

SUPPORTING INFORMATION

Synthesis of highly ordered L1₀ MPt alloys (M=Fe, Co, Ni) from crystalline salts: an in-situ study of the Pre-Ordered Precursor Reduction strategy

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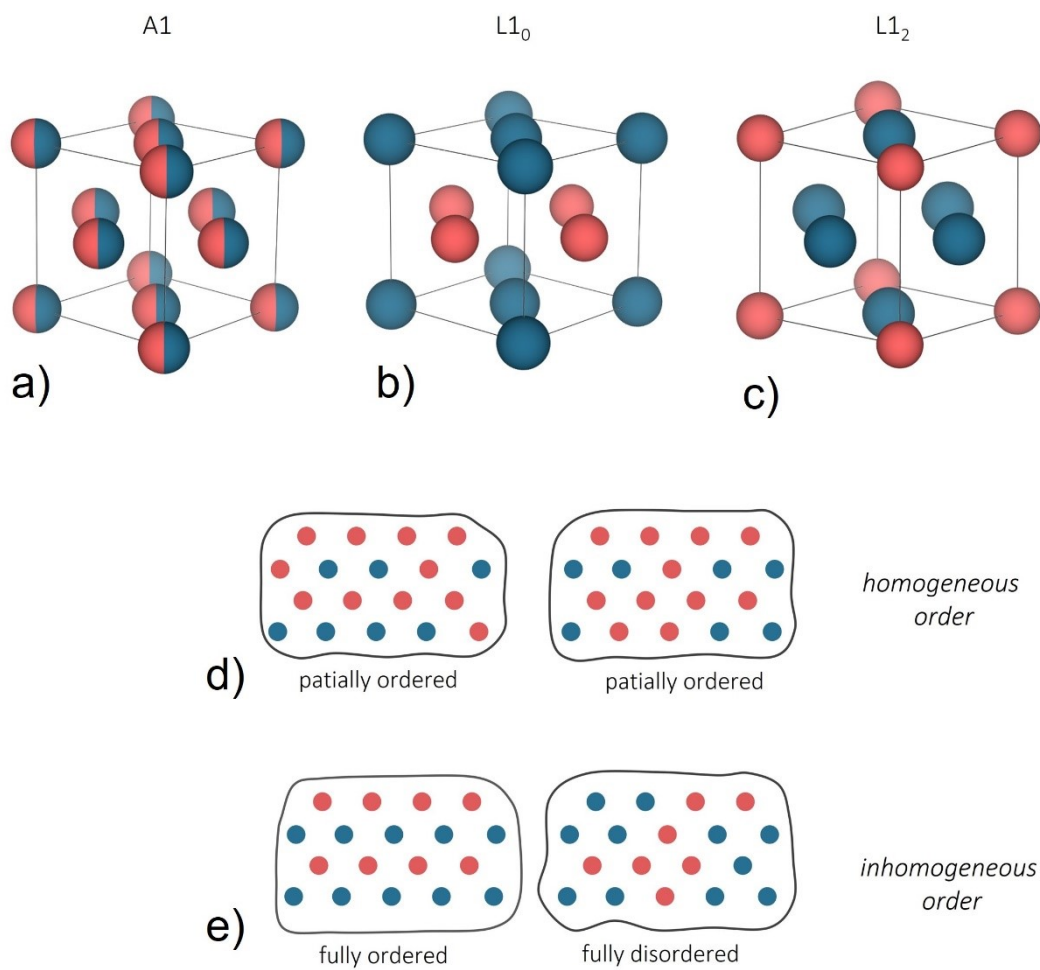
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Figure S1. (a-c) Sketches of the disordered (50:50, A1) and ordered (50:50 L1₀, 25:75 L1₂) crystallographic phases of the MPt (M = Fe, Co, Ni) alloys. (d-e) Differences in homogeneous and inhomogeneous partial order in a granular MPt (50:50) system. In a homogeneous system, each grain is characterized by a L1₀ arrangement with a certain amount of atoms in a wrong position (partial order); in an inhomogeneous system, some grains are fully ordered and some grains are fully disordered. In the present study we assume that the PPR process produces homogeneous FePt and CoPt and inhomogeneous NiPt alloys.¹



¹ A. Cebollada, R.F.C. Farrow, M.F. Toney, *Structure and magnetic properties of chemically ordered magnetic binary alloys in thin film form*, in: H.S. Nalwa (Ed.), *Magn. Nanostructures*, American Scientific Publishers, 2002, p. 93.

