

Amorphous magnetic materials

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Fundamental research on the physics of amorphous magnetic materials is reviewed. The atomic and magnetic structure and the magnetic properties are discussed. Theoretical results on the nature of the magnetic ordering and the spectrum of elementary excitations are reported. Applications are discussed.

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1. INTRODUCTION

Rapid progress in research on magnetically ordered crystals has greatly strengthened and extended our understanding of the nature of the condensed state in general and of magnetic ordering in particular. This progress has also resulted in the widespread technological use of magnetic materials. In contrast, there has been comparatively little study of magnetic ordering in noncrystalline media (amorphous media, polymers, and liquids). Interest in these media has increased sharply in recent years, however, to the point where we can speak of the appearance of the new field of amorphous magnetism. Work on the problem of amorphous magnetism began back in 1960 with Gubanov's original paper¹ regarding the possible existence of amorphous and liquid ferromagnets. Since then, several international and national conferences have been devoted to amorphous magnetism,²⁻⁵ and this field has also assumed an important place in recent years in the traditional conferences on magnetism.⁶

The reason for the high level of interest in this problem apparently lies in the very logic of fundamental and applied research in solid state physics. Contemporary solid state physics is primarily a field of the physics of crystals, i. e., of systems in which the atoms are

arranged periodically in a crystal lattice. The translational invariance of such systems has made it possible to develop an elegant theory. It is thus a natural tendency for researchers to take up the physics of more complicated systems, such as amorphous media or polymers. On the basis of general considerations we may expect that the distinctive features of amorphous systems, such as the absence of long-range order and the lack of equilibrium, will lead to important, nontrivial consequences. The task of the theoreticians is to analyze these consequences and to predict the properties of such materials.

The outlook for applications depends on how well the amorphous materials lend themselves to technological processing and on whether qualitatively new materials can be developed. It is already clear from experience with semiconductors,⁷ magnetic materials,⁸ and laser materials that the use of glassy materials not only reduces the cost of the devices but also allows a greater range of compositions and thus a greater range of properties. The amorphous metal-metalloid alloys illustrate the progress which has already been achieved in the field of magnetic materials. Table I shows certain properties of these materials, along with the properties of several traditional crystalline materials.⁸

TABLE I. Comparison of the magnetic properties of certain crystalline and amorphous alloys (300 K).

	Composition	$4\pi M$, kG	T_C , °C	H_C , Oe	$10^{-8} \Omega \cdot \text{cm}$	$\lambda_S \cdot 10^6$
Commercial crystalline alloys	80Ni16Fe4Mo	7.8	460	0.025	55	~ 0
	80Ni20Fe	8.2	400	0.005	65	~ 0
	50Ni50Fe	16.0	480	0.10	45	40
	96,8Fe3,2Si	20.3	730	0.5	50	4
Amorphous alloys	$\text{Fe}_{80}\text{CO}_{12}\text{P}_{14}\text{B}_4\text{Al}_3$	6.3	260	0.015	140	~ 0
	$\text{Fe}_{80}\text{P}_{14}\text{B}_4$	13.6	344	0.10	—	26
	$\text{Fe}_{80}\text{P}_{12}\text{C}_2\text{B}_1$	14.9	292	0.05	150	30
	$\text{Fe}_{80}\text{B}_{20}$	16.0	374	0.04	140	30

These amorphous alloys can be produced in the form of tapes with a thickness of about 0.05 mm and a width up to several centimeters by forcing a stream of the molten alloy through an orifice onto the lateral surface of a spinning drum.⁹ It can be seen from Table I that, in terms of magnetic properties, the amorphous alloys already available are nearly or actually the equal of the better crystalline materials—materials which require complicated and expensive operations such as rolling and special annealing. Interestingly, amorphous alloys also exhibit extremely low magnetic and acoustic losses and an unprecedented corrosion resistance.¹⁰

Another important class of amorphous materials is found in the alloys of rare earths with 3d metals. These materials have been used to produce amorphous magnetic films with magnetic bubbles¹¹ and high-coercivity materials.¹²

Substantial progress has already been achieved in research on the fundamental physics of amorphous magnetic materials. Some of these results have been covered in several recent reviews,¹³⁻¹⁹ but the pace of research on amorphous magnetism is so great that many fundamentally new results have been obtained in the past two or three years. Our intention in this review is to summarize the major results on most aspects of the problem which are of fundamental interest. We will also discuss the outlook for technological applications.

2. ATOMIC STRUCTURE OF AN AMORPHOUS MAGNETIC MATERIAL

a) Definition of an amorphous solid

From the standpoint of atomic structure, an amorphous material is analogous to a liquid, having the following main features: macroscopic isotropy, a short-range order in the arrangement of atoms, but no long-range order. In real amorphous materials the macroscopic isotropy may be imperfect because of various external, primarily technological, factors. The resulting macroscopic anisotropy is not an internal property of an amorphous material. By "short-range order" here we mean the atomic order which is seen over a few interatomic distances. The simplest case of short-range order is that in which certain values of the interatomic distance r are more common than others. "Long-range order," in contrast, means a periodicity in the arrangement of atoms. Since liquids also exhibit short-range order without long-range

order,²⁰ some characteristic is needed to make a definite distinction between amorphous materials and liquids. This characteristic is the deviation of an amorphous material from thermodynamic equilibrium.

A melt and the corresponding crystalline phase which results from cooling are both equilibrium states; i.e., their free energy F is at a minimum for the given conditions. An amorphous material is in a nonequilibrium state with a higher value of F . Although not an equilibrium state, this state may persist for a certain time (the "stability time") in a certain temperature interval (the "stability interval"). It is thus clear that a fundamental problem here is that of the stability of an amorphous material, and this problem includes the questions of crystallization and structural relaxation.^{18,19}

At the melting point, T_m , the enthalpy H of a crystal increases discontinuously by an amount ΔQ , which is the latent heat of fusion (Fig. 1). This heat is expended on disrupting the long-range crystalline order (ΔQ_1) and on changing the short-range order (ΔQ_2) in accordance with the structure of a melt at the temperature T_m . The components ΔQ_1 and ΔQ_2 are usually comparable in magnitude.²¹ When a melt is cooled rapidly however, without crystallization, there is only a continuous change in the short-range order, accompanied by the evolution of the heat ΔQ_2 . There is a certain temperature T_g (the "glass transition temperature") at which the viscosity becomes able to prevent rearrangement of the atoms to lower-energy configurations. Cooling below this temperature simply reduces the thermal vibrations (as in a crystal). For this reason, curves 1 and 2 in Fig. 1 run parallel to each other at temperatures below T_g . The crystallization of an amorphous material must therefore be accompanied by the evolution of an amount of heat of the order of $\Delta Q/2$. A study of the thermodynamic characteristics (calorimetry, for example) thus furnishes important fundamental information on the configurational entropy, structural-relaxation effects, etc.^{18,19}

b) Structure of an amorphous material

Because of the macroscopic isotropy of an amorphous material, its atomic structure can be characterized as a function of a single variable, which is the radial distribution function (RDF) of the atomic density, $\rho(r)$. This is the atomic density expressed as a function of the distance r from some arbitrary atom. The RDF can be found experimentally from the diffraction of x rays, neutrons, or electrons with a subsequent Fourier transformation of the interference function $S(Q)$, where $Q = 4\pi(\sin \theta)/\lambda$, λ is the wavelength of the radiation, and θ is the scattering angle. Therefore,

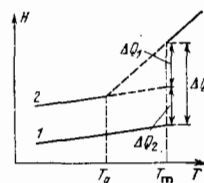


FIG. 1. Temperature dependence of the enthalpy 1—in the crystalline material; 2—in the amorphous material.

