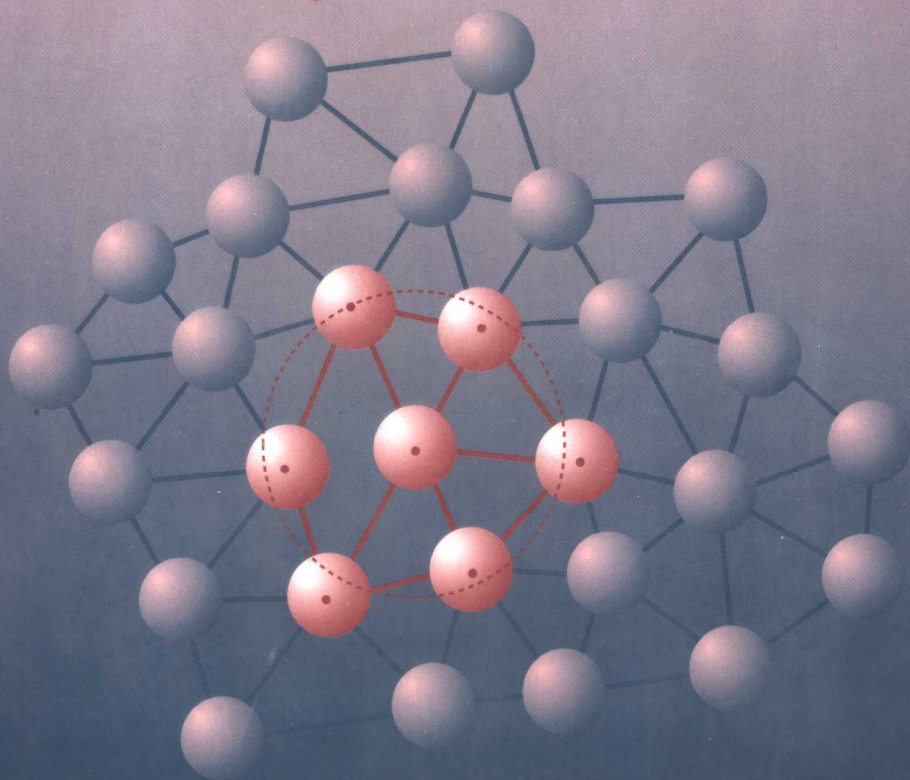


THE MAGNETISM OF AMORPHOUS METALS AND ALLOYS

Editors

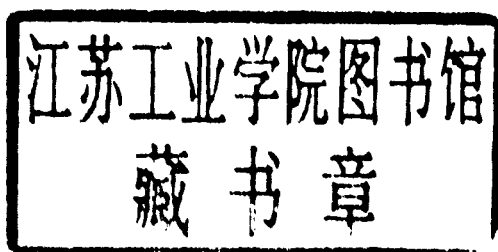
J. A. Fernandez-Baca

Wai-Yim Ching



World Scientific

THE MAGNETISM OF AMORPHOUS METALS AND ALLOYS



Editors

J. A. Fernandez-Baca

Oak Ridge National Laboratory, USA

Wai-Yim Ching

University of Missouri - Kansas City, USA



World Scientific

Singapore • New Jersey • London • Hong Kong

Published by

World Scientific Publishing Co. Pte. Ltd.

P O Box 128, Farrer Road, Singapore 9128

USA office: Suite 1B, 1060 Main Street, River Edge, NJ 07661

UK office: 57 Shelton Street, Covent Garden, London WC2H 9HE

THE MAGNETISM OF AMORPHOUS METALS AND ALLOYS

Copyright © 1995 by World Scientific Publishing Co. Pte. Ltd.

All rights reserved. This book, or parts thereof, may not be reproduced in any form or by any means, electronic or mechanical, including photocopying, recording or any information storage and retrieval system now known or to be invented, without written permission from the Publisher.

For photocopying of material in this volume, please pay a copying fee through the Copyright Clearance Center, Inc., 222 Rosewood Drive, Danvers, Massachusetts 01923, USA.

