Figure S1. $^2$H NMR spectra (in CH$_3$CN, 76.75 MHz, 300K) of (a) $m$N3, (b) $m$N4, (c) $m$I3, and (d) $m$I4.
Figure S2. ORTEP (10% probability level) of trans-[Cr(acac)₂(py)₂]+ in tpy. Selected bond lengths (Å): Cr–O(1) 1.950(2), Cr–N(1) 2.080(5).
Figure S3. Absorption spectra of pyridine complexes.
Figure S4. UV-vis absorption spectra of (a) tI3 (———), cI3 (······), and IM3py (— · — ·); (b) tI4 (———), cI4 (······), and IM4py (— · — ·) in acetonitrile at room temperature.
Figure S5. The difference absorption spectra of the NIT3py and IM3py complexes from the corresponding pyridine complexes.
Figure S6. The difference absorption spectra of the NIT4py and IM4py complexes from the corresponding pyridine complexes.
Figure S7. Temperature dependence of the magnetic susceptibilities in the form of $\chi_mT$ vs. $T$ for (a) $tN3$ (O) and $tN4$ (■), (b) $cI3$ (O) and $cI4$ (■), and (c) $tI3$ (O) and $tI4$ (■).