Tight-binding spin dynamics and tight-binding Monte Carlo: a study on BCC Fe

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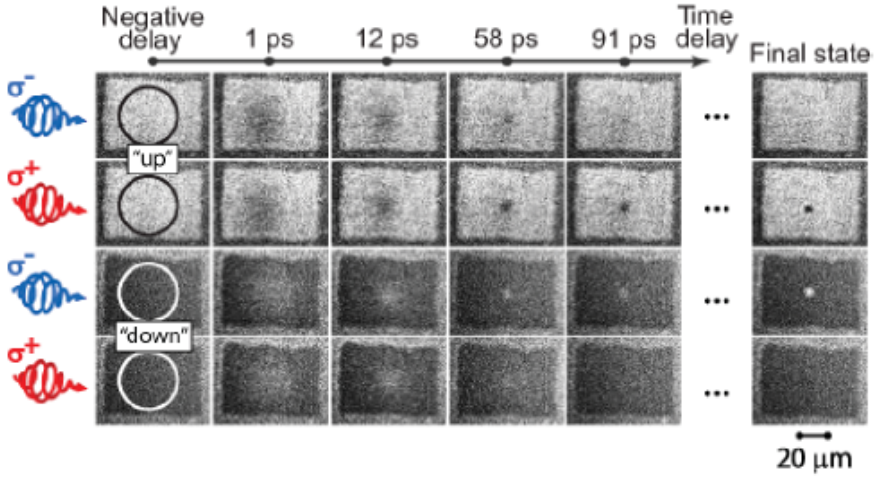
Motivation

- **Study of laser-induced magnetization dynamics**
  - Scientific interest: excitation on the time scale of exchange
  - Technological interest: magnetization reversal within 100ps



K. Vahaplar *et al*, 2009 Phys Rev Lett **103**, 117201

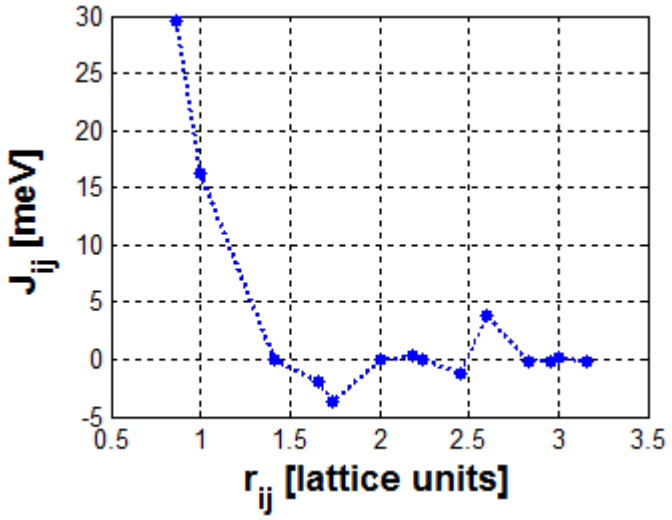
→ **Accurate atomistic description outside equilibrium needed**

- System studied: BCC Fe for T>0K

Localized versus delocalized description

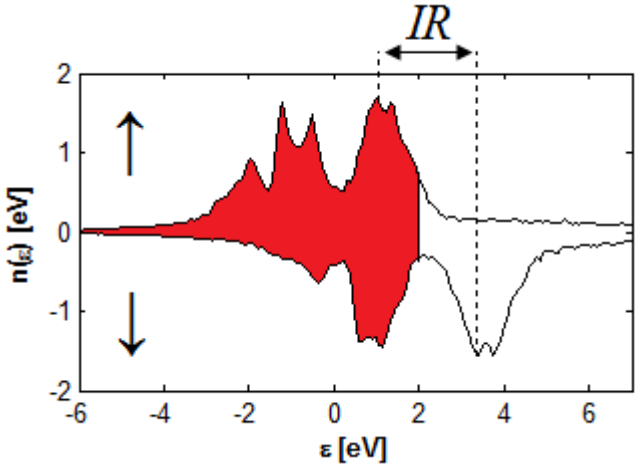
Heisenberg model

$$E = -\sum_i \sum_{j \neq i} J_{ij} \vec{m}_i \cdot \vec{m}_j$$



Stoner model

$$E_{\downarrow}\left(\vec{k}\right) = E\left(\vec{k}\right) + \frac{IR}{2} \quad \textbf{and:} \quad E_{\uparrow}\left(\vec{k}\right) = E\left(\vec{k}\right) - \frac{IR}{2}$$