ISBN 981-02-1033-7

Printed in Singapore.

FOREWORD

About four years ago we were invited by the editors of World Scientific Publishing Co. Pte. Ltd. to edit a review volume on selected topics of randomness in magnetism. This subject was later narrowed down to the magnetism of amorphous metallic alloys. Of course the first question that came to our minds is "Why bother in editing a volume on a subject that has already excellent and comprehensive review books and articles like those of Moorjani and Coey¹ and J. Chappert.²?" The immediate answer was that perhaps it was worthwhile to make an update to the literature in this field, in the form of a book that would gather a selection of topics written by the people that had made the most recent progress. This would offer current reviews from the point of view of the specialists who would concentrate on their most recent progress while not neglecting to refer the reader to the already abundant existing literature. With this in mind we invited a selected group of specialists to review their progress in this subject, some of them accepted this challenge and this book is the result of their hard work. We thank all of them for their dedication and for their commitment. Because of the way that this book came about, this volume is by no means a comprehensive review of all the progress in the subject. Rather it is a collection of selected topics in the recent progress on the study of the magnetism of amorphous metals and alloys. Finally, we recognize the title of this book is very ambitious because while a great deal has been learned about the magnetism of amorphous metallic alloys, the study of the magnetism of the pure amorphous metals is still a subject of intense activity. We hope that the content of this book will help to motivate research in this direction. We trust that the reader will find in this volume a worthwhile update of some of the progress in the field of amorphous magnetism.

J. A. Fernandez-Baca

W. Y. Ching

¹K. Moorjani and J. M. D. Coey, *Magnetic Glasses*, (Elsevier, 1984).

²J. Chappert, "Magnetism of Amorphous Metallic Alloys", in *Magnetism of Metals and Alloys*, ed. M. Cyrot (North Holland, 1982)

CONTENTS

Foreword

xi

1. THEORY OF MAGNETISM IN AMORPHOUS TRANSITION METALS AND ALLOYS

by Yoshiro Kakehashi and Hiroshi Tanka

1. Introduction	1
2. Theoretical Approach to Amorphous Metallic Magnetism	3
2.1. Basic problems and developments	3
2.1.1. Amorphous structure	4
2.1.2. Electronic structure calculations	7
2.1.3. Calculating magnetic properties	8
2.2. LMTO-recursion method	9
2.2.1. Muffin-tin method and linear method	9
2.2.2. Tight-binding LMTO method	11
2.2.3. Recursion method	17
2.3. Finite-temperature theory of amorphous metallic magnetism	19
2.3.1. Functional integral method	19
2.3.2. Local moment in an effective medium	22
2.3.3. Structural disorder and electronic states	24
2.3.4. Distribution-function method	26
2.3.5. Extension to amorphous alloys	29
3. Magnetic Properties of Amorphous Transition Metals	32
3.1. General survey	32
3.2. Amorphous Fe	37
3.3. Amorphous Co and Ni	44
4. TM Amorphous Alloys	48
4.1. TM-TM amorphous alloys	48
4.1.1. General aspects	48
4.1.2. Fe-Zr amorphous alloys	50
4.1.3. Fe-Ni amorphous alloys	52
4.1.4. Co-Y amorphous alloys	54
4.2. RE-TM amorphous alloys	56
4.2.1. General aspects	56

4.2.2. Gd-Fe alloys	59
4.2.3. Gd-Co alloys	63
4.3. TM-metalloid alloys	67
4.3.1. General aspects	67
4.3.2. Fe-B alloys	70
4.3.3. Co-B alloys	72
References	76

