Single ionic properties of $R_2Fe_{14}B$, and RRu_2Si_2 (R: rare earth element) calculated with Atomic Matters and Atomic Matters MFA computation systems

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Atomic Matters MFA is an application for predicting properties of defined materials in a magnetic ordered state and around phase transition temperature. The application is based on an atomic, localized-electron approach and Mean Field Approximation (MFA) methodology.

Atomic Matters MFA provides magnetic, calorimetric and electronic properties of atomic-like localized electron systems under the influence of:

- **Crystal Electric Field (CEF)**,
- **Spin-Orbit coupling (LS coupling)**,
- **Magnetic interactions (Zeeman Effect)**, taken as both Mean Field Approximation (MFA) and the influence of external, user-defined magnetic fields.

The application combines quantum electronic properties of correlated electron systems with calculations for properties of bulk crystals of a defined material. The application has an intuitive, interactive graphic interface with many unique tools for for easy, fast and fruitful work. The 3D Visualization of CEF potentials, easy import and export of data and rapid calculation results gives users a unique chance to form an opinion on the effect of defined interactions, simplifications, calculation spaces etc. for the macroscopic properties of crystals of a defined material.

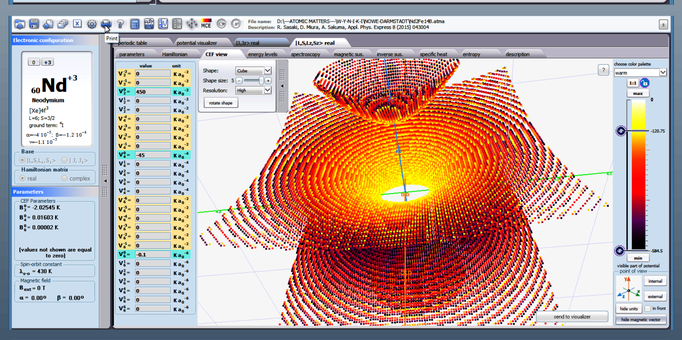
Atomic Matters MFA provides macroscopic properties of materials in defined temperature regions, especially around phase transition temperature, such as:

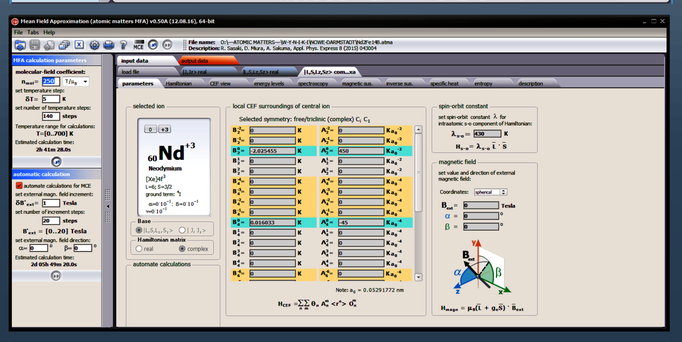
- magnetic moment in ordered state: **m**(T) (spin, orbit and directional components),
- magnetocrystalline anisotropy constants: $K_1(T)$, $K_2(T)$, $K'_2(T)$, $K_3(T)$, $K_4(T)$
- thermal evolution of energy level positions, eigenstates: $E^i(T)$, $\Gamma^i(T)$,
- free energy temperature dependencies in a definable temperature region: F(T),
- localized electron specific heat: $c_{el}(T)$ (with Debye, crystal lattice contribution),
- localized electron entropy: S(T) with a useful toolset for curve comparison and $-\Delta S(T)$ calculations for **Magnetocaloric Effect (MCE)** research and more...

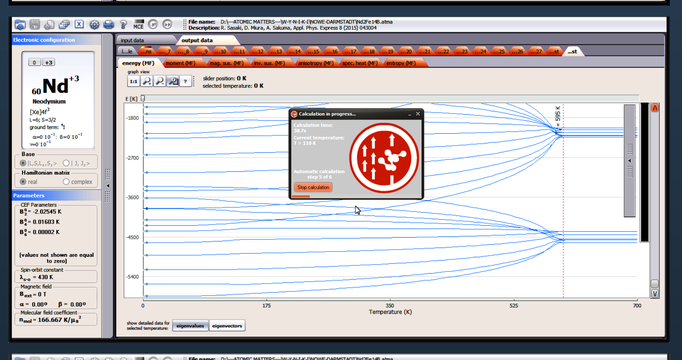
Atomic Matters MFA is an extension of the **Atomic Matters** application, a system for describing the Fine Electronic Structure according to modern CEF approach and predicting basic magnetic and spectral properties of materials in a stable paramagnetic state. The magnetic face transition can be simulated by **Atomic Matters MFA** according to Mean Field Approximation methodology. The calculations are realized using only one user-defined 'free parameter' n_{mol} which defines molecular magnetic field strength $\mathbf{B}_{mol}$ according to the simple relation of ion magnetic moment **m** at the current temperature:

