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Ultrafast photochemical processes in the complex *trans*,*trans*,*trans*-[$Pt^{IV}(py)_2(N_3)_2(OH)_2$]

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S1. Experimental details.

Experiments on the ultrafast TA spectroscopy with the registration in visible spectral region were performed using the setup described earlier.[S1,S2] The initial source of light was the Ti-sapphire laser. Samples were excited by *ca.* 100 fs pulses at 320 nm (4^{th} harmonic of the signal wave of TOPAS parametric amplifier). Energy of exciting pulses was *ca.* 1 μJ at the repetition frequency 1 kHz. A portion of exciting laser beam was focused on a cell with heavy water to generate a probe radiation (continuum). The studied solution (total volume 20 ml) was pumped through a 1 mm optical cell. ExciPro program (CDP System Corporation) was used for corrections of the group velocity dispersion (chirp). The corrected experimental data (45 kinetic curves at different wavelengths) were globally fitted with a single set of kinetic parameters using PigSpec software.[S3] This software allows one to simulate instrument response function (IRF) as a Gaussian pulse. The width of the instrument response function is added as the global fitting parameter. Usually the typical width was 140–180 fs. Typically, a three-exponential function was used for fitting.

S2. Ultrafast TA absorption of complex 1 in aqueous solutions

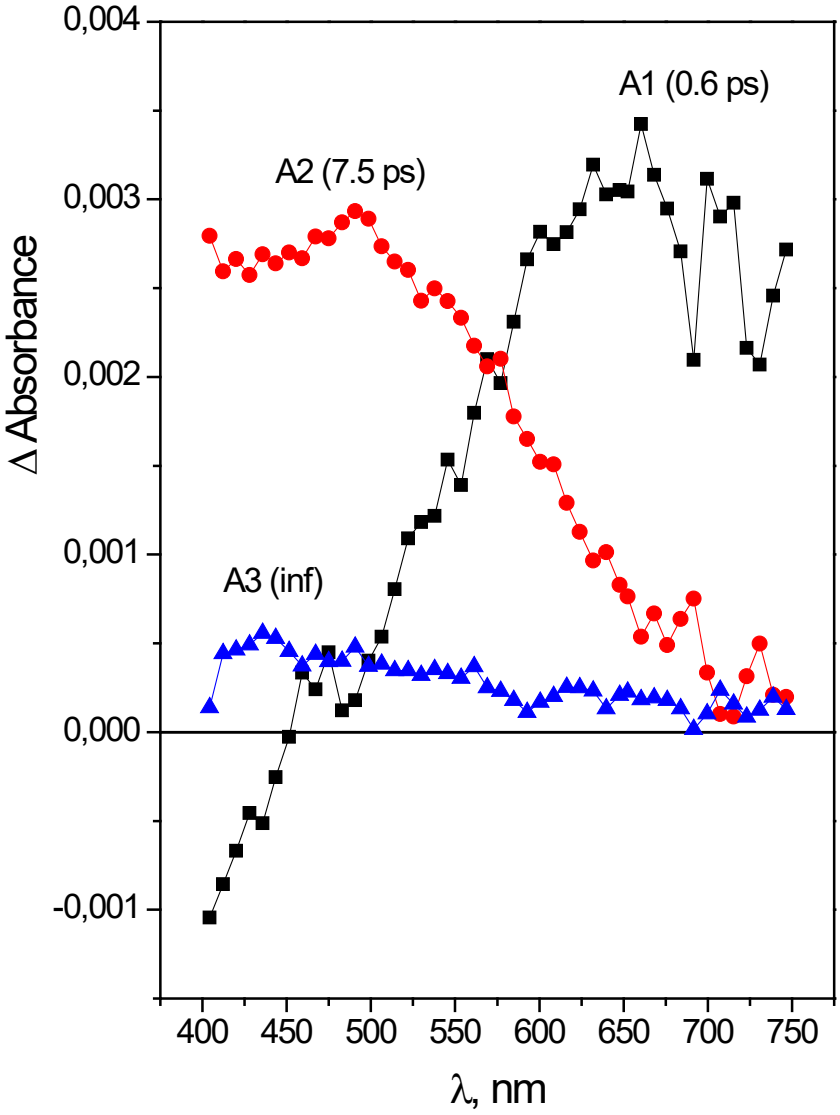

Figure S1 Results of the experiment on the ultrafast kinetic spectroscopy (320 nm) of complex **1** (1.0 mM, 1 mm cell) in water. Amplitudes of the 3-exponential global fitting with the residual [Equation (S1)].