2. ELECTRONIC STRUCTURE CALCULATIONS IN MAGNETIC METTALIC GLASSES

by *W.Y. Ching*

1. Introduction	85
2. The Orthogonalized Linear Combination of Atomic Orbitals Method	87
2.1. The atomic basis functions	87
2.2. The site-decomposed potential functions	88
2.3. The technique of gaussian transformation	91
2.4. The technique of core-orthogonalization	96
2.5. The secular equation	98
2.6. Spin-polarized calculations	99
2.7. Scalar relativistic correction	100
2.8. Spin-orbit coupling	102
2.9. Calculation of physical observables	104
3. Application to Metallic Glasses	109
3.1. Periodic structure models	109
3.2. Amorphous Ni and Ni-P glasses	112
3.3. Amorphous Cu-Zr glasses	114
3.4. Amorphous Mg-Zn glasses	116
3.5. Transport properties	117
3.6. Optical properties	123
4. Application to Magnetic Glasses	125
4.1. Crystalline FeB, Fe ₂ B and Fe ₃ B compounds	125
4.2. Amorphous Fe model	127
4.3. Amorphous Fe ₈₀ B ₂₀ model	132
5. Some Conclusions and Future Directions	136

Acknowledgements	137
6. References	137

3. RANDOM ANISOTROPY IN AMORPHOUS ALLOYS

by Eugene M. Chudnovsky

1. The Random Anisotropy Model	143
2. The Correlated Spin Glass	150
2.1. The Imry-Ma arguments	150
2.2. The random-anisotropy XY model	152
2.3. The random-anisotropy mean-spherical model	155
2.4. Experimental evidence of the correlated spin glass	157
2.5. The phase diagram	159
2.6. Coherent anisotropy	160
3. Behavior in a Magnetic Field	162
3.1. Zero-field susceptibility of the CSG	163
3.2. Magnetic disorder in the presence of a field	166
3.3. The magnetization curve	168
3.4. Dimensional crossover	171
Acknowledgements	173
References	173

4. MAGNETIC PROPERTIES OF VAPOR-QUENCHED AMORPHOUS AND METASTABLE CRYSTALLINE ALLOYS

by C.L. Chien

1. Introduction	175
2. Rapid Quenching Processes and Vapor Quenching Methods	178
3. Characterizations and Measurements of Magnetic Properties	180
4. Magnetic Properties of Crystalline Fe and Co	181
5. Composition Ranges for Amorphous Alloys	183
6. Magnetic Properties of TM-Metalloid Systems	185
7. Magnetic properties of TM-ET Systems	194
8. Toward the Amorphous Pure Metal Limit	202
9. Metastable Crystalline Systems	204

10. Conclusions	212
Acknowledgements	214
References	215

5. NEUTRON SCATTERING STUDIES OF THE SPIN DYNAMICS OF AMORPHOUS ALLOYS

by Jeffrey W. Lynn and Jamie A. Fernandez-Baca

1. Introduction	221
2. Neutron Scattering	223
2.1. Unpolarized neutron scattering cross sections	223
2.2. Polarized neutron cross sections	226
2.3. Neutron scattering from amorphous alloys	229
3. Spin Dynamics of Isotropic Ferromagnets	231
3.1. Small q	231
3.2. Invar systems	239
3.3. Large q excitations	243
4. Spin Dynamics of Amorphous Frustrated Magnets	246
4.1. Diluted magnets	247
4.2. Iron-rich systems	249
5. Future Directions	254
Acknowledgements	255
References	255

6. NUMERICAL STUDIES OF MAGNONS IN AMORPHOUS MAGNETS

by D. L. Huber

1. Introduction	261
1.1. Overview	261
1.2. Magnons	262
1.3. Eigenvalues and eigenvectors	263
1.4. Magnetic materials and model Hamiltonians	264
2. Amorphous Ferromagnets	265
2.1. Overview	265
2.2. Calculations	266

