

INTERNATIONAL SEMINAR ON AMORPHOUS MATERIALS: A NEW SCIENCE AND A NOVEL TECHNOLOGY—A MEETING REPORT

They conduct themselves in a disorderly and unruly fashion with little regard for established rules. They merrily flout our heritage, patiently and painfully acquired from past generations. They mock all that we have learnt to respect, cherish and obey. Ladies and gentlemen, it is time again to impose law and order, and impose it we shall, no matter what the cost. K.M.

Amorphous materials, their science and their technology, have finally come of age. A legitimate question would be, why so late?

There are several reasons for this late flowering and recognition, one of them being the preponderance of terrestrial minerals that originate from the slow cooling of lava. The ubiquitous presence of water reduces the viscosity of terrestrial magmas and increases the diffusion rate for oxides dissolved in them, thus favoring the growth of crystals.¹ Because of the rarity of water on the moon, one finds, in contrast, only a few minerals but many types of glasses. Undoubtedly, space geologists will have to acquire greater knowledge of glasses and lesser of minerals, opposite of their emphasis in their present habitat.

In spite of the relative abundance of crystalline materials, there was a fair amount of interest in amorphous solids until the discovery by W. H. and W. L. Bragg, in 1912, of X-ray diffraction, which directed the attention of physicists to crystalline solids for the next two generations. Combined with the predilection of science for simplicity of formulation, crystalline materials gained primacy, and a theoretical framework, based on F. Bloch's theorem for a periodic lattice and the concept of reciprocal space, emerged.

However, the fact that many phenomena are essentially unaltered by the absence of a periodic lattice has forced a reappraisal of the existing theoretical framework. The paradox has obliged physicists to deepen their understanding of solids, leading them to a better appreciation of chemical concepts and of the relation between bonds and bands. Combined with the novel phenomena induced by disorder, the stage is now set for intense activity in amorphous structures. The quiet revolution is not confined to materials, but extends to geology, to biology where disorder abounds, and to mathematics with its recent interest in fractals and chaos.

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The extraordinary richness of the field was evident at the International Seminar on Amorphous Materials: A New Science and a Novel Technology, held in Ft. Lauderdale, Florida, on February 13-15, 1985, under the auspices of Florida Atlantic University in cooperation with S. P. Wolsky of Ansum Enterprises, Inc. The purpose of the seminar was to bring together individuals and groups interested in the science and increasing applications of amorphous materials for diverse technologies ranging from photovoltaics and Xerography to metallurgy, and for devices exploiting electronic, optical, and magnetic phenomena. A wide spectrum of topics was discussed, including amorphous semiconductors and metals and dealing mainly with their technologically important aspects. Detailed and comprehensive talks by leaders in the field, drawn from universities and governmental and industrial laboratories, were geared toward educating the novice and bringing experts to the leading edge of amorphous materials technology. The seminar participants possessed equally broad backgrounds and, besides researchers and technologists from the fields of condensed matter physics, materials science, metallurgy, and electronic engineering, included managers whose common concern lay mainly in applications of fundamental concepts to the solution of complex technological problems.

I served as chairman of the seminar and delivered the opening talk, "Amorphous Materials: Introduction and Overview."

Amorphous materials—whether semiconductors, insulators, or metals—are inherently complex and present concomitant challenges for their understanding at a level comparable to that for crystals. First, their atomic structure cannot be characterized simply from a finite number of space symmetry groups. There are literally infinite possibilities! The atomic structures of noncrystalline solids are obviously not completely random, like those of gases, because of the constraint that chemical bonds must be formed between atoms. Atoms are obliged to be in contact with one another, and a certain regularity is thereby imposed on the nearest neighbor shells. In a monatomic ideal amorphous solid, the cluster of an atom and its neighbors corresponds, in a sense, to the unit cell in a crystal.