Figure S2. Comparison between data acquired at the Pt-L_{III} (left) and Fe-K (right) edge on the L₁₀ FePt final alloy synthesized with the PPR process in the home furnace (@lab) and in the EXAFS chamber (@LISA). The comparison shows the high reproducibility of the process.

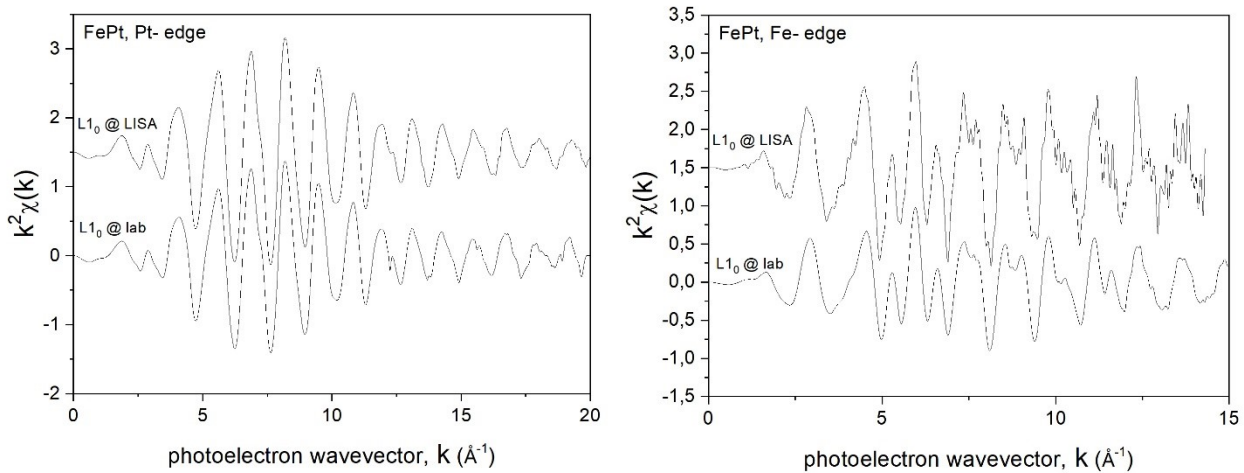
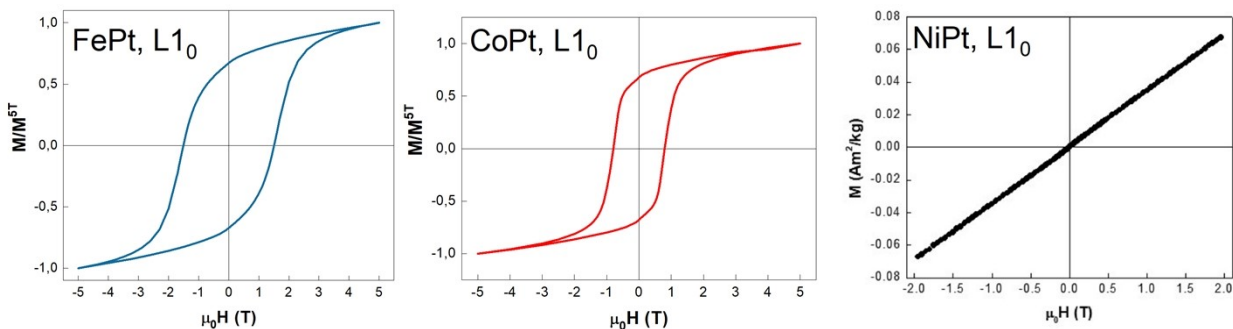


