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TITLE "ATOMISTIC MODELING OF MATERIALS - BEYOND PAIR POTENTIALS"

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AN EMBEDDED ATOM POTENTIAL FOR BCC IRON

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We have used the embedded atom method (EAM)[1-4] to construct interatomic potentials for use with BCC iron. Our original motivation for this work was to model the grain boundaries in iron[5]. The version of the EAM we have used is essentially the same as that described in references 3 and 4, where the total energy was given as the sum of two body terms summed over pairs of atoms i,j, together with an embedding term given by the sum of embedding functions whose arguments are the total electronic charge density at the sites i. A Morse potential

$$phi(r_{ij}) = v_1[1 - \exp[(-v_3(r_{ij}-v_2)]]^2 - v_1$$

with the three parameters v_1, v_2 and v_3, is used to represent the two-body terms. The total electronic charge densities at each individual site which are the arguments of the embedding functions, are obtained from the linear superposition of atomic 4s Slater function charge densities of form

$$r_{ij}^6[\exp[-v_4 r_{ij}] + 512 \exp[-2v_4 r_{ij}]].$$

The fifth parameter v_5 is taken as the cutoff distance for the atomic charge densities and for $phi(r_{ij})$, which are adjusted for smoothness at cutoff. The embedding function itself is determined by the fit of the cohesive energy of bcc iron as a function of interatomic distance to the "universal" cohesive energy function described by Rose et al.[6-8]. In

energy function is matched to the experimental cohesive energy, the bulk modulus B, and equilibrium lattice constant, a_0, but introducing no additional explicit matching parameters. The parameters v_1 through v_5 are selected by fitting (using a simplex optimization procedure[9]) to selected experimental properties of iron that we shall now describe. The first are the remaining elastic constants, either c_{11} or c_{12} (since $3B = c_{11} + 2c_{12}$) and c_{44}. We next try to match the observed vacancy formation energy. This is known somewhat less precisely than are the elastic constants, but it is important for matching purposes since the deviation from this value is a measure of the importance of many body forces due to the fact that the unrelaxed vacancy energy would be equal to the cohesive energy if only pair forces operated.

For a magnetic material such as iron there is a contribution to the vacancy energy arising from the altered magnetism on the neighboring atoms to the vacancy. There are also important contributions arising from magnetic effects in the energy difference between ferromagnetic bcc and nonmagnetic fcc and hcp iron. Our EAM calculation takes no explicit account of spin polarization effects, whereas quantum computations including these effects[10] indicate that they are crucial in providing phase stability of the bcc phase. However just as the adjustment of fitting parameters to vacancy energy may account for some of the magnetic energy in an empirical way, we try to make this fit also provide phase stability. Therefore we have made it a requirement that the minimum energies of the fcc or hcp phases should always be higher than that of the bcc phase of iron. This requirement was deemed essential in order to be able to utilize the resulting potential for the computation of defect configurations by energy minimization techniques. It turned out that this constraint on ensuring the stability of the bcc phase is quite restrictive and prevented us from finding a choice of parameters which exactly fit the elastic constants and vacancy energy. We might have gotten some additional freedom in fitting if we had chosen a two body potential with additional parameters. However we hesitated to do so since we wanted to avoid unrealistic structure in the potential which might arise from too much freedom in curve fitting.

In TABLE I we show some results with computed EAM potentials. We list the experimental and calculated values of elastic constants and vacancy energy for the bcc phase, and the calculated cohesive energies

We also tabulate the values of the fitting parameters for the different potentials.

The column marked FCA is for the potential published in reference 5 where we tabulated interplanar spacings near the surface as well as surface and grain boundary energies obtained with its use. Cheung and Yip[11] also used this potential in preliminary computations studying thermal instability by means of molecular dynamics techniques. They found that a density discontinuity occurred for a 4x4x4 periodic cell of 128 atoms maintained at zero pressure using the Parinello-Rahman boundary conditions[12], at about 1750 K. This is somewhat lower than the melting temperature of iron (1809 K), while one might expect an instability temperature to be higher than melting. An additional molecular dynamics "experiment" was done on the uniaxial deformation of iron with this potential, in an attempt to simulate the martensitic Bain transformation to the fcc phase[13]. A phase transformation to the hcp phase occurred rather than to the expected fcc phase. This result may be due to the fact that the hcp phase for this potential is only 0.009 ev above the bcc ground state, while the fcc phase is 0.02 ev above bcc. This closeness of another phase might even have contributed to the lower than expected instability temperature. We therefore tried to compute other potentials which would show greater separation from the ground state and possibly reverse the order of the fcc and hcp phases. As the results listed in the other columns show we have increased the separation of the energies of the hcp and fcc phases from that of the bcc phase, although we have not been able to reverse the order. There is also a somewhat better fit to the elastic constants. For the potentials FBC and FCD, the energies of the hcp and fcc are identical, a consequence of the fact that the cutoff distance is between second and third neighbors; any potential with this property will give this result since the distances and numbers of first and second neighbors are the same for hcp and fcc. It is suggestive that perhaps a short range many body potential coupled with a two body potential having a longer range and additional structure might give an optimal description.

We might conclude this presentation of EAM potentials with the remark that as empirical potentials their test must be their robustness in describing various types of dynamical as well as static structural phenomena. We have not tried to review other related work, but we must

[illegible] whose iron potential has already been subjected to various tests[16].

TABLE 1

FITTING PARAMETERS FOR EAM POTENTIALS FOR BCC IRON

Lattice constant a_0 : 2.670 A Cohesive energy : 4.28 ev
Bulk modulus : 1.73×10^{12} dyn/cm^2 EXACTLY FIT BY ALL POTENTIALS

Elastic constants (10^{12} dyn/cm^2)	Experimental	FEA (previous)	FEB	FBC (short range)	FED
c_{11}	2.432	1.93	2.131	2.178	2.299
c_{12}	1.381	1.63	1.534	1.507	1.447
c_{44}	1.219	1.05	1.165	1.175	1.179
vacancy energy					
unrelaxed		1.93	2.623	1.953	2.207
relaxed	1.79	1.64	2.181	1.792	2.032
Parameters:					
v_1 $\times 10^{-2}$ Hartrees		2.32299	2.849	2.634	2.771
v_2 angstroms		2.09238	2.721	2.690	2.852
v_3 1/(angstroms)		1.34321	0.746	1.225	1.293
v_4 1/(angstroms)		6.26195	8.508	6.580	6.423
v_5 (cutoff distance, A.)		4.52432	4.5096	3 625	3.626
Phase separation:					
$E_{fcc} - E_{bcc}$ (ev)		0.02	0.057	0.074	0.108
$E_{hcp} - E_{bcc}$ (ev)		0.009	0.048	0.074	0.108

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