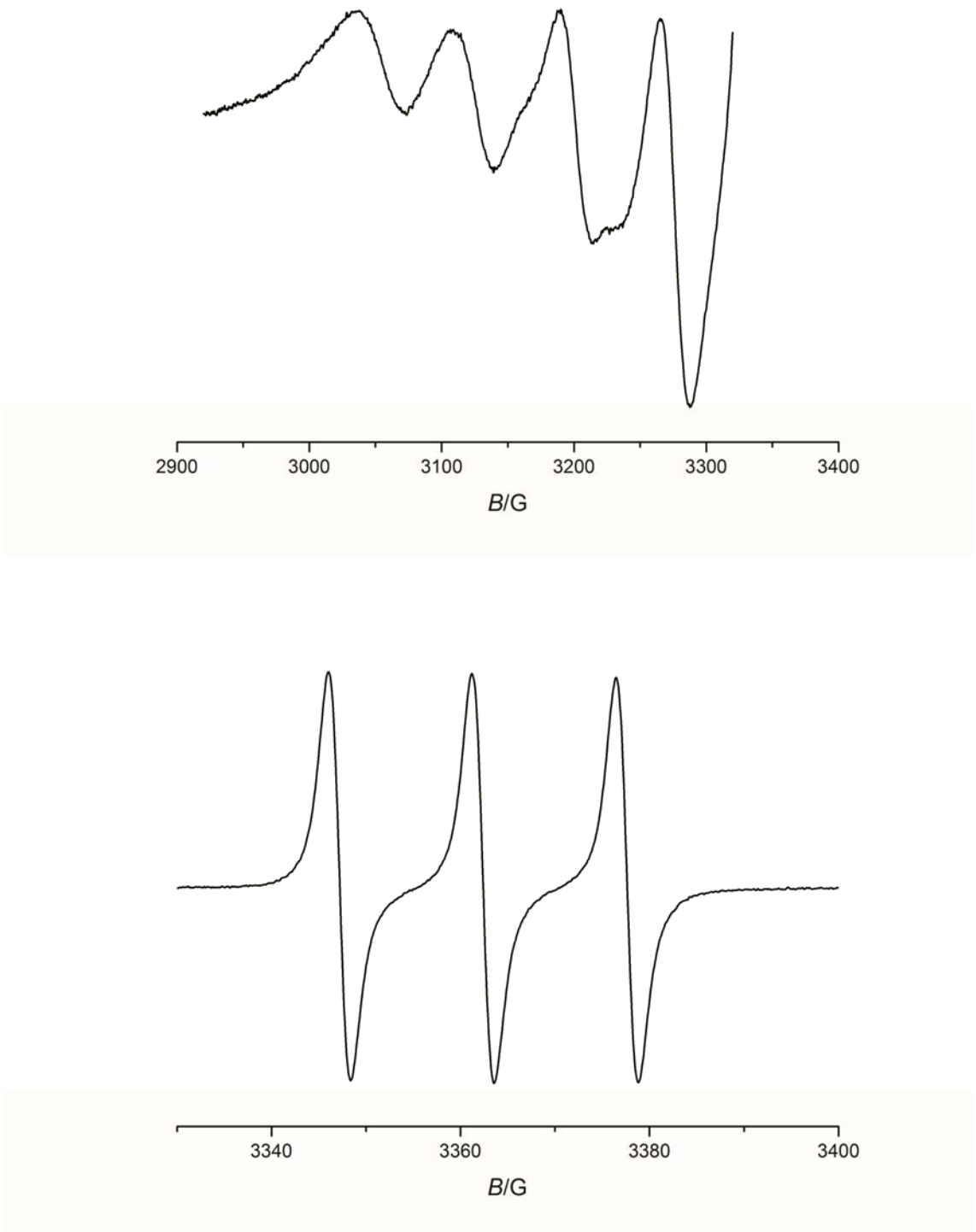

Figure S1: EPR spectra of $[Cu(hfac)_2(N_3TEMPO)]_n$ in *n*-heptane solution. Zoom in the $[Cu(hfac)_2]$ signal (top) and the nitroxide radical signal (bottom).

Table S1: Powder X-band simulated spectral parameters for $[Cu(hfac)_2(N_3TEMPO)]_n$ at 293, 40, 20, 10 and 5 K. The values of hyperfine coupling are given in Gauss (G).