$$\Delta A(\lambda,t) = A_1(\lambda)e^{-\frac{t}{\tau_1}} + A_2(\lambda)e^{-\frac{t}{\tau_2}} + A_3(\lambda)e^{-\frac{t}{\tau_3}} \qquad \text{(S1)}$$

S3. Ultrafast TA absorption of complex 1 in acetonitrile

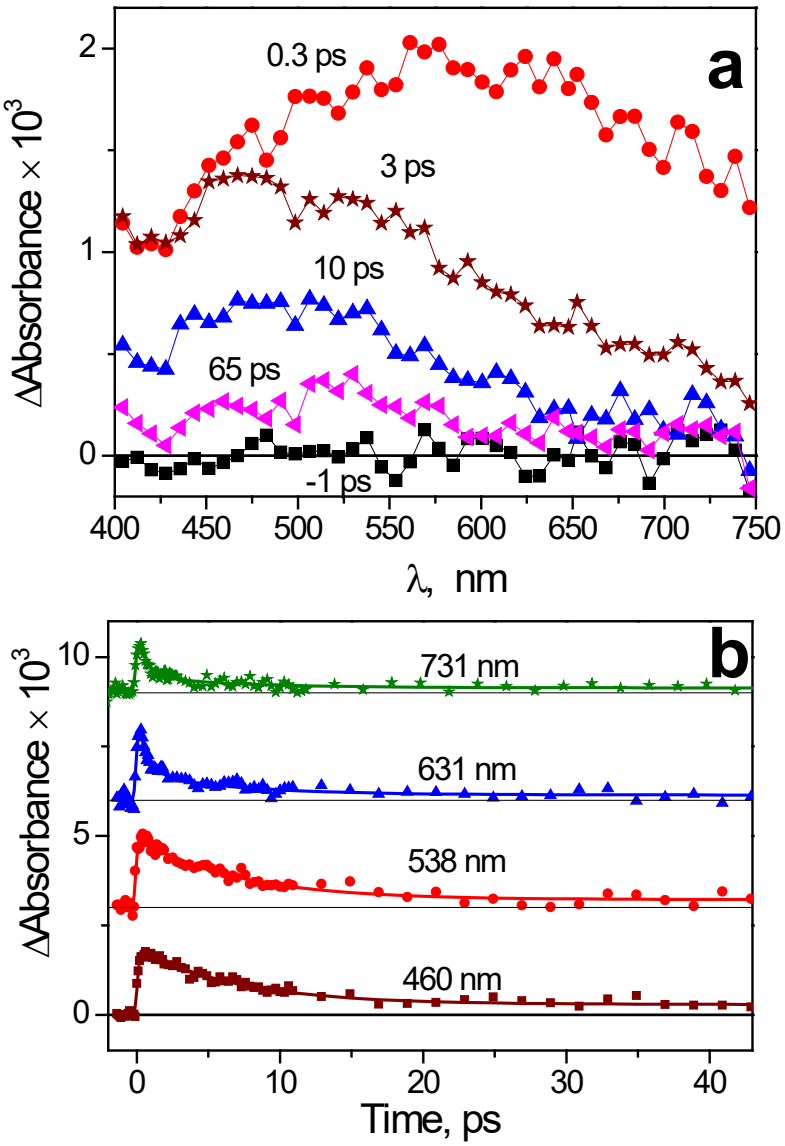

Figure S2 Results of ultrafast TA spectroscopy experiment (excitation at 320 nm) with complex **1** in acetonitrile (0.91 mM, 1 mm cell): (**a**) time evolution of intermediate absorption spectrum; (**b**) experimental kinetic curves (dots) and results of global fitting calculation (solid lines) using 3-exponental function [equation (S1)]. Characteristic lifetimes are $\tau_1 = 0.7 \pm 0.3$ ps, $\tau_2 = 6.9 \pm 1.6$ ps and $\tau_3 = 10000$ ps (infinity).