$$\text{RDF} = 4\pi r^2 \rho(r) = 4\pi r^2 \rho_0 + \frac{2r}{\pi} \int_0^\infty Q [S(Q) - 1] \sin(Qr) dQ. \quad (2.1)$$

Here ρ_0 is the average atomic density of the material. An analogous structure function can be introduced in the more complicated case of a structure with atoms of different species.²²

Figure 2 shows some typical radial distribution functions for certain amorphous ferromagnetic alloys.²³ The curve of $4\pi r^2 \rho(r)$ oscillates around the parabola $4\pi r^2 \rho_0$, which corresponds to a disordered arrangement of atoms. The positions of the peaks and the areas under the peaks are measures of the average radius and the average number of atoms of the given coordination sphere, respectively. The second peak is split in two. For a liquid metal (say, cobalt) the second peak in the radial distribution function is not split. The typical value of the first coordination number for amorphous metallic ferromagnets is $Z_1 \sim 12-13$. Study of the radial distribution functions of amorphous materials has shown that positional correlations of the atoms are distinguishable only at distances r up to ~ 15 Å.

The radial distribution function gives only the positional dependence of the average atomic density; it does not describe the details of the atomic structure, e.g., the topology of the short-range order. More-detailed information on the submicroscopically inhomogeneous structure can be obtained by small-angle x-ray scattering,^{13,23} for example.

c) Modeling of amorphous structures

Two basic models²² are used to describe amorphous structures: the microcrystalline model and the topologically disordered model. In a microcrystalline model it is assumed that most of the atoms lie in small ordered regions (~ 15 Å), so that the structural disorder results from the fact that these regions are oriented at random. The microscopic regions do not, however, have surfaces, and this circumstance greatly distinguishes an amorphous structure from a fine-grain polycrystalline system with a grain size of the order of 50–150 Å. Figure 3a is a schematic representation of

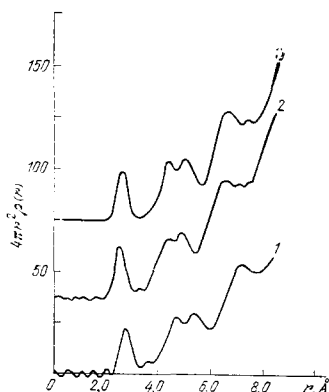


FIG. 2. Radial distribution function $4\pi r^2 \rho(r)$ for certain amorphous alloys. 1— $\text{Pd}_{80}\text{Si}_{20}$; 2— $\text{Ni}_{81.4}\text{P}_{18.6}$; 3— $\text{Fe}_{80}\text{P}_{18}\text{C}_2$.

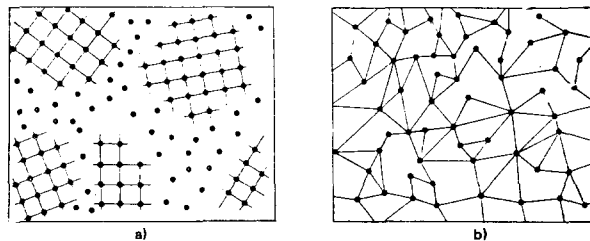


FIG. 3. Schematic diagrams of models for an amorphous structure. a—Microcrystalline model; b—topologically disordered model.

an amorphous material consisting of disoriented regions of this type. A detailed study of microcrystalline models yields results which disagree with experiment.

In the topologically disordered model, the atoms are packed in a continuous liquid-like fashion without any abrupt structural discontinuities (Fig. 3b). The earliest model of this type to be proposed was that of Zachariasen,²⁴ and this model has proved successful in describing the structures of amorphous silicon and germanium. Detailed structural studies by Cargill¹³ have shown that a satisfactory model for (3d-metal)-metalloid amorphous ferromagnetic alloys is the model of a close random packing of hard spheres, which is known as the Bernal model.²⁵ In this model the structures are formed by hard spheres which are closely packed in the sense that they leave no voids big enough for another sphere. The packing is random in the sense that there are only slight correlations between spheres separated by five or more diameters. This model can be used to calculate the radial distribution function and the packing factor, $\eta = 4\pi(\sigma/2)^3(\rho_0/3)$, where σ is the diameter of a hard sphere. Here, of course, it is necessary to know the atomic radii. Cargill's analysis¹³ shows that the best radii for metal-metalloid amorphous alloys are the Goldschmidt radius of the metal in a twelfold coordination and the tetrahedral covalent radius for the metalloid. In the Bernal model the small spheres represent the metalloid atoms in this case; the metalloid atoms occupy the largest voids in the random close packing of the larger metal spheres. The number of such voids is about 20% of the total number of atoms; thus we have the typical amorphous-alloy composition $\text{Fe}_{80}\text{B}_{20}$. The calculations yield a qualitatively correct expression for the radial distribution function,¹³ a packing factor $\eta \approx 0.7$, and a density which agrees within $\pm 5\%$ with the experimental density.¹⁸ So far, there has been no analytic formulation of the Bernal model, and all the specific results have been found numerically (by the Monte Carlo method, for example). A nucleation cluster is adopted and then used to generate statistically a large cluster of about 10^3 atoms. Since a real system contains 10^{22} atoms, the calculations cannot be completed without adopting some simplifying approach of the cluster-replication type, and the adoption of this approach is one of the most important approximations of the model.

d) Methods for synthesizing amorphous materials

The basic characteristics of the methods used to syn-

thesize thermodynamically nonequilibrium amorphous materials stem from the need to avoid the thermodynamically preferred crystallization. The necessary conditions are analyzed in reviews by Jones²⁶ and Luborsky,¹⁸ for example. Here we will simply discuss some of the methods used to synthesize amorphous magnetic materials.

Several of the methods make use of rapid cooling of a melt. The cooling rate must be such that the temperature interval between the melting point T_m and the glass transition temperature T_g is traversed in a time too short to allow crystallization. Clearly, then, we would like to raise the reduced glass transition temperature $\tau_g = T_g/T_m$ and to reduce the dimensions of the object. A simple analysis shows²⁶ that for $\tau_g \geq 2/3$ the glassy state is easily reached even by comparatively slow cooling. A decrease in τ_g makes it necessary to raise the cooling rate. For metals the typical values of τ_g are in the range 0.45–0.6, and a cooling rate $\sim 10^8$ deg/s is required to form a glass. The value of τ_g is increased by admixtures of glass-forming elements. For alloys of 3d metals with metalloids, for example, a composition of the type M_4M_0 is used (M is the 3d metal and M_0 is the metalloid). The reason for the increase in τ_g in this case is the pronounced lowering of the melting point T_m .

A technologically important method embodying the principle of fast cooling of a melt is to force a stream of the molten material through an orifice onto the lateral surface of a spinning drum⁹ (Fig. 4). If the characteristics of the system are chosen correctly,²⁷ the melt can be cooled at a rate $\sim 10^5$ deg/s.

