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Electronic Supplementary Information

Tuning the magneto-optical response of $TbPc_2$ single molecule magnets by the choice of the substrate

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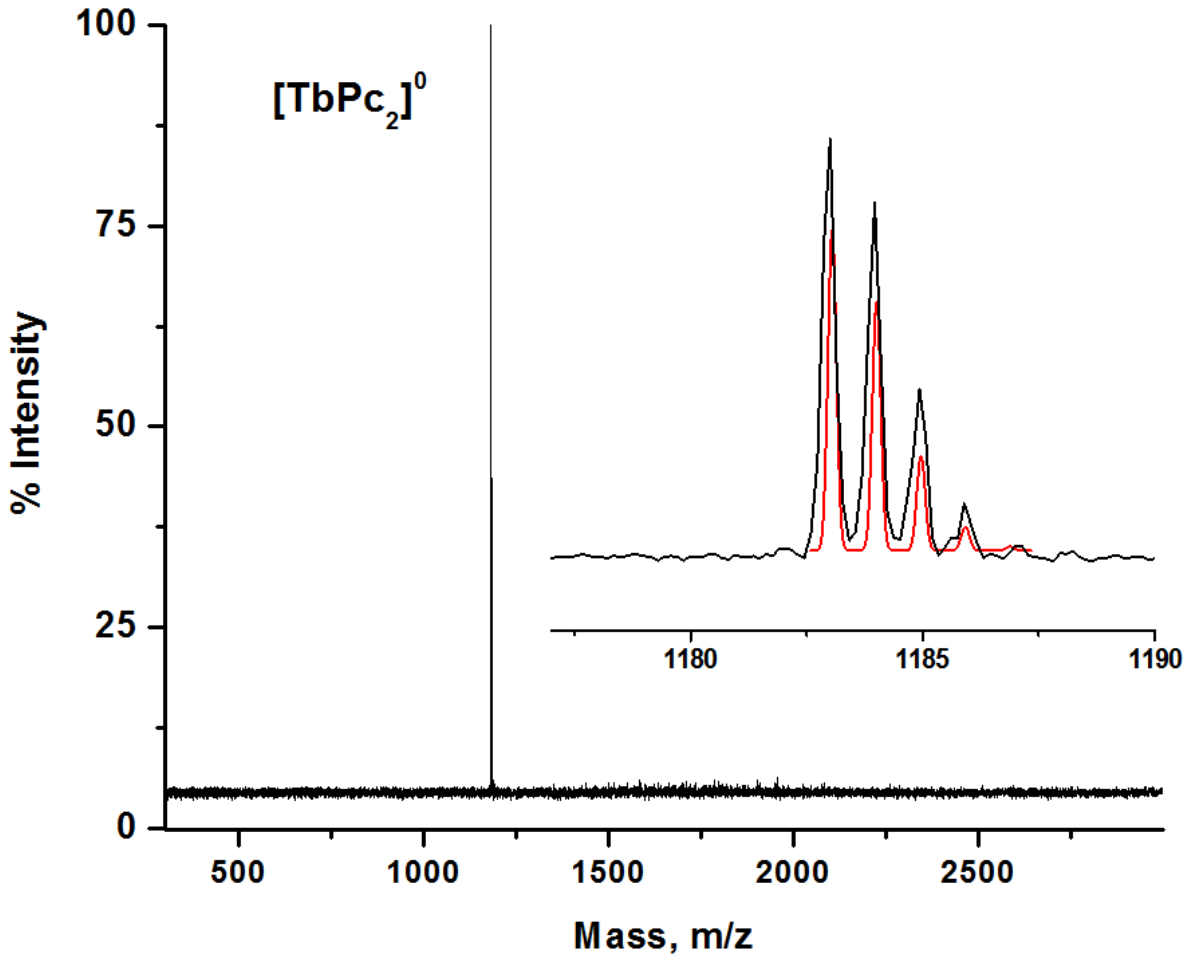

Fig. S1. Maldi-ToF spectrum of $[TbPc_2]^0$ complex shows the parent ions at *m/z* = 1183. An expanded view of the peak shows an isotope distribution, which is in very good agreement with theoretical predictions.