	I[a]			**II**				
Temperature (K)	g_1	g_2	g_3	g_1	g_2	g_3	$A_{\|\|}$	$A_\perp$
293	2.04	2.05	2.1	2.06	2.09	2.33	150	10
40	2.04	2.05	2.1	2.07	2.07	2.3	150	10
20	2.04	2.04	2.09	2.07	2.07	2.3	150	10
10	2.04	2.04	2.09	2.07	2.07	2.3	150	10
5	2.04	2.04	2.07	2.07	2.07	2.3	150	10

[a] = These spectra were calculated considering S = ½.

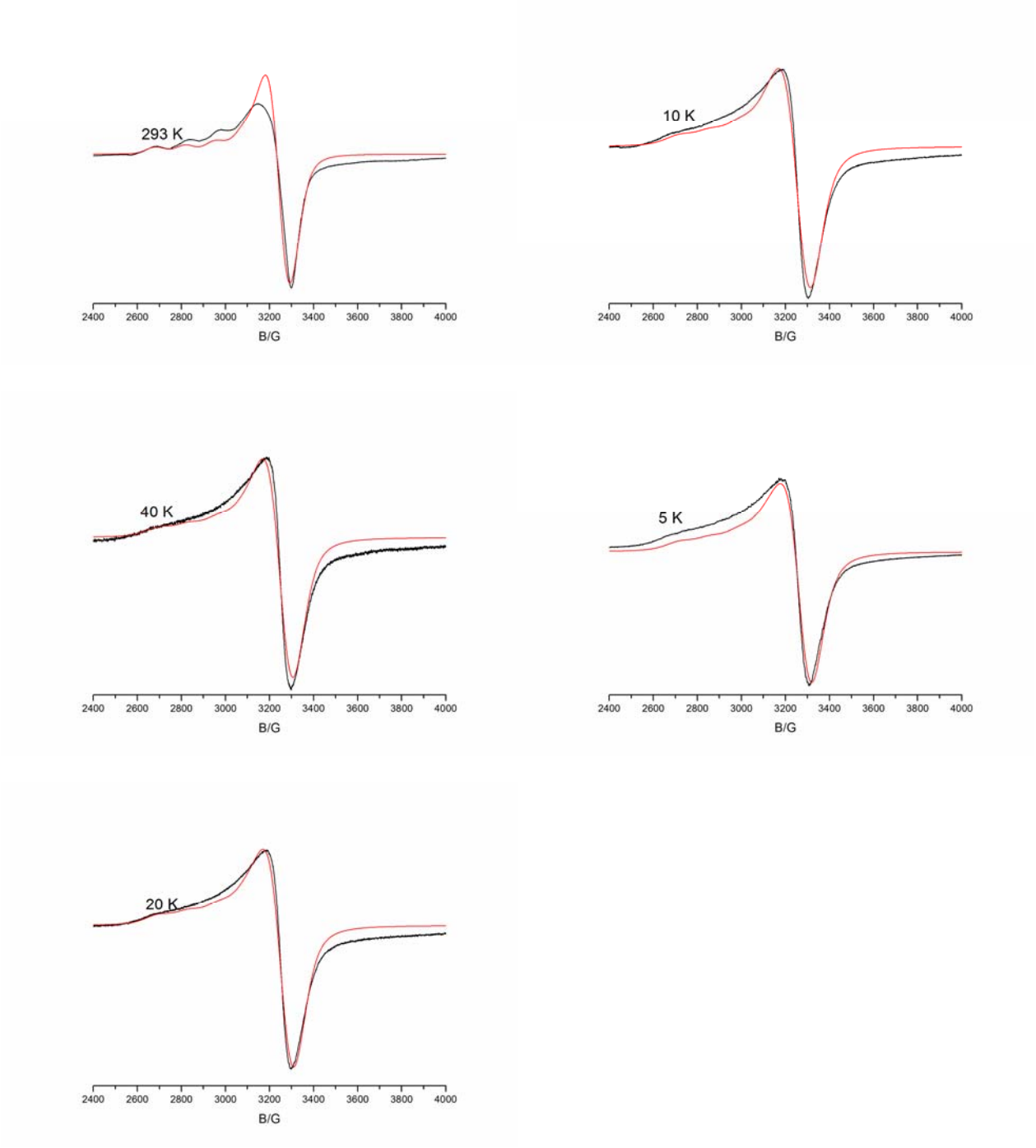

Figure S2: Powder X- Band EPR spectrum of $[Cu(hfac)_2(N_3TEMPO)]_n$ recorded at 293, 40, 20, 10 and 5K (black lines) with the best simulation curves (red lines).