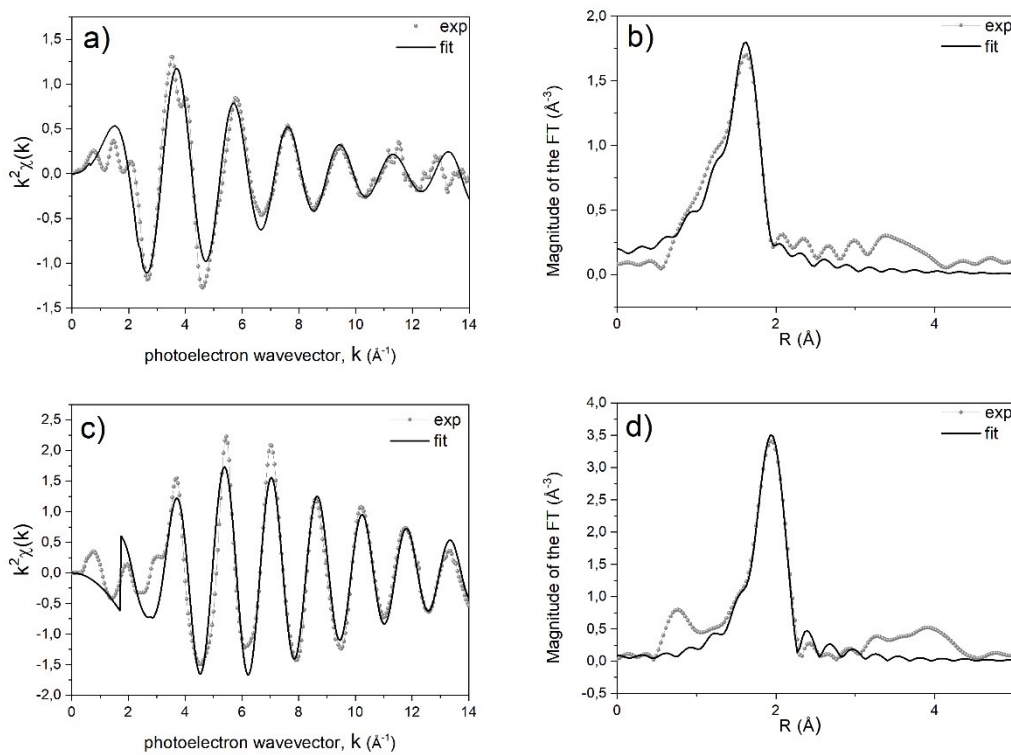
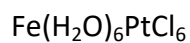
Figure S3: Characteristic room temperature hysteresis loop of the L₁₀ phases obtained in the best PPR experimental conditions for each compound. L₁₀ FePt and CoPt show the typical hysteresis loop of hard ferromagnetic materials², while NiPt present a linear anhysteretic curve characteristic of a paramagnetic material as expected for this alloy³.



