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Structure/Property Relationships for Metal/Metal Interfaces

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Symposium held April 29-May 1, 1991, Anaheim, California, U.S.A.

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Structure/Property Relationships for Metal/Metal Interfaces

Preface

This volume contains the Proceedings of the Symposium on Structure/Property Relationships for Metal/Metal Interfaces held at the 1991 Spring Meeting of the Materials Research Society. The forty-four papers published here, of which thirteen were invited, address this topic by means of experimental and theoretical investigations on a variety of interfacial systems that are found or can be fabricated in metals.

The central theme of the Symposium was the relationship between the physical properties of metallic materials containing interfaces and the detailed atomic structure of those interfaces. As is evidenced by the contributions to this volume, this theme actually encompasses quite a number of active research fields. When categorized by an interest in interfacial structure, these fields include metal/vacuum surface physics and surface layer characterization, x-ray diffractometry of films and superlattices, TEM characterization of orientational relationships in superlattices, grain boundaries, and dissimilar metal interfaces, and analytical microscopy of grain boundaries and dissimilar metal interfaces. As categorized by an interest in properties, these fields include elastic and plastic properties of superlattices, grain boundaries, and generic interfaces, dislocation motion through interfaces, fracture, segregation, and precipitation at grain boundaries and at dissimilar metal interfaces, x-ray optical properties of multilayers, and magnetic properties of superlattices.

From this diversity of subject matter it is clear that the role interfaces play in determining the properties of materials can be considerable. This is apparent in both basic science investigations and modern technological applications especially where metallic thin films, superlattices and nanocrystals are involved since their structures are composed mainly of interfaces. Due to the sheer breadth of the subject matter presented here and at the symposium, it is difficult to simply summarize all of the outstanding work and new observations. Nevertheless, one underlying aspect continues to dominate our attempts at understanding the impact of interfacial structure on properties. This is the ongoing development and improvement of our characterization tools, which allow us to clearly "see" the atomic structure of the interface. Progress in obtaining a detailed microscopic picture of interfaces, particularly in high-density-interface materials, will continue to be central to our understanding and control of the new physical properties which are observed.

A.D. Romig
D.E. Fowler
P.D. Bristowe

June 1991

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Their generosity is greatly appreciated and it allowed the organizers to focus on making the symposium a successful and smoothly functioning experience for all. Last, but not least, we would like to acknowledge the assistance of Ms. Sandy Ung in the preparation of the Proceedings.

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Structure/Property Relationships for Metal/Metal Interfaces

RECONSTRUCTIONS AND MORPHOLOGICAL INSTABILITIES OF FCC (110) SURFACES

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ABSTRACT

We compare the ground state energy of the following structures of fcc (110) surfaces: the (1×1) flat surface, the (n×1) reconstruction ($2 \leq n < \infty$), and the (∞×1) structure where the surface is faceted into macroscopic (111) surfaces. We map out the phase diagram using a lattice model with pair and triplet interactions. We argue that it is unlikely that any realist pair potential will give a reconstructed ground state. We also doubt that including, in addition, three body interactions will stabilize the reconstructed structure for interatomic interactions derived from the embedded atom method.

INTRODUCTION

It has been clearly established that, at low temperatures, the (110) surface of certain fcc metals (Au, Pt, Ir) undergoes reconstructions into (n×1) structures,[1] while many other fcc metals do not. A wide variety of theoretical techniques have been used to investigate the ground state structure of these surfaces and to compare with experimental results. On the one hand, it is useful to perform such calculations to test the success of theoretical techniques and models. On the other hand, one would like to have a more qualitative understanding of which structure is expected. The latter approach is taken here.

Two recent papers[2,3] put forward simple arguments which give criteria to determine which of the (1×1) or (2×1) structures has the lower energy. Foiles[2] argues that the repulsive three body interactions stabilize the reconstructed phase, while Chen and Voter[3] argue that a large second moment of missing bond distribution favors reconstruction. In both cases, it is argued that reconstruction occurs when the Cauchy discrepancy $C_{12} - C_{44}$ is large. Both authors argue that in those cases in which (2×1) is lower in energy than (1×1), (n×1) reconstructions with n>2 have a lower energy yet. Here we seek to investigate in more detail the lowest energy structure of (110) fcc surfaces.