There are also other methods for synthesizing amorphous materials which make use of fast cooling of a melt: the shock-wave method,^{28,29} the anvil method,^{30,32} the centrifuge method,³³ the catapult method,³⁴ and the method of continuous expulsion of melt.^{35,36}

Other technologically important methods make use of the condensation of a vapor on a cooled substrate,^{37,38} electrodeposition and chemical deposition,^{39,40} and deposition by sputtering.⁴¹⁻⁴³

3. MAGNETIC STRUCTURE OF AMORPHOUS MAGNETIC MATERIALS

The most important question in research on amorphous magnetic materials is that of their magnetic structure and the role played by the amorphous structure in the estab-

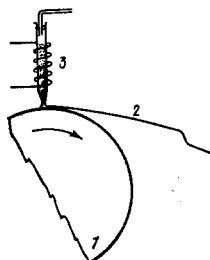


FIG. 4. Device for fabricating amorphous tapes. 1—Rotating metal disk; 2—amorphous tape; 3—quartz cell holding the melt.

lishment of magnetic order. There is no longer any doubt that magnetic order does exist in an amorphous material: This point has been demonstrated by many experimental and theoretical studies. However, the effort to construct a general classification of amorphous magnetic materials on the basis of their magnetic structures is still in a beginning stage.^{18,44}

a) Magnetic structures

Andreev⁴⁴ has reported a phenomenological analysis of the types of magnetic order which are possible in principle in spatially disordered materials. Working from symmetry considerations and the assumption that exchange forces are dominant in the establishment of magnetic order, he showed that a macroscopically isotropic amorphous structure is consistent with only four types of magnetic structures: an ordered ferromagnet, a disordered ferromagnet, a disordered antiferromagnet with three mutually perpendicular antiferromagnetic moments, and a spin glass. From the microscopic standpoint, three of these structures can be described clearly and classified¹⁸ as in Fig. 5. Also shown in Fig. 5 are some structures which have been observed for amorphous magnetic materials. We might point out that there has been no reliable experimental observation of amorphous magnetic materials with an antiferromagnetic long-range order. For example, although the susceptibility⁴⁵ of amorphous Fe_2O_3 exhibits features characteristic of an antiferromagnet the temperature dependence of the hyperfine field is not what would be expected.

Coe¹⁶ has carried out a qualitative microscopic analysis of magnetic structures and has shown that the magnetic order in an amorphous magnetic material is determined by the distribution of the exchange interactions $J(r_{ij})$, the local single-ion anisotropy D_i , and the ratio of these quantities. The simplest model Hamiltonian is

$$\mathcal{H} = - \sum_{i,j} J(r_{ij}) S_i S_j - \sum_i D_i S_i^z. \quad (3.1)$$

When the exchange interactions $J(r_{ij})$ are primarily positive and $|J| \gg |D|$, a ferromagnetic state arises in the system. Such ferromagnets have been observed in both ionic materials⁴⁶ (FeF_2) and metallic materials. In the case $|J| \leq |D|$, a disordered-ferromagnetic state arises in a system with $D > 0$ and $J > 0$, according to theoretical and experimental studies.^{47,48} This behavior is exhibited, for example, by the amorphous alloys

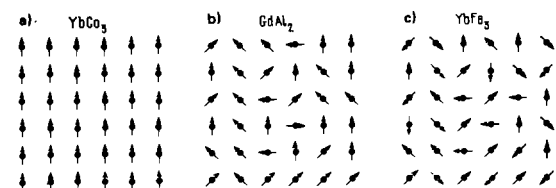


FIG. 5. Magnetic structures of amorphous magnetic materials. a—Ferromagnet ($S_i \neq 0$, $S_i S_j = S_i^2$, $\langle S_i S_j \rangle = S_i^2$); b—disordered ferromagnet ($S_i \neq 0$, $-S_i^2 < S_i S_j \leq S_i^2$, $0 < \langle S_i S_j \rangle < S_i^2$); c—spin glass ($S_i \neq 0$, $-S_i^2 \leq S_i S_j \leq S_i^2$, $\langle S_i S_j \rangle = 0$).

DyNi₃ (Ref. 48), YbFe₃ (Ref. 48), and TbAg (Ref. 49). Because of the strong effect of the pronounced single-ion anisotropy, magnetic fields $\sim 10^6$ Oe are required for the magnetization of such systems.¹⁶

A case more difficult to analyze is that in which the exchange interactions $J(r_{ij})$ of the nearest neighbors are primarily negative. In such a case, since the amorphous structure is not an alternating structure, a situation with an antiferromagnetic long-range order cannot be conceived.^{50,51} Analysis¹⁶ of experimental studies shows that magnetic states of the disordered-antiferromagnet or spin-glass type occur in such systems, in agreement with theoretical considerations. Typical structures of this type are amorphous oxide compounds⁵² and polymers.⁵³⁻⁵⁵

More complicated magnetic structures arise in amorphous magnetic materials which contain several different species of magnetic atoms. Figure 6 shows microscopic pictures of two such structures, with atoms of two species, A and B, for cases with $|J_{BB}| > |J_{AB}| \sim |D_A|$. Such structures arise, for example, in the compounds¹⁶ DyCo₃ and NdCo₃.

The magnetic structure of an amorphous magnetic material is thus determined by the distribution of exchange interactions $J(r_{ij})$ and the anisotropy. We should emphasize in particular that the ordered ferromagnetic state is unstable in the presence of a random anisotropy.^{12,56}

b) Effect of the amorphous structure on the magnetic state of a material

The most important consequence of the switch to an amorphous structure is the disappearance of the periodic crystal structure. Because of this circumstance, a theoretical description of amorphous magnetic materials runs into serious fundamental difficulties. Since there is no translational invariance in an amorphous material, the traditional methods of solid state theory, worked out for ideal crystals, cannot be applied.

The chemical and structural atomic disorder of amorphous materials leads to fluctuations in all the characteristics of the magnetic subsystems. The magnetic state is affected most strongly by fluctuations of the exchange interactions, of the magnetic moment, and of the single-ion anisotropy. Fluctuations of the exchange interactions result from the strong dependence of the

exchange integrals on the distance between the interacting atoms and on the configuration of the bonds of these atoms. Fluctuations of the anisotropy result from fluctuations of the crystal fields. The amorphous nature of a structure affects the magnetic moments most strongly in the case of 3d metals, because of the pronounced delocalization of their unpaired electrons. In this case the local magnetic moments are determined not only by the positions but also by the number and type of nearest neighbors. The fluctuations of the local magnetic moments are less important in rare earth compounds (because of the strong screening of the 4f electrons) and in ionic insulators, in which the magnetic moments are not changed substantially by the covalent admixture of ligand wave functions. The distribution of magnetic moments in magnetic materials can be characterized by the hyperfine fields, which can be measured by NMR or Mossbauer methods. Figure 7 shows some results from studies of this type for the three groups of magnetic materials listed above.¹⁸

Although we lack a systematic theory of magnetic order in a real amorphous structure, there are several papers in which the role played by fluctuations in the characteristics of an amorphous medium has been analyzed on the basis of simple models. To develop an approximate description of the properties of an amorphous material, for example, we could begin by assuming that an average translational invariance is established after an averaging over all possible cases. The material can then be described as an ideal crystal with certain effective properties. The procedure for taking the average over all cases should in general incorporate fluctuations in the positions of the atoms and in the characteristics of the interaction in the system. The procedure should also take the fluctuations of thermodynamic quantities into account in a self-consistent manner. Because of the exceptional difficulty of this problem, the most common approach has been to adopt a simpler approximation: that the most important effect of the disorder of the atomic structure on the magnetic subsystem can be taken into account by a "stochastic lattice model." In this approach it is assumed that the spins are localized in a regular crystal lattice, while the exchange integrals are stochastic quantities.^{57,58} This model naturally ignores the topological disorder.⁵⁹

The simplest method for analyzing the properties of such a model is to adopt the molecular-field approximation. Intense studies have been carried out in this

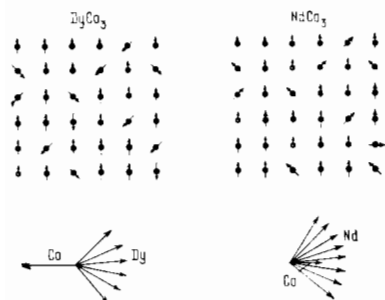


FIG. 6. Magnetic structures of amorphous alloys of a rare earth with a 3d metal.