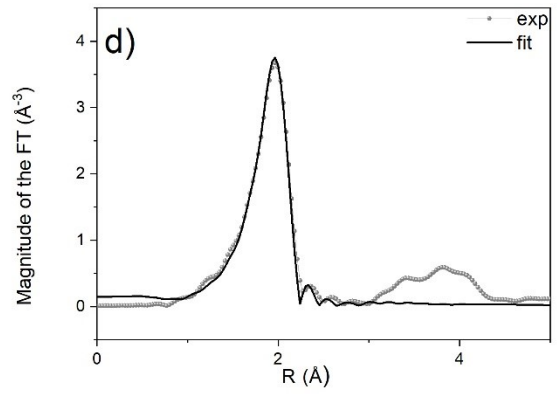
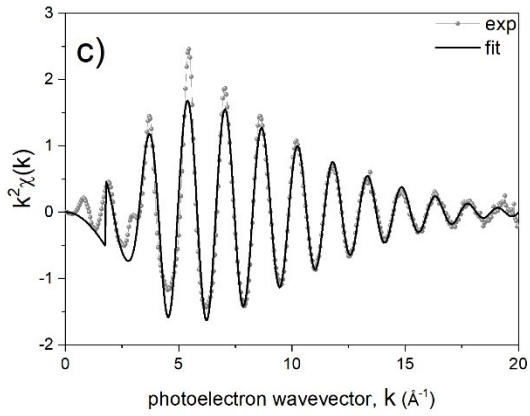
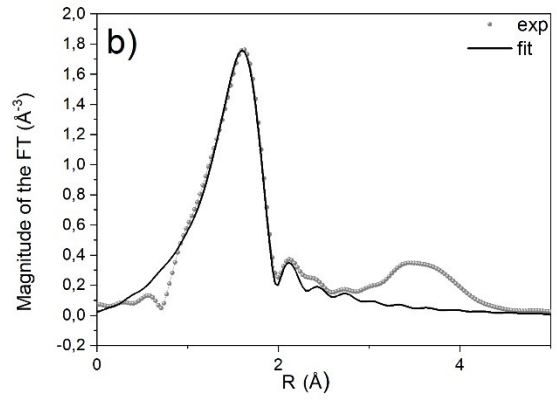
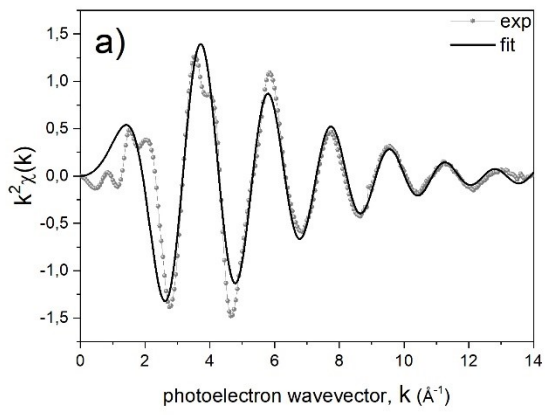
² A. Cebollada, R.F.C. Farrow, M.F. Toney, *Structure and magnetic properties of chemically ordered magnetic binary alloys in thin film form*, in: H.S. Nalwa (Ed.), *Magn. Nanostructures*, American Scientific Publishers, 2002, p. 93.

³ U. Kumar, P.K. Mukhopadhyay, B. Sanyal, O. Eriksson, P. Nordblad, D. Paudyal, K. Tarafder, A. Mookerjee, *experimental and theoretical study of annealed Ni-Pt alloys*, *Phys. Rev. B* 74 (2006), 064401, <https://doi.org/10.1103/PhysRevB.74.064401>; U. Kumar, K.G. Padmalekha, P.K. Mukhopadhyay, D. Paudyal, A. Mookerjee, *Magnetic transition in NiPt alloy systems: experiment and theory*, *J. Magn. Magn Mater.* 292 (2005) 234e240, <https://doi.org/10.1016/j.jmmm.2004.10.116>.

Figure S4. Experimental and fitted EXAFS signal (a, c) and corresponding Fourier Transform, FT (b, d) at the Fe-K edge (a, b) and Pt-L_{III} edge (c, d) of the initial precursors.