2.3. Discussion	268
3. Ferromagnets with Random Axis Anisotropy	269
3.1. Overview	269
3.2. Calculations	270
3.3. Discussion	273
4. Summary and Conclusions	276
5. References	276
6. Appendix	277
6.1. Overview	277
6.2. Recursion relations	277
6.3. Equation-of-motion method	279
 7. MAGNETISM AND MAGNETO-OPTICS OF RARE EARTH-TRANSITION METAL GLASSES AND MULTILAYERS	
<i>by D. J. Sellmyer, R.D. Kirby and S. S. Jaswal</i>	
1. Introduction and Scope	281
2. Overview of Magnetic Interactions and Structures	283
3. Perpendicular Anisotropy in Films and Multilayers	288
4. Electronic Structure and Magneto-Optic Effects	298
5. Domain Dynamics and Magnetization Reversal	305
5.1. Experimental results	307
5.1.1. Direct observations of magnetic domains	307
5.1.2. Time dependence of remanence decay and magnetization reversal	311
5.2. Models of magnetization reversal	315
5.2.1. Street and Woolley model	315
5.2.2. Single barrier height approach	317
5.2.3. The Fatuzzo model	320
5.2.4. Micromagnetic calculations and simulations	324
6. Concluding Remarks	330
Acknowledgements	332
References	332

CHAPTER ONE

THEORY OF MAGNETISM IN AMORPHOUS TRANSITION METALS AND ALLOYS

Yoshiro KAKEHASHI

Department of Physics, Hokkaido Institute of Technology
Maeda, Teine-ku, Sapporo 006, Japan

Hiroshi TANAKA

IBM Research, Tokyo Research Laboratory,
IBM Japan Ltd., 1623-14, Shimotsuruma, Yamato, Kanagawa 242, Japan

1. Introduction

In the past two decades, a large number of amorphous transition metal alloys showing fascinating magnetic properties have been found with the development of rapid quenching techniques [1-10]. Their physical properties, such as amorphous structure, magnetic properties, electronic states, transport properties, and magneto-optical properties, have been extensively investigated by means of microscopic experimental techniques. A variety of magnetism caused by structural disorder has opened a new field of "amorphous metallic magnetism", in which structural disorder and metallic magnetism are closely related each other *via* electronic structure. This chapter reviews theoretical aspects of recent developments in amorphous metallic magnetism of transition metals (TM) and alloys.

Needless to say, the most essential character of amorphous systems is structural disorder. In the past, experimental data for amorphous magnetic alloys have often prevented us from understanding the effects of structural disorder, because of the nonexistence of amorphous pure transition metals and the limited range of concentration in amorphous alloys. Amorphous transition metal alloys containing considerable amount of metalloids (typically 20 at.% B or P), were first systematically investigated

by Mizoguchi *et al.* [11-12], Hasegawa *et al.* [13], and O'Handley *et al.* [14-15] in the 1970's. The data showed that the magnetization and Curie temperature (T_C) were uniformly lower than those of crystalline alloys when they were plotted as a function of average d -electron numbers. Amorphous $\text{Fe}_{80}\text{B}_{10}\text{P}_{10}$ alloy [15], for example, shows ferromagnetism with the ground-state magnetization $M=2.1 \mu_B$ and $T_C=640$ K, while bcc Fe has $M=2.2 \mu_B$ and $T_C=1040$ K. Amorphous $\text{Co}_{80}\text{B}_{10}\text{P}_{10}$ alloy [15] also shows ferromagnetism with $M=1.1 \mu_B$ and $T_C=770$ K, which may be compared with $M=1.7 \mu_B$ and $T_C=1400$ K in fcc Co.

The magnetization vs. concentration curves were analyzed by using a concept of generalized Slater-Pauling curves suitable for the strong ferromagnets [16-19]. This method is based on the following identity:

$$M = Z_m + 2n_{\text{sp}\uparrow} \quad (1.1)$$

Here $n_{\text{sp}\uparrow}$ is the averaged number of sp -electrons with up spin. Z_m is the averaged magnetic valence defined by $Z_m = \sum_{\alpha} c_{\alpha}(2n_{\alpha\uparrow} - Z_{\alpha})$. c_{α} , $n_{\alpha\uparrow}$, and Z_{α} are the concentration, d -electron number with up spin, and the chemical valence for the constituent atom α , respectively. Since Z_m and $n_{\text{sp}\uparrow}$ are linear with respect to the concentration in transition metal alloys with strong ferromagnetism, we have a linear relation between M and Z_m .

