Structural complexity in binary bcc ground states: The case of bcc Mo-Ta

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alloy system. In the present work, we apply a three-level, iterative construction scheme:

(i) For a given number n_p of pairs and a set of nonpair interactions J_{MB}, one first obtains an optimum interaction set J_f by minimizing[8,20]

$$s_{\mathrm{MBCE}} = \sum_{\sigma} w_{\sigma} \left| \tilde{H}_{\mathrm{LDA}}(\boldsymbol{\sigma}) - \tilde{H}_{\mathrm{CE}}(\boldsymbol{\sigma}) \right|^2 + \frac{t}{\Lambda} \sum_{p}^{n_p} R_p D_p J_p^2 . \quad (3)$$

Here, the usual least-squares sum is amended by an additional constraint per pair. This allows for an unlimited number of pair interactions, and avoids an unphysical cutoff dictated by the finite number of input structures. The proper spatial decay of J_p with pair distance R_p is enforced by weight factors R_p, with t a Lagrangian multiplier and $\Lambda = (\sum_{p}^{n_p} \sqrt{R_p/D_p})^2$ a normalization factor.

(ii

Carlo simulations, cell sizes $20\times20\times20$ or larger, 2000 or 4000 flips per site) based on the converged MBCE.

Ground states with sufficiently large are identified as large circles in Fig. 2. Here, the Mo-Ta system is divided into four structurally distinct regions. i) In *region 1*, the Mo-rich range below 20% Ta, we find several possible, very shallow ground states. However, with $\lesssim 2$ meV, they are very close to the phase-separated limit of pure Mo and Mo_4Ta, so that we did not investigate this range in greater detail. ii) In *region 2*, on the Ta-rich side above 75% Ta), no ground states are found. The interesting regions are 3 and 4: iii) In *region 3* roughly 20–60% Ta) we find five distinct small-cell ground states, denoted A_4B, A_2B, A_3B_2, AB, and A_2B_3 in Fig. 2 A=Mo, B=Ta). These structures are depicted in Fig. 3. Each is a superlattice SL) of pure 100) planes: the well-known $B2$ and $C11_b$ structures AB and A_2B 100) SL's, respectively], the larger A_4B 100) SL and the hybrid A_2BAB 100) SL's of composition Mo_3Ta_2 and Mo_2Ta_3. In addition, the MBCE predicts an exceedingly low enthalpy for the inclusion of antiphase defects between the basic 100) superlattice units. Therefore, region 3 contains a quasicontinuum of more complex 100)

reciprocal space for 79% Mo and 63% Mo solid solutions.[32] This is confirmed by simulations based on our converged MBCE.

The ground-state structures of regions 3 and 4 are unsuspected in that they cannot exist within a few-interaction ground-state enumeration scheme:[12,13]

Necessity of high-order pairs: The 100