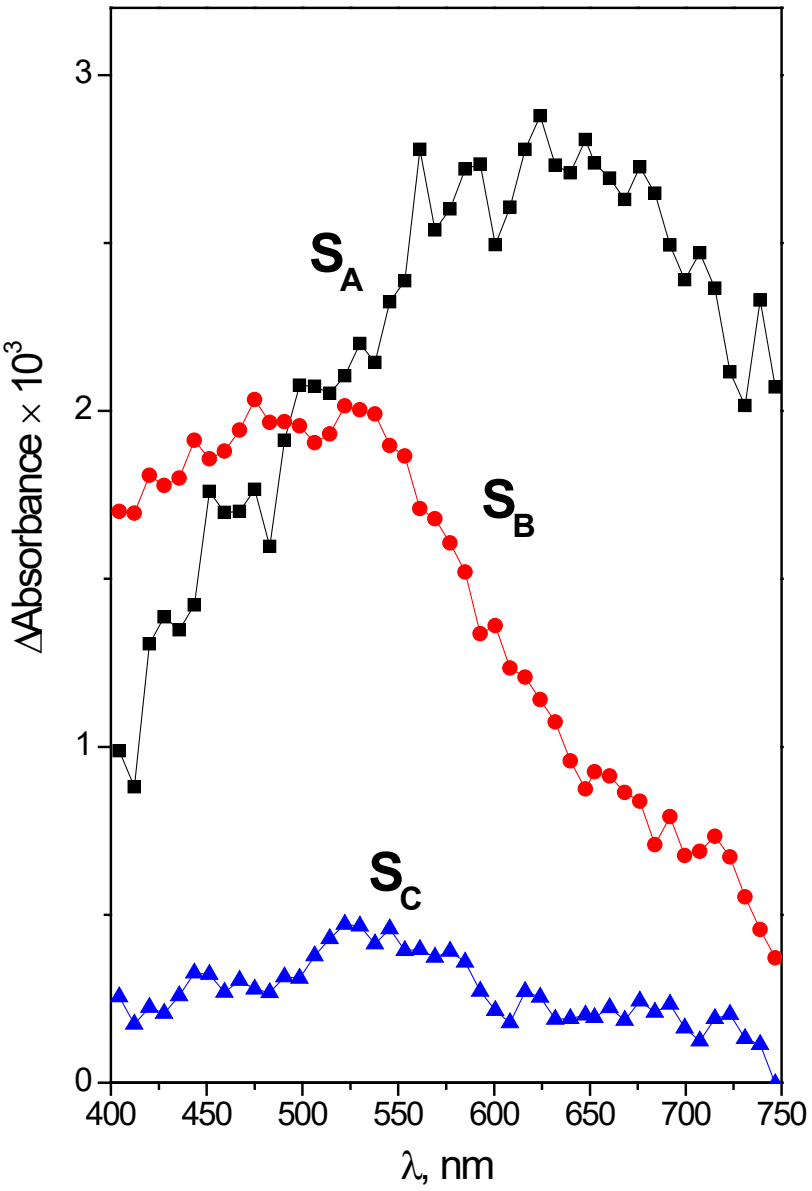

Figure S3 Results of ultrafast TA spectroscopy experiment (excitation at 320 nm) with complex **1** in acetonitrile (0.91 mM, 1 mm cell). Species Associated Difference Spectra (SADS) corresponding to the sequential decay of the transient absorption **A** → **B** → **C**.

References

S1 S. V. Chekalin, *Phys.-Usp.*, 2006, **49**, 634 (*Usp. Fiz. Nauk*, 2006, **176**, 657).

S2 E. M. Glebov, I. P. Pozdnyakov, V. P. Chernetsov, V. P. Grivin, A. B. Venediktov, A. A. Melnikov, S. V. Chekalin and V. F. Plyusnin, *Russ. Chem. Bull.*, 2017, **66**, 418.

S3 N. V. Tkachenko, *Optical Spectroscopy: Methods and Instrumentations*, Elsevier, Amsterdam, 2006.