Although the bond distances and angles in the cluster are not uniquely defined and take a certain distribution of values, the cluster is the unit that is repeated,

within the limits of these distributions, at an infinite number of sites throughout the noncrystalline structure. However, unlike the unit cell in a crystal, the repetition scheme is not periodic, and the orientation of the characteristic structural cluster is different at every site. As a result, any amorphous solid is infinitely more varied than a crystal. Plausible structural models can therefore provide equally valid fits to easily measurable average properties, such as radial distribution functions and density. Hence it is important to characterize models by higher order correlation functions in order to compare them with experiments that supplement the diffraction data. In appropriate circumstances, neutron diffraction is a powerful tool in obtaining three correlation functions for a binary magnetic alloy. Techniques that focus on the local environment (such as extended X-ray absorption fine structure) or resonant spectroscopies (such as the Mössbauer effect and nuclear magnetic resonance) are useful for investigating the short-range order in certain amorphous materials. The details of these techniques, the relative strengths and weaknesses of various structural models, and the attempts to go beyond the radial distribution function were discussed by M. Gerl (Centre Nationale de la Recherche Scientifique, France) at the seminar.

The next issue concerns chemical composition. The methods for preparing amorphous solids make it essential that the composition and homogeneity of the final product be ascertained. While liquid-quenched alloys can be trusted to possess the composition of the liquid from which they were quenched, the vapor-deposited films often differ in composition from the starting material. This is due not only to differing sputtering rates or vapor pressures of materials, but also to the unavoidable incorporation of impurities and gases present during the deposition process. The two interfaces, film/substrate and film/air, are likely to have compositions and homogeneities that are unlike each other and different from the bulk. Various techniques are available for elucidating these features; Fig. 1, for example, shows the secondary-ion mass spectrum of an amorphous film obtained from sputtering a powder of composition iron boride (FeB).

Subtle complexities are also found in rapidly quenched metallic ribbons, where the surface in contact with the chill block may have somewhat different properties than the outside surface. Slight variations in quenching parameters may lead to ribbons that, though prepared in the same batch, possess different microstructures and therefore different macroscopic properties. A vivid example is presented in Fig. 2 for the rapidly quenched iron-aluminum-boron ($\text{Fe}_{60}\text{Al}_{28}\text{B}_{12}$) alloy. The section of the ribbon where an amorphous phase is embedded in crystalline FeAl behaves as a soft ferromagnet, while the part where Fe_2B has crystallized appears not to order magnetically. The ability to control the microstructure is actually leading to fruitful exploitation of rapidly quenched metals in areas traditionally left to conventional metallurgy.

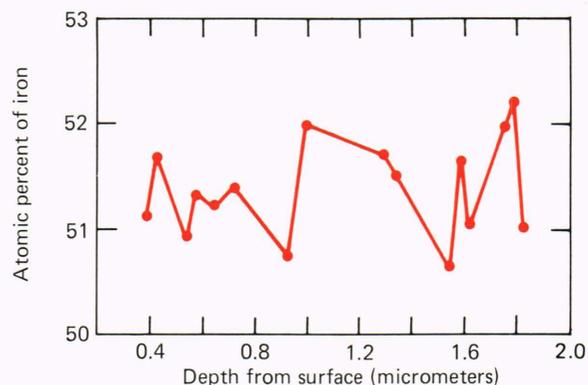


Figure 1—Variations with depth of iron concentration in a typical sputtered iron boride film; the average atomic percent of iron is 51.4 ± 0.4 (from Ref. 2).