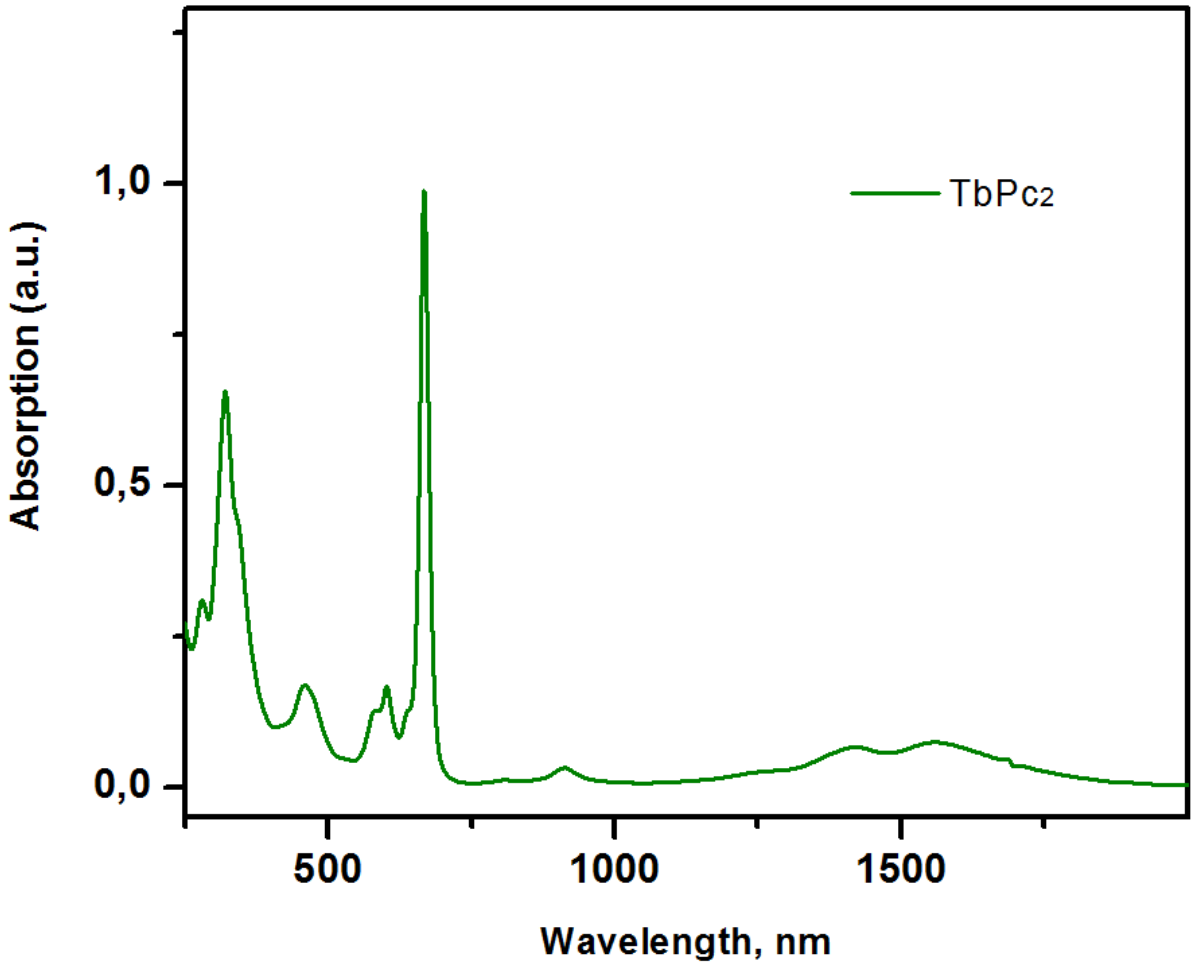

Fig. S2. The UV/vis/nIR absorption spectrum is similar to the neutral green form of $[LnPc_2]^0$ spectra [D. Markovitsi, T.-H. Tran-Thi, *Chem. Phys. Letters*, *1987*, **137**, 107-112] with the most intense band - the Q-band - at 672 nm. In the near-infrared region two main bands are observed at about 906 nm and 1300-1800 nm. The shorter wavelength band is related to the radical part and attributed