Stoner criterium (T=0K): $I\tilde{D}\left(E_F\right) > 1$

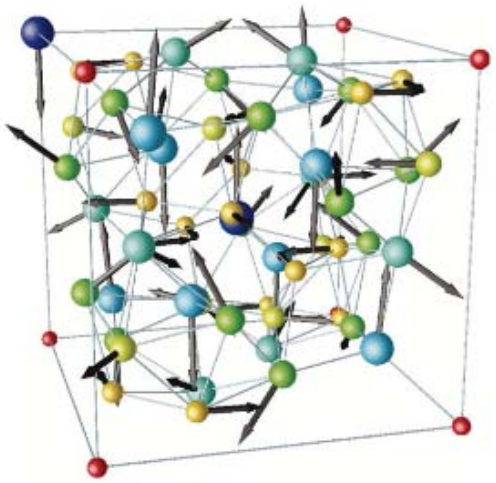
Characteristics

Heisenberg model	*Stoner model*
• Reasonable T_C • Qualitative correct χ above T_C	• Failure description for T>0K • No Curie-Weiss law above T_C

- **However Heisenberg model lacks…**
  - Itinerant character of electrons around ε_F
  - Large cohesive energies and specific heat

  $\rightarrow \quad E \neq -\sum_i \sum_{j \neq i} J_{ij} \vec{m}_i \cdot \vec{m}_j$

- **Unified approach needed:**
  - Rigorous description of non-collinear state
  - Electronic structure via tight binding



Picture taken from:
Noncollinear Magnetism,
David Hobbs and Jürgen hafner

Tight binding

- **Free energy:**

Pinning of charge *Constraint field*

$$F = E_0 + \frac{U_{LCN}}{2}\sum_i \left(n_i - n_i^0\right)^2 - \frac{1}{4}\sum_i\sum_\mu I_{i\mu}\left|\vec{m}_{id}\right|\left|\vec{m}_{i\mu}\right| - \sum_i\sum_\mu \vec{B}_{i\mu}\cdot\vec{m}_{i\mu}$$

spin independent kinetic + potential energy *Stoner model*

- ***F* in terms of *ψ*:**

$$E_0 = \sum_{\vec{k},n} f\left(\varepsilon_{\vec{k},n}\right)\sum_{\mu,\nu}\sum_{i,j}\left(\psi^\dagger_{\vec{k},n}\right)_{i\mu}\cdot\left(\psi_{\vec{k},n}\right)_{j\nu}\cdot\left[\hat{H}_0\right]^{j\nu}_{i\mu}$$

$$\rho^{\sigma\sigma'}_{i\mu} = \sum_{\vec{k},n} f\left(\varepsilon_{\vec{k},n},\varepsilon_F\right)\left(\psi^\dagger_{\vec{k},n}\right)^{\sigma}_{i\mu}\left(\underline{\underline{S}}_{\vec{k}}\cdot\psi_{\vec{k},n}\right)^{\sigma'}_{i\mu}$$

$$\rightarrow$$

$$c_i = tr\left[\hat{\rho}_{i\mu}\right]$$

$$\left[\vec{m}_{i\mu}\right]_\alpha = tr\left[\hat{\rho}_{i\mu}\cdot\hat{\sigma}_\alpha\right]$$

Parametrization of $\hat{H}_0$: M. J. Mehl and D.A. Papaconstantopoulos, 1996, Phys. Rev. B, **54** ***and*** J. C. Slater and G. F. Koster, 1954, Phys Rev **94**, 1498-1524