$\text{Co}(\text{H}_2\text{O})_6\text{PtCl}_6$



$\text{Ni}(\text{H}_2\text{O})_6\text{PtCl}_6$

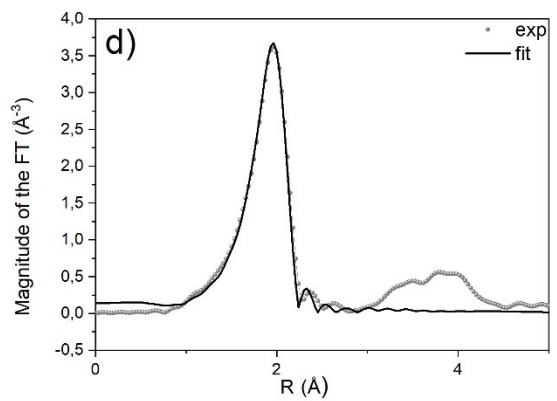
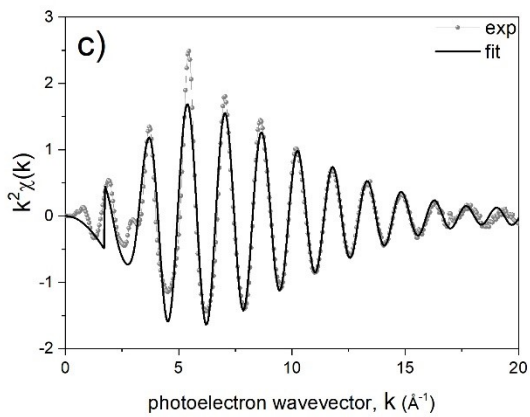
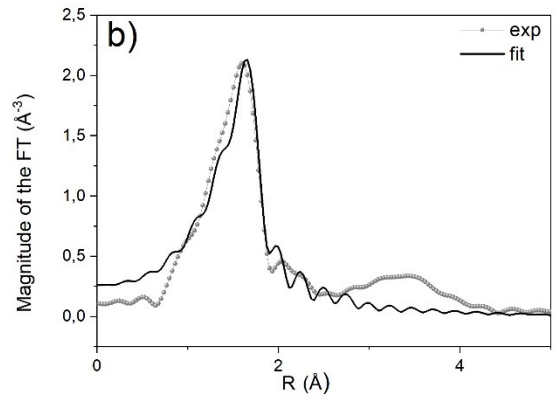
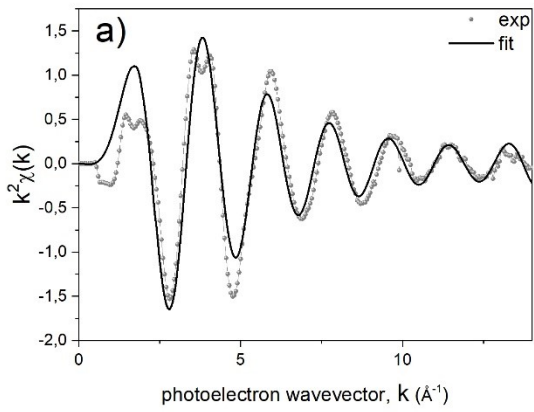


Figure S5. Experimental and fitted EXAFS signal (a, c) and corresponding Fourier Transform, FT (b, d) at the Pt-L_{III} edge of the CoPt and NiPt alloys synthesized by the PPR approach.

