Ann. Phys. (Leipzig) **10** (2001) 1–2, 89 – 94

Quantum architecture of novel solids

A. Zunger

National Renewable Energy Laboratory, 1617 Cole Blvd., Golden, CO 80401
USA
`alex_zunger@nrel.gov`

Received 25 Sep. 2000, accepted 22 Dec. 2000 by C. Thomsen

The current status of our understanding of Quantum Mechanics is that if one specifies the chemical formula of a compound (e.g., CuAu, or GaAs, or NiPt) it is still impossible to predict if this material is a superconductor or not, but it is now possible to predict its crystal structure. This is a nontrivial accomplishment for there are as many as 2^N possible structures for a binary compound. This article reviews this classic question of structural chemistry and condensed matter physics: How can one figure out which of the astronomic number of possible crystal structures is selected by Nature?

Crystal structures, Local Density Approximation, cluster expansion, structural selectivity

61.50.Ah, 64.10.+h, 71.15.Ap

1 Introduction

. 1 Schematic illustration of how the LEGO idea works. See Refs. [4, 5]

than the interaction between nearer atoms, then the expansion of $E(\sigma)$ could require fewer than 2^N interaction energies J_G . Thus, an LDA calculation of the total energies $E_{LDA}(\sigma)$ of N_σ ordered configurations could be used to determine N_σ interaction energies [2], thus according examination of the rate of convergence of the series E_{LEG} (σ). If the series converges after $\approx 10-20$ terms (as it does in many cases, see below) then one can calculate $E(\sigma)$ for ***any*** configuration σ by just summing over that many terms. Furthermore, one could combine such an expansion allows us to calculate the excess energy $\Delta E(\sigma)$ of any arbitrary atomic configuration, (even consisting of more than 100,000 atoms) and it includes automatically the energy lowering due to atomic relaxations. Formulation of the method requires as input the $T = 0$ K excess energies ΔE_{LDA} of 20-30 ordered compounds A_pB_q consisting of only 2-16 atoms per unit cell. The excess energy $\Delta E(A_pB_q, (\sigma)$ of such ordered A_pB_q bulk compounds is defined as the energy gain or loss with respect to the bulk constituents at their equilibrium lattice constants:

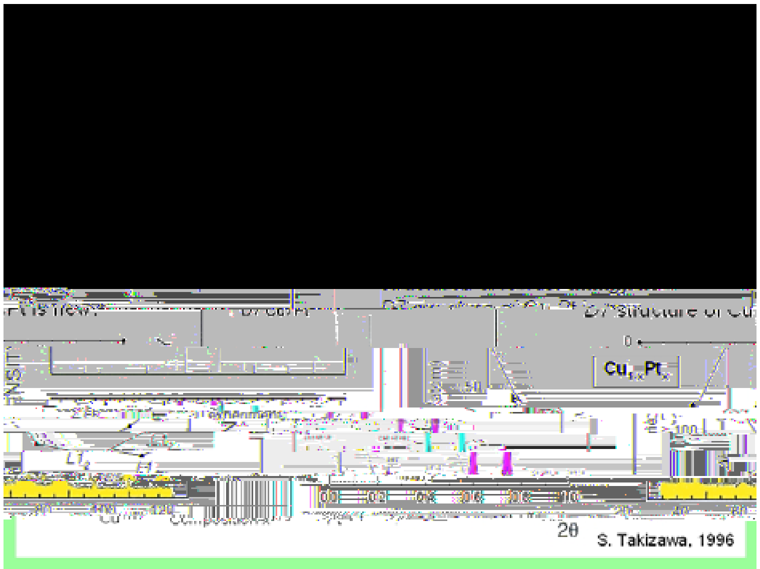
$$\Delta E_{LDA}(A_pB_q;\sigma) = E^{tot}(A_pB_q,\sigma) - xE_A^{tot}(a_A) - (1-x)E_B^{tot}(a_B). \tag{1}$$

Here, σ denotes the type of ordered structure, $x = p/(p+q)$, and a_A and a_B are the equilibrium lattice constants of the bulk elements A and B. $E^{tot}(a_A)$ and $E^{tot}(a_B)$ are the total energies of A and B, respectively. The formation energies of such small unit cell structures can be easily calculated within the local-density approximation

1 These calculations provide comprehensive state-of-the-art first-principles description of *ground state structures*, *phase stability*, and *short-range order* in these systems.

Pd-Pt	[6]	Ni-V	[7,8]	Cu-Pt	[12,13]
Rh-Pt	[6]	Ag-Au	[9,10]		
Cu-Au	[11]	Ni-Au	[11]		

The new Cu Pt "D7-type" ground state structure was predicted [12,13], and subsequently found experimentally by S. Takizawa (1996). This is illustrated in Fig. 2 and demonstrates the power of first-principles theory to predict previously unsuspected structures!



S. Takizawa, 1996

Illustration for prediction (Ref. [12,13]) and subsequent verification of a new structure for Cu-Pt.

We are now at the beginning of the process of solving one of the classic problems of Quantum Theory of Solids: We are transforming our qualitative and semi-quantitative understanding of sohesion into a predictive theory of new materials!

References

[1] P. Villars and L.D. Calvert, *Pearson's Handbook of Crystalographic Data for Intermetallic Phases*, Amer. Soc. Metals, Metals Park, OH (1985)

[2] J. W. Connolly and A. R. Williams, Phys. Rev. (1983) 5169