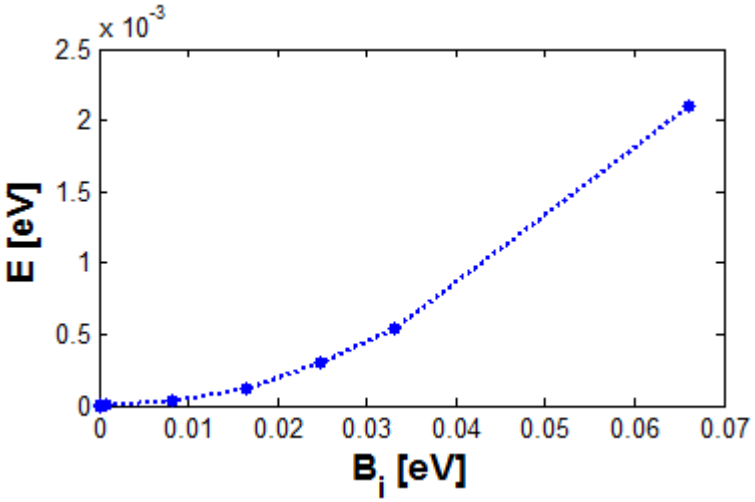
Variational Hamiltonian

- **Minimalisation free energy:**

$$\frac{\partial F}{\partial \left(\psi_{\vec{k},n}\right)_{i\mu}^{\sigma}} = 0 \qquad \textit{under the constraint:} \qquad \sum_{i\mu\sigma} \left(\psi_{\vec{k},n}^{\dagger}\right)_{i\mu}^{\sigma} \left(\psi_{\vec{k},n}\right)_{i\mu}^{\sigma} = 1$$

- **Obtained Hamiltonian:**

$$\left[\hat{H}_{TB}\right]_{i\mu}^{j\nu} = \left[\hat{H}_{0}\right]_{i\mu}^{j\nu} + U \cdot \left(n_i - n_i^0\right)\delta_{ij}\delta_{j\nu} - \frac{1}{4}\left(I_{i\mu}\vec{m}_{i\mu} \cdot \hat{\sigma} - \vec{B}_i \cdot \hat{\sigma}\right) - \frac{1}{4}\hat{S} \cdot \left(I_{i\mu}\vec{m}_{i\mu} \cdot \hat{\sigma} - \vec{B}_i \cdot \hat{\sigma}\right)$$

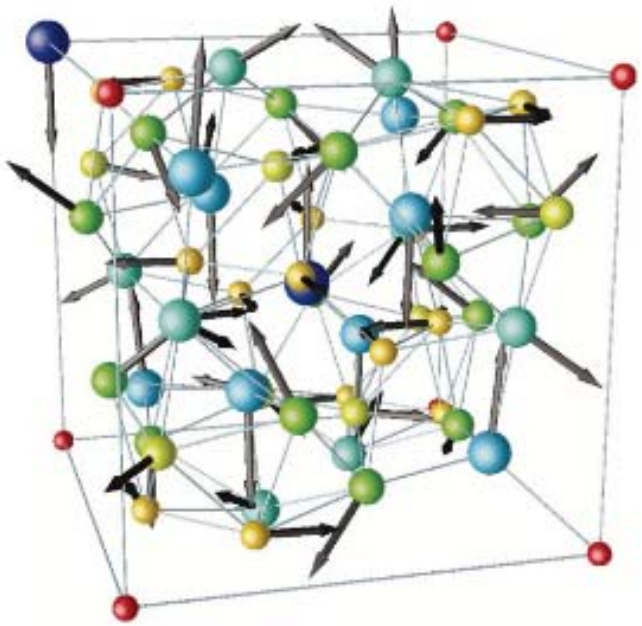


Description of a NC state

- **Problem to be solved:**

$$\hat{H}_{TB} \cdot \vec{c} = E\hat{S} \cdot \vec{c}$$

$$|\psi\rangle = \sum_{\vec{n},i,\upsilon} c_{\vec{n}i\upsilon} \cdot |\vec{n},i,\upsilon\rangle$$