CoPt, NiPt L1₀ alloys

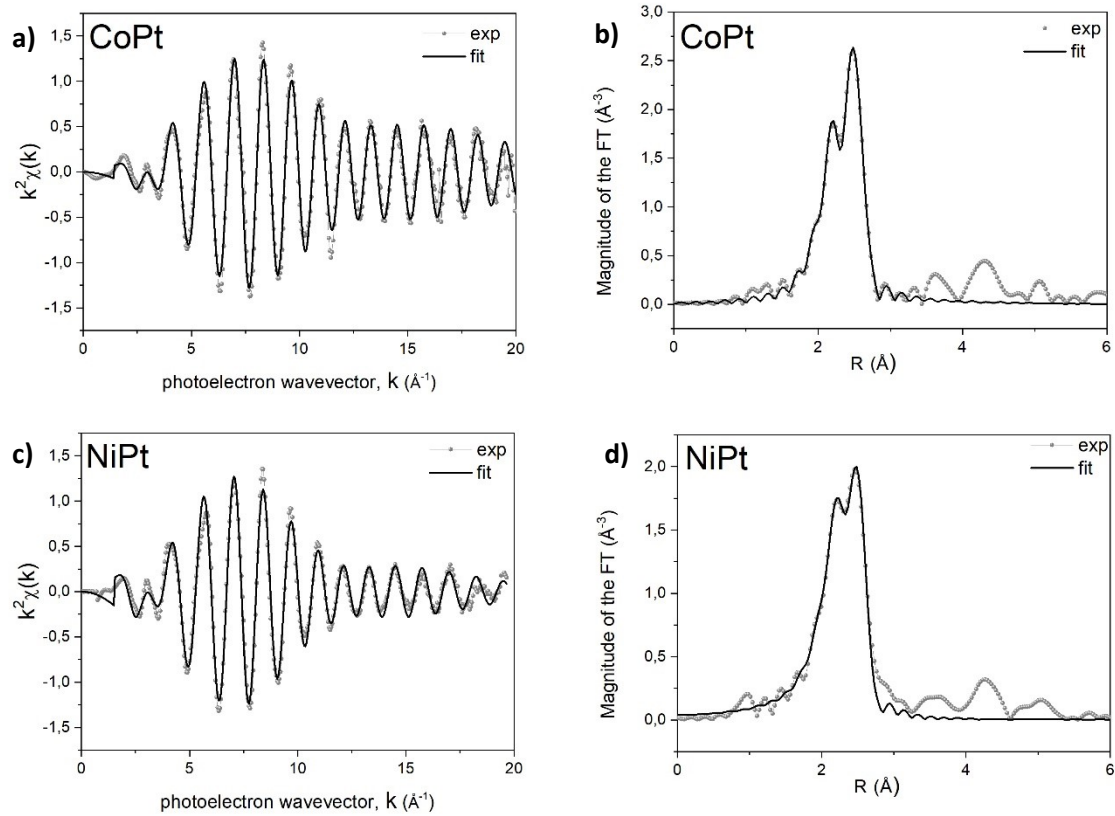


Figure S6. Contribution of the Fe-Pt and Fe-O scattering paths on the *in-situ* EXAFS oscillation (Fe-K edge) acquired at 300°C during the PPR process.

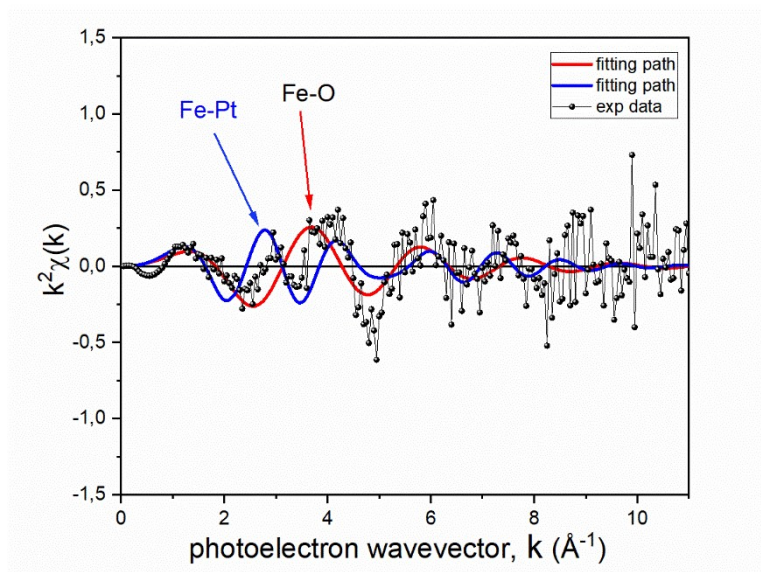


Figure S7. In-situ experimental and fitted EXAFS signal at the Pt-L_{III} edge of during the PPR treatment on the Fe-based system.

