AD-A156 039 CLUSTER VARIATION STUDY OF COHERENT ORDERING IN FCC AND BCC SOLID SOLUTIO..(U) CALIFORNIA UNIV BERKELEY DEPT OF MATERIALS SCIENCE AND MINERA.. D DE FONTAINE MAY 85 1/1

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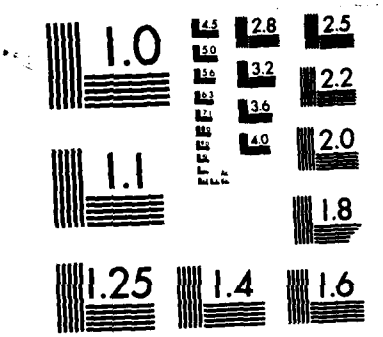

Final Report for DAAG29-80-K-0044
GF 25517 - 6/15/80-5/15/84

CLUSTER VARIATION STUDY OF COHERENT ORDERING IN FCC AND BCC SOLID SOLUTIONS

Principal Investigator - Didier de Fontaine

PRIMARY OBJECTIVE

The primary objective of this investigaiton was the derivation of binary phase diagrams by means of a statistical mechanical model called the "Cluster Variation Method" (CVM). In particular, phase equilibrium as a function of the ratio $\alpha=V_2/V_1$ between the values of second and first neighbor effective pair interactions, α being the only physical input to the computer codes. The resulting phase diagrams are important for the purpose of analyzing experimentally determined alloy phase diagrams.

SUMMARY OF RESULTS

The calculated phase diagrams are called prototype ordering phase diagrams: "prototype" because no attempt was made initially to reproduce experimental diagrams of real alloy systems, "ordering" because only equilibrium between ordered superstructures and the parent lattice (in this case fcc) were investigated. The CVM was used in the so-called tetrahedron-octahedron approximation, which required considerable computer code development. It had been determined earlier that Bragg-Williams approximations were quite inadequate. The CVM, however, performed extremely well in those cases where a comparison could be made, for instance with Monte Carlo simulations.

The only input parameter was $\alpha=V_2/V_1$, the ratio of second to first-neighbor effective pair interactions in fcc cyrstals. These interactions suffice to stabilize certain ground states of order, many of which are experimentally found in real intermetallic compounds. Complete temperature - composition phase diagrams were obtained for seven values of α: 0.0, 0.25, 0.35, 0.45, 0.55, -0.2, -1.0. Hence, for the first time, an ordering prototype phase diagram with first and second neighbor interactions was derived for the fcc lattice with, as parameters, the temperature T, the average composition c, and the pair interaction ratio α. Along the way, the CVM technique was extended and computer codes developed, ground state analysis was generalized, and, importantly, CVM equations were developed to predict short-range order intensity in partially ordered systems.

The stated objectives of the original proposal were fully met, resulting in 16 publications (see attached list). In particular, Ref. 14, published as an "Overview" in Acta Metallurgica (1985), constitutes an extensive survey of work performed under the ARO(D) Contract. The text of this article is included in this final report.

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The primary objective of this investigation was the derivation of binary phase diagrams by means of a statistical mechanical model called the "Cluster Variation Method." In particular, phase equilibrium as a function of the ratio between the values of second and first neighbor effective pair interactions, the ratio being the only physical input to the computer codes. The resulting phase diagrams are important for the purpose of analyzing experimentally determined alloy phase diagrams.

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The work that has been accomplished can be regarded as an essential first step towards a general "Alloy Theory", i.e. one that would combine both electronic band structure and statistical thermodynamic calculations in one coherent whole, for the purpose of deriving phase diagrams from first principles. A new proposal was therefore submitted to ARO(D) with this objective in mind. As of this writing, the proposal is still in limbo, due, we are told, to lack of funds.

"Alloy theory" was the subject of the 1984 Physical Metallurgy Gordon Research Conference, under the Chairmanship of D. de Fontaine, and with ARO(D) financial backing. A report was sent to ARO(D) in late 1984 concerning this Conference.

PUBLICATIONS

1. J. M. Sanchez, R. Kikuchi, H. Tamauchi and D. de Fontaine, "Cluster Approach to Order and Disorder, "Theory of Alloy Phase Formation, Ed. by L. H. Bennett, The Metallurgical Society of AIME (1980), pp. 289-301.

2. J. M. Sanchez and D. de Fontaine, "Ordering in fcc Lattices with First and Second Neighbor Interactions," Physical Review B, 21:216, 1980.

3. J. M. Sanchez and D. de Fontaine, "Theoretical Prediction of Ordered Superstructures in Metallic Alloys," in Structure and Bonding in Crystals II, M. O'Keeffe and A. Navrotsky, Eds., pp. 117-132, Academic Press, 1981.

4. D. de Fontaine, "Ordering Instabilities and Pretransitional Effects," Met. Trans., A 12A, 559-566 (1981).

5. D. de Fontaine, "Spinodal and Equilibrium Reactions," Physica, 103B, 57-66 (1981).

6. D. de Fontaine, "Compositional Instabilities," Proc. of International School of Physics "Enrico Fermi" on Mech. and Thermal Behav. of Metallic Materials, July (1981), Course LXXXII, Ed. by G. Caglioti and A. Ferro Milone, North Holland, (1982) pp. 449-471.

7. J. M. Sanchez, "Pair Correlations in the Cluster Variation Approximation," Physica 111A, 200 (1982).

8. J. M. Sanchez, D. Gratias and D. de Fontaine, "Special-Point Ordering in General Crystal Structures," Acta Cryst. A 38, 214 (1982).