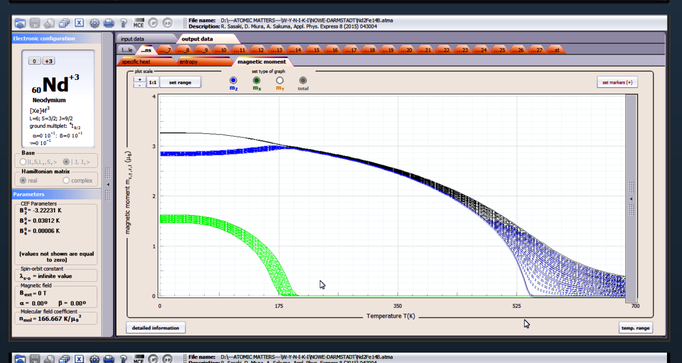
$$\mathbf{B}_{mol}(T) = n_{mol}\,\mathbf{m}(T)$$

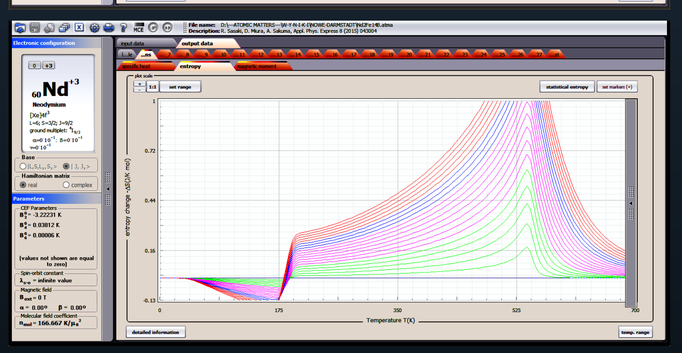
The value of parameter n_{mol} defines phase transition temperature T_c for self-consistent magnetic interactions in defined ionic systems.











Screenshots of **Atomic Matters** **Atomic Matters MFA** software. Calculations of properies of Nd^{3+} ions in $Nd_2Fe_{14}B$ under the influence of external magnetic field: B_{ext}=0 .. 10 T .

THEORETICAL APPLICATION BACKGROUND

Atomic Matters Hamiltonian: **Calculation space:**

$$H_{J\,CEF} = \sum_n \sum_m B_n^m \hat{O}_n^m(J, J_z) + g_L\,\mu_B\,\mathbf{J}\cdot\mathbf{B}_{ext} \qquad |J, J_z\rangle$$

$$H_{LS\,CEF} = \sum_n \sum_m B_n^m \hat{O}_n^m(L, L_z) + \lambda\mathbf{L}\cdot\mathbf{S} + \mu_B(\mathbf{L} + g_e\mathbf{S})\cdot\mathbf{B}_{ext} \qquad |L,S,L_z\,S_z\rangle$$

Atomic Matters MFA Hamiltonian:

$$H_{J\,mol} = \sum_n \sum_m B_n^m \hat{O}_n^m(J, J_z) + n_{mol}\,g_L^2\,\mu_B^2\left(-\mathbf{J}<\mathbf{J}>+\frac{1}{2}<\mathbf{J}>^2\right) + g_L\,\mu_B\,\mathbf{J}\cdot\mathbf{B}_{ext} \qquad |J, J_z\rangle$$

$$H_{LS\,mol} = \sum_n \sum_m B_n^m \hat{O}_n^m(L, L_z) + \lambda\mathbf{L}\cdot\mathbf{S} + n_{mol}\,\mu_B^2\left(-(\mathbf{L}+g_e\mathbf{S})<\mathbf{L}+g_e\mathbf{S}>+\frac{1}{2}<\mathbf{L}+g_e\mathbf{S}>^2\right) + \mu_B(\mathbf{L}+g_e\mathbf{S})\cdot\mathbf{B}_{ext} \qquad |L,S,L_z\,S_z\rangle$$