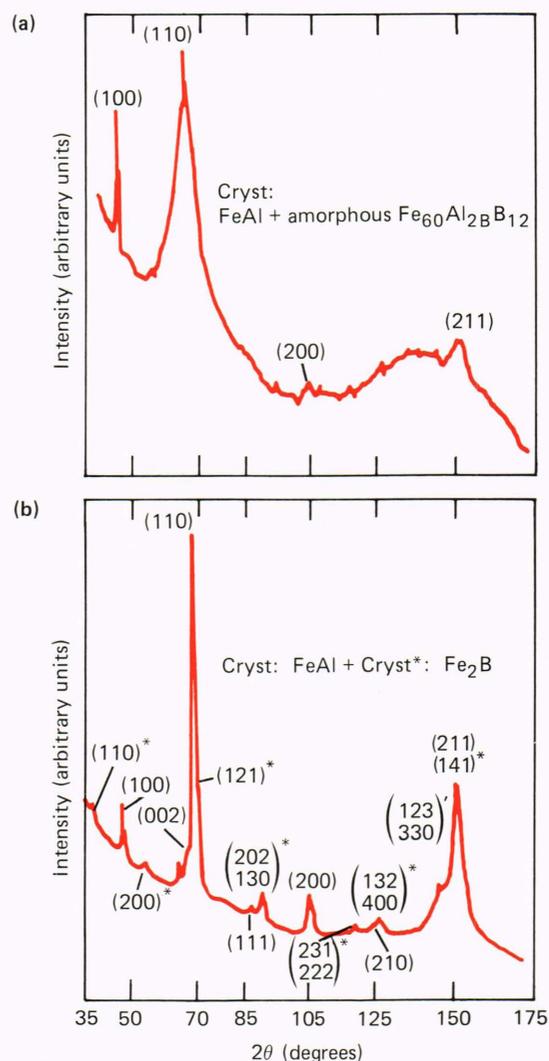


Figure 2—Room temperature X-ray scattering patterns from two sections of the liquid-quenched ribbon of $\text{Fe}_{60}\text{Al}_{28}\text{B}_{12}$ (from Ref. 3).

The homogeneity of the sample can be ascertained from depth profiling by secondary-ion mass spectroscopy as seen in Fig. 1. Also, variations in chemical

composition and structural inhomogeneities owing to defects and voids cause fluctuation in electron density, electrostatic potential, and density of nuclei that can be investigated by scattering of X rays, electrons, and neutrons, respectively. In particular, the spatial extent and magnitude of the inhomogeneities can be estimated from the intensity and angular dependence of the small-angle scattering.⁴ Results on amorphous metallic alloys have been interpreted as evidence for inhomogeneities that vary in size from 15 to 2000 angstroms.^{5,6} A summary of information obtained from small- and large-angle scattering of neutrons from amorphous iron boron ($\text{Fe}_{80}\text{B}_{20}$) alloy is illustrated in Fig. 3. The presence of a columnar structure caused by the shadowing of regions by materials already deposited has been found in small-angle scattering from many metallic and semiconducting amorphous films.⁷

The determination of microscopic structure and chemical composition and the degree of homogeneity of amorphous solids, though quite challenging, provide enormous possibilities for manipulating these features to obtain materials whose properties can be controllably fine-tuned over a broad range.

When I first became interested in amorphous semiconductors in 1962, the journal literature on the subject was almost exclusively from the Soviet Union.⁸ The major exception concerned the studies of amorphous selenium⁹ then being conducted in Great Britain and the United States, primarily for the potential use of the material in Xerography. This can be seen in the review paper, "Electrical Properties of Amorphous Semiconductors," published in 1964.¹⁰ The review also served as the basis of a proposal, submitted to the National Aeronautics and Space Administration, that was funded at a grand total of \$10,000 per year, making it the first and only United States government contract in the field of amorphous semiconductors at that time.¹¹ The paper was well received, judging from the copious requests for reprints; but neither that review nor the extended review by the Nobel Laureate N. F. Mott on "Electrons in Disordered Structures,"¹² published three years later, could be said to have sparked the wide interest that amorphous semiconductors were soon to attract. That was achieved by S. R. Ovshinsky's paper in 1968 on reversible switching in chalcogenide glasses¹³.

Of course, marking events chronologically gives history a linear appearance that is often, if not always, fallacious. The very paper that brought amorphous semiconductors to the attention of the condensed matter physics community also revealed earlier work that had been buried in the patent literature¹⁴ and later brought focus on earlier papers in journals (for example, Ref. 15), that have proved to be seminal to the present understanding of amorphous semiconductors. The contemporary history of science offers abundant examples of scientific progress being hastened and guided by technological developments.

