Magnetic Bistability: from Microscopic to Macroscopic Understandings of Hysteretic Behavior using \textit{ab initio} Calculations

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\section*{Introduction}

- Magnetic bistability: \textit{spin-crossover} phenomenon
- Thermal hysteresis often attributed to weak intermolecular interactions (i.e. Van der Waals, H-bonds, \textit{x}-stacking,...)
- Regular solution-based theories introduce an intermolecular parameter $\gamma$
- $\gamma_{\text{LAT}}$ compatible with $\Delta T \approx 50K$?

\section*{Microscopic inspection}

- A preliminary study on a model complex: $[\text{Fe(NCH}_3\text{)}_2]^+$
- Wavefunction-based calculations (CAS8CPP/CASPT2)
- Determination of the electronic distribution
- Observations confirmed on synthetic compounds

\section*{Temperature induced electron trapping}

\begin{align*}
G &= G_{\text{out}} + G_{\text{in}} + G_{\text{trapping}}
\end{align*}

Physical origin of the hysteresis:
- LS Madelung field tends to favor the charge transfer
- HS Madelung field tends to block the opposite transfer: the charge is trapped

\section*{Application to synthetic systems}

- 3 polymorphs for 1 complex with very different behaviors
- X-ray structures at high-temperature for polymorphs A, B and C

\begin{align*}
\Delta V_{\text{LS}} &= 165 \text{ cm}^{-1} \\
\Delta V_{\text{HS}} &= -370 \text{ cm}^{-1}
\end{align*}

No electron-trapping for polymer A at high-temperature

\section*{Conclusions and perspectives}

- Proposed model gives an account of the hysteresis cycle with a physical interpretation of the phenomenon: electron trapping
- All the parameters are accessible by means of \textit{ab initio} calculations
- The electrostatic contributions cannot be neglected
- Experimentation of the model on synthetic model, but lack of experimental data on low-temperature structure

\section*{How can one act on $\gamma$?}

\begin{itemize}
  \item i) via $V_{\text{e}}$ i.e. packing
  \item ii) via $Q_{\text{e}}$ i.e. coordination sphere
\end{itemize}

About FeN$_2$O$_2$ systems?

\section*{Improvement of the model}

- Distorted embedding: $\gamma$ scaling?
- Towards 1D and 2D-like environments
- Beyond mean field approximation: Monte-Carlo simulations