Adaptive Crystal Structures of Au-Cu alloy

Keywords: UNCLE, alloy, cluster expansion

The stable phases in the gold-copper alloy were identified using the cluster expansion method as implemented in the *MedeA UNCLE* module. The well-established AuCu $L1_0$ and $AuCu_3$ $L1_2$ phases were found by means of an iterative cluster expansion ground-state search. In addition to these two well-established phases an adaptive crystal structure around the Au_3Cu stoichiometry range, made up of repeat units of pure-Au and pure-Cu planes along (001), and a Au_7Cu_5 P4/mmm phase was identified.

1 Experimental Results and Previous Computed Results

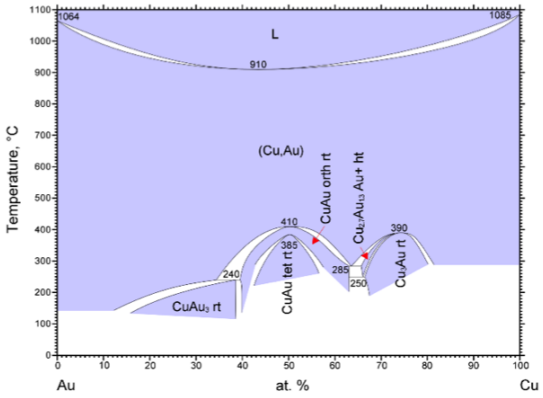


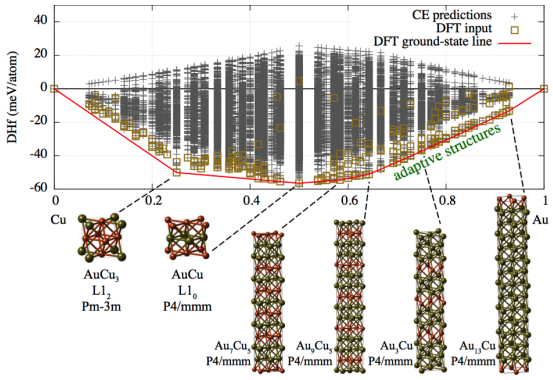
Figure 1: H.Okamoto, D.J. Chakrabarti, D.E. Laughlin, and T.B. Massalski, Binary Alloy Phase Diagrams, Second Edition, Ed. T.B. Massalski, ASM International Ohio **1**, 358-362 (1990)

Pure gold, pure copper, the gold-copper compounds, and the alloy are found on the same face-centered cubic/tetragonal lattice. There are two well-established phases in the Au-Cu alloy: at stoichiometry AuCu the $L1_0$ (P4/mmm) phase and at composition $AuCu_3$ the $L1_2$ (Pm-3m) phase. The high temperature AuCu Imma phase is a long-range ordered structure consisting of alternating $AuCu_3$ and Au_3Cu $L1_2$ units.

Beyond these phases a low temperature phase is suggested at Au_3Cu. It is stable in a comparatively wide concentration range and is classified as $L1_2$ (Pm-3m). However, its formation temperature (240 °C) is much lower than that of AuCu (385 °C) and $AuCu_3$ (390 °C). A cluster expansion ground state search by Sanati *et al.* [1] noticed that the structure around Au_3Cu resembles that of an adaptive crystal structure made up of repeat units of pure-Au and pure-Cu (001) planes. In such an infinitely adaptive crystal structure any composition within a certain concentration range is realized by a fully ordered crystal structure without defects [2].

2 Computed Results

A cluster expansion ground state search in a configuration space of 34368 structures does indeed find the two well established phases, the AuCu $L1_0$ and the $AuCu_3$ $L1_2$ phase. Instead of an Au_3Cu $L1_2$ phase an adaptive crystal structure, composed of repeat units of pure Au- and Cu-planes along (001), is identified in the Au concentration range of 0.65-0.92. In between the concentration range of the adaptive structures and the $L1_0$ AuCu phase an isolated Au_7Cu_5 P4/mmm phase that has the adaptive structures plane-wise ordering along (001) is also found.



[1] M. Sanati *et al.*, "Adaptive Crystal Structures: CuAu and NiPt", *Phys. Rev. Lett.* **90**, 045502 (2003) link

[2] J. S. Anderson, "On infinitely adaptive structures", *J. Chem. Soc. Dalton Trans.* **10**, 1107 (1973) link

3 Significance

The cluster expansion method of *MedeA UNCLE* is the perfect complementary tool to analyze and understand phase stability. By quickly scanning through a large number of configurations with first principles accuracy stable ground state structures at *T=0* K can be identified automatically. These phases can be more complex than originally contemplated and expected.

4 Comments

The adaptive crystal structure has been found using DFT calculations at *T=0* K. Entropic effects, such as mixing and vibrational entropy, might significantly alter the stable phases around Au_3Cu composition at higher temperatures and may lead to the $L1_2$ phase being more stable (e.g. at room temperature and above). Such an effect could explain the discrepancy between experimental data and DFT studies.

MedeA modules

- *MedeA Environment*
- *MedeA VASP*
- *MedeA UNCLE*

APPLICATION NOTE