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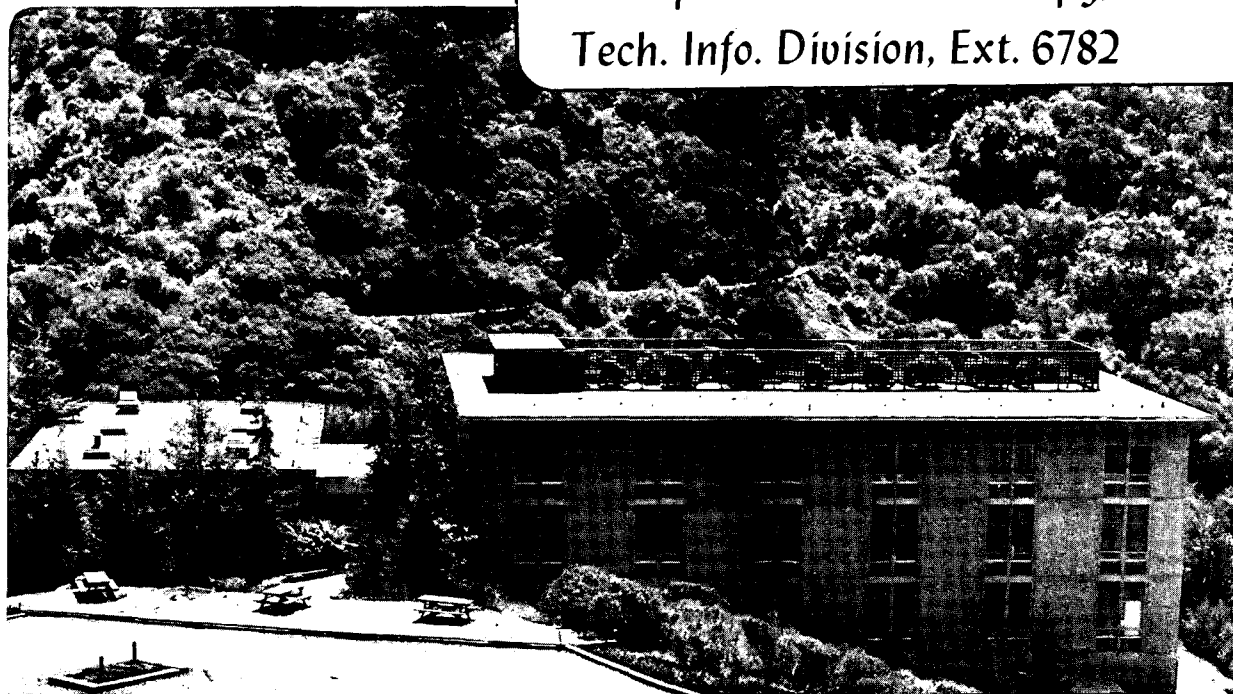
SUMMARY: DISORDERED SYSTEMS IN PERSPECTIVE

L.M. Falicov

September 1981

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## SUMMARY: DISORDERED SYSTEMS IN PERSPECTIVE

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## INTRODUCTION

The summary of an Advanced Institute like this should ideally report the progress made in the field and the problems which remain unsolved, and minimize as much as possible the prejudices of the summarizer and the platitudes inherent in a general talk. I will try to the best of my abilities to emphasize the first two and reduce the last two to their irreducible minimum.

The field of disordered systems has had an explosive development<sup>1-3</sup> in the 1970s, and it is starting the decade of the 1980s with uncommon vigor and in good state of health. It is a field of enormous interest to a variety of constituencies: from the topological aspects of the structures which fascinate the mathematician, to the statistical way of describing the phenomena, to the variety of materials which occupy the materials scientist, to the richness of physical phenomena which involve sophisticated experiments with a variety of techniques, to the design, fabrication and successful production of devices and industrial products, of which catalysts and solar cells are only two examples, the field of disordered systems is a truly interdisciplinary endeavor. It is impossible of course to cover all these areas adequately in an Institute like this. But we have managed to encompass a large amount of territory, and the rapid progress has been in evidence in the enthusiasm of the speakers, the new results reported here and the enormous promise for the future.

## PROPERTIES OF DISORDERED SYSTEMS AS MEASURED AND/OR CALCULATED

The determination of STRUCTURAL properties remains by far the

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central problem of the field. How do we describe a disordered system? Have we developed a formalism to characterize a given system? When we take averages, what kind of a population do we choose for that purpose? Are long- and short-range orders of equal importance? Can we influence the various forms of disorder by preparation techniques and wise control of external agents?