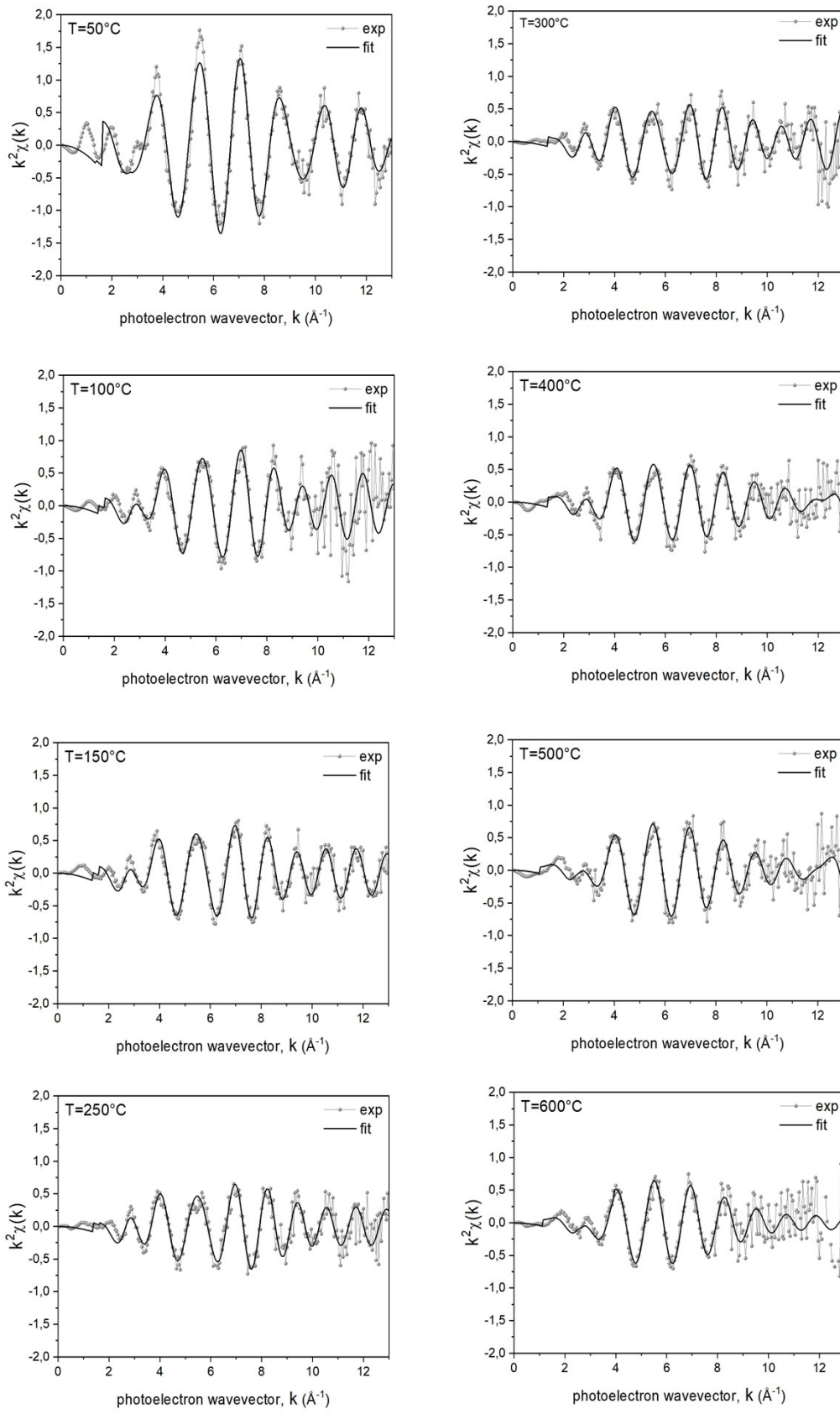


Figure S8. in-situ experimental and fitted EXAFS signal at the Fe-K edge during the treatment of the Fe-based system. Up to 150°C, the fitting model is based on 6 Fe-O scattering paths with the $r_{\text{Fe-O}}$ distance that slightly changes with temperature (distorted octahedral) (I model). In the intermediate range of temperatures, up to 350°C, the fitting model is based on a combination of Fe-O and Fe-Pt scattering paths with no evidence of Fe-Fe distances (II model). Above 400°C the III model is based on pure FePt alloy with Fe-Fe and Fe-Pt scattering paths, consistently with a $L1_0$ phase.

