Ab initio calculation of variable saddle point energies for atom jumps in L1_2 ordered Ni_3Al

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Monte-Carlo simulation: How sensitive is saddle point energy to atomic environment?

The 4-atom window and the single atom jump

Geometric relations in the L1_2 structure:

- L1_2 structure
- Window configuration
- 4-atom window

The single atom jump:

- Vacancy mechanism
- Main stages of the jump: 1. Initial equilibrium position. 2. Saddle point state. 3. Final equilibrium position.
- Transition state theory: \( \Gamma_1 = \Gamma_3 \exp \left( \frac{A}{RT} \right) \)

Results for \( E_{\text{diff}} \):

- \( E_{\text{diff}} \) in eV
- \( \frac{\Delta E_{\text{diff}}}{\Delta F_{\text{diff}}} \)

The importance of the 4-atom window:

Variation of distance of one antiflu to jumping atom

Classification and results

Classification

Jump types

Window types

Results for \( E_{\text{diff}} \):

Barrier height [eV]

Left

Right

Classification

Jump types

Window types

Comparison of 3x3x3 and 2x2x2 supercells

Details of VASP calculations

- GGA (PW91), ECVT 50-AV
- 2x2x2 and 3x3x3 supercells (6x6x6 and 4x4x4 k points, respectively)
- NEB (Nudged-elastic band) method was used where necessary
- atoms located on the surface of the supercell are fixed

Long range order parameter

\( q(t) = 4 \exp \left( -\frac{t}{\tau_2} \right) + [1 - 4 \exp \left( -\frac{t}{\tau_1} \right)] < \)

with \( 0 \leq A = t / \tau_2 < t_1 \)

Jump statistics

Window statistics

Saddle point energies are very sensitive to atomic environment!

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