We have heard various points of view in this Institute. The description of disordered alloys presented by the various speakers is a good example of the variety of meanings that the word disorder may evoke. It is fair to say that in describing structural properties of disordered systems somebody's noise becomes somebody else's signal.

The promising feature which has emerged out of our proceedings is that we are making considerable progress not only in developing a common language to describe the structure of disordered systems, but also have now experiments which measure these parameters, and even some theories which make a valiant attempt to calculate them from either first principles or from microscopic, independent input parameters.<sup>4,5</sup>

Since the field is dominated by traditional solid-state physicists, the most common properties to be determined in these materials, either experimentally or theoretically, are the staple bread and butter of the crystalline solid-stater: ELECTRONIC,<sup>3-9</sup> ELASTIC<sup>10</sup> and VIBRONIC<sup>1,11-14</sup> properties, ELECTROMAGNETIC<sup>2,9,15-17</sup> responses, THERMODYNAMIC<sup>18,19</sup> behavior and TRANSPORT<sup>2,3</sup> properties. In varying degrees of accuracy, with varying degrees of specificity and with varying degrees of difficulty, all these properties give us now a much more consistent picture of what disordered systems are and how their excitations behave under a set of different conditions.

#### EXPERIMENTAL TECHNIQUES

The experimental tools which have helped us understand disordered materials represent the most spectacular progress made in the field. For the purpose of the discussion we can classify them in four different groups.

##### (1) Techniques which reveal information in $\vec{k}$ and frequency spaces

These yield of course the most detailed and useful pieces of information to be obtained. They give us insight on the dynamics of the system and how these dynamic modes are distributed throughout the disordered system. Of these techniques, NEUTRON SCATTERING is still the king.<sup>20,21</sup> As we have seen in the various talks in this Institute, it tells us a lot about magnetic, vibronic, and electronic excitations. It gives us excitation energies, decay times, space resolution, etc. It is not difficult to predict that neutron scat-

tering will remain the most important technique in the field for many years to come. The development of new sources of neutrons, with higher fluxes and better monochromaticity, coupled to better detectors with better resolution assure the predominance of neutron-scattering experiments.

The by now classical X-RAY SCATTERING experiments, with their almost 80 years of distinguished service to condensed-matter physicists, are in more senses than one having a renaissance. The existence of tunable sources of synchrotron radiation,<sup>22</sup> which are sprouting all over the world, guarantees in the future new and exciting experimental data, not easily available with the old techniques.

The same synchrotron sources will facilitate obtention of ANGLE-RESOLVED PHOTOEMISSION<sup>22,23</sup> data both in the ultraviolet and in the X-ray range. This technique was modestly represented in this Institute<sup>23</sup> and we will hear much more from it, as applied to disordered systems, in the years to come.

(2) Techniques which yield frequency information averaged over large regions of space ( $\vec{k} = 0$ )

This is a traditional area of the solid-state experimentalists. The measurements are the ones described in any solid state physics textbook:

- conductivity, susceptibilities and dielectric responses;
- thermodynamic quantities (specific heat in particular);
- optical properties;
- integrated photoemission spectra;
- Raman scattering.<sup>13,14</sup>

Their usefulness is selfevident by looking at the current literature and at the proceedings of this Institute.

(3) Techniques which yield local (small regions) information

These include some classical techniques

- nuclear magnetical resonance (NMR);
- impurity Raman scattering;
- X-ray absorption and emission;

as well as some techniques borrowed from nuclear physics

- muon-spin rotation ( $\mu$ SR)

and some new techniques made possible by the new tunable X-ray sources<sup>22</sup>

- extended X-ray absorption fine structure (EXAFS).

The first three are well-known and tested;  $\mu$ SR is a fascinating new technique<sup>24,25</sup> which potentially can give information not available otherwise. In all fairness however, it should be said that up to this moment the information obtained from  $\mu$ SR in a large variety of systems, almost exclusively crystalline, could only be disentangled if precise information was available from other techniques (X-ray or

neutron scattering). On the other hand EXAFS has already proved to be<sup>26,27</sup> a unique opportunity in determining, with great accuracy, the local environment of a given atomic species; the information obtained from EXAFS, without being all encompassing, is accurate and unique.