The simple behaviors of M and T_C in TM-metalloids alloys were often regarded as inherent properties of amorphous structure rather than as effects of the presence of metalloids. It was concluded that the structural disorder merely introduced such simple behaviors into the magnetic properties [4]. However, the above picture changed with the appearance of amorphous transition metal alloys containing early transition metals or rare-earth (RE) metals in the 1980's [10,20-27]. In particular, Hiroyoshi and Fukamichi [22], Saito *et al.* [23], Coey *et al.* [24-25], Wakabayashi *et al.* [26], and Fukamichi *et al.* [21] found that the ferromagnetism in Fe-rich amorphous alloys collapses completely beyond 90 at.% Fe, and that a new spin-glass (SG) phase appears in which the transition temperatures are 120 K irrespective of the second elements, contradicting the early data on alloys containing metalloids. Furthermore, Fukamichi, Goto, and Mizutani [27] found that the Curie temperatures in Co-rich Co-Y amorphous alloys are enhanced as compared with those in their crystalline counterparts. It was suggested that the Curie temperature extrapolated to amorphous pure Co should reach 1850 K, which is 450 K higher than for fcc Co. These drastic changes of magnetism in the vicinity of amorphous pure metals have revealed the important role of structural disorder in amorphous metallic magnetism. It is our main purpose to describe such a new feature of amorphous metallic magnetism from the theoretical point of view.

There are two directions in the development of solid state physics. One is to include more and more electron correlations in order to achieve microscopic understanding of magnetism, metal-insulator transition, and high- T_C super conductors [28-29]. In particular, theoretical description of the magnetism in transition metals and alloys

has been the main part of correlation problems, since the transition metal alloys show both itinerant and localized behaviors in their magnetic properties [30]. The other direction is to take account of more and more disorder [31]. The theory of transition metals and alloys in this direction has been developed in the order of magnetic impurities, substitutional alloys, and amorphous alloys [32-33]. A quantitative theory of amorphous metallic magnetism is, therefore, a goal that these two lines of investigation should reach at the end of the development.

This chapter is organized as follows. We discuss in the following section the theoretical problems and historical developments in amorphous metallic magnetism. There are discussions on the amorphous structure, electronic structure, and magnetism in Sec. 2.1. The most important advances in the recent theories of amorphous metallic magnetism have been the first-principles band calculation of the ground-state amorphous electronic structure and the development of a finite-temperature theory. The former was established by Fujiwara [34-35] through a combination of the linear-muffin-tin-orbital (LMTO) method [36-37] with the recursion method [38-39] in the framework of the band theory, which will be reviewed in Sec. 2.2. The latter was recently proposed by Kakehashi [40-42] on the basis of the functional integral method [43-45] and the distribution-function method [46-47]. The finite-temperature theory is reviewed in Sec. 2.3. Section 3 is devoted to the magnetism in amorphous transition metals, in particular, amorphous Fe, Co, and Ni. Numerical examples for these systems will demonstrate the basic effects of structural disorder. The magnetic properties of transition metal alloys are governed by both structural and configurational disorders. In Sec. 4, we discuss the magnetic properties of TM-TM alloys (Sec. 4.1), TM-RE alloys (Sec. 4.2), and TM-metalloid alloys (Sec. 4.3) on the basis of their electronic structures, which have been obtained in the last decade.

2. Theoretical Approach to Amorphous Metallic Magnetism

2.1. Basic Problems and Developments

Amorphous transition metals and alloys are characterized by structural disorder and itinerant magnetism. In the latter, electrons move from site to site with the electron hopping integrals which form electronic band structure. The magnetism is known to be caused by competition between the electron hopping and the electron-electron Coulomb interactions. The magnetic properties are sensitive to the band structure rather than the Coulomb interactions in metals, because the screened Coulomb interactions act mainly between the on-site electrons, and thus depend very little on the structure of solids. It is therefore essential to take into account the effect of the structural disorder on the electronic band structure. This implies that it is difficult to understand the magnetic properties of amorphous transition metals and alloys without taking the following regular steps:

- (1) Construction of amorphous structure

(2) Calculation of electronic structure

(3) Constructing a theory of amorphous magnetism on the basis of the electronic structure

In what follows, we briefly discuss the theoretical problems in these three steps, as well as historical developments.