to the $1e_g(\pi) \rightarrow a_{1u}(\pi)$ transition; the lower-energy band is assigned to an intramolecular charge transfer (CTI). Those signals are the fingerprints of the neutral species $[LnPc_2]^0$ and, thus, confirm their nature. All the near-infrared bands disappear upon reduction by hydrazine hydrate (1% vv).

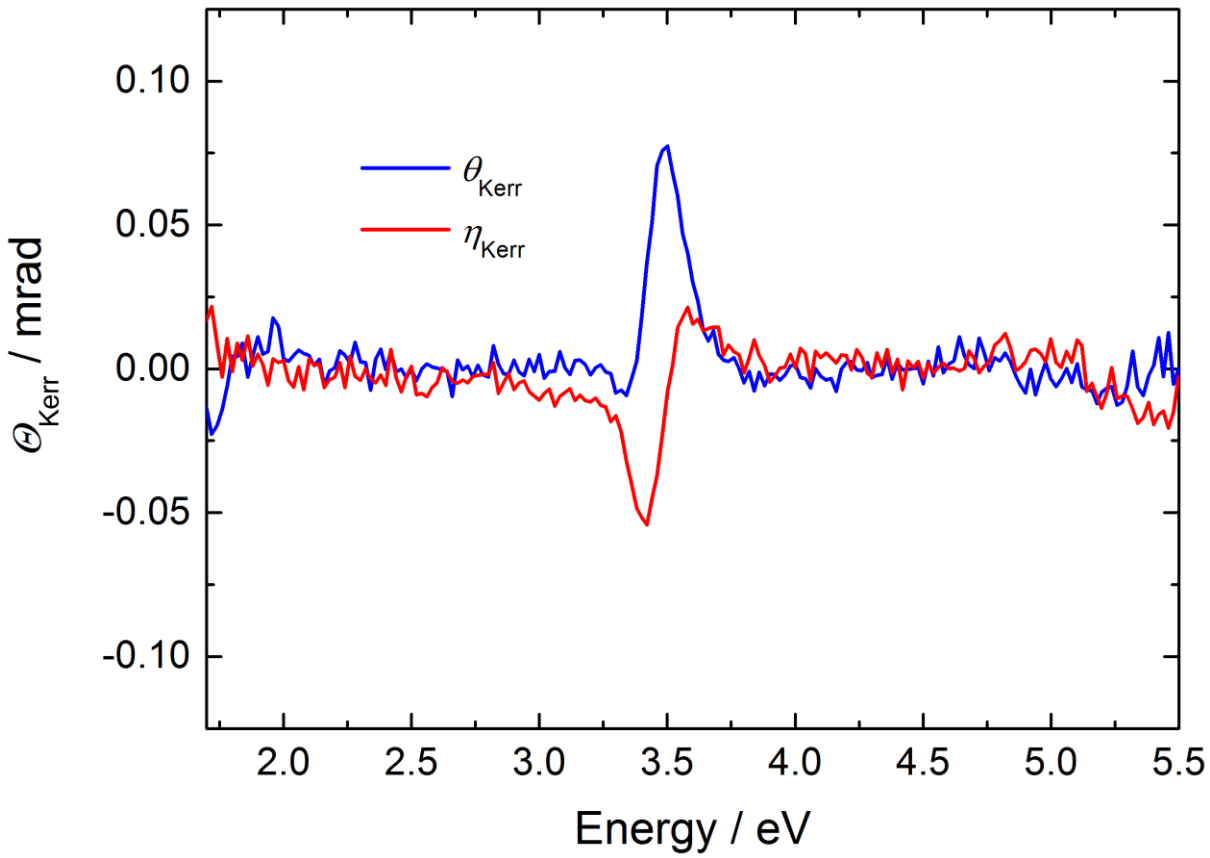

Fig. S3. MOKE spectrum of a blank piece of Si(111) covered with native oxide.

