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**Supporting Information** 

## Thermostructural Behaviour of Ni-Cr Materials: Modelling Bulk and Nanoparticle Systems

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## S1. Equations used to calculate the properties of the materials

The cohesive energy is calculated as follows:  $E_c = \frac{E_{bulk}}{n} - E_{at}$ 

The bulk modulus is calculated with Birch-Murnaghan's equation of state:

$$B = \frac{9 \ \partial^2 E}{4a \partial a^2} \qquad E(V) = E_0 + \frac{9V_0 B_0}{16} \left\{ \left[ \left( \frac{V_0}{V} \right)^2 - 1 \right]^3 B'_0 + \left[ \left( \frac{V_0}{V} \right)^2 - 1 \right]^2 \left[ 6 - 4 \left( \frac{V_0}{V} \right)^2 \right] \right\}$$

where *a* is the lattice parameter,  $E_0$  is the equilibrium energy,  $V_0$  is the equilibrium volume,  $B_0$  is the Bulk modulus and  $B'_0$  is the derivative of the bulk modulus with respect to pressure.

The elastic constants are calculated with this set of three strain tensors. The first isotropic deformation is related to the Bulk Modulus, the second and the third, volume conserved deformations, are related to C' and  $C_{44}$  respectively. Fitting to the equations given by Kart *et al.*<sup>1</sup>:

$$\begin{bmatrix} \delta & 0 & 0 \\ 0 & \delta & 0 \\ 0 & 0 & \delta \end{bmatrix} \begin{bmatrix} \delta & 0 & 0 \\ 0 & \delta & 0 \\ 0 & 0 & -\delta^2 - 2\delta \\ 0 & 0 & -\delta^2 - 2\delta \\ 0 & 0 & -\delta^2 \\ 0 & 0 & -\delta^2 \\ 0 & 0 & -\delta^2 \end{bmatrix}$$

 $C_{11}$  and  $C_{12}$ :

$$E(\delta) = E_0 - 6V_0C\delta^2 + O(\delta^4)$$

$$B = \frac{(C_{11} + 2C_{12})}{3}$$

$$C' = \frac{(C_{11} - C_{12})}{2}$$

*C*<sub>44</sub>:

$$E(\delta) = E_0 + 2V_0 C_{44} \delta^2 + O(\delta^4)$$

To calculate the thermal expansion coefficients we have carried out several molecular dynamics simulations at different temperatures. We fit these points with a third order polynomial equation like this:

$$f(x) = a \cdot x^3 + b \cdot x^2 + c \cdot x + d$$

where *a*, *b*, *c* and *d* are fitted parameters.

1. Özdemir Kart, S.; Uludogan, M.; Karaman, I.; Çagın, T., Dft Studies on Structure, Mechanics and Phase Behavior of Magnetic Shape Memory Alloys: Ni2MnGa. Physica Status Solidi (A) 2008, 205, 1026-1035.

## S2. Radial Distribution Functions of the core and outer layers of the nanoparticles

Ni-Ni RDFs of the core and of the outer 1nm thick layer of the Ni nanoparticles of sizes (from top to bottom) 3 nm, 5 nm, 7 nm, and 10 nm. The RDFs are obtained from the last structures of the simulations at 300 K, which accounts for the high level of noise. In order to get a better insight into how the RDFs change, a Gaussian function has been fitted to each of the peaks, and the positions of the center of the Gaussians are shown as vertical lines. The RDF of the fcc Ni bulk structure at 300 K is also shown for comparison.



3 nm







5 nm