At the seminar, S. R. Ovshinsky (Energy Conversion Devices), who has long had a commitment to dis-

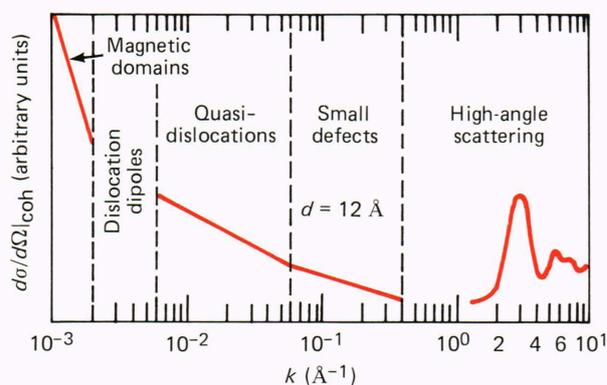


Figure 3—Scattering behavior observed in an amorphous iron boride alloy; the small-angle scattering reveals four subregions as indicated (from Ref. 6).

order, its science and its exploitation in technology, spoke on the "Relationship of Science and Technology of Amorphous Materials." A talk on the "Fundamentals of Technologically Important Amorphous Semiconductors" was delivered by D. Adler (Massachusetts Institute of Technology).

The seminal paper that I just referred to¹⁵ is the one by Nobel Laureate P. W. Anderson on localization. He considered an initially localized electron at 0 K placed in a crystalline lattice of potential wells with random depths. The mathematical analysis of the model shows that, for a sufficient amount of disorder, the solutions of the Schrödinger equation are localized in space. That is, the electron remains localized for all times provided the disorder in potential depths is large enough. This idea was later made relevant to electronic transport at finite temperatures by taking the time interval between displacements of the electron to be random,¹⁶ so as to reflect the randomness in potential energies. The motion of the electron is thus governed by a distribution of waiting times.

Another concept that is finding increasing applications in the subject of amorphous solids is that of fractional dimensionality or fractals,¹⁷ which are mathematical curves that delineate order within seeming chaos; they may prove fruitful in modeling the atomic structure of amorphous materials. Fractional dimensionality, d_f , can be defined from the relation, mass $\propto (\text{length})^{d_f}$, where integer values of d_f define the familiar Euclidean space. M. F. Shlesinger (Office of Naval Research), who has extended this concept to fractal time and combined it with the waiting time distribution approach to formulate a theory of charge transport in amorphous solids, described his research in a paper, "Fractal Time Transport and Relaxation in Amorphous Solids."

Numerous technological applications of amorphous semiconductors were detailed at the seminar. One particular application that was emphasized concerned amorphous silicon solar cell technology since it has made dramatic progress in the last decade, and still continues to offer solid opportunities for significant

improvements, and because, in the long run, it is likely to affect the well-being of a large fraction of humanity.

The possibility of doping amorphous silicon arose accidentally. The behavior of amorphous silicon obtained from the decomposition of silane, SiH_4 , in an RF glow discharge was remarkably different from that prepared by vacuum evaporation.¹⁸ In particular, the density of states in the band gap was over two orders of magnitude lower in the samples obtained by glow discharge. This was eventually traced to the presence in these samples of large amounts of hydrogen, which played the role, among others, of terminating the dangling bonds inherent to amorphous forms of tetrahedrally coordinated elements. The low density of gap states made it possible to chemically dope and therefore control the charge transport in hydrogenated amorphous silicon. Almost immediately, this led to the fabrication of the first photovoltaic cell based on an amorphous Si:H alloy.¹⁹