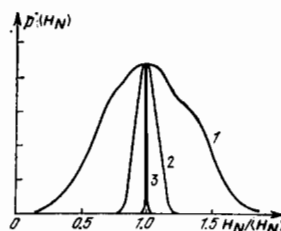


FIG. 7. Distribution of hyperfine magnetic fields $p(H_N)$ in several amorphous magnetic materials. 1—Co₈₁P₁₉; 2—Fe(OH)₃nH₂O; 3—DyNi₃.

approximation by Handrich,^{60,61,63} for example. The initial Hamiltonian in the Heisenberg model can be written in the form

$$\mathcal{H} = - \sum_{i,j} J_{ij} S_i S_j - \mu_B g H \sum_i S_{iz}, \quad (3.2)$$

where S_i and S_{iz} are the operators representing the i th spin and its z component, respectively; J_{ij} are fluctuating exchange integrals; g is the Landé factor; μ_B is the Bohr magneton; and H is the external field, which is directed along the z axis. It is assumed in (3.2) that J_{ij} depends only on the distance between adjacent spins. Even this assumption imposes serious limitations since J_{ij} actually depends in addition on the topology of the surroundings.

In the molecular-field approximation the average value of the z component of the spin at lattice site i is described by the Brillouin function B_S :

$$\bar{S}_{iz} = S B_S \left[\frac{S}{kT} \left(\mu_B g H + \sum_j J_{ij} \bar{S}_{jz} \right) \right], \quad (3.3)$$

where it is assumed for simplicity that $S_i = S$. Expanding the Brillouin function in a series in the fluctuational exchange, we find the normalized magnetization to be

$$\sigma = \frac{1}{2} [B_S((1+\Delta)x) + B_S((1-\Delta)x)], \quad (3.4)$$

where

$$x = 3S(S+1) \frac{T_c \sigma}{T}, \quad \sigma = \frac{\langle \bar{S}_z \rangle}{S}, \quad \Delta^2 = \frac{\langle \Delta J_{ij}^2 \rangle}{\langle J_{ij} \rangle^2};$$

and the angle brackets denote a configurational average. The fluctuations of the exchange increase the rate at which the magnetization decreases with increasing temperature, as can be seen from (3.4). Figure 8 shows the temperature dependence¹⁴ of the magnetization found from (3.4) for the two cases $\Delta = 0$ and $\Delta = 0.5$, along with experimental results for the amorphous alloy $\text{Co}_{70}\text{B}_{20}\text{P}_{10}$. For this alloy, $\Delta = \sqrt{\langle \Delta J_{ij}^2 \rangle} / \langle J_{ij} \rangle \sim 0.3$. When the fluctuations Δ reach a certain critical level the ferromagnetism disappears altogether.⁶⁰

Handrich⁶¹ has carried out a general analysis of the effect of fluctuations on the magnetic state of an amorphous ferromagnetic, working from the Bogolyubov theorem⁶⁴ for the free energy F . For the Hamiltonian (3.2), this theorem can be written

$$F = F(\mathcal{H}) \leq F(\mathcal{H}_0) \equiv F_0, \quad \Delta F = F - F_0 \leq 0, \quad (3.5)$$

where \mathcal{H}_0 describes the corresponding pseudocrystal with $J_{ij} = \langle J_{ij} \rangle$. The Bogolyubov theorem thus takes the following form: The structural fluctuations incor-

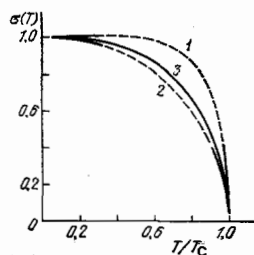


FIG. 8. Temperature dependence of the magnetization. 1—Theory for $S = 1/2$, $\Delta = 0$; 2—theory for $S = 1/2$, $\Delta = 0.5$; 3—experiment for $\text{Co}_{70}\text{B}_{20}\text{P}_{10}$.

porated in the Hamiltonian (3.2) either reduce the free energy of the magnetic system or do not affect it. We should emphasize that we are discussing here the fluctuational lowering of the free energy, reckoned from some ideal system (the pseudocrystal) with an averaged exchange parameter.

Analysis of the consequences of the Bogolyubov theorem leads to the conclusion that, first, the structural fluctuations increase the state density of the magnetic system at low and high energies. We thus again conclude that there is a faster lowering of the magnetization at low temperatures. Second, in amorphous ferromagnets there is a sharp magnetic phase transition; structural fluctuations lead to a reduction of the Curie temperature T_C in comparison with T_C^0 , that of the pseudocrystal. This statement does not mean, of course, that in all cases in which a real crystal becomes amorphous the temperature of the magnetic phase transition will decrease.⁶⁵

Experimental study of amorphous ferromagnets has basically confirmed these conclusions regarding the effect of fluctuations on the magnetic state.^{4,14,19} At the same time, it has been shown in several cases that the switch to an amorphous structure in magnetically ordered crystals may be accompanied by a change in the type of magnetic order and by an increase in the temperature of the magnetic phase transition. Table II summarizes these results. We emphasize that these effects have been established for both ionic compounds and metals. The possibility of a change in the type of magnetic order and of an increase in the Curie temperature T_C upon switching to an amorphous structure has been studied theoretically by Kuz'min and Petrakovskii^{65,70} for nonconducting magnetic materials and by Kaneyoshi⁷¹ for metals.

Kuz'min and Petrakovskii^{65,70} have studied switching to an amorphous structure of magnetic dielectric crystals, whose magnetic structure is strongly affected by the geometry of the exchange-bond distribution. This is the situation, for example, in quasi-few-dimensional magnetic materials.⁷² At least two different exchange parameters must be introduced in order to describe magnetic materials of this type. The type of magnetic order and the temperature of the magnetic phase transition in quasi-few-dimensional magnetic materials are determined by the weak exchange which binds the magnetic chains or layers.⁷³ These characteristics of

TABLE II. Comparison of the characteristics of crystalline (Cr) and amorphous (Am) magnetic materials.

Composition	Type of magnetic order		Temperature of magnetic phase transition		Reference
	Cr	Am	Cr	Am	
FeF_2	AFM	FM	78	21	46
FeCl_2	AFM	FM	24	21	46
FeBr_2	AFM	?	41	21	46
$\text{Bi}_2\text{Fe}_4\text{O}_9$	AFM	FM	265	600	66
GdAl_2	AFM	FM	138	122	67
GdAl_2	FM	Spin glass	170	15.8	68
GdCu_2	AFM	FM	41	75	68
MnPt	AFM	FM	—	—	69

the amorphous material, on the other hand, are determined primarily by some average exchange. Accordingly, switching of quasi-few-dimensional systems to an amorphous structure can lead to a change in the type of magnetic order as well as to a substantial increase in the temperature of the magnetic phase transition.