#### (4) Purely spatial (static) information

The information in this category is a special limit of the other three and in principle should be the starting point which gives us the structure and properties of the ground state. In the case of crystalline solid state physics, this information was the foundation stone which allowed us to understand solids in general, and metals in particular. The information goes from the very general

- density measurements,
- mechanical properties,

through the classical diffraction experiments

- X-ray diffraction,
- neutron diffraction,

to the more detailed experiments which yield accurate information on the electron properties

- de Haas-van Alphen effect,<sup>9,10</sup>
- positron annihilation,<sup>28,29</sup>
- Compton scattering.<sup>30</sup>

The information obtained from the global density and mechanical properties is by necessity restricted to isolated numbers. The information obtained from the diffraction experiments is almost exclusively restricted to pair-correlation functions, and partial and total structure factors.

The de Haas-van Alphen effect,<sup>31</sup> which proved to be such a magnificent tool for pure crystalline metals has, in the area of disordered materials, very restricted applicability: it can only be observed in very dilute alloys of very specific metals.<sup>9,10</sup> Even so, as we have heard in this Institute, the information obtained is detailed and susceptible to easy interpretation and to prompt comparison with theory.

Positron annihilation<sup>28,29</sup> and Compton scattering<sup>30</sup> experiments are (and have been for more than a decade) still the most promising techniques to give static information. Their promise remains still a promise. Information obtained from them is available in the literature but the rate at which it appears is slow, and the effort required to extract the useful details from the obvious uninteresting bulk data is somewhat overwhelming for all but the very sturdy and committed experimentalists.

#### DISORDERED MATERIALS

The field of disordered systems is in fact a paradise for the

materials scientists. The variety and diversity of the materials of interest boggle the imagination and challenge the ingenuity of any materials-oriented physicist or chemist. The list of the studied materials makes in itself the index of a substantial treatise.

-Alloys

These are by far the deans of the field, probably the most widely studied systems in nature, of interest to mankind since the end of the stone age. It is therefore not surprising that in this Institute they have been the subject of the most intensive studies.<sup>4,6-9,16,32,33</sup>

-Metallic Glasses<sup>34</sup>

These are relatively new systems in which structural disorder is superimposed to constitutional disorder. It is a sad irony of nature that no single elemental metallic glass is known. However, the progress made in the understanding of the structural, electronic, thermodynamic and transport properties of binary and ternary metallic glasses augurs well for the future of this new subfield.

-Insulating Glasses<sup>3,12,14,35,36</sup>

These are, with alloys, the standard disordered materials. In this case, the disorder is purely structural. Silica glass has been known to mankind since the beginning of civilization. Its microscopic properties, however, are just beginning to be understood. Its structural complexities,<sup>36</sup> its vibronic modes<sup>14</sup> and its thermodynamic properties make fertile ground for theoretical simulations,<sup>12</sup> experimental measurements and comparison between the two.

-Amorphous semiconductors<sup>37,38</sup>

They constitute the most challenging materials of today, mainly because of their potential applications as photovoltaic cells.<sup>37</sup> Their simple, and at the same time complex structure, the possibility of passivation of their active centers by the addition of hydrogen or fluorine, the presence of mobility edges<sup>3</sup> and band gaps makes this field the industrial moving force in the area of disordered solids.

-Highly-doped semiconductors<sup>3,15,39-41</sup>

These systems constitute an ideal situation to study metal-insulator transitions<sup>15,39,41</sup> and other properties of disordered systems. Since its basic materials are the well developed crystalline semiconductors to which impurities are added, they have been studied in the last 20 years with more care and devotion than probably any other disordered system.

-Disordered magnetic systems,  
which include, among others,

- disordered ferromagnets,<sup>35,42-44</sup>
- disordered antiferromagnets,<sup>21,42,43</sup>
- disordered magnetic alloys,<sup>33</sup>
- spin glasses.<sup>18,19,36,46-49</sup>



Their popularity has increased exponentially in the last six years, both theoretically and experimentally, and some of the new physical concepts introduced to describe them are discussed below.

-Liquids<sup>5,50</sup>

They constitute a class of their own and are the subject of intensive studies, mostly by physical chemists.

Other systems which have been mentioned in this Institute and to which microscopic methods or their macroscopic extensions are applied are:

- polymers,<sup>51</sup>
- small particles,<sup>52</sup>
- composites,<sup>10,17</sup>
- rocks,<sup>10,17</sup>
- disordered Quantum Solids.<sup>53</sup>

This collection constitutes only a partial list of the kinds of materials that are already of interest to physicists, chemists and materials scientists working on disordered systems. There is no doubt in my mind that other disordered materials will be developed and studied intensively in the very near future.