9. D. Gratias, J. M. Sanchez and D. de Fontaine, "Application of Group Theory to the Calculation of the Configurational Entropy in the Cluster Variation Method," Physica 13A, 315 (1982).

10. J. M. Sanchez and D. de Fontaine, "Ising Model Phase Diagram Calculations in the fcc Lattice with First- and Second-Neighbor Interactions," Phys. Rev. B 25, 1759 (1982).

11. D. de Fontaine, "Studies of the Thermodynamics of Ordering by the Cluster Variation Method," Proc. of Int'l Conf. on Solid-Solid Phase Transformations, Pittsburgh, PA. Aug. 1981, Ed. by H. I. Aaronson, D. E. Laughlin, R. F. Sekerka and C. M. Wayman, Mat. Soc. AIME (1982), pp. 25-47.

12. J. M. Sanchez, D. de Fontaine, W. Teitler, "Comparison of Approximate Methods for the Study of Antiferromagnetism in the fcc Lattice," Physical Review, B 26, 2465 (1982).

13. D. de Fontaine, "Coherent Phase Diagrams in the Cluster Variation Approximation," in Alloy Phase Diagrams, Symp. Proc. Vol. 19, Mat. Res. Soc., Ed. by L. H. Bennett, T. B. Massalski and B. C. Giessen, North Holland (1983), pp. 149-162.

14. M-O. Lafon, "X-ray Study of Ordering in $Ni_3(Mo,X)$ Ternary Alloys," M.S. Thesis, U. C. Berkeley, December 1983.

15. D. de Fontaine, "Composition Modulations in Solid Solutions," Proc. NATO Advanced Study Inst. on Modulated Structures 1983, Crete, in Modulated Structure Materials, ed. T. Tsakalakos, 1984, pp. 43-80.

16. D. de Fontaine, "Prototype Ordering Phase Diagrams," Proc. Conf. High-Temperature Alloys: Theory and Design, The Metallurgical Society of AIME, ed. J. O. Stiegler, pp. 73-82 (1984).

17. T. Mohri, J. M. Sanchez and D. de Fontaine, "Binary Ordering Prototype Phase Diagrams in the Cluster Variation Approximation," Acta Metallurgica, (in press).

18. T. Mohri, D. de Fontaine and J. M. Sanchez, "Short Range Order Hardening with Second Neighbor Interactions in fcc Solid Solutions," accepted in Met. Trans. AIME, December 1984.

19. T. Mohri, J. M. Sanchez and D. de Fontaine, "Short Range Order Diffuse Intensity Calculations in the Cluster Variation Method," Acta Met., accepted, Feb. 1985.

ORAL PRESENTATIONS

1. D. de Fontaine, "Coherent Phase Diagrams in the Cluster Variation Approximation," Alloy Theory and Development Workshop, Los Alamos, N. M., September 8-10, 1982.

2. D. de Fontaine, "First-Principles Phase Diagram Calculations, a Possibility?" Seminar at ALCOA, Pittsburgh, PA, Octobert 13, 1982.

3. D. de Fontaine, "First-Principles Phase Diagram Calculations, a Possibility?" Department Seminar at U. C. Berkeley, October 21, 1982.

4. J. M. Sanchez, B. Davies and D. de Fontaine, "Theoretical Study of Coherent Phase Equilibria in fcc Binary Systems," TMS-AIME 1983 Meeting, Atlanta, Georgia, March 6-10, 1983,

5. D. de Fontaine, "Ordering at Special and Not-So-Special Points," Seminar at Carnegie-Melon University, Pittsburgh, PA, March 11, 1983.

6. D. de Fontaine, "Composition Modulations in Solid Solutions," NATO Advanced Institute and the International Conference on Modulated Structure, Crete, Greece, June 27-30, 1983.

7. J. M. Sanchez, T. Mohri and D. de Fontaine, "Prototype Binary Phase Diagrams in the Cluster Variation Approximation," TMS-AIME 1984 Annual Meeting, Los Angeles, CA, February 26-March 1, 1984.

8. D. de Fontaine, "Prototype Ordering Phase Diagrams," High-Temperature Alloys: Theory and Design," Bethesda, Maryland, April 9-11, 1984.

PERSONNEL

Name	Employment Dates	Degree Earned
Dr. Juan M. Sanchez	6/81 to 7/81	Post-Doc
Dr. Winston Teitler	6/81 to 5/82	Programmer
Prof. D. de Fontaine	7/81, 7/82, 8/83	Prin. Investigator
Beverly Nagaue	11/81 to 2/82, 4/82, 7/82, 9/82	Clerical
Barrett Davis	3/82 to 6/82, 8/82, 11/82 to 3/83, 5/83 to 11/83	U. G. Coder
Marie-Odile Lafon	9/82 to 12/83	Grad. Student-M.S.
Dr. Tetsuo Mohri	1/83 to 12/83	Post-Doc

BINARY ORDERING PROTOTYPE PHASE DIAGRAMS IN THE CLUSTER VARIATION APPROXIMATION

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ABSTRACT

The Tetrahedron Octahedron Cluster Variation Method (TO-CVM) has been employed to investigate fcc based order-disorder phase diagrams with first (positive) and second nearest neighbor pair interactions. The investigation covers the entire range of α values, the ratio of second to first nearest neighbor pair interactions. Ground state structures are discussed in detail and phase diagrams for seven different α values are presented. Good qualitative (topological features) and reasonable quantitative (transition temperatures) agreement is obtained with available Monte Carlo results, thereby confirm the reliability of the TO-CVM phase diagram calculations. The present study concludes the first global phase diagram investigation of fcc-based Ising model with first and second nearest neighbor pair interactions, and can be regarded as a precursor to more elaborate first principles phase diagram calculations.

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