The magnetic state of nonconducting magnetic materials can be described by the Hamiltonian

$$\mathcal{H} = - \sum_{j,m} A_{j,m} S_j S_m. \quad (3.6)$$

where $A_{j,m}$ are exchange parameters which are nonzero only for nearest neighbors. In an ideal crystal we would have $A_{j,m} \rightarrow A_0(f-m)$, while in the general case of an amorphous magnetic material the $A_{j,m}$ fluctuate in magnitude and sign (the lattice model). The ideal crystal may be characterized by an anisotropic distribution of exchange bonds, e.g., $A_0(h_1) = J$, $A_0(h_2) = K$, where $\lambda = K/J$ is a measure of this anisotropy. In quasi-few-dimensional magnetic materials we have $|\lambda| \ll 1$, and the magnetic-ordering temperature is $T = T_C(|\lambda|)$, where $T_C(0) = 0$. It is natural to assume that the properties become isotropic upon switching to an amorphous structure and that, on the average, the short-range order¹³ characteristic of the original crystal is preserved. If the fluctuating exchange parameters are of the same sign, the ground state is evidently ferromagnetic in the case $A_{j,m} > 0$ or antiferromagnetic in the case $A_{j,m} < 0$ (for alternating lattices). If the exchange bonds fluctuate in both magnitude and sign the problem of finding the ground state becomes a complicated one.⁷⁴⁻⁸⁰

In the simplest case it can be assumed that the conversion of a crystal to an amorphous structure leads to a mixing of the J and K exchanges with concentrations $1 - \nu_K$ and ν_K respectively. An amorphous ferromagnet corresponds to a certain region of the parameters (λ, ν_K) with a relative magnetization $\bar{\sigma} = \bar{\sigma}(\lambda, \nu_K, z)$. The problem of finding $\bar{\sigma}$ is solved by the numerical Monte Carlo method. Figure 9 illustrates the results with our Monte Carlo calculations of $\bar{\sigma}$ for a cubic Ising lattice ($10 \times 10 \times 10$) with periodic boundary conditions. The exchange interactions are specified by the distribution function

$$\rho(A) = \nu_K \delta(A - K) + (1 - \nu_K) \delta(A - J). \quad (3.7)$$

The coherent-potential method^{81,82} was used in Ref.

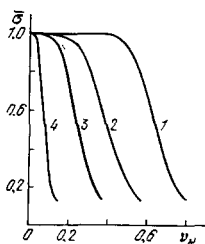


FIG. 9. Results of a numerical (Monte Carlo) calculation of the concentration dependence of the magnetization for various values of the parameter λ . 1— $\lambda = 0$; 2— $\lambda = -0.5$; 3— $\lambda = -1$; 4— $\lambda = -10$.

70 to describe the properties of an amorphous ferromagnet. According to this approximation, the disordered system is approximated by a translationally invariant crystal in which the exchange is implemented by a coherent exchange parameter $A_c(f-m, E)$ which depends on the excitation energy E of the magnetic system. The coherent parameter is found in the following manner: The equation for $S_f^z (S = 1/2)$ is treated in the Tyablikov approximation⁸⁴ using the substitution $\langle 2S_f^z \rangle = \bar{\sigma}(T)$, and the exchange-parameter matrix is broken up into coherent and fluctuational parts, $A_{f,m} = A_c(f-m) + \xi_{f,m}$. The equation for the Green's function $\hat{G}(E) = G_{f,m}(E) = \langle \langle S_f^z / S_m^- \rangle \rangle E$ is

$$(\hat{T}\Omega - \hat{H}_c - \hat{V}) \hat{G}(\Omega) = \hat{I}, \quad \Omega = E/\bar{\sigma}, \quad (3.8)$$

where $A_c(f-m)$ forms the translationally invariant matrix \hat{H}_c , and $\xi_{f,m}$ forms the fluctuational matrix \hat{V} . The zeroth approximation corresponds to the Green's function

$$\hat{G}_c(\Omega) = (\hat{T}\Omega - \hat{H}_c)^{-1} = \frac{1}{N} \sum_k e^{ik(f-m)} (\Omega - z A_c \epsilon_k)^{-1}, \quad (3.9)$$

$$\epsilon_k = 1 - \gamma_k, \quad \gamma_k = \frac{1}{z} \sum_h e^{ikh}.$$

The solution of (3.8) can be expressed in terms of a T matrix, and its configurational average can be taken:

$$\langle \hat{G} \rangle = \hat{G}_c + \hat{G}_c \langle \hat{T} \rangle \hat{G}_c. \quad (3.10)$$

The properties of the system are described by \hat{G}_c if $\langle \hat{T} \rangle = 0$. Since $\hat{T} = \hat{V} + \hat{V} \hat{G}_c \hat{T}$, then \hat{T} is a function of $G_c(\Omega)$, so that $\langle \hat{T} \rangle = 0$ is an equation for determining $A_c = A_c(\Omega)$. In the approximation of independent fluctuations of the exchange bonds we have $\hat{T} \approx \sum_\alpha \hat{t}_\alpha$, where \hat{t}_α is the partial scattering matrix for any pair α of nearest neighbors, and the condition $\langle \hat{T} \rangle = 0$ transforms to $\langle \hat{t}_\alpha \rangle = 0$. The matrix \hat{t} is found exactly, and after a configurational averaging with the distribution function (3.7) we find an integral equation for $A_c(\Omega)$:

$$\frac{1}{N} \sum_k \epsilon_k / (\omega - X(\omega) \epsilon_k) = \Psi[X(\omega), \lambda, \nu_K], \quad (3.11)$$

$$\Psi = \frac{z}{2} [1 - X - (1 - \lambda) \nu_K] [(1 - X)(\lambda - X)]^{-1},$$

where $\omega = \Omega/zJ$, and $X = A_c/J$ is a dimensionless coherent parameter.

The ground state is ferromagnetic if $\text{Re} X(0) > 0$ and $\sigma(0) > 0$. Figure 10 shows the upper limit for the existence of an amorphous ferromagnet as a function of λ and ν_K , according to the condition $\text{Re} X(0) = 0$.

The solutions for $X(\omega)$ determine the dispersion law for spin waves in an effective translationally invariant medium. The corresponding state density $g_c(\omega)$ is a

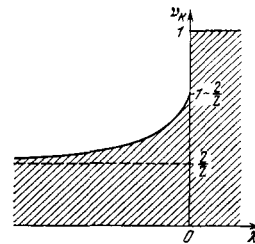


FIG. 10. Ferromagnetism occurs in the hatched region.

complicated function of ν_k and λ . This density $g_e(\omega)$ has been used to evaluate the Curie temperature of an amorphous ferromagnet.⁷⁰ The results for $z=6$ and $\nu_k=1/3$ are shown in Fig. 11. We see that for $|\lambda| \ll 1$ the switch to an amorphous structure gives us $T_C^{am} \gg T_C^0$, and there may be a conversion from an antiferromagnetic crystal to a disordered ferromagnet. Effects of this type have been observed experimentally.^{46,66}

The effect of an amorphous structure on the magnetic state of metals was studied in Refs. 71, 83, and 84. Richter and Kobe⁸⁴ carried out calculations using the model of close random packing of hard spheres. Tyagov *et al.*⁸³ and Kaneyoshi⁷¹ demonstrated that there may be a change in magnetic order upon switching to an amorphous structure. The physical reason for this effect lies in modifications of the Ruderman-Kittel exchange interaction⁷¹ and of the density of spin-wave states⁸³ when the crystal is converted to an amorphous state. A change from antiferromagnetic to ferromagnetic order upon the conversion of a crystal to an atomically disordered state has been observed experimentally in the alloys $Gd_{54}Ag_{46}$ (Ref. 67), $MnPt$ (Ref. 69), and $GdCu$ (Ref. 68). Interesting results have been obtained, for example, by Mizoguchi *et al.*⁶⁶ on the conversion of the compounds $GdAl_2$ and $GdCu_2$ to an amorphous structure. Crystalline $GdCu_2$, for example, is an antiferromagnet with a Néel temperature $T_N=41$ K, while the alloy $GdCu_2$ in the amorphous state is a ferromagnet with $T_C \approx 75$ K.

In summary, switching to an amorphous structure can fundamentally change the magnetic properties of a material and may even change the type of magnetic order.