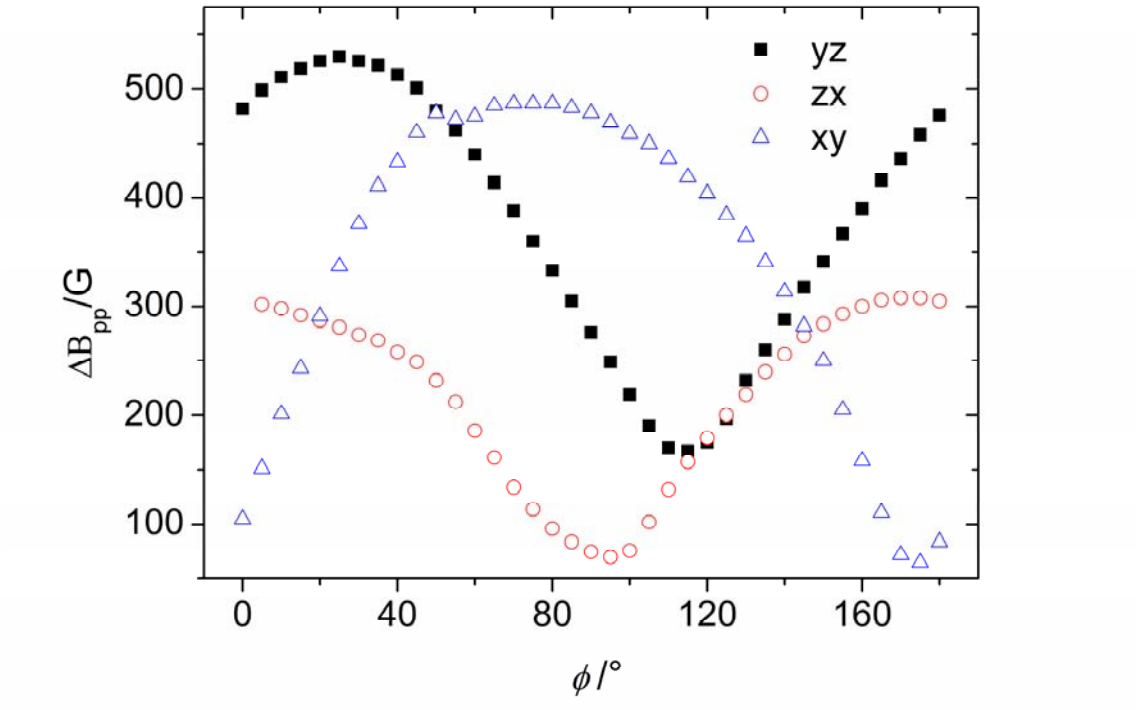

Figure S3: Room temperature angular dependence of the peak-to-peak linewidths for **II** at X-band in the three orthogonal laboratory axes.