The present scientific and technological status, described in detail by S. K. Deb (Solar Energy Research Institute), who spoke on "SERI Program on Hydrogenated Amorphous Silicon Solar Cells," and M. Izu (Energy Conversion Devices), whose subject dealt with "Manufacture of Amorphous Silicon Alloy Solar Cells by a Continuous Process," is indeed a happy one. Some of the highlights include over 10 percent efficient photovoltaic cells in single- or multi-junction stacked configuration, and achievement of multiple-gap stacked solar cells where the top one can absorb photons of energy corresponding to its band gap, allowing the low-energy photons to be absorbed by the lower cells in the stacked configuration. Efficiencies of 25 to 30 percent are realistic possibilities. The problem of photoinduced instability (Staebler-Wronski effect) no longer poses a serious threat to the cell lifetime; technologically, it is exciting that stable, high-efficiency, multiple-gap, multiple-function solar cells based on amorphous semiconducting alloys deposited on inexpensive stainless steel substrates are being produced at low cost on a large scale by a continuous roll-to-roll process developed at Energy Conversion Devices. Alternative methods were covered by V. Dalal (SPIRE Corp.) in his talk, "Equipment for Fabrication of Amorphous Silicon."

Amorphous metals, like amorphous semiconductors, are of a relatively recent vintage in contrast to amorphous insulators, which have been known since antiquity. The former have had a history that is not very different from that of amorphous semiconductors. Amorphous metals obtained by melt quenching appeared in 1960,²⁰ but long before that, information on the process of melt quenching was buried in the patent literature and not accessed by the scientific community at large. (It is interesting to note that nature has always known about rapid solidification because fulgurites, formed where lightning strikes soil or rock, represent examples of rapidly formed glasses.) The history and present status of melt quenching were described by its brilliant inventor, R. B. Pond (The Johns

Hopkins University), whose talk on "Rapid Solidification: History and Promise" was accompanied by a demonstration of the melt-quenching technique.

Another rapidly growing area of noncrystalline solids discussed in detail at the seminar concerned magnetic glasses.²¹ Magnetism, like many other properties of solids, has no need for the periodic arrangement of atoms on a crystalline lattice. For a material to be magnetic, all that is necessary is the existence of a magnetic moment (and therefore uncompensated spin) on an atom and the requirement that when these atoms are assembled in a solid, they interact via the exchange forces. However, it was only in 1960 that A. I. Gubanov²² foresaw the existence of ferromagnetism in an amorphous solid. But in repetition of the now-familiar history of scientific progress, his paper went largely unnoticed, and it was nearly a decade later that the technological possibilities of certain amorphous ferromagnets excited interest in the subject. In the process, new scientific concepts have emerged.

A particularly useful concept is that of frustration.²³ In magnetic solids, frustration arises from the conflict caused by the simultaneous presence and ensuing competition between ferromagnetic and antiferromagnetic exchange interactions. It has been fruitfully applied to the study of the ground state and low-lying excited states of random magnetic structures, termed spin glasses.

Consider Ising spins, $S_i = \pm 1$ at corners of a square interacting via nearest neighbor exchange interaction, J_{ij} . For $J_{ij} > 0$, one obtains the ferromagnetic configuration of spins and for $J_{ij} < 0$, the antiferromagnetic configuration results. Both these configurations have the ground state energy $-4J(J_{ij} = J)$. The ferromagnetic configuration is nondegenerate, while the antiferromagnetic configuration is doubly degenerate corresponding to two directions of the starting spin. The ground state energy remains unchanged if two of the bonds are ferromagnetic and two are antiferromagnetic. In all these cases, the directions of the spins can be easily chosen to satisfy the constraints imposed by the signs of the bonds, and the overall energy is minimized. However, the situation changes drastically if an odd number of bonds is antiferromagnetic. One of the bonds then remains necessarily unsatisfied; the ground state energy is raised to $-2J$, and the degeneracy increases to 8, corresponding to two ways of choosing the direction of the initial spin and four ways of placing the unsatisfied bond. This simple illustration of frustration demonstrates how competing exchange interactions raise the ground state energy and degeneracy. Equivalent results are obtained even in the absence of competing interactions provided one has structures that impose constraints leading to misfitting bonds. The simplest example is the two-dimensional triangular lattice with antiferromagnetically interacting Ising spins, a problem studied long before the subject of spin glasses became important.²⁴ Its kinship to spin glasses lies in the observation that a disordered arrangement of atoms in an amorphous solid can lead to constraints that do not

permit an antiferromagnetic structure and consequently lead to misfitted bonds.