4. MAGNETIC EXCITATIONS IN AMORPHOUS FERROMAGNETS

In a translationally invariant medium, the elementary magnetic excitations are of course described by Bloch plane waves. An amorphous medium is not translationally invariant over interatomic distances, as we have already mentioned. It is thus obvious that plane waves are generally not good eigenfunctions for such structures.⁸⁵ In general, it would be better to

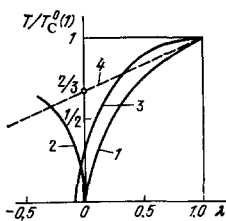


FIG. 11. Ratio of the magnetic-ordering temperature to the Curie temperature of an ideal, isotropic ferromagnet, $T_C^0(\lambda=1)$, as a function of λ for $\nu_k=1/3$. 1— $T_C^0(\lambda)/T_C^0(1)$ [$T_C^0(\lambda)$ is the Curie temperature of an ideal ferromagnet, $\lambda \geq 0$]; 2— $T_N^0(\lambda)/T_C^0(1)$ [$T_N^0(\lambda)$ is the Néel temperature of an ideal antiferromagnet, $\lambda \leq 0$]; 3— $T_C^{am}(\lambda)/T_C^0(1)$ [$T_C^{am}(\lambda)$ is the Curie temperature of an amorphous ferromagnet]; 4— $\bar{T}_C(\lambda)/T_C^0(1)$ [$\bar{T}_C(\lambda)$ is the Curie temperature of an amorphous ferromagnet in the approximation of an average exchange parameter $\bar{A} = (2/3)J(1+\lambda/2)$].

describe each excited state by a packet of spin waves, so that we cannot speak of a spin-wave spectrum in the strict sense of the term. It is clear, however, that if the amorphous structure is averaged over a sufficiently large volume the macroscopic homogeneity will lead to a translational invariance over distances corresponding to this large volume, and the description of excited states by plane waves becomes valid again. In other words, when the waves are sufficiently long in comparison with the radius of the first coordination sphere they do not sense the discrete nature of the atomic structure, and the approximation of a continuous medium is valid. In this limit the problem of the spin-wave spectrum is solved phenomenologically.

a) Phenomenological theory of spin waves

In the phenomenological approach, certain steady-state, random functions of the coordinates are adopted as models of the magnetic inhomogeneities of the amorphous structure. The first study of the spin-wave problem in an amorphous ferromagnet by this approach was carried out by Henderson and de Graaf.⁸⁶ They considered only the fluctuations of the exchange interaction and found that the nature of the dispersion law does not change in an amorphous ferromagnet, while the "magnetic hardness" of the system is reduced. This problem was pursued by Ignatchenko and Iskhakov.⁸⁷ They considered not only the exchange fluctuations but also fluctuations of the anisotropy, and they took into account the magnetic dipole-dipole interaction and the finite correlation radius r_0 for the fluctuations of the corresponding parameter.

The density of the phenomenological Hamiltonian of an amorphous ideal ferromagnet can be written

$$\mathcal{H} = \frac{1}{2} \alpha (\nabla M)^2 + \frac{1}{2} \beta (Ml)^2 - HM + H_m^2/8\pi, \quad (4.1)$$

where M is the magnetization, α is the exchange constant, β is the anisotropy constant, l is the unit vector along the easy axis of the anisotropy, H is the external magnetic field, and H_m is the magnetic dipole field, which is related to the magnetization by Maxwell's equations. The structural and chemical disorder of an amorphous ferromagnet causes the parameters α , β , M , and l to become functions of the coordinates. Ignatchenko and Iskhakov⁸⁷ write the correlation functions for the fluctuating quantities and the spectral densities (related to these fluctuating quantities by a Fourier transformation) in the form

$$K(x-x') = e^{-k_0(x-x')}, \\ S(k) = \frac{1}{\pi} k_0 (k^2 + k_0^2)^{-1}, \quad (4.2)$$

where k_0 is a typical wave number. Then the Landau-Lifshitz equation of motion for the magnetization can be used to write a dispersion relation⁸⁷ for waves propagating along the direction of the equilibrium magnetization, M , as follows:

$$\omega = \omega_0 + \alpha g M k^2 \left[1 - \left(\frac{\Delta \alpha}{\alpha} \right)^2 J(k) \right], \quad (4.3)$$

where $J(k)$ is some complicated function of k and the parameters of the system. Equation (4.3) is plotted in Fig. 12. Asymptotes 1 and 2, for $k=0$ for $k \gg k_0/2$,

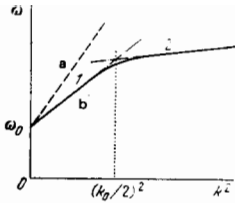


FIG. 12. Plot of dispersion relation (4.3) for spin waves. a—Crystal; b—inhomogeneous medium with fluctuating exchange α .

respectively, are given by Eq. (4.3) with

$$J_{1,2} \approx \begin{cases} 1 - \frac{k_M^2}{2(k_0 + k_M)^2}, \\ \frac{S}{4} - \frac{(k_0/2)^2 - k_M^2}{4k_M^2}. \end{cases} \quad (4.4)$$

The parameter k_s , corresponding to the slope change of the dispersion curve, is

$$k_s^2 = \left[\left(\frac{k_0}{2} \right)^2 - k_M^2 \right] \left[1 + \frac{2k_M^2}{(k_0 + k_M)^2} \right]^{-1}, \quad (4.5)$$

$$k_M = \sqrt{\frac{4\pi}{\alpha}}.$$

A general phenomenological theory of spin waves for amorphous magnetic materials has been derived by Andreev⁴⁴ and by Volkov and Zheltukhin.^{88,89}

b) Low-temperature behavior of the magnetization and spin-wave size-effect

The existence of spin waves in amorphous ferromagnets has now been demonstrated experimentally and theoretically. Spin waves are studied experimentally by primarily three methods: an indirect method involving measurement of the temperature dependence of the magnetization at low temperatures, inelastic neutron scattering, and the spin-wave size-effect resonance. The first study of the low-temperature behavior of the magnetization, $M(T)$, of the amorphous alloy⁹⁰ Co-P verified the prediction

$$M(T) = M_0 (1 - BT^{3/2} - CT^{5/2} - \dots) \quad (4.6)$$

or

$$\frac{\Delta M}{M(0)} = \frac{M(0) - M(T)}{M(0)} = B_{3/2} \left(\frac{T}{T_C} \right)^{3/2} + C_{5/2} \left(\frac{T}{T_C} \right)^{5/2}$$

of the spin-wave theory. This experimental result was the basis for the suggestion of a spin-wave dispersion law

$$\epsilon_k = Dk^2 + Ek^4 \quad (4.7)$$

with the familiar spin-wave coupling of B and D ,

$$B = \frac{2.612g\mu_B}{M_0} \left(\frac{k_B}{4\pi D} \right)^{3/2}, \quad (4.8)$$

where D is the dispersion parameter, a measure of some average exchange in the amorphous material. Expression (4.3) shows that the magnetic "hardness" becomes "softer" in an amorphous medium, so that it can be expected that the coefficient B in an amorphous material will be greater than in the corresponding crystal. This conclusion has been confirmed qualitatively in many experiments on the low-temperature magnetization and also on the hyperfine fields in amorphous magnetic materials.⁹¹⁻⁹⁴ Table III shows measurements of the coefficients B and C by various meth-

TABLE III. Spin-wave characteristics of certain alloys.

