Magnetocaloric properties of gadolinium based heterometallic molecules studied by the finite-temperature Lanczos method

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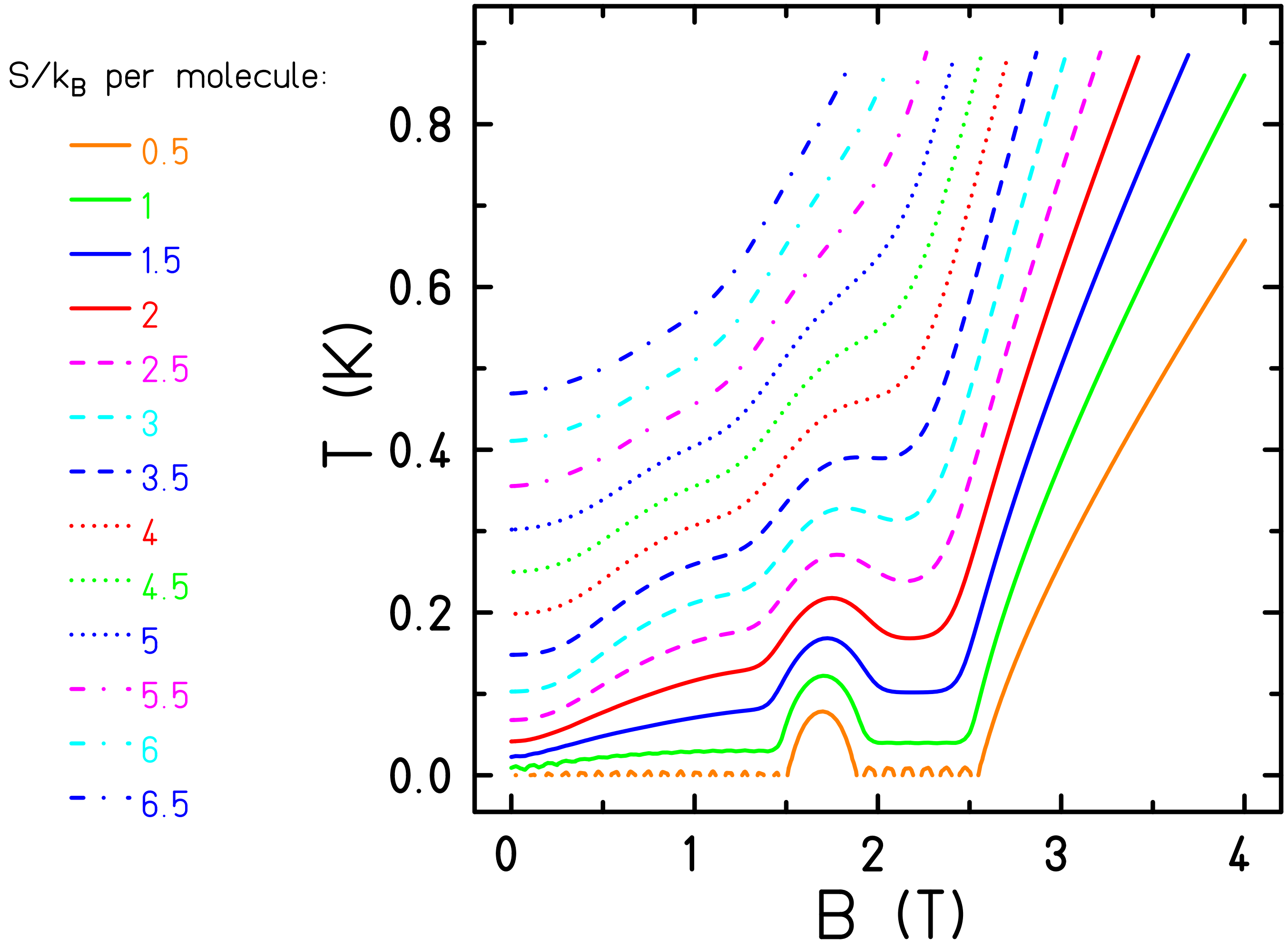
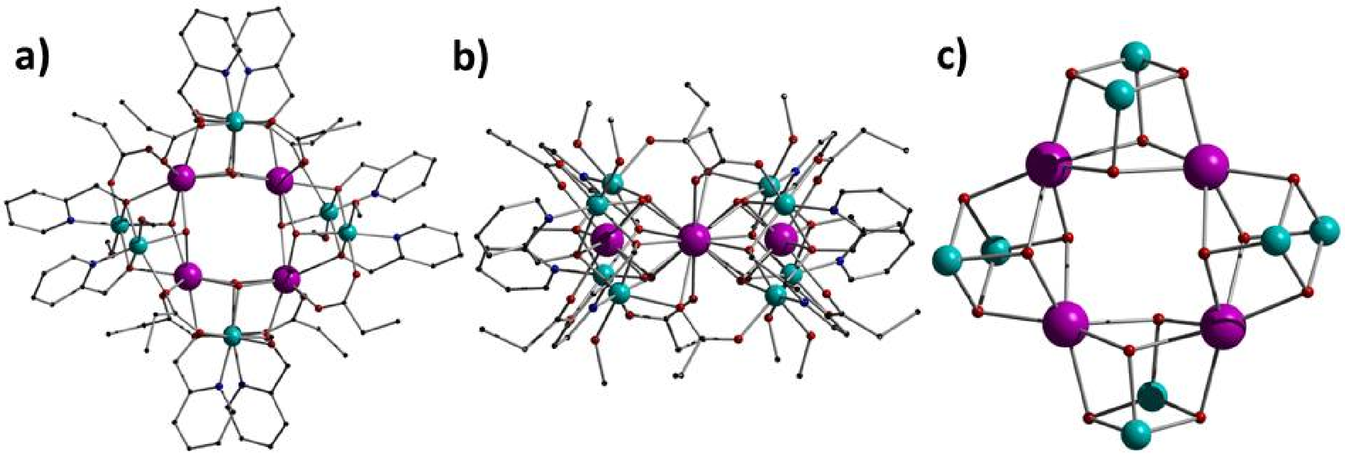
Magnetocaloric effect – cooling rate