#### MODELS AND THEORETICAL METHODS

It is obvious from the content of the lectures in this Institute that the theorists are at a stage of rather explosive developments. The methods and models developed by them are moving in many, sometimes opposite, directions. It is difficult, if not impossible, to classify them in a unique way. A rough way of grouping them will be as follows:

(1)  $\vec{k}$  space methods

-Rigid band models,<sup>9</sup>

old, almost obsolete but sometimes unbelievably useful in interpreting simple (or not so simple) properties of alloys.

-Virtual crystal methods

These are the favorite of the traditional solid staters who, with a single stroke, restore periodicity to a nonperiodic system. Although they have been discredited because of their naivete, they surprisingly reappear as zeroth order approximation in more sophisticated situations,<sup>54</sup> some of them discussed in this Institute.

-Scattering methods

These have been the favorite approach of the theorists since the middle of the 1960s, and the reason for the theoretical renaissance in the field of electronic and vibronic properties of disordered systems. They include the by now familiar average t-matrix<sup>1,7</sup> (with its various stages and approximations) and the queen of the theoret-

ical methods, the coherent potential approximation (CPA), which has proved to be the most successful single-site scattering formulation.<sup>1, 6-9, 11, 54-57</sup> The extension of CPA to include more than one center and, if possible, short-range correlations has not been however developed at the same intensive rate. Some prospects are nonetheless promising<sup>54, 57</sup> at this point and I have no doubt that new avenues will be found to extend this highly successful approach to more complicated situations.

## (2) Real space methods

These methods are advocated by those who believe that in dealing with disordered systems,  $\vec{k}$ -space should be thrown away.<sup>58</sup> The different varieties of this approach include

- cluster calculations,<sup>4, 5, 51</sup>
- continued-fraction methods,<sup>59-62</sup>
- the recursion method,<sup>63-65</sup>
- transfer-matrix methods,<sup>4, 5, 33, 42, 51, 66, 67</sup>

all of which focus their attention on the immediate neighborhood of a given point and find ways of extrapolating the local properties to infinity to cover systems in the thermodynamic limit. Their success has been repeatedly proved by the various contributions to this Institute.

These real space approaches have the additional advantage of permitting the study of a variety of physical effects: surfaces,<sup>33, 42</sup> local defects,<sup>42, 51, 68, 69</sup> short-range order,<sup>4, 5, 67</sup> and the transfer of electronic charge<sup>4, 5, 67</sup> between various chemical species or between atoms in various environments. These effects are important in designing catalysts<sup>42, 70</sup> and in studying the stability of a variety of systems.

## (3) Effective medium theories<sup>32</sup>

These are general methods, some of which are included in (1) and (2) above, which replace the whole system or those parts of it which are outside a central cluster by a simpler homogeneous medium determined either selfconsistently or from experimental measurements of structure factors or pair-correlation functions.

## (4) Numerically intensive methods<sup>12, 71</sup>

These are essentially computer experiments which solve numerical-ly equations of motion, renormalization equations, or density-matrix equations, taking full advantage of the capabilities of the huge memory of the latest generation of "number crunchers". These methods include

- simulation techniques,<sup>12, 43</sup>
- renormalization techniques,<sup>44, 72</sup>
- molecular dynamics calculations.<sup>73-75</sup>

Their success is at this point beyond question. With the proper selection of systems, number of particles, number of chemical components, degrees of freedom, intermolecular forces and proper boundary conditions, a large variety of physical effects can be described, explained, analyzed, and even predicted.<sup>75</sup> In a sense, these theoretical methods have become the numerical experiments which constitute the probing grounds for more analytically-oriented theoretical models.<sup>1</sup>

#### (5) Idealized models

These topological constructions constitute less realistic but more mathematically-oriented approaches to the study of systems, disordered in nature, but with some inherent structure, particularly stressed by the approach. Among others they include

- networks,<sup>76</sup>
- Bethe lattices,<sup>4, 33, 68, 69, 77</sup>
- Husimi cacti,<sup>77</sup>
- curved-space topological models.<sup>78</sup>

Their lack of reality is more than compensated by their intrinsic mathematical beauty and by the fact that they can explain in simple, elegant formulas effects that are difficult to abstract from more cumbersome, realistic methods.