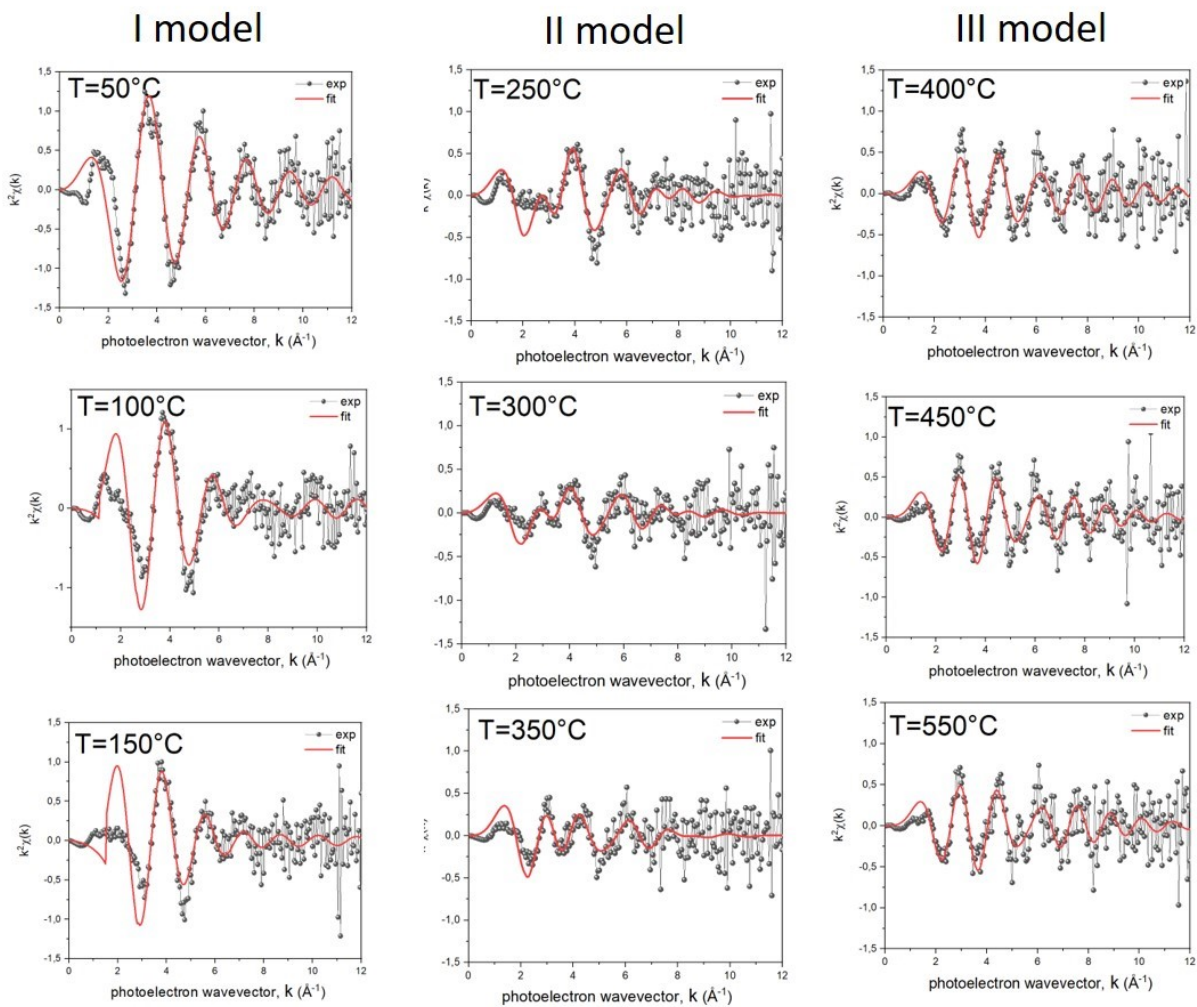


Figure S9. Comparison of the EXAFS signals of the three compounds, collected at the intermediate temperature of 300°C.