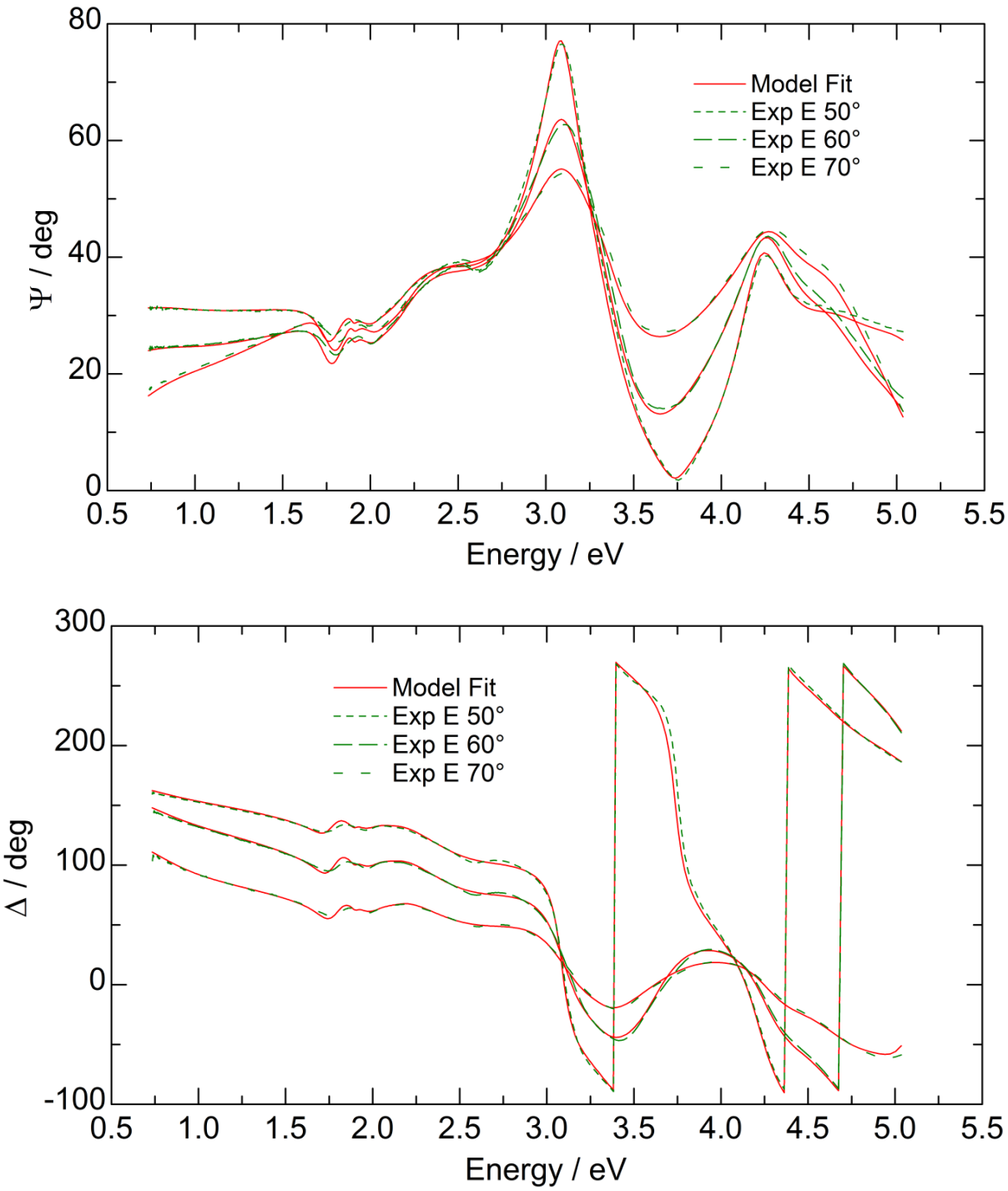

Fig. S4. VASE (Variable Angle Spectroscopic Ellipsometry) spectra of the 62 nm $TbPc_2$ on SiO_2/Si sample. The green dashed lines belong to the experimental data and the red solid lines represent the model fit. The model consists of eleven Gaussian oscillators and a uniaxial anisotropy of the optical properties is assumed.