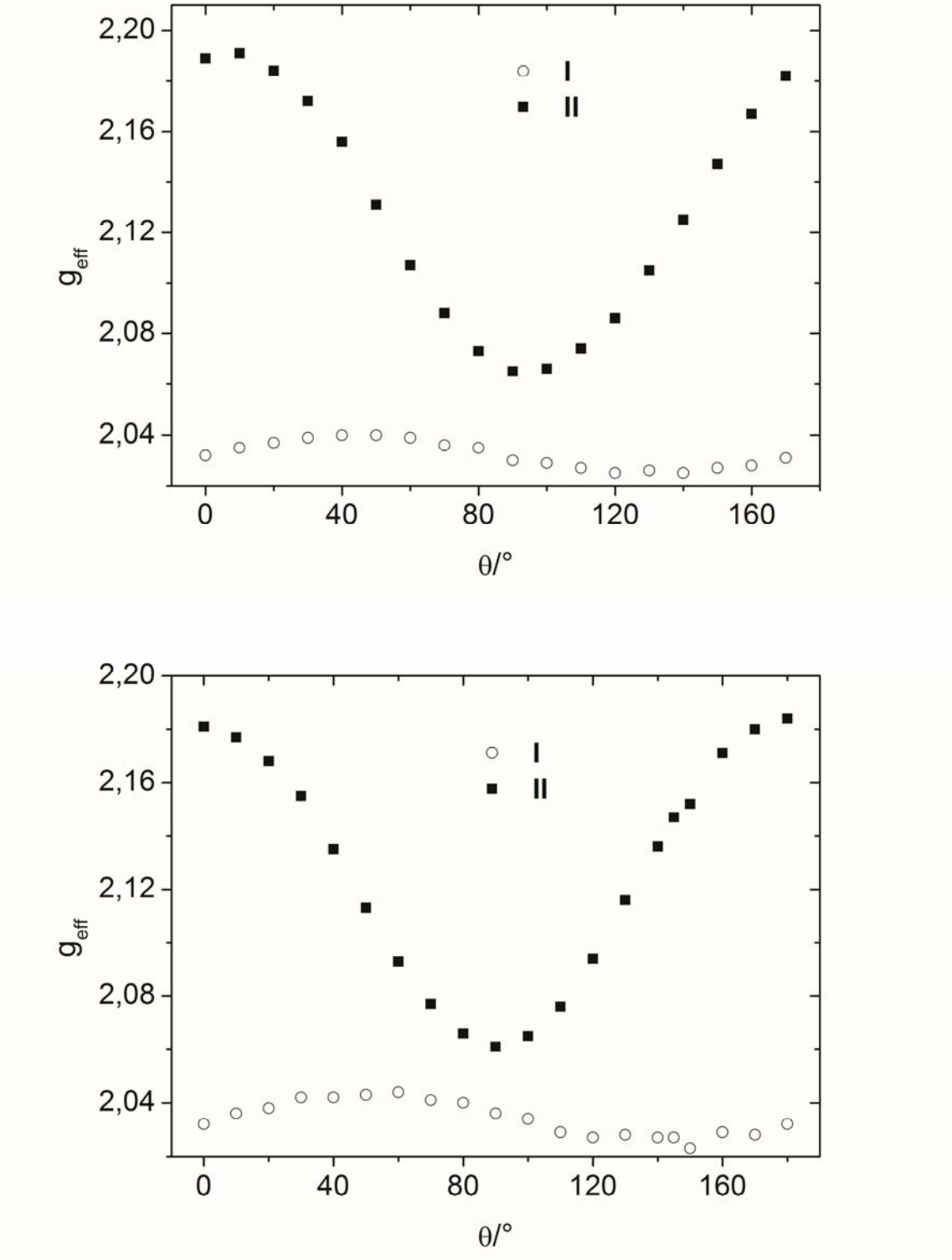

Figure S4: Angular dependence of g_{eff} for the two signals at W-band at 293 K (top) and 40 K (bottom). The rotation was performed around the direction perpendicular to the (11-1) face; θ = 0° corresponds to magnetic field making an angle of 20° with the intersection of the (011) and (11-1) faces.