2.1.1. Amorphous structure

In crystalline systems, the crystal structure and lattice constants can be determined by analyzing the Bragg peaks in X-ray or neutron diffraction, and thus the electronic structure can be calculated on the basis of the Bloch theory [37,48]. This procedure is no longer available for amorphous metals and alloys, because of the lack of translational symmetry. X-ray experimental techniques [49,50] give us only the pair distribution functions (PDF) $g_{\alpha\gamma}(R)$, which are defined as follows:

$$g_{\alpha\gamma}(R) = \frac{\rho_{\alpha\gamma}(R)}{\rho_{\gamma}} , \quad (1.2)$$

where $\rho_{\alpha\gamma}(R)$ is the density of a γ atom at a distance R from an α atom, and ρ_{γ} is the density of the γ atom. The distribution function $g_{\alpha\gamma}(R)$ converges to 1 as R approaches infinity. Reduced PDF $G_{\alpha\gamma}(R)$ are also used, and are defined as follows.

$$G_{\alpha\gamma}(R) = 4\pi R^2 \{g_{\alpha\gamma}(R) - 1\} . \quad (1.3)$$

Although we can obtain further information on the amorphous structure from EXAFS (extended X-ray absorption fine structure) and neutron measurements [9], it is not possible to determine experimentally all the atomic positions in the amorphous structure, though these are indispensable for electronic-structure calculations of amorphous systems and microscopic understanding of magnetic properties. This difficulty imposes us on the above theoretical problem (1), namely, how to construct a reasonable model for an amorphous structure.

The simplest model for amorphous metals and alloys is the dense random packing of hard spheres (DRPHS) model. The model was first proposed by Bernal [51-52]. He constructed the model by squeezing and kneading rubber bladders filled with ball bearings of the same size. He found that the DRPHS model consists of only the five types of polyhedra (so-called 'Bernal holes') shown in Fig. 1.1. Finney [53] constructed a much larger cluster model by using the same method as Bernal, and obtained a PDF. Bennett [54] constructed a DRPHS model by using a computer. His algorithm for generating the cluster is as follows: (1) make an equilateral triangle consisting of three hard spheres touching each other, (2) list up all the pockets in which new sphere could be added in hard contact with three spheres which already exist in the cluster, (3) add the new sphere to the nearest pockets from the origin of

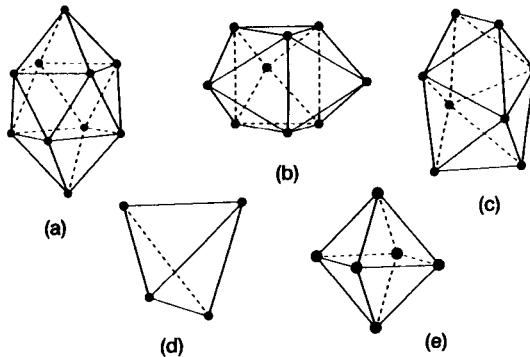


Fig. 1.1: Five types of polyhedra in the DRPHS model: (a) Archimedean antiprism, (b) trigonal prism, (c) tetragonal dodecahedron, (d) tetrahedron, and (e) octahedron

the cluster, and (4) return to the step (3). The PDF calculated from this computer-simulated cluster model was quite similar to that of the ball bearing model. It was however somewhat different from the experimental one. In particular, the splitting of a second peak into two peaks and their relative amplitudes could not be reproduced.