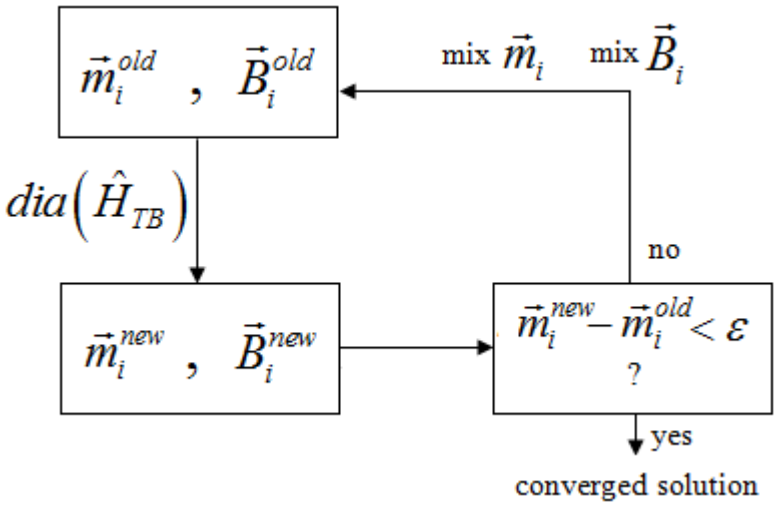


- **Magnetic moment directions:**

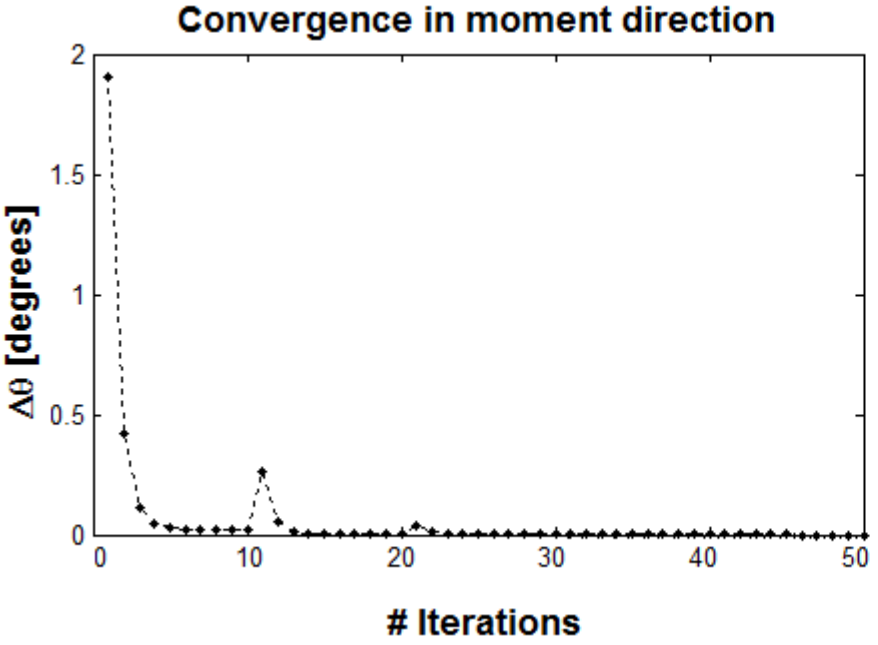
$$\hat{\rho}_{i\upsilon}^{\sigma\sigma'} = \sum_{k} f\left(\varepsilon_k, \varepsilon_F\right)\left(\vec{c}_k^{\dagger}\right)_{i\upsilon}^{\sigma} \cdot \left(\hat{S}\vec{c}_k\right)_{i\upsilon}^{\sigma'}$$

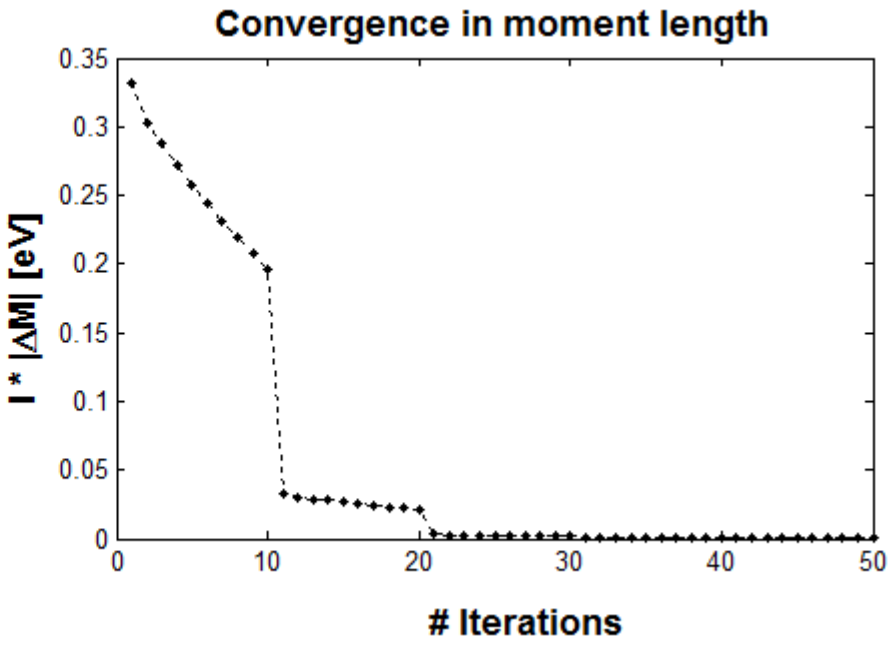
$$\left[\vec{m}_{i\upsilon}\right]_{\alpha} = tr\left[\hat{\rho}_{i\upsilon} \cdot \hat{\sigma}_{\alpha}\right]$$

