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Title: "Multi-Body Forces and the Energetics of Transition Metals, Alloys, and Semiconductors"

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Content

RESEARCH PROGRESS

The bulk of the research progress has been concentrated in the areas of transition-metal interatomic-potential functions on the one hand, and complex structures and quasicrystals on the other hand.

(1) Transition-metal interatomic potentials (papers 2 and 6). Using the basic physics of Fermi-level "quasigaps" vs. peaks in the electron density of states (DOS), developed in the previous grant period, we have developed a semiempirical angular-force method for Group VI transition metals. This method is based on the electronic energy as a function of the moments

$$\mu_n = \int_{-\infty}^{\infty} E^n \rho(E) dE$$

of the electron DOS, using up through μ_4. The method contains only four adjustable parameters, which is a much smaller number than is typically included in "embedded-atom" type methods. For the Group VI transition metals, these are least-squares fitted to five input quantities, which include the cohesive energy, the equilibrium lattice constant, and the three independent cubic elastic constants. The total-energy function is sufficiently transparent that one can obtain analytic expressions for the forces, *i.e.*, it is not necessary to virtually displace an atom in order to calculate the force on it.

The calibration tests that have been performed are quite encouraging. Both vacancy-formation energies and bcc-fcc structural-energy differences are obtained well by the method. The former result is expected, since a large part of the vacancy-formation energy is generally obtained already at the "μ_2" level. However the accuracy of the structural energies is surprising, since no information whatsoever about the fcc structure is included in the input database.

We have used the angular-force method to study the c (2×2) reconstruction that occurs on the W(100) surface. A non-zero reconstruction amplitude is found for all three different types of fitting methods that we have investigated. Although the amplitude depends on the fitting method, a value consistent with previous *ab-initio* calculations is obtained for sensible values of the parameters. The reconstruction is due to the μ_4-terms, which in an approximate fashion describe the destabilizing effect of a partly filled band of surface states in the unreconstructed surface. This physics is reflected in a bond-strength enhancement at the surface which exceeds that obtained by embedded-atom type methods.

(2) Complex structures and quasicrystals (papers 1,3,4, and 5). We have completed the work on the energy difference between the $Al_{12}W$ and Cu_3Au structures in transition-metal aluminides, that was begun in the preceding grant period. In order to evaluate long-ranged interaction effects between transition-metal impurities in Al, we have evaluated the energy of hypothetical $Al_{15}T$ compound (T=transition metal) on an underlying fcc lattice and compared it to the appropriate combination of Al and Cu_3Au-structure Al_3T. We find that there is a long-ranged interaction energy which is roughly half as large as the energy favoring the $Al_{12}W$ structure for nearly half-filled d-bands. Further *ab-initio* calculations have confirmed the importance of

effects beyond the nearest-neighbor shell by comparing the electronic DOS distributions in two distinct A15-based structures, both of which have icosahedral coordination for the Mn sites, but different longer-ranged environments. Finally, we have begun Anderson model-based perturbative calculations of the interaction energy between transition-metal impurities in Al. At this point, the Al is simply treated as a uniform electron gas; however, already roughly 50% of the *ab-initio* values for structural-energy differences between simple competing arrangements of T atoms are obtained.

We have also treated the structure and energy of transition-metal quasicrystals. With emphasis on the Ti–Mn system, we have developed a method for improving structures derived from 6d–3d projections. Such structures tend to have either short bond lengths, a low density, or many pairs of atoms having too few common neighbors. We have used this number of common neighbors, or "bond spindle", to locate positions for additional atoms. In the case of Ti–Mn, the original projected structure had a number of twofold bond spindles; in general, one expects to have mainly four– and five–fold spindles. The procedure is to locate the lowest bond spindles, and to add atoms in their vicinity via a simple geometric construction. This procedure greatly reduces the number of bad bond spindles, and significantly lowers an estimate of the total energy based on pair-potential calculations. We have also performed electronic total-energy calculations for this model using a d–orbital tight-binding method. These are the first electronic structural-energy calculations for transition-metal quasicrystals. They show a reduction in total energy which is roughly proportional to the number of twofold bond spindles that are removed. We expect that this "annealing" procedure should be useful in a variety of other problems involving incommensurate structures. In particular grain boundaries contain regions of "bad" crystal, and this might be a useful way of finding such regions.

For the Al–Mn quasicrystal, we have performed electronic DOS calculations using an optimized tight-binding model. Using the calculated DOS distributions, we have obtained the magnetic moments on the Mn sites. We find a large distribution of magnetic moments, consistent with the results of several experimental measurements; the mean-squared moment is close to the value deduced from high-temperature susceptibility experiments. In addition, the DOS around the Fermi level does not display the type of "quasigap" effects that are seen in *ab-initio* calculations for stable Al–Mn compounds.

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