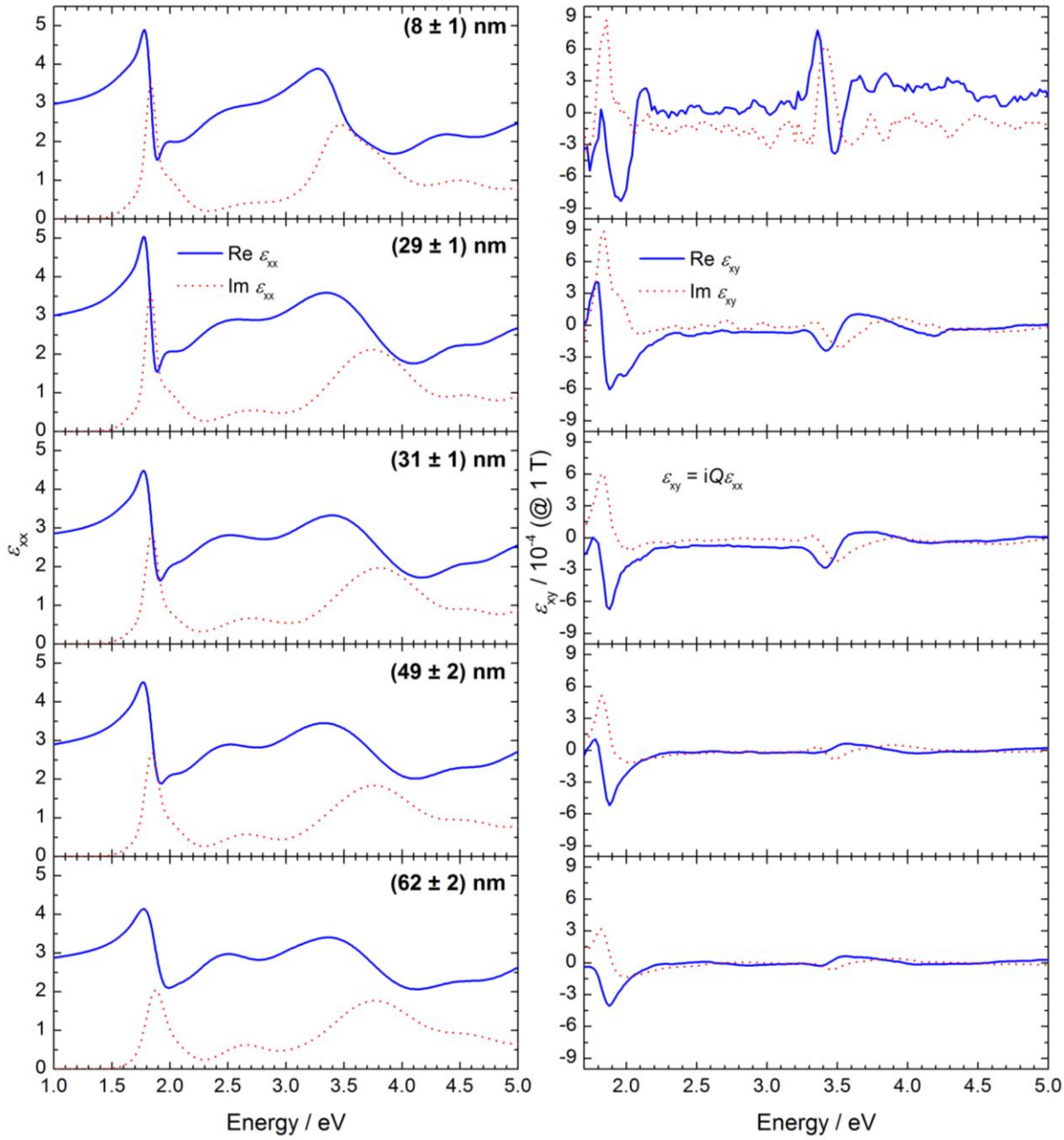

Fig. S5. Diagonal elements of the dielectric tensor in the plane (left) and the calculated off-diagonal elements (right) of $TbPc_2$ thin films on SiO_2/Si with different thicknesses. The