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Metastable state of the photomagnetic Prussian blue analog $K_{0.3}Co[Fe(CN)_6]_{0.77}\cdot 3.6H_2O$, investigated by various physical techniques.

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The thermodynamic properties of the photomagnetic Prussian blue analog K 0.3 Co[Fe(CN) 6] 0.77 · 3.6H 2 O, which exhibits a charge-transfer-induced spin transition (CTIST), are reported. According to the thermal history of the sample, different low-temperature states are obtained: a quenched high-spin state (Q), a low-spin state (LT) achieved through the decay of the Q state, and an intermediate state (IM) combining low-spin (LS) and high-spin (HS) local states. The IM state is obtained by slow cooling and displays a slight kinetic dependence. The long lifetime of the IM state, which persists up to the vicinity of the CTIST temperature, is unusual and differs from all metastable states reported so far for photomagnetic Prussian blue analogs. The properties of the different states and of their photoexcited counterparts are investigated by magnetic and photomagnetic measurements, calorimetry, diffuse reflectance, and x-ray diffraction. A key feature of the IM state revealed by x-ray diffraction is the onset of phase separation between a LT-type fraction and a mixed HS-LS fraction (approximately 50 : 50). X-ray patterns of the IM state obtained during successive cooling and heating stages document irreversible transformations. The formation and properties of the IM state suggest a self-organization process between low- and high-spin sites in response to internal structural stresses, and this effect is hindered by irradiation with visible light. The relaxation kinetics of the Q and photoexcited states are analyzed by using a two-variable model, which accounts for the onset of correlations due to short-range elastic interactions. A quantitative comparison to the analogous sodium compound Na 0.32 Co[Fe(CN) 6] 0.74 · 3.4H 2 O confirms the nonstandard behavior of the title potassium compound and supports the structural origin of the self-organization processes.

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