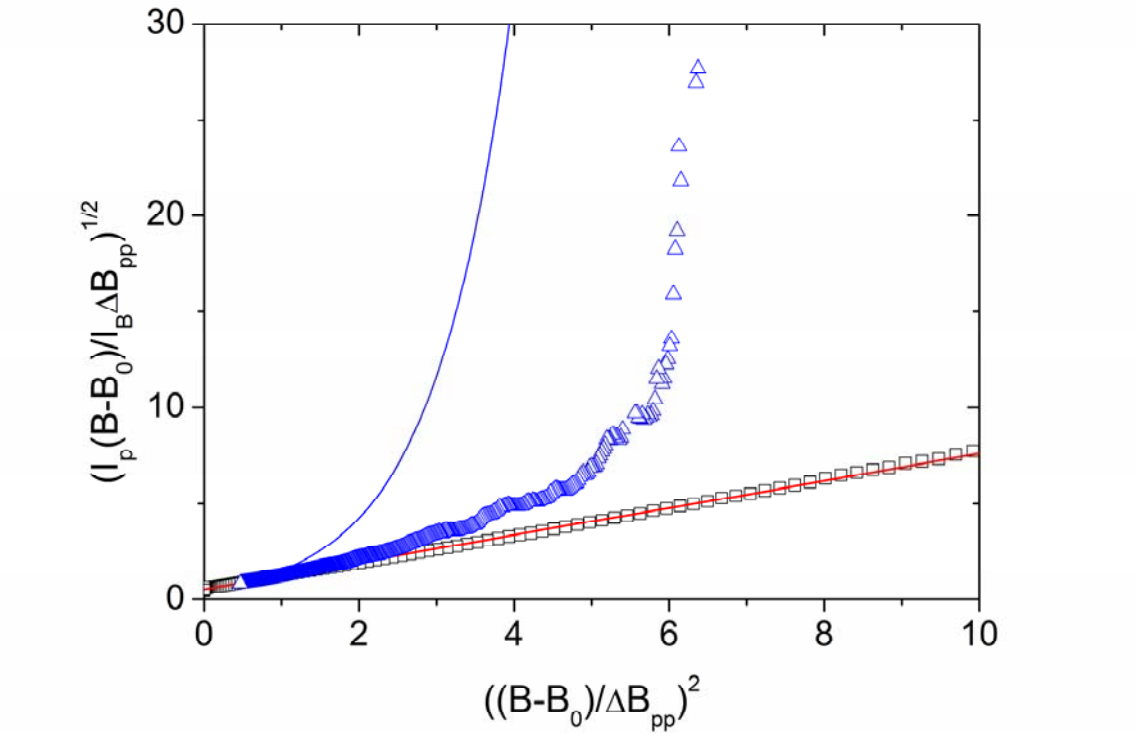

Figure S5: Dietz's Plot analysis of the EPR line shape at W-band and at room temperature of the signal **I**. Blue triangles: field applied close to the chain direction (θ = 0°); empty squares: field applied at 60° with respect to the chain direction (θ = 60°). The solid red line represents the Lorentzian curve calculated assuming the same line width as that observed for field applied at 60° and the blue line represents the Gaussian curve calculated assuming the same line width as that observed for field applied close to the chain direction.

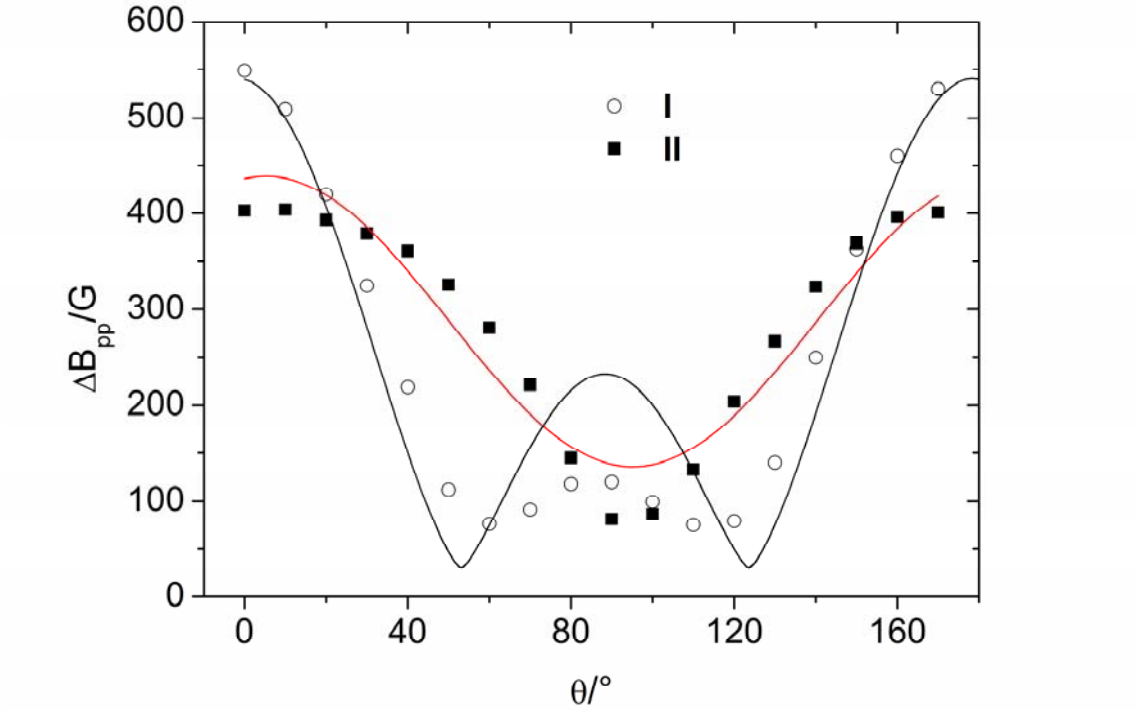

Figure S6: Room temperature angular dependence of linewidths for both signals at W-band. The black line is the best fit curve obtained with the expression $\Delta B_{pp} = \alpha + \beta|\ 3\cos^2\theta\text{-}1\ |^{4/3}$ (see text) for **I**. The red line represents the best fit curve for **II** obtained with the expression $\Delta B_{pp} = \delta + \gamma\cos^2\theta$ ($\delta = 742.2$ G, $\gamma = -303.2$ G). The rotation was performed around the direction perpendicular to the (11-1) face. $\theta = 0°$ corresponds to magnetic field making an angle of 20° with the intersection of the (011) and (11-1) faces.