- **Self-consistency for fixed moment directions!**



Torques





- **Validity adiabatic approximation?**
  - → Spin wave frequencies whole BZ ~ 0.1eV/ħ – 1eV/ħ (~ $k_B T_C/\hbar$)
  - → Electron hopping frequency *3d* bandwidth (W/ħ) (W_{Fe} ~ 5eV)
  - → Better look at*: $\vec{B}_i$

- **Torque**:** $\frac{1}{2}\frac{d}{dt}\langle\hat{\sigma}_i\rangle = \vec{m}_i \times \left(-\vec{B}_i\right)$ → $\frac{\hbar}{2}\frac{d}{dt}\langle\hat{\sigma}_i\rangle = \frac{i}{2}\left\langle\left[\hat{H}_{Hub}, \hat{\sigma}_i\right]\right\rangle$

* D. M. Edwards, JMMM 45 (1984) 151-156

** L.M. Small and V. Heine., 1984 *J. Phys. F: Met. Phys.* **14** 3041

Spin dynamics and Monte Carlo

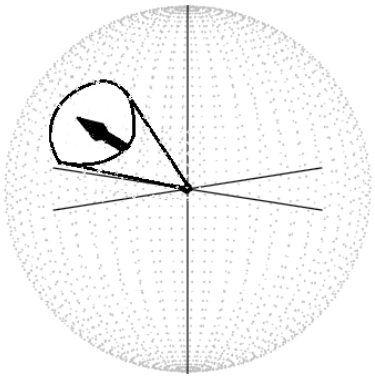
- **Add temperature b_i and damping λ:**

$$\frac{d\vec{e}_i}{dt} = -\left[\vec{e}_i \times \left(-\vec{B}_i + b_i\right)\right] - \lambda\left[\vec{e}_i \times \left(\vec{e}_i \times \left(-\vec{B}_i + \vec{b}_i\right)\right)\right]$$

$$\left\langle \vec{b}_{fluc}^{\,i,\mu}(t) \right\rangle = 0 \quad \textbf{and} \quad \left\langle b_{i,\mu}^{fluc}(t)\, b_{j,\nu}^{fluc}(s) \right\rangle = 2D\delta_{ij}\delta_{\mu\upsilon}\delta(t-s) \quad \textbf{with:} \quad D = \frac{\lambda k_B T}{\gamma |m|}$$

- **Computational effort:**
  - Number of time steps: 10^3 - 10^6
  - Per time step: ~ 10 iterations
  - Per iteration: diagonalization of $\hat{H} \sim N_K \cdot N_A^3$

- **Thermodynamic properties: Monte Carlo**
  - Reduces number of time steps
  - Metropolis criterion: $\exp\left(\left(E_{old} - E_{new}\right) / k_B T\right) > \nu$