Ichikawa [55] suggested that this splitting was related to the tetrahedral local atomic structure, and improved Bennett's algorithm. He introduced a parameter Λ which measured the perfection of the tetrahedral structure. It is given in the form

$$\Lambda = \frac{r_{ij}^{\max}}{R_i + R_j} . \quad (1.4)$$

where each i or j denotes one of the three spheres in the cluster forming a new pocket in step (2) of Bennett's algorithm. r_{ij} is the distance between the spheres i and j . R_i is the radius of the sphere i . Obviously, Λ changes from 1 to 2 for a single-size sphere system in Bennett's algorithm. The deviation from a perfect tetrahedral structure is evaluated as the deviation from the condition $\Lambda = 1$.

Ichikawa constructed a DRPHS model with several Λ values, and found that the PDF was well reproduced when Λ was 1.2. Although the PDF was well reproduced, the packing fraction of the model was smaller than in the experimental data because of porosity of the model structure.

These disadvantages were overcome by a process for structural relaxation of a DRPHS model through the atomic force produced by appropriate pair potentials [56,57]. This method, the so-called relaxed DRPHS model, was proposed by Cargill [49].

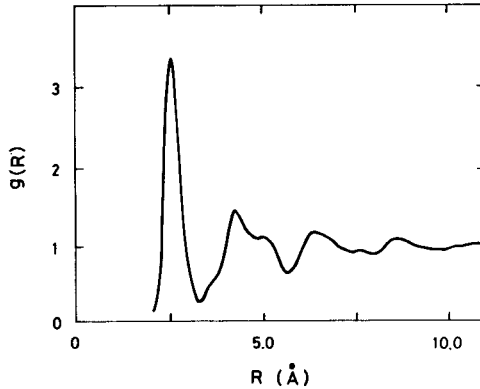


Fig. 1.2: Pair-distribution function of computer-generated amorphous iron [58]

Figure 1.2 shows an example of a pair-distribution function for amorphous Fe calculated by Yamamoto and Doyama [58], using the relaxed DRPHS model. The first peak at $r_1 = 2.54 \text{ \AA}$ is sharp, and is clearly separated from the second and third ones. This means that there exists a well-defined nearest-neighbor (NN) shell even in amorphous systems. The ratio of the fluctuation of the NN interatomic distance to the average NN distance is estimated from the width of the first peak to be 0.067, which is in agreement with recent experimental data for Fe-rich amorphous alloys [59]. The second peak, at $r_2 = 1.67r_1$, is considered to originate in the local structures of the rhombi, each of which consists of two regular triangles with a side r_1 , and the hexahedra, each of which consists of two tetrahedra with a side r_1 . The third peak at $r_3 = 2r_1$ is associated with three contact atoms on a line.

The thermodynamical molecular-dynamics (MD) method [60] is a more sophisticated way of constructing an amorphous structure. In this method, the constituent atoms are distributed in a box with a periodic boundary condition, and the Newton equations of atomic motion are solved by assuming appropriate short-range interatomic pair potentials. Rapid quenching is simulated by reducing the kinetic energy, which is proportional to the temperature, at constant time intervals, under the condition that either the pressure or volume is constant. The method aims to simulate the formation of an amorphous structure according to the physical principle by using computers.

More recently, an *ab-initio* MD method was developed by Car and Parrinello [61]. In this method, interatomic forces are calculated directly from electronic structure and atomic structure without any empirical parameters, so that both the electronic and atomic structure are treated on an equal footing. Car and Parrinello greatly

accelerated the *ab-initio* MD calculation by solving the equations of motions for both atoms and electrons at the same time. The method is most efficient when combined with the pseudo-potential technique and plain wave orbitals, and has been applied to amorphous semiconductors [62-63], surfaces of semiconductors, and interdiffusion in semiconductors [64].

Although MD calculations are very effective for constructing an atomic structure model of amorphous materials, there are some limitations at the present stage. The MD calculations are limited to rather small systems (100 ~ 1000 atoms in a box) because of the insufficient efficiency of computers. Moreover, the minimum cooling rate in the MD is about 10^{11} (K/s), which is too large as compared with the experimental rates ($\simeq 10^6$ (K/s)). We do not enter into the details of these problems because they fall outside the scope of our discussion.

