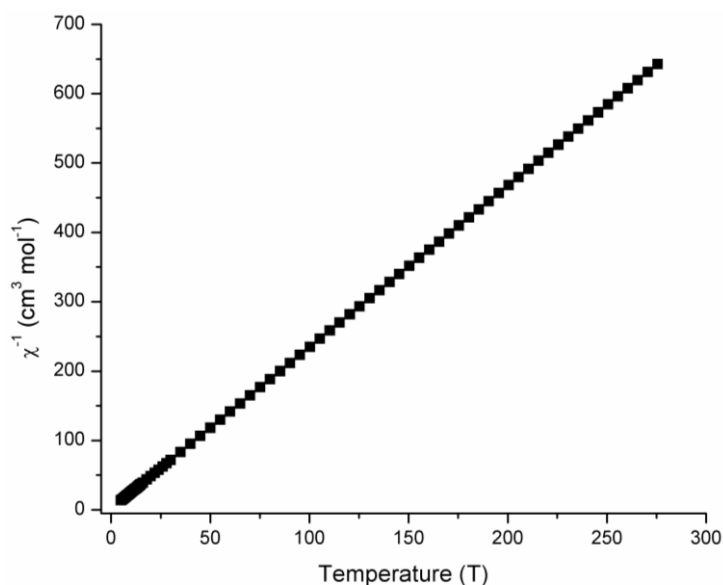
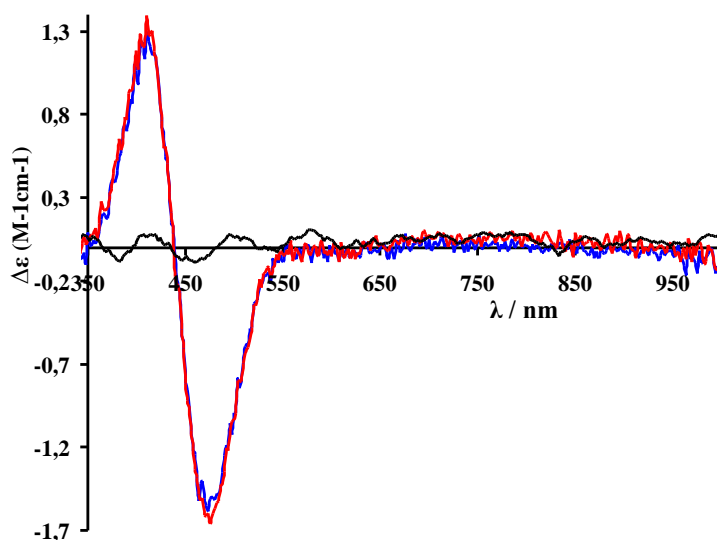


### Supplementary Information

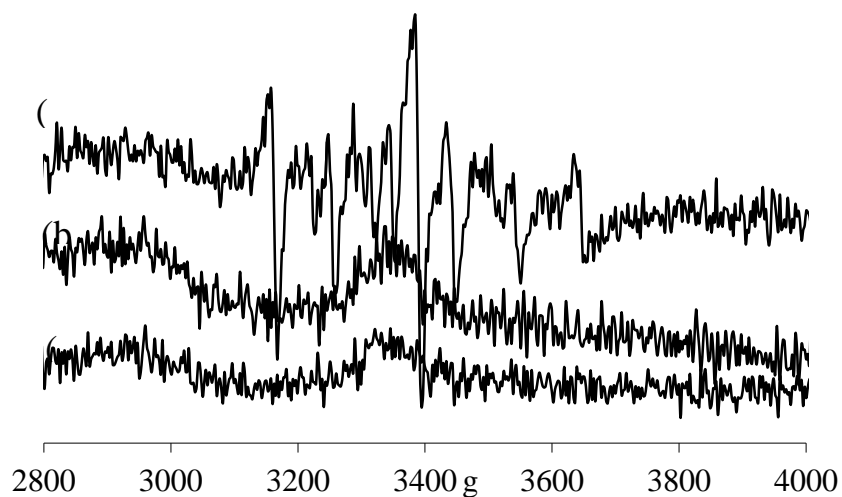


**Figure S1.** Change of the reciprocal magnetic susceptibility of **2** against temperature.

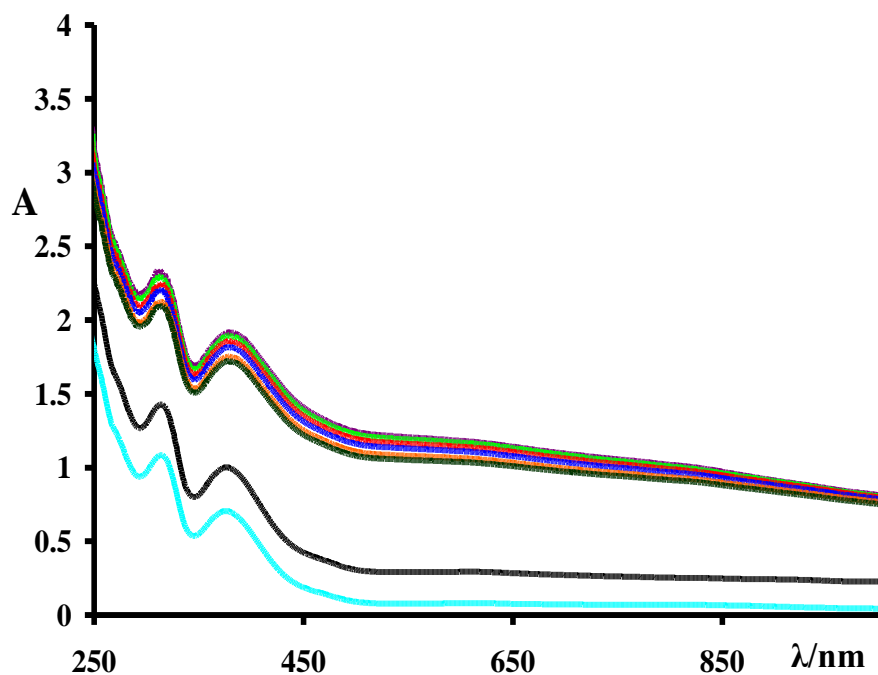
CD and EPR of solutions containing human serum albumin and either  $[\text{Ni}^{\text{II}}(\text{pydmedpt})]$  or  $\text{NiCl}_2$ :



