A new bidentate Schiff base ligand N'-(2-pyridylmethylene)-3,5-dimethylaniline (PM-3,5-DMA) was prepared and reacted with [Fe(NCS)2] precursor complex in mixed MeOH/EtOH or MeOH/DCM afforded two polymorphisms of a mononuclear iron(II) complex with the formula [Fe(NCS)2(PM-3,5-DMA)2] (1 and 1'). Both polymorphs 1 and 1' crystallize in the same triclinic space group P-1 and contain four (Z = 8) and six (Z = 24) molecules in the asymmetric unit, respectively. The structure of 1 was solved at three temperatures 296, 175, and 100 K, whereas, the structure for 1' was determined at 296 and 85 K. The main differences between the structures of the two polymorphs are found in their crystal packing. Polymorph 1 exhibits gradual, one-step spin transition with T1/2 ~ 150 K. and also displays LIESST effect when irradiated at 10 K temperature.

Keywords: iron(II), Schiff base, spin-crossover