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FORMULATION

In the Embedded Atom Method (EAM) the energy of a system of identical atoms located at positions $\mathbf{r}_i$ may be written[4]

$$E = \sum_m F[\sum_{i(\neq m)} \rho(R_{mi})\ ] + \frac{1}{2} \sum_{\substack{i,j \\ (i \neq j)}} \phi(R_{ij}) \ , \qquad (1)$$

where F is the embedding function, $\rho(R_{ij})$ is the electron density at atom j a distance $R_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$ from atom i, and $\phi(R_{ij})$ is the core-core pair repulsion. This may be rewritten[2]

$$E = E_0 + \sum_m \Big[\sum_i J(R_{mi}) + \sum_i \sum_j T(R_{mi}, R_{mj}) + \sum_i \sum_j \sum_k Q(R_{mi}, R_{mj}, R_{mk}) + \cdots \Big] \ , \qquad (2)$$

where n, i, j, k, ... refer to distinct atomic sites and J, T, Q, ... may be expressed in terms of F and ϕ. Graphically the interactions are of the "star" type, with site m in the middle and the "legs" going out to sites i, j, k, ...

In this paper we make the major assumption that the atomic positions are restricted to the sites of an fcc lattice. Foiles[2] has found that this assumption has a small effect on the difference in energies between competing structures. In terms of the lattice gas variables $n = \pm 1$, Eq. (2) becomes

$$E = - \sum_m n_m \Big[\sum_{i=1}^{\infty} J_i \sum_{p_i} n_{p_i} + \sum_{i=1}^{\infty} \sum_{j=1}^{\infty} T_{ij} \sum_{p_i} \sum_{p_j} n_{p_i} n_{p_j} + \sum_{i=1}^{\infty} \sum_{j=1}^{\infty} \sum_{k=1}^{\infty} Q_{ijk} \sum_{p_i} \sum_{p_j} \sum_{p_k} n_{p_i} n_{p_j} n_{p_k} + \cdots \Big] \ . \qquad (3)$$

We require coexistence by demanding that both the solid phase (all $n_i = 1$) and the vapor phase (all $n_i = 0$) have the same reduced energy $E - \mu N$. From this we can determine the chemical potential μ. Then we calculate $E - \mu N$ with this μ for various surface structures. This gives the surface energy γ as a function the parameters J_i, T_{ij}, Q_{ijk}, ...

The goal here is to investigate which phase of the (1×n) structures has the lowest reduced energy at each point in coupling parameter space. To make this problem feasible and to increase our understanding of the various couplings, we look at small subsets of the full parameter set $\{J_i, T_{ij}, Q_{ijk}, \ldots\}$. Foiles[2] has put forth a procedure by which these parameters may be determined using the EAM potentials. It will turn out that the (1×1) and (∞×1) phases are found in a large volume of

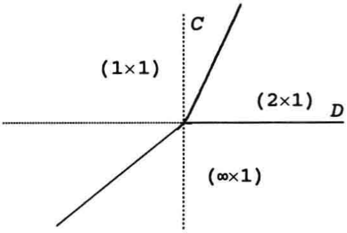

Fig. 1. Phase diagram of fcc (110) surfaces with pair interactions only, neglecting 9th and higher interactions. The model parameters C and D are defined in Eq. (5).

parameter space. Thus, a question we will address in detail is which parameters are needed to stabilize the (1×2) and (1×3) phases.

For convenience, instead of finding the true interface free energies ("surface tensions") per unit area $\tilde{\gamma}$, we find $\gamma = \tilde{\gamma}\mathcal{A}$, where $\mathcal{A}$ is the area of the unit cell of the (1×1) surface, namely, $a^2/\sqrt{2}$, where a is the lattice constant.

PAIR INTERACTIONS ONLY

We begin by including pair (two body) interactions only, setting $T_{ij} = Q_{ijk} = \ldots = 0$ in Eq. (3). We include pair interaction up to eighth neighbor, corresponding to a distance $2a$, which is $2\sqrt{2}$ longer than the nearest neighbor separation distance $a/\sqrt{2}$. In this case, bond counting gives

$$\gamma_{(1\mathrm{x}1)} = \gamma_{(\infty\mathrm{x}1)} - C + D \,, \tag{4a}$$

$$\gamma_{(n\mathrm{x}1)} = \gamma_{(\infty\mathrm{x}1)} - C/n \,, \tag{4b}$$

where

$$\gamma_{(\infty\mathrm{x}1)} = 6J_1 + 6J_2 + 24J_3 + 12J_4 + 36J_5 + 12J_6 + 72J_7 + 12J_8 \,, \tag{5a}$$

$$C = 2J_2 + 4J_3 + 16J_5 + 4J_6 + 16J_7 + 8J_8 \,, \tag{5b}$$

$$D = 4J_5 + 8J_7 + 2J_8 \,. \tag{5c}$$

Note that, of the many model parameters $J_1, \ldots, J_8$, the parameters arise in such a combination that only two parameters, C and D, are needed to determine the phase. This greatly simplifies the analysis. The resulting phase diagram is given in Fig. 1.

Several conclusions may be drawn from these results. It appears unlikely any realistic model of an fcc metal will arise which has pair interactions only and has a (2×1) ground state. Such a model must satisfy $D > \frac{1}{2}C > 0$. Note that D depends on fifth neighbor pairs and higher. At the fifth neighbor