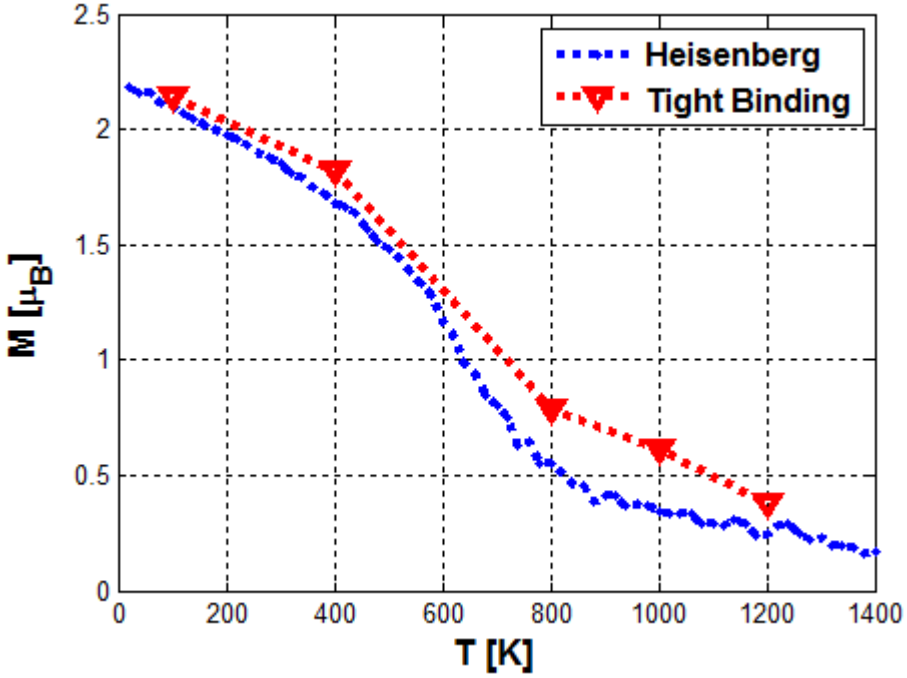


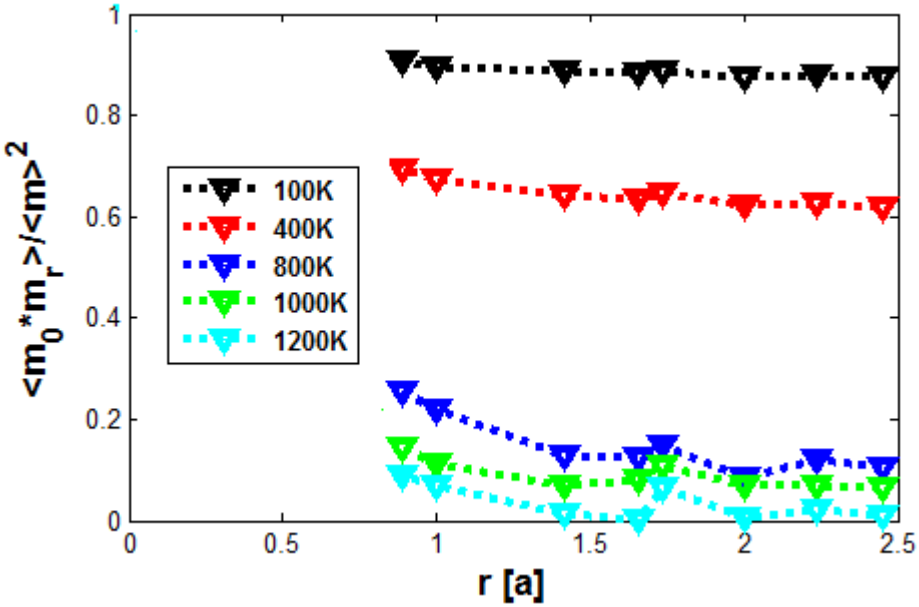
Monte Carlo results

System:

- 125 atoms per unit cell
- 8 k-points
- 500MC steps per T
- 4 averages

T [K]	<|m|> [μ_B]	<|B|> [eV]
100	2.271 ± 0.026	0.031 ± 0.016
400	2.279 ± 0.032	0.056 ± 0.028
800	2.115 ± 0.080	0.064 ± 0.034
1000	2.066 ± 0.083	0.059 ± 0.031
1200	2.049 ± 0.088	0.054 ± 0.029





Conclusions

- **We developed a TB spin dynamics and Monte Carlo code**
- **Self-consistent constraining fields are the torques in SD**
- **Thermodynamics properties via tight – binding MC**
- **Outlook: use of large computer systems**

Aknowledgements

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