values of the components of the dielectric tensor are normalized to a magnetic field of 1 T.

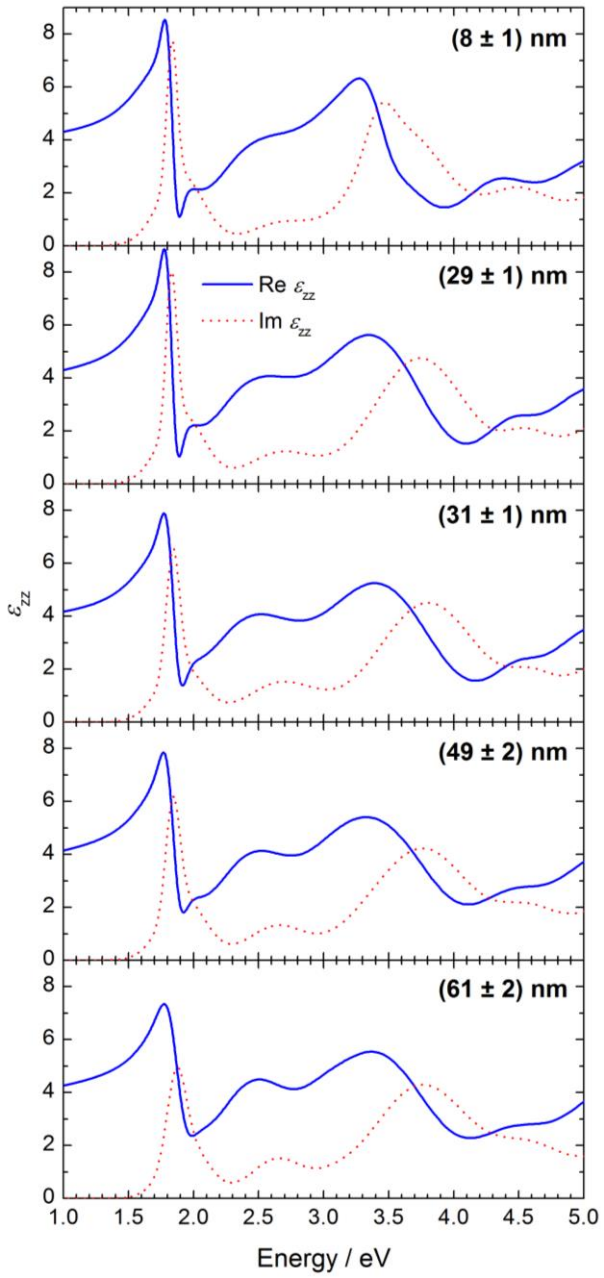

Fig. S6. Out-of-plane components of the dielectric tensor of five $TbPc_2$ films on SiO_2/Si.