Composition	T_C , K	$\frac{B}{10^{-6}}$ K ^{-3/2}	$B_{3/2}$	$\frac{C}{10^{-9}}$ K ^{-5/2}	$C_{5/2}$	Reference
Fe ₄₀ Ni ₄₀ P ₁₄ B ₆	537	38	0.47	1.2	0.08	95
Fe ₇₅ P ₁₅ C ₁₀	619	23	0.36	0.8	0.08	96
Fe ₈₀ B ₂₀	685	22	0.40	1.2	0.17	96
Fe ₈₀ P ₁₅ B ₅	590	25	0.36	1.5	0.13	96
Fe ₈₀ P ₁₅ B ₅ Si ₂	377	65	0.48	1.5	0.04	96
(Fe ₈₀ Mo ₇) ₈₀ B ₁₀ P ₁₀	450	45	0.43	3.5	0.15	98
(Fe ₈₀ Mo ₇) ₈₀ B ₂₀	585	28	0.40	—	—	94
Co ₁₀ B ₁₀ Si ₁₅	660	16.9	0.29	—	—	97
Co ₁₀ B ₁₀ Si ₁₅ (crystal)	675	12.7	0.22	—	—	97
Fe (crystal)	1042	3.4	0.114	0.1	0.04	96
Ni (crystal)	627	7.5	0.117	1.5	0.15	96
Co (crystal)	1394	3.3	0.17	—	—	98
Fe ₈₀ B ₁₀ (crystal)	916	4.3	0.12	—	—	98

ods for crystalline and amorphous ferromagnets; comparison of the values of $B_{3/2}$ shows that this coefficient is much higher for the amorphous alloys. Very important information regarding the nature of this anomalous increase in $B_{3/2}$ came from a study of the dispersion parameter D by the methods of magnetic neutron diffraction and the spin-wave size-effect resonance. These two methods permit direct measurements of D and E in the spin-wave dispersion law, (4.7). The low-temperature behavior of the magnetization, on the other hand, is determined by the joint effects of various magnetic excitations, including local excitations.^{14,99,100}

The dispersion law for spin waves of amorphous magnetic materials has been studied¹⁰¹⁻¹⁰⁴ by the method of the ferromagnetic spin-wave resonance in thin magnetic films. These experiments have revealed a dependence of the resonant field H_n of the spin wave resonance mode n on the wave number of this mode, k_n . This relationship is easily found from (4.3) and the relationship among k_n , n , and the film thickness d :

$$k_n = \frac{n\pi}{d}. \quad (4.9)$$

The general result of this work was to confirm the quadratic dispersion law for long spin waves ($k \sim 10^5 - 10^6$ cm⁻¹). As expected theoretically, the dispersion parameter of an amorphous ferromagnet turned out to be considerably lower than that for a crystal.

Using data on the spin-wave size-effect resonance, we can easily calculate B in (4.6) and compare the results with measurements of the low-temperature magnetization. This comparison shows^{19,83} that the parameter $B(M)$ is systematically higher than the parameter $B(\text{SWR})$ for amorphous ferromagnets.

c) Neutron-diffraction study of the magnetic-excitation spectrum. Roton nature of the spectrum

Inelastic neutron scattering is a versatile method for studying spin-wave dispersion laws. It has been used for amorphous ferromagnets in Refs. 105-109. Fundamental results were obtained in experiments by Mook^{105,110} in a study of inelastic neutron scattering by the amorphous alloys Co₄P and Fe_{0.75}P_{0.15}C_{0.10} (Figs. 13 and 14). The basic result of that work was that the spin-wave spectrum is quadratic in k for long spin waves ($k < 0.5$ Å⁻¹), while the short-wave region ($k \sim 3$ Å⁻¹) has a "roton" spectrum with a minimum energy near the maximum of the structure factor (as in the case of ⁴He). In the case of amorphous ferromagnets, however, the sharp maximum in the structure factor

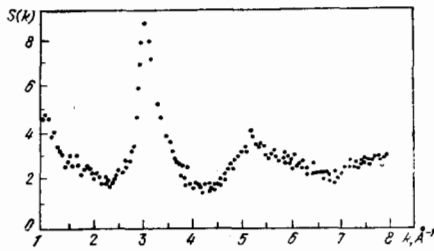


FIG. 13. Static structure factor $S(k)$ (in arbitrary units) of amorphous $\text{Fe}_{0.75}\text{P}_{0.15}\text{C}_{0.10}$.

(Fig. 13) is more likely to be accompanied by a diffuse minimum in the magnetic-excitation spectrum. This fact is not explained by the analog of the Feynman theory¹¹¹ of the roton spectrum of ^4He . At present we do not have a systematic analytic theory for the roton spectrum of spin waves in amorphous ferromagnets. The results obtained by Mook *et al.*^{105,110} have been explained qualitatively in Refs. 112–115. For example, Takahashi and Shimizu¹¹² introduce highly localized magnons to explain the roton nature of the spin waves near the maximum of the structure factor. They postulate the following wave function for such excitations:

$$\psi_{\mathbf{k}} = \lambda \sum_i e^{-\mathbf{k} \cdot \mathbf{R}_i} \exp [i(\mathbf{k} - \mathbf{Q}) \cdot \mathbf{k}_0 \mathbf{R}_i] a_i, \quad (4.10)$$

where λ is a normalization constant, \mathbf{R}_i is the radius vector of spin i , \mathbf{k}_0 is a unit vector along \mathbf{k} , \mathbf{Q} is determined by the average distance between spins, and a_i is the state function of spin i . The excitation spectrum for small values of $|\mathbf{k} - \mathbf{Q}|$ is then

$$\epsilon_{\mathbf{k}} = \Delta + D_1 (\mathbf{k} - \mathbf{Q})^2, \quad (4.11)$$

where Δ and D_1 are determined by the parameters of the wave function (4.10) and by the Heisenberg-exchange distribution function. The dashed curve in Fig. 14 corresponds to Eq. (4.11).

Studies of the low-temperature behavior of the magnetization, of the spin-wave resonance, and of inelastic neutron scattering by amorphous alloys of the type $\text{Fe}_{80}\text{B}_{20}$ has thus shown that, although the dispersion law is quadratic for low spin waves, the density of ordinary spin-wave states is not sufficient to explain the rapid decay of the macroscopic low-temperature magnetization of amorphous magnetic materials. Calculations by Coey¹⁶ show that this state density is roughly 30% lower than that required to explain the measured values of the coefficient B in Eq. (4.6). At present we do not have a well-grounded argument regarding which other magnetic excitations are responsible for the in-

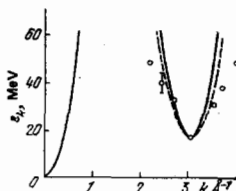


FIG. 14. Spin-wave spectrum of amorphous $\text{Fe}_{0.75}\text{P}_{0.15}\text{C}_{0.10}$. Solid curve— $\epsilon_{\mathbf{k}} = 120k^2$; dashed curve—theory of Takahashi and Shimizu.¹¹²

crease in B in amorphous ferromagnets. There is the suggestion, however, first advanced by Mizoguchi¹⁴ that diffusive oscillation modes must be taken into account. Such modes result from longitudinal spin deflections, and their existence is the result of the non-collinear structure of a disordered amorphous ferromagnet.¹¹⁶

d) Local magnetic excitations

Alloys in which one component is a rare earth metal constitute a special class of amorphous magnetic materials. They have attracted the attention of researchers not only because of their interesting magnetic properties¹⁹ but also because of their potential technological applications as magnetic-bubble materials,^{117–119} magnetically hard materials,^{56,120} and materials for magneto-optical applications.¹²¹ The unusual magnetic properties of these materials are determined by the large value of the single-ion anisotropy parameter D of the rare earth ions.