$$\left(\frac{\partial\, T}{\partial\, B}\right)_S = -\frac{T}{C}\left(\frac{\partial\, S}{\partial\, B}\right)_T$$

Magnetocaloric effect – isentropes

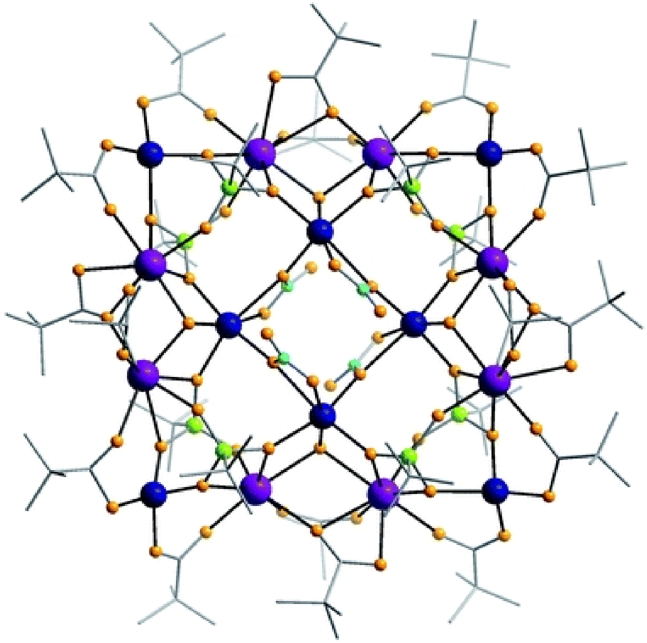


Magnetocaloric effect – Gd_4Ni_8



T. N. Hooper, J. Schnack, St. Piligkos, M. Evangelisti, E. K. Brechin, Angew. Chem. Int. Ed. (2012) in print.

Magnetocaloric effect – Why Gd compounds?



- High spin of $s = 7/2$;
- Weak exchange $\Rightarrow$ high density of states;
- Can vary the entropy with moderate fields.
- But large Hilbert spaces!
  Exact modeling impossible.