2.1.2. *Electronic structure calculations*

Amorphous systems lose translational symmetry and the Bravais lattice. This makes it impossible to calculate their electronic structures by making use of the Bloch theorem. Much theoretical effort, therefore, has been concentrated on this problem in the past twenty years.

The best single-site approximation was first established by Roth [65-66]. She introduced a k -dependent effective selfenergy caused by the structural disorder. The selfconsistent equations for the selfenergy were obtained from a single-site decoupling to the coupled equations for the averaged T-matrices. This is called the effective medium approximation, and is regarded as a natural extension of coherent potential approximation (CPA) in disordered substitutional alloys [67-68].

The single-site approximation does not describe the details of the local environment effects (LEE) on the densities of states and the local magnetic moments, which are in particular important for transition metal alloys. Fujiwara [34] developed a general method going beyond single-site approximation. He combined the tight-binding linear muffin-tin orbital method (LMTO) [36] with the recursion method [38-39] in electronic-structure calculations. The former gives an effective tight-binding Hamiltonian which greatly simplifies the band-structure calculations within the framework of the local-spin density functional theory [69-71]. The latter is powerful for the electronic structure calculations of disordered systems when their Hamiltonians can be described in a tight-binding form. The method provide us with first-principles electronic structure calculations for a given amorphous structure, and is regarded as the best method for calculating the electronic structure of amorphous transition metals and alloys. We will review the theory briefly in Sec. 2.2.

An alternative approach which also aims at the first-principles calculations of amorphous metals and alloys is to simulate the amorphous structure by means of a crystal with a large unit cell. This "supercell" approach [72-76] was made effective by the development of the linear method [36-37] and super computers. It is now possible

to perform first-principles calculations for an amorphous “compound” with 50 to 100 atoms in a unit cell.

2.1.3. Calculating magnetic properties

When we calculate magnetic properties on the basis of electronic structure, we have to take into account the spin degrees of freedom. This causes two major difficulties in the theoretical investigations of amorphous transition metals and alloys. First, the local magnetic moments (LM) change their directions as well as their amplitudes according to their local environments, as a result of the structural disorder. The question is then how one determines the LM configuration selfconsistently. This is not an easy problem even at the ground state when the ferro- and antiferro-magnetic interactions compete with each other in the disordered system, because many local minima in energy are expected, and finding the lowest energy is beyond the trial-and-error calculations. The problem is essential for the description of spin glasses, in which LM's are spatially disordered with no net magnetization, but are ordered or frozen thermodynamically [77-78].

As far as the ground state is concerned, one possibility may be to use the simulated annealing method [79]. In this method, the total energy E is minimized relative to the spin densities $\{\langle m_i \rangle\}$ by generating a succession of $\{\langle m_i \rangle\}$'s with a Boltzmann-type probability for a fictitious temperature T in a Monte-Carlo method, or by solving the equations of motion for classical particles with a “potential energy” E in the MD method. When $T \rightarrow 0$, the state with the lowest energy may be reached. However, these methods have not been applied yet.

The second problem is that transition metals show local-moment as well as itinerant-electron behaviors in their magnetism [80]. The band theory based on the Stoner model explains the non-integer ground-state magnetization, the existence of the Fermi surface, and the T -linear specific heat at low temperatures. But it does not lead to the local-moment behaviors such as a reasonable T_C , the Curie-Weiss susceptibility, and a large specific heat at T_C in Fe, Co, and Ni. Although the difficulty has been a long-standing problem in the theory of itinerant magnetism, it has been clarified in the past decade that it can be solved by taking into account the thermal spin fluctuations missing in the Stoner model [29-30]. Such a spin fluctuation theory has recently been applied to amorphous metallic systems by Kakehashi [40-42]. He took into account thermal spin fluctuations by making use of the functional integral method developed by Cyrot [43], Hubbard [44], and Hasegawa [45], and determined the LM distribution due to the structural disorder using the distribution-function method developed by Matsubara and Katsura [46-47]. This is the only theory which is available at finite temperatures at present. It will be reviewed in detail in Sec. 2.3.