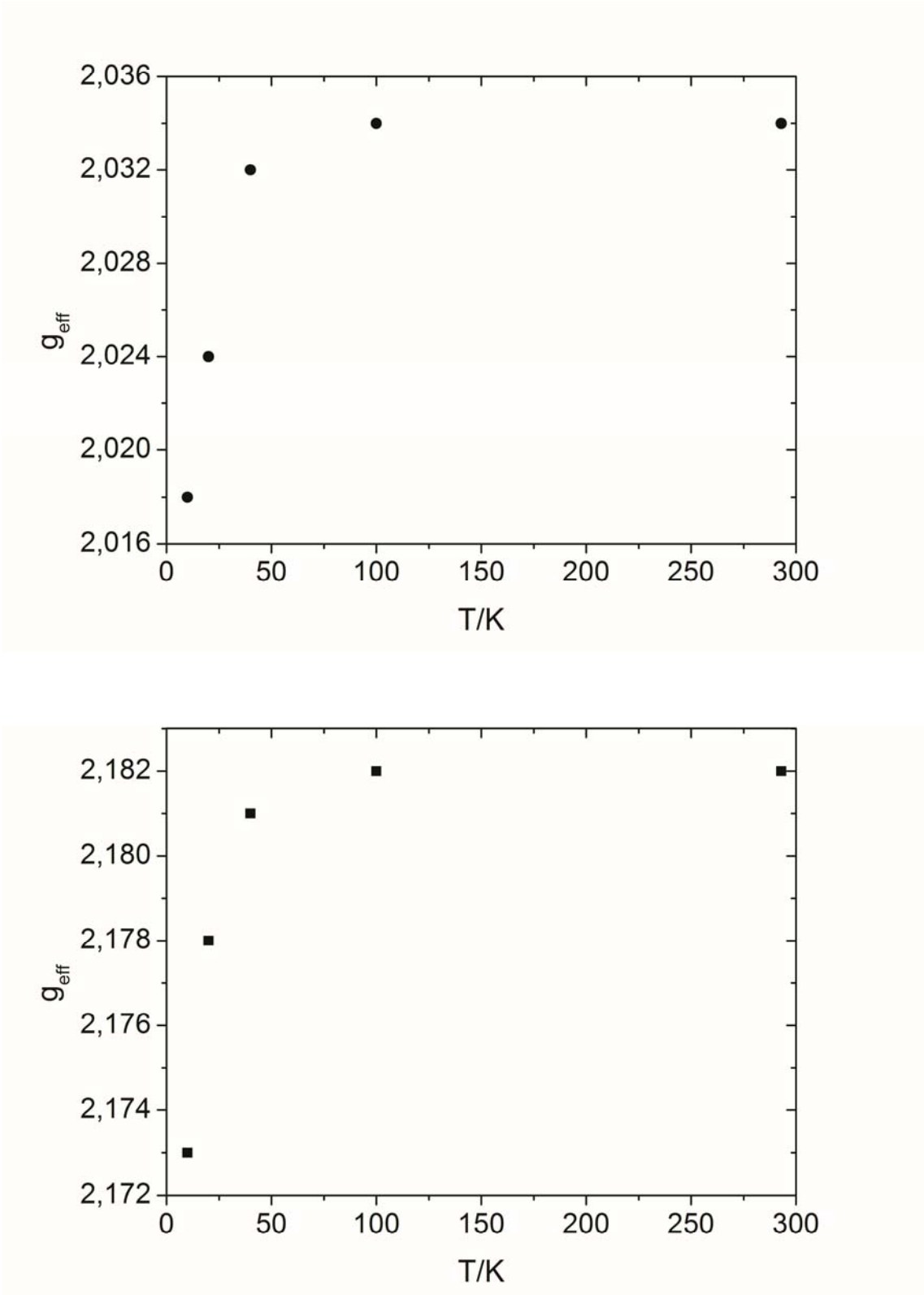

Figure S7: Temperature dependence of observed ***g*** values for **I** (top) and **II** (bottom) at W-band at θ = 0°.