Measurements of the low-temperature specific heat¹⁶ of DyCu (Fig. 15) have shown it to be a linear function of the temperature. Coey¹⁶ has suggested that this behavior is a consequence of local magnetic excitations which arise because of the particular electronic structure of rare earth ions and the atomic disorder of the amorphous material. A theory of local magnetic excitations and of the thermodynamic properties of amorphous magnetic materials of this type has been derived.^{99,100} This theory is based on the Hamiltonian (3.1) in the molecular-field approximation. Harris *et al.*⁴⁷ have shown that in systems with a strong single-ion anisotropy ($|D| \sim |J|$) a magnetic ground state is established for which the spins of the rare earth ions fluctuate in direction. As a result, the molecular field acting on an ion in the amorphous alloy has a random orientation. For spins whose local anisotropy axis runs nearly perpendicular to the molecular field, the ground spin doublet of a rare earth ion is split by that component of the molecular field which is directed perpendicular to the anisotropy axis. This splitting, Δ , is small not only in comparison with the splitting in a crystal field ($\sim DS^2$) but also in comparison with the molecular field energy ($\sim HS$). Korenblit and Shender¹⁰⁰ have found

$$\Delta \sim DS^2 \left(\frac{H \sin \theta}{DS} \right)^{2S}, \quad (4.12)$$

where H is the molecular field, and θ is the angle between the local anisotropy axis and the molecular field.

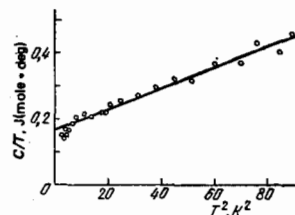


FIG. 15. Magnetic specific heat of the amorphous alloy $\text{DyCu}_{1.44}\text{Ar}_{0.05}\text{O}_{0.23}$.

The existence of local low-energy excitations with a density nearly independent of the energy leads to a linear temperature dependence of the specific heat of the magnetic system. The magnetization in such systems is a quadratic function of the temperature.

5. CRITICAL PHENOMENA IN AMORPHOUS MAGNETIC MATERIALS

The general interest in amorphous materials has spurred some recent studies of the critical properties of "random" magnetic materials.¹⁾ The disorder of the atomic structure of such materials greatly complicates both a theoretical analysis and the determination of reliable experimental results. These difficulties are apparently the reason why there has been nothing approaching a comprehensive study of critical phenomena in amorphous magnetic materials. Several theoretical studies have been carried out for atomically disordered systems consisting of a diamagnet and a dilute ferromagnet (or dilute antiferromagnet). It has been assumed in this work that the crystal lattice is preserved and that the disorder results from either a disordered substitution of diamagnetic ions for paramagnetic ions or a rupture of exchange bonds at random places in the lattice. Theoretical work on disordered magnetic materials has been reviewed by Korenblit and Shender¹²² and by Wortis *et al.*¹²³ Other papers¹²⁴⁻¹²⁸ have been published on the theory of phase transitions in such systems. Experimental work on critical phenomena in amorphous magnetic materials has been reported in Refs. 14 and 127-129.

We know that the following asymptotic relations should hold near the Curie temperature T_C of ferromagnets:

$$\left. \begin{aligned} \lim_{H \rightarrow 0} M &= M_S \sim (T_C - T)^\beta & \text{at } T < T_C, \\ \lim_{H \rightarrow 0} \frac{H}{M} &= \chi_0^{-1} \sim (T - T_C)^\gamma & \text{at } T > T_C, \\ M &\sim H^{1/\delta} & \text{at } T = T_C. \end{aligned} \right\} \quad (5.1)$$

The theoretical values¹³⁰ of the critical indices for Heisenberg-magnet crystals are $\beta = 1/3$ and $\gamma = 4/3$. Moorjani and Ghatak¹²⁶ used the cluster molecular-field method (the Bethe-Peierls-Weiss method) and found that the standard result of molecular-field theory, $\beta = 1/2$, $\gamma = 1$, emerges for disordered ferromagnets.

Malmhäll *et al.*¹²⁸ have reported an experimental study of the critical behavior of the amorphous ferromagnetic alloy $\text{Fe}_{29}\text{Ni}_{49}\text{P}_{14}\text{B}_6\text{Si}_{12}$. Figure 16 shows the results on the critical behavior of the susceptibility. We see that the susceptibility is described by (5.1) with $\gamma = 1.7 \pm 0.1$. For the other critical indices Malmhäll *et al.* found $\beta = 0.41 \pm 0.01$ and $\delta = 5.25 \pm 0.1$. Corresponding results were found in Ref. 131 for amorphous $\text{Co}_{70}\text{B}_{20}\text{P}_{10}$: $\gamma = 1.342 \pm 0.025$, $\beta = 0.402 \pm 0.007$, $\delta = 4.39 \pm 0.05$. Figure 17 shows the typical behavior of the specific heat C_p near T_C for $\text{Fe}_{29}\text{Ni}_{49}\text{P}_{14}\text{B}_6\text{Si}_{12}$ (Ref. 128).

A study by Lienard and Rebouillat¹³² is interesting in

1) That is, magnetic materials having a stochastic distribution of parameters.

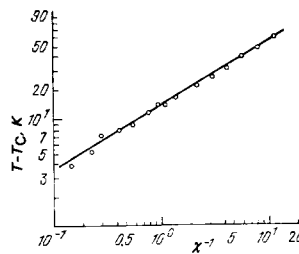


FIG. 16. Temperature dependence of the initial susceptibility of amorphous $\text{Fe}_{29}\text{Ni}_{49}\text{P}_{14}\text{B}_6\text{Si}_{12}$.

connection with concentration phase transitions in amorphous magnetic materials. Figure 18 plots the Curie temperature of the amorphous ferromagnet $\text{Y}_x\text{Ni}_{1-x}$ against the yttrium concentration; the plot is nearly linear as a function of $x_c - x$, where x_c is the percolation concentration ($x_c = 16.7$ at. %).

6. SOME PHYSICAL PROPERTIES OF AMORPHOUS MAGNETIC MATERIALS

a) Anisotropy

Ideally, an amorphous material is isotropic on the macroscopic scale, so that there is no macroscopic magnetic anisotropy. Again in this case, however, a local anisotropy (for example, that resulting from the spin-orbit interaction or the anisotropy of the local crystal field) has important effects on the ground state and the coercivity of the amorphous magnetic material. An explanation for this effect is now taking shape. This effect is important for the development of materials for applications (materials for magnetic-bubble technology and magnetically soft and magnetically hard materials).

The simplest model Hamiltonian for an exchange-coupled system with a fluctuating uniaxial anisotropy is that in (3.1). As was mentioned earlier, fluctuations of the anisotropy axes of an amorphous magnetic material lead to a noncollinear magnetic structure. A homogeneous ferromagnetism is unstable with respect to even a slight random anisotropy; a so-called stochastic magnetic structure arises.^{56,87}

Clark¹³³ and Rhyne *et al.*¹³⁴ reported the first experimental observations of an effect of a random anisotropy on the low-temperature behavior of the magnetic properties of amorphous alloys of Tb, Dy, or Ho with Fe and Co. These alloys have magnetic moments much lower than those of their collinear ferromagnetic analogs, and they have a high coercive force (up to 30 kOe). Further research has shown that the lowering of the

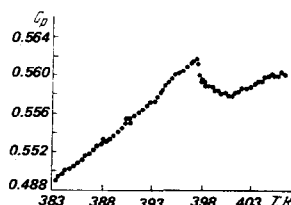


FIG. 17. Temperature dependence of the specific heat C_p (in arbitrary units) of amorphous $\text{Fe}_{29}