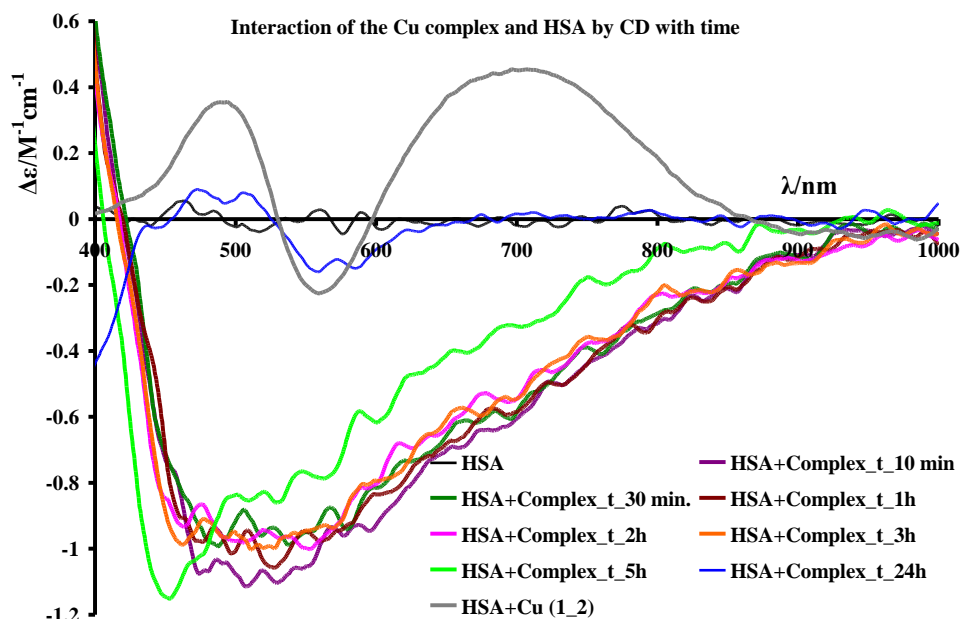
**Figure S2.** Circular dichroism spectra of solutions at pH 7.4 (PBS buffer). Black: CD spectrum of  $[\text{Ni}^{\text{II}}\text{Cl}_2]$  (800  $\mu\text{M}$ ); Blue: CD spectra of a solution containing HSA (400  $\mu\text{M}$ ) and  $[\text{Ni}^{\text{II}}\text{Cl}_2]$  (800  $\mu\text{M}$ ) after 30 min of addition of the Ni salt; red: CD spectra of a solution containing HSA (400  $\mu\text{M}$ ) and  $[\text{Ni}^{\text{II}}(\text{pydmedpt})]$  (800  $\mu\text{M}$ ) after 24 h of addition of the Ni salt.



**Figure S3.** X-band EPR spectra in H<sub>2</sub>O for [Ni<sup>II</sup>(pydmedpt)] (800 μM); (a) [Ni<sup>II</sup>(pydmedpt)] (800 μM); (b) [Ni<sup>II</sup>(pydmedpt)] (800 μM) and HSA (400 μM) after 30 min; (c) [Ni<sup>III</sup>Cl<sub>2</sub>] (800 μM) and HSA (400 μM) after 24 hrs.



**Figure S4.** UV-Vis spectra of a solution of compound **1** (1.0 mM) in PBS buffer at pH 7.4 recorded with time. Pink: immediately after dissolving (ca. 10 min.); green: after ca. 1 h of preparation; red: after ca. 2 h of preparation; blue: after ca. 3 h of preparation; orange: after ca. 4 h of preparation; dark green: after ca. 5 h of preparation; black: after ca. 24 h of preparation; light blue: after ca. 36 h of preparation of the buffered solution.



**Figure S5.** Circular dichroism spectra of solutions in PBS buffer at pH 7.4. Black: CD spectrum of a HSA solution (500  $\mu\text{M}$ ); grey:  $[\text{Cu}^{\text{II}}\text{Cl}_2]$  (800  $\mu\text{M}$ ) and HSA (400  $\mu\text{M}$ ) after ca. 24 hrs of mixture. The other CD spectra were measured with a solution containing HSA (500  $\mu\text{M}$ ) and  $[\text{Cu}^{\text{II}}(\text{pydmedpt})]$  **1** (1.00 mM); these were measured with time after addition of **1** to a solution of HSA. Some of these CD spectra are the same of those presented in Figure 12, but restricted to the wavelength range 400-1000 nm. The spectra of the solutions containing **1** and HSA were recorded with a 5 mm optical path quartz cell.