THERMODYNAMICS & MACROSCOPIC PROPERTIES

- statistical Sum of States: $Z(T) = \mathrm{Tr}\left(\exp\left(-\frac{\hat{H}}{k_B T}\right)\right) = \sum_i \exp\left(-\frac{E_i(T)}{k_B T}\right)$
- Helmoltz Free Energy: $F(T) = -k_B T \ln Z(T)$
- localized electron specific heat: $c_{mol}(T) = -T\left(\frac{\partial^2 F(T)}{\partial T^2}\right)$
- entropy of localized electron system: $S(T) = S(0) + \int_0^T \frac{c(T)}{T}\,dT$
- magnetic moment in ordered state: $m_J^\alpha(T) = \frac{g_L\mu_B}{Z(T)}\sum_i <J_\alpha^i> \exp\left(-\frac{E_i(T)}{k_B T}\right)$

$$m_{LS}^\alpha(T) = \frac{\mu_B}{Z(T)}\sum_i <L_\alpha^i + g_e S_\alpha^i> \exp\left(-\frac{E_i(T)}{k_B T}\right)$$

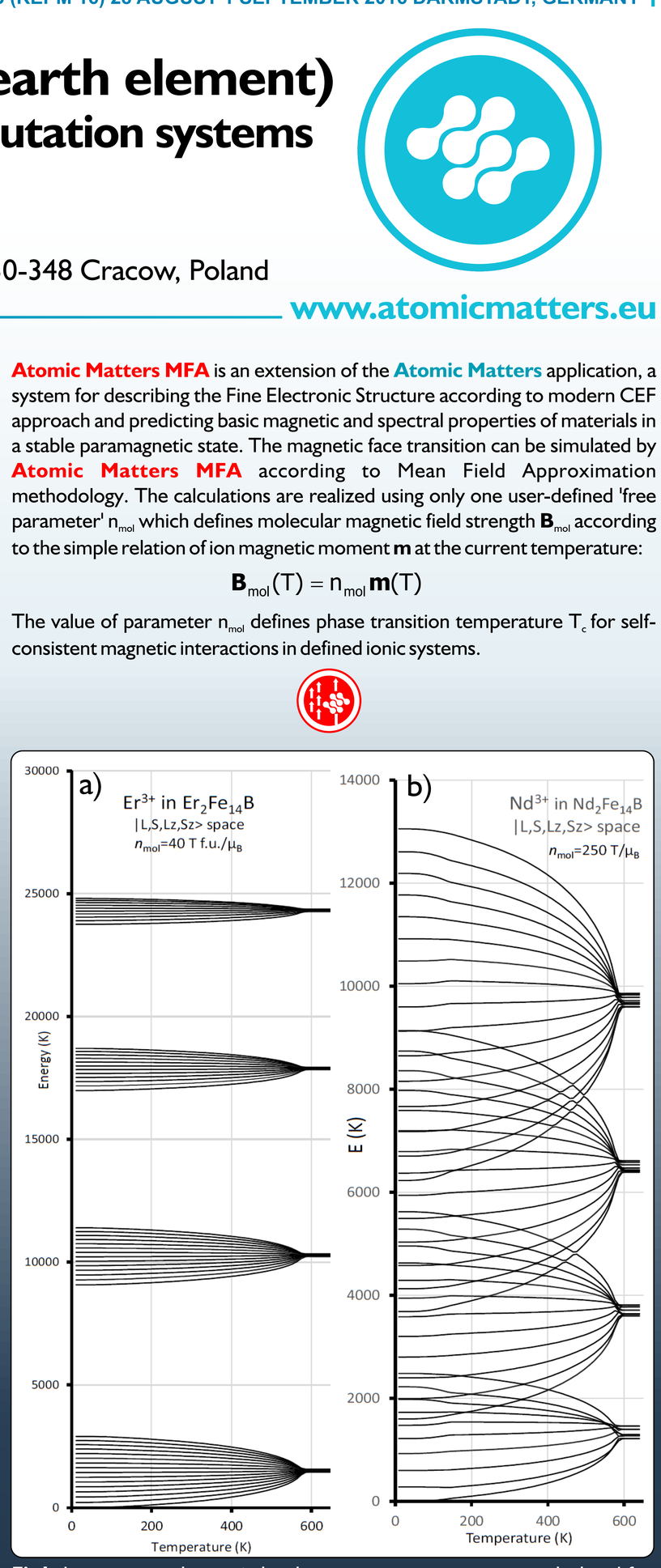


Fig1. Low energy electronic level structure vs. temperature, calculated for:
a) Er^{3+} ions in $Er_2Fe_{14}B$ in $|L,S,L_z,S_z\rangle$ calculation space under the influence of a molecular field: n_{mol}=40 T/μ_B, CEF parameters from [1]