Yan-Zhen Zheng, Marco Evangelisti, Richard E. P. Winpenny, Chem. Sci. **2**, 99-102 (2011)

Model Hamiltonian

$$\underset{\sim}{H} = -2 \sum_{i<j} J_{ij}\, \underset{\sim}{\vec{s}}(i) \cdot \underset{\sim}{\vec{s}}(j) \quad + \quad g\, \mu_B\, B \sum_{i}^{N} \underset{\sim}{s}_z(i)$$

Heisenberg Zeeman

Finite-temperature Lanczos Method I

$$Z(T,B) = \sum_\nu \langle\,\nu\,|\,\exp\left\{-\beta \underset{\sim}{H}\right\}|\,\nu\,\rangle$$

$$\langle\,\nu\,|\,\exp\left\{-\beta \underset{\sim}{H}\right\}|\,\nu\,\rangle \approx \sum_n \langle\,\nu\,|\,n(\nu)\,\rangle \exp\left\{-\beta\epsilon_n\right\} \langle\,n(\nu)\,|\,\nu\,\rangle$$

$$Z(T,B) \approx \frac{\text{dim}(\mathcal{H})}{R}\sum_{\nu=1}^{R}\sum_{n=1}^{N_L}\exp\left\{-\beta\epsilon_n\right\}|\langle\,n(\nu)\,|\,\nu\,\rangle|^2$$

- $|\,n(\nu)\,\rangle$ n-th Lanczos eigenvector starting from $|\,\nu\,\rangle$
- Partition function replaced by a small sum: $R = 1\ldots 10, N_L \approx 100$.