The concept of frustration has proved to be a rich one. The importance of local symmetries in the model²³ and their relationship to lattice gauge field theories²⁵⁻²⁷ previously introduced in connection with the quark confinement model^{28,29} have been investigated. It is also proving useful in phenomena as varied as the roughening transition in crystal growth theory,³⁰ orientation of complex molecules in solids,³¹ incommensurate structures,³² and content-addressable memory in neural networks.³³

The large degeneracy or near-degeneracy in experimental examples of frustrated structures has led to the suggestion that the spin glass state is inherently nonergodic.³⁴ Many roughly equivalent, free-energy minima with significant barriers between them exist so that some of the minima are inaccessible during the approach to equilibrium. The system can therefore get locked into a state of "local" equilibrium in which the spin configurations and the consequent internal fields are quite different from those in the "true" equilibrium state.

Evidently, old order parameters, such as magnetization or sublattice magnetization, are of little use in a description of a glass phase characterized by multiple energy states separated by barriers. To remedy this lacuna, several different order parameters appropriate to the spin glass state have been suggested, the most general one being related to the probability distribution of the overlap of magnetizations between different states.³⁵

Theoretically, the spin glass state is an idealized magnetic structure where all directions are equally probable so the structure is truly random; in practice, this is rarely so. The competition between ferromagnetic and antiferromagnetic exchange interactions is never exact, so net correlations of either variety can dominate. Moreover, for certain amorphous alloys, random anisotropy plays a dominant role in determining the resulting magnetic structure. Consequently, an entirely new lexicon has emerged to describe a variety of noncollinear magnetic structures; it was described in detail by its talented originator, J. M. D. Coey (Trinity College, Dublin) in his talk at the seminar on "Fundamentals of Technologically Important Amorphous Magnetic Materials."

Although noncollinear magnetic structures and their dynamics present exciting developments in the science of amorphous magnets, they represent only a part of a larger spectrum. Conventional ferromagnetism, seen in a great variety of amorphous magnets, is actually what is exploited in many technological applications. The advantage over crystalline ferromagnets lies in the ease and economy of production combined with the possibility of fine-tuning desired properties by chemical tailoring. The soft magnetic properties of amorphous ferromagnets, arising from the absence of magnetocrystalline anisotropy, have been exploited successfully in distribution transformers that are being operated by various utility companies in the United

States and Japan. Other varied applications range from amorphous metal cores, with a decreased signal distortion and lower noise than the Permalloy® core used in a phonograph cartridge, to amorphous heads for computer recording equipment and hard magnetic materials for permanent magnets. These and many other potential commercial applications of amorphous magnets were described in a talk on "Technology of Amorphous Magnetic Materials" by F. E. Luborsky (General Electric Corporate Research and Development Center), who has had first-hand experience of the technology of amorphous magnets at General Electric. The theory of magnetoelastic effects in amorphous ferromagnetic materials and their uses in strain gauges and accelerometers, their nonlinear moduli and torsion-sensing applications, were covered by H. T. Savage (Naval Surface Weapons Center), who has been at the forefront of the research on this subject, in a talk entitled, "Theory and Applications of Magnetoelastic Effects in Amorphous Magnets."

In spite of the broad scope of the seminar, the allotted time was not sufficient to cover many fascinating aspects of the study of disordered materials. I will note that many hopeful avenues still need to be explored. An example lies within the framework of broken ergodicity and finite time statistical mechanics and hierarchically constrained dynamics recently proposed in condensed matter physics and specifically explored for glasses.³⁶ Experimental investigations of amorphous superlayers have just begun and offer exciting opportunities in the fabrication of man-made materials.³⁷

The study of disorder has also refined the concept of order, order that is not confined to spatial configuration but includes time and has far-reaching ramifications in many areas, including living matter. We are familiar with the idea that coherence can result from collectivity; it is becoming evident that so can chaos. But then, chaos is just one harmony too many.

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