b) Nd^{3+} ions in $Nd_2Fe_{14}B$ in $|L,S,L_z,S_z\rangle$ space under the influence of a molecular field: n_{mol}=250 T/μ_B , CEF parameters from [2]. The spin-orbit constant $\lambda_{S\text{-}O}$ values were taken as a free ion value for the relevant ion.

A few exampes of **Atomic Matters MFA** *calculation results obtained for different rare-earth magnetic compounds*

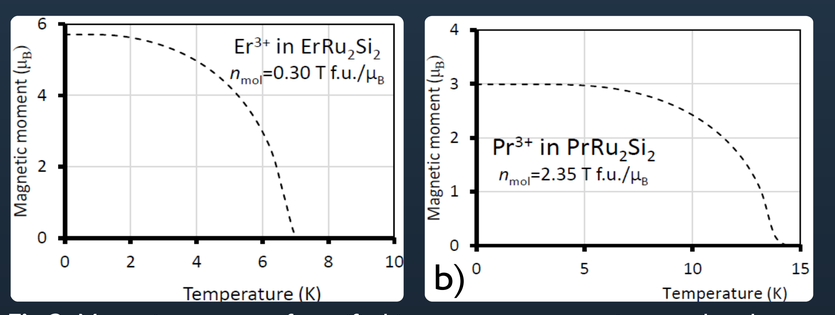


Fig 2. Magnetic moment of specified ions vs. temperature in an ordered state calculated according to Mean Field Approximation methodology for:
a) Er^{3+} ions in $ErRu_2Si_2$ in $|L,S,L_z,S_z\rangle$ space, molecular field constant n_{mol}=0.3 T/μ_B, CEF parameters taken from [3].
b) Pr^{3+} ions in $PrRu_2Si_2$ in $|L,S,L_z,S_z\rangle$, n_{mol}=2.35 T/μ_B, CEF parameters from [4].
c) Er^{3+} ions in $Er_2Fe_{14}B$ in $|L,S,L_z,S_z\rangle$, n_{mol}=40 T/μ_B, CEF parameters from [1].
d) Nd^{3+} ions in $Nd_2Fe_{14}B$ in $|L,S,L_z,S_z\rangle$, n_{mol}=250 T/μ_B , CEF parameters from [2].