### IMPORTANT PHYSICAL EFFECTS IN DISORDERED SYSTEMS

The existence of disorder brings some new properties to condensed-matter systems which are interesting in themselves, and naturally the subject of the most intensive study. I will list some of them here.

#### (1) Localization of excitations due to disorder

This has been the most striking effect of the lack of periodicity and Anderson's contribution of 1958, which opened up this new area,<sup>79</sup> has been well rewarded by a Nobel prize. The field has had a renaissance in the last few years due to the interest in systems with restricted dimensionality.<sup>81, 84</sup> The phenomenon of localization in one and two dimensions has occupied some of the most active minds in the field, and was well represented in this Institute.<sup>82, 84</sup> The problems in the area however are by no means solved, and the activity in the future will no doubt continue at a frantic pace.

#### (2) Phase transitions

The presence of disorder brings a new and challenging component into the area of phase transitions. Disorder is in itself the cause of some phase transitions but it influences, and sometimes drastically, other critical phenomena. The most interesting transitions

to study are

- the glass transition itself;<sup>26</sup>
- metal-insulator transitions;<sup>3,15</sup>
- order-disorder transformations;<sup>4,33,67</sup>
- magnetic transitions (including ferro, ferri, antiferro and spin-glass transitions);<sup>19,21,33,45-49</sup>
- percolation transitions as they influence any and all of the above;<sup>2,21,44</sup>
- influence of disorder on other transitions (e.g. superconductivity<sup>16</sup>).

(3) Abundance, variety and instability of low frequency excitation modes

It has become now an almost fixed rule the fact that in every meeting in which disordered systems are discussed, at least one new low frequency mode (vibronic, elastic, magnetic, electronic) is reported. Their number and variety seem to be endless and they are responsible for the richness of phenomena observed in disordered materials. Just to mention a few, we have heard in this meeting of two longitudinal acoustic waves in composite materials;<sup>10,17</sup> splitting of optical modes are common and sometimes puzzling;<sup>12,14</sup> famous tunneling modes<sup>35,36</sup> due to two-level systems in most glasses have been postulated for sometime now, are considered responsible for the linear specific heat of glasses,<sup>36</sup> but unfortunately have not yet been observed directly by neutron diffraction or any other scattering technique.

(4) Thermodynamic foundations

The disordered systems which we consider open up a new area of study of thermodynamic properties. In particular the distinction between equilibrium and nonequilibrium process, between stability and longtime metastability have become blurred.<sup>18</sup> It is obvious that we are dealing here with systems very different from either crystalline solids or liquids. The new areas of statistical mechanics which open up under these conditions have barely been explored. New and challenging concepts are being discussed. These include the possibility of non-ergodicity,<sup>18</sup> nonanalytic behavior of the thermodynamic functions (with a consequent breakdown of the ordinary expansions)<sup>36</sup> and even the approach to chaotic behavior through the presence of nonlinear forces.<sup>85</sup>

In this respect numerical methods will play a fundamental role in our understanding of this behavior. As an example we have heard in this Institute the report of a new "classical" phenomenon, the existence of "quench" echoes<sup>75</sup> which appears, clearly and beautifully, in molecular dynamics calculations.

## IMPORTANT NEW PHYSICAL CONCEPTS

In the studies of disordered systems some new concepts have been developed which have gone beyond the restricted interest of the area and have influenced science in general, physics in particular. I will simply mention them here as a simple way to satisfy our egos for these achievements.

- (1) The concepts of percolation<sup>2,36</sup> and localization;<sup>36,79-84</sup>
- (2) The concept of frustration, a new way of classifying degenerate ground states;<sup>19,47-49</sup>
- (3) The concepts of mobility edges, mobility gaps and band tails;<sup>3,15,40,85,86</sup>
- (4) The concept that there is a fundamental interplay between dimensionality and the role of disorder.<sup>80,81,84</sup>

## EPILOGUE

I would like to finish my summary by expressing, on behalf of my colleagues and myself, our special thanks to Mike Thorpe for his skillful organization of the meeting, his judicious and tasteful choice of subjects and his patience, energy, and good humor. Special thanks are also due to Ms. Dolores Sullivan, to Michigan State University, to NATO, to the industrial sponsors, for having taken good care of us; and to the musicians, dancers, singers, actors, windsurfers, drivers and tour guides who made of our lighter afternoons and evenings enjoyable experiences.

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