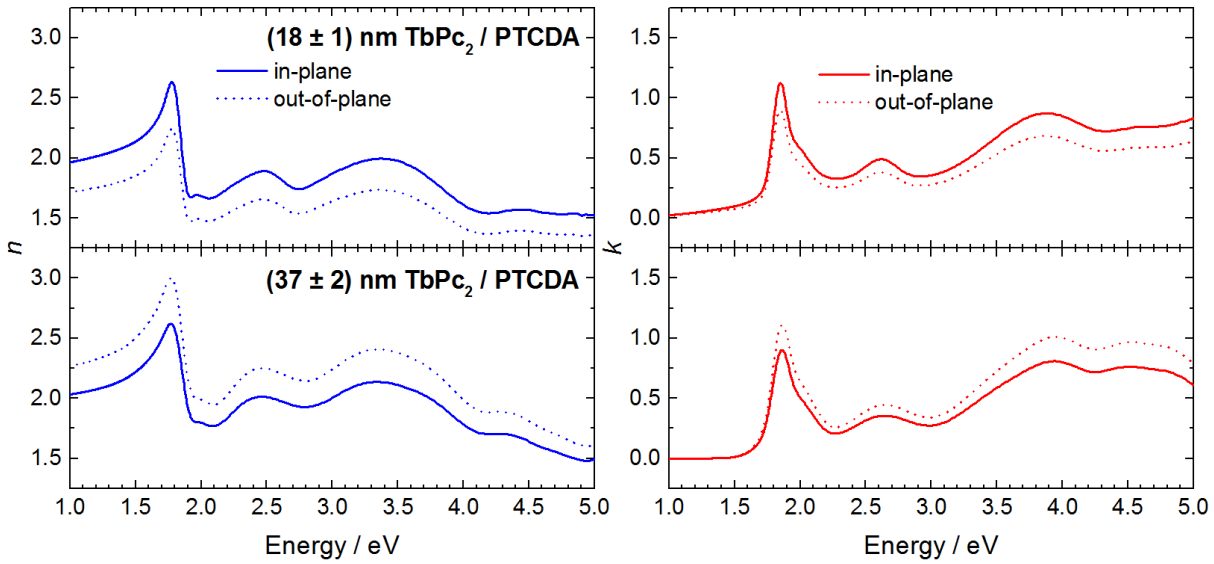

Fig. S7. Complex refractive index of $TbPc_2$ films on PTCDA/SiO_2/Si.