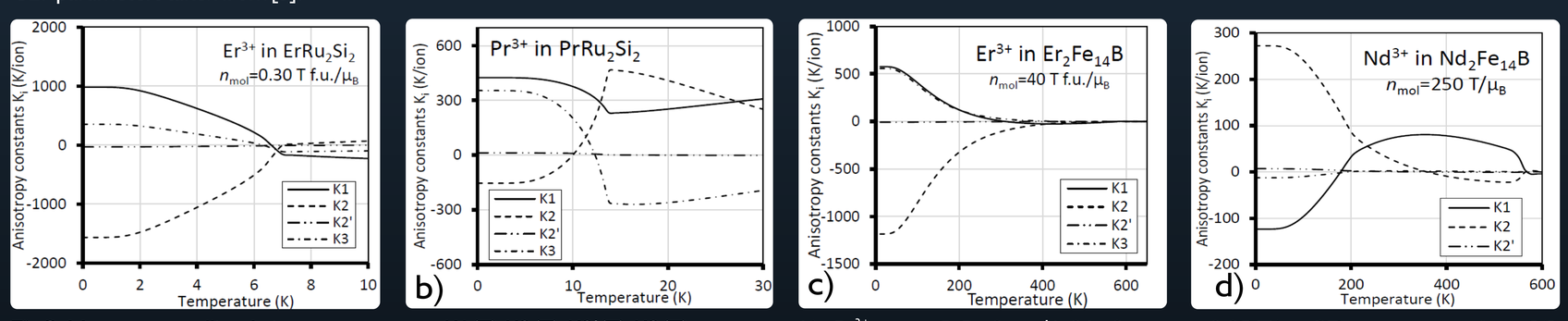


Fig 3. Magnetocrystalline Anisotropy constants K1(T),K2(T),K2'(T),K3(T), calculated according to Mean Field Approximation methodology for:
a) Er^{3+} ions in $ErRu_2Si_2$ in $|L,S,L_z,S_z\rangle$ space, n_{mol}=0.3 T/μ_B, CEF parameters from [3].
b) Pr^{3+} ions in $PrRu_2Si_2$ in $|L,S,L_z,S_z\rangle$, n_{mol}=2.35 T/μ_B, CEF parameters from [4].
c) Er^{3+} ions in $Er_2Fe_{14}B$ in $|L,S,L_z,S_z\rangle$, n_{mol}=40 T/μ_B, CEF parameters from [1].
d) Nd^{3+} ions in $Nd_2Fe_{14}B$ in $|L,S,L_z,S_z\rangle$, n_{mol}=250 T/μ_B , CEF parameters from [2].

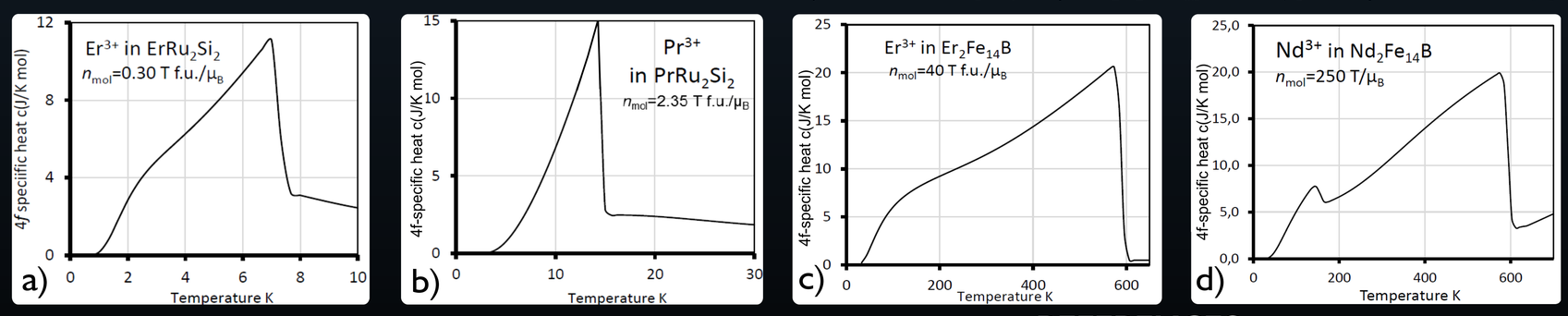


Fig 4. *4f*-component of molar specific vs. temperature :
a) Er^{3+} ions in $ErRu_2Si_2$ in $|L,S,L_z,S_z\rangle$ space, n_{mol}=0.3 T/μ_B, CEF parameters from [3].
b) Pr^{3+} ions in $PrRu_2Si_2$ in $|L,S,L_z,S_z\rangle$, n_{mol}=2.35 T/μ_B, CEF parameters from [4].
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d) Nd^{3+} ions in $Nd_2Fe_{14}B$ in $|L,S,L_z,S_z\rangle$, n_{mol}=250 T/μ_B, CEF parameters from [2].

REFERENCES

[1] B.F. Bogacz, A. T. Pedziwiatr, *Nukleonika* **58** (2013) 31-33-
[2] R. Sasaki, D. Miura, A. Sakuma, *Appl. Phys. Express* **8** (2015) 043004
[3] R.J. Radwanski, R. Michalski, Z. Ropka, *Physica B* **319** (2002) 78-89
[4] R. Michalski, Z.Ropka, R. J. Radwanski. *J. Phys. Condens. Matter* **12** (2000) 7609

We kindly invite your cooperation in testing and developing **Atomic Matters** **Atomic Matters MFA** software. Do not hesitate to contact us: r.michalski@induforce.eu