J. Jaklic and P. Prelovsek, Phys. Rev. B **49**, 5065 (1994).

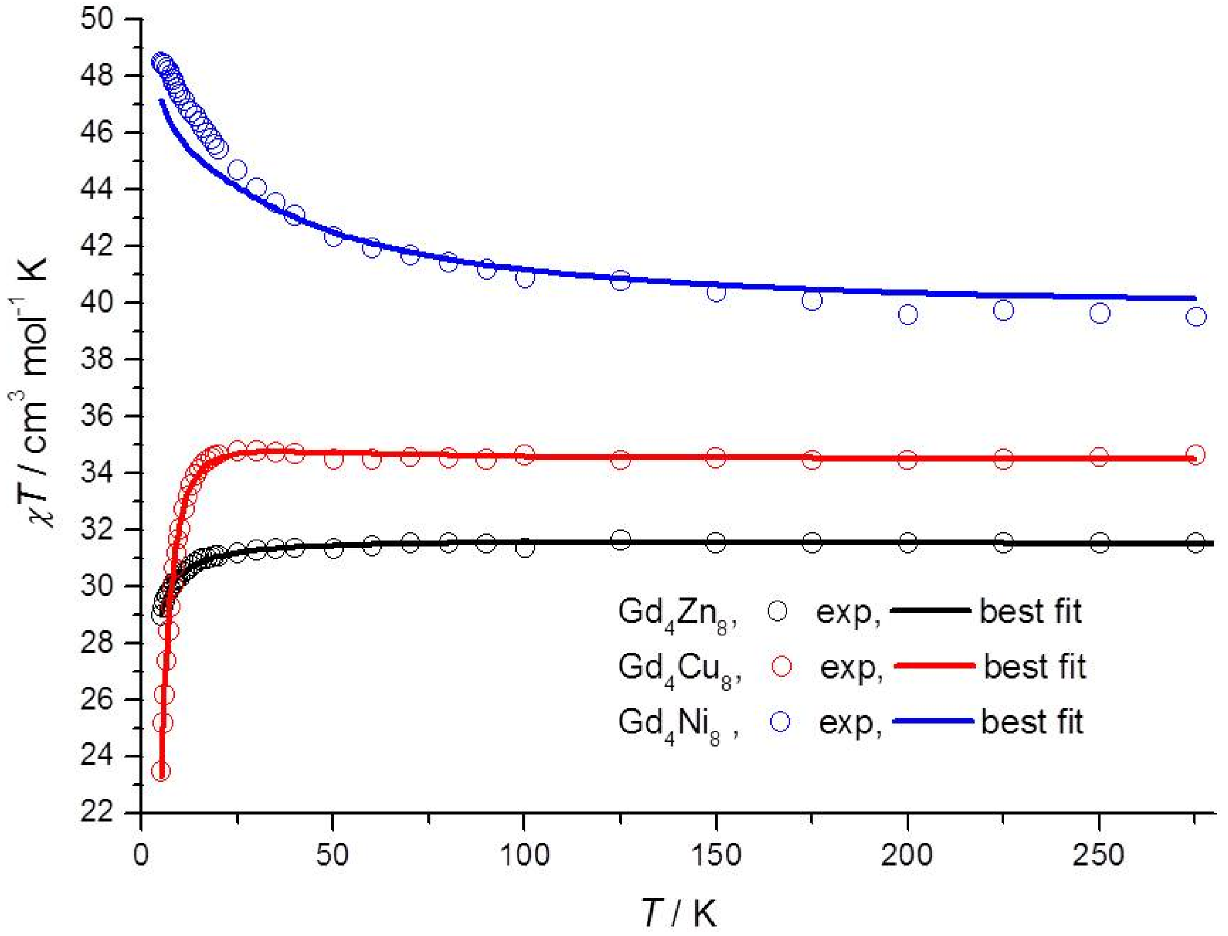
Finite-temperature Lanczos Method II

$$Z(T,B) \approx \sum_{\Gamma} \frac{\dim(\mathcal{H}(\Gamma))}{R_\Gamma} \sum_{\nu=1}^{R_\Gamma} \sum_{n=1}^{N_L} \exp\{-\beta\epsilon_n\} |\langle\, n(\nu,\Gamma)\,|\,\nu,\Gamma\,\rangle|^2$$

- Approximation better if symmetries taken into account.
- Γ denotes the used irreducible representations.

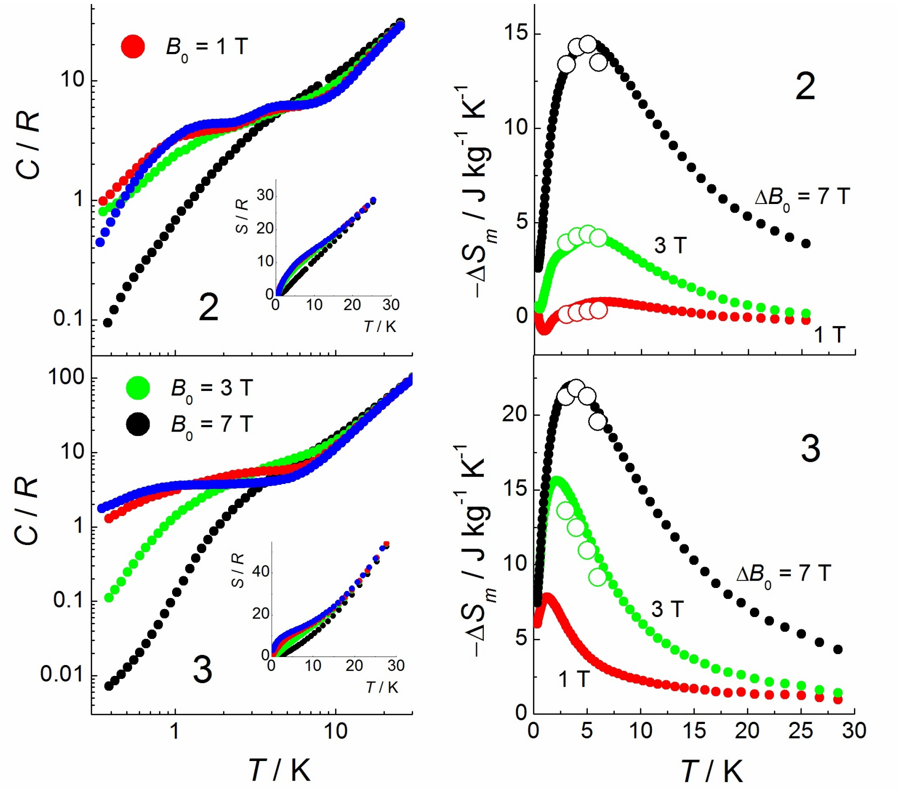
J. Schnack and O. Wendland, Eur. Phys. J. B **78** (2010) 535-541

Gd_4M_8 – Susceptibility



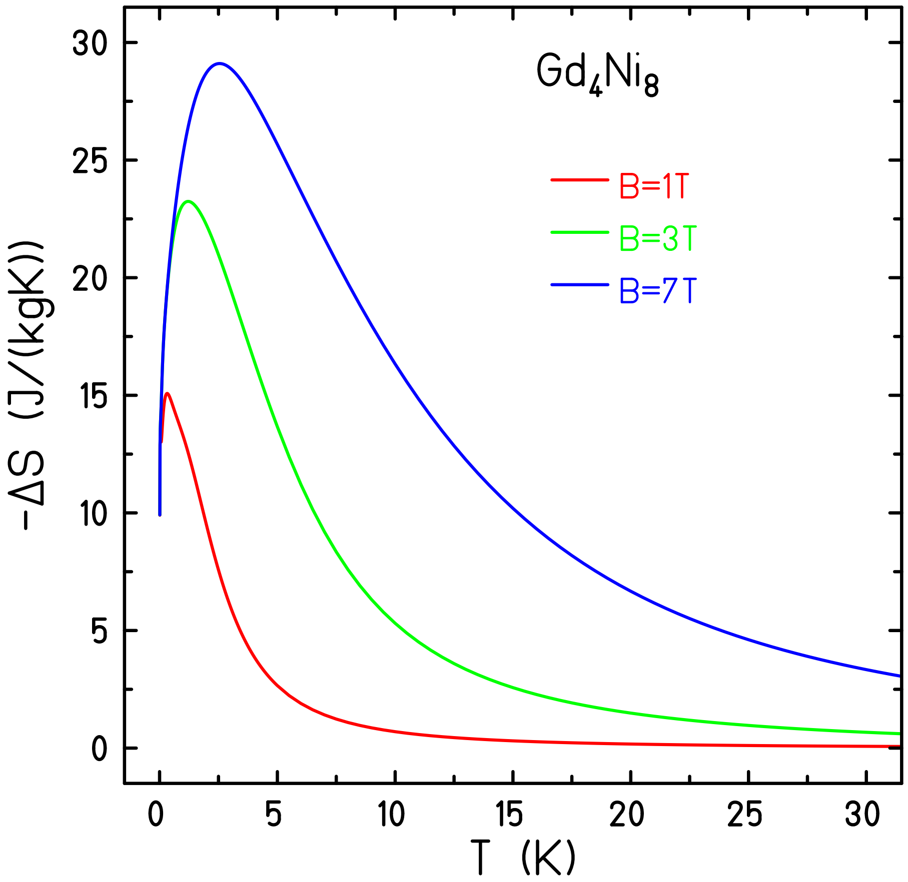
T. N. Hooper, J. Schnack, St. Piligkos, M. Evangelisti, E. K. Brechin, Angew. Chem. Int. Ed. (2012) in print.

Gd$_4$M$_8$ – experimental $C(T,B)$ and $S(T,B)$

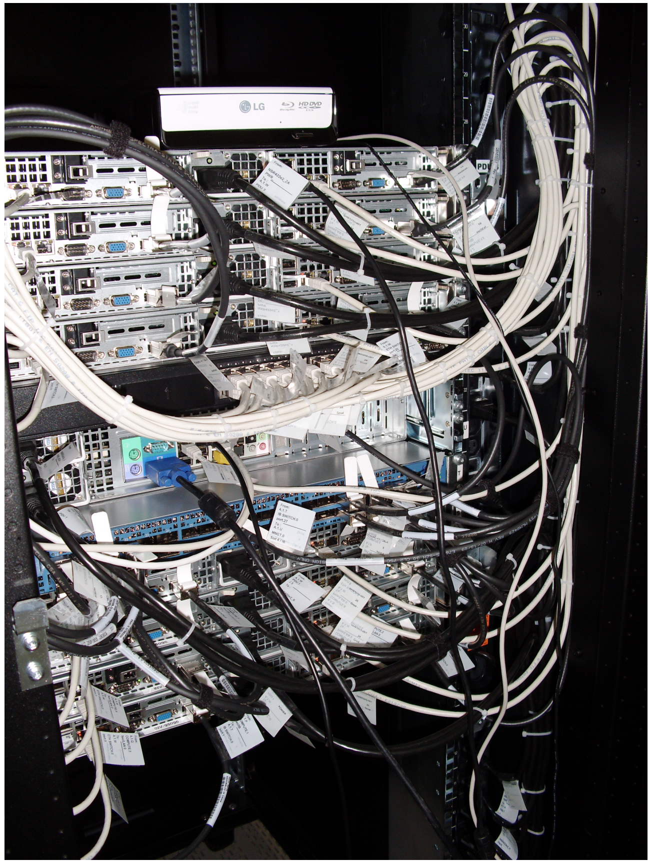


T. N. Hooper, J. Schnack, St. Piligkos, M. Evangelisti, E. K. Brechin, Angew. Chem. Int. Ed. (2012) in print.

Gd$_4$Ni$_8$ – theoretical $S(T, B)$



Summary



- Magnetocalorics is an interesting application for magnetic molecules.
- Problem: Large dimension of Hilbert spaces.
- Finite-temperature Lanczos is a good approximate method for Hilbert space dimensions smaller than 10^{10}.
- Extension towards anisotropic Hamiltonians under way.

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Thank you very much for your attention.

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