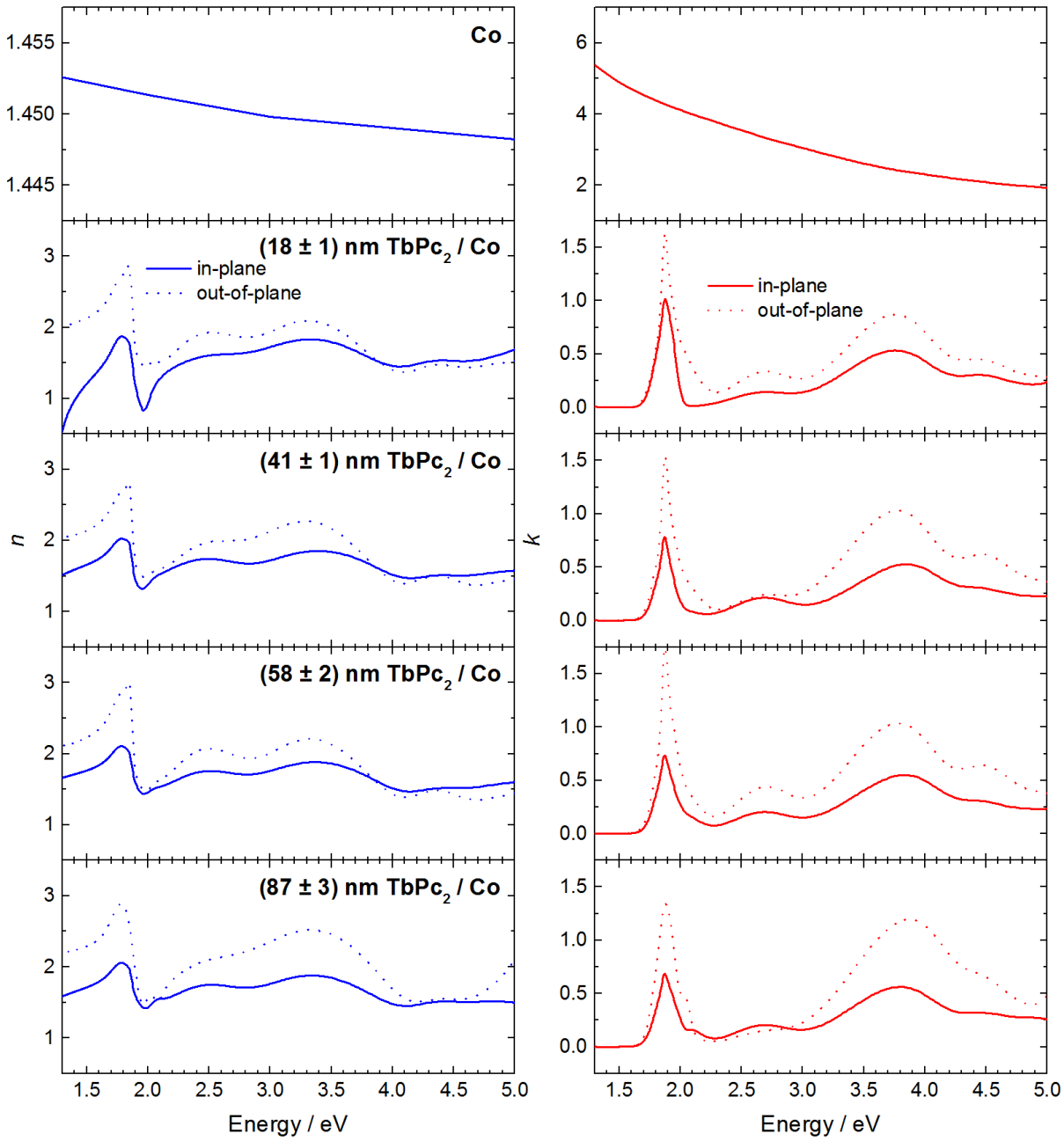

Fig. S8. Complex refractive index of $TbPc_2$ films on $Co/SiO_2/Si$.

Table S1. Summary of the obtained Landé factors and ligand field (LF) parameters from fitting of the hysteresis loops at 1.8 K using the Hamiltonians below. For comparison the parameters for simulation from Ishikawa *et al.* [Ishikawa, N.; Sugita, M.; Okubo, T.; Tanaka, N.; Iino, T.; Kaizu, Y. *Inorg. Chem.*, *2003*, **42**, 2440-2446] are shown. The Stevens coefficients used for the Tb^{3+} ion are $\alpha = -1/99$, $\beta = 2/16335$ and $\gamma = -1/891891$. [Stevens, K.W.H. *Proc. Phys. Soc. A*, *1952*, **65**, 209] For having reasonable LF parameters it was necessary to include the Landé factor as a free parameter in the fitting procedure. Remarkable are the much higher LF parameters in the $TbPc_2$ film on silicon compared to the powder measurement and the simulation. This might be related to lower inter-molecular interaction in the film, compared to the powder.

$$\widehat{H}_{TbPc2} = \widehat{H}_{Zeeman} + \widehat{H}_{lf}$$

$$\widehat{H}_{Zeeman} = g_J \mu_0 \mu_B \boldsymbol{J} \cdot \boldsymbol{H}$$

$$\widehat{H}_{lf} = A_2^0 < r^2 > \alpha O_2^0 + A_4^0 < r^4 > \beta O_4^0 + A_6^0 < r^6 > \gamma O_6^0$$

	Simulation	**43 nm $TbPc_2$/Si (Fit)**	**$TbPc_2$ Powder (Fit)**	**37 nm $TbPc_2$ /PTCDA (Fit)**
$\boldsymbol{g_J}$	1.50	1.58	1.40	0.86
$\boldsymbol{A_2^0}$ / cm^{-1}	415	849	153	849
$\boldsymbol{A_4^0}$ / cm^{-1}	-230	175	59	175
$\boldsymbol{A_6^0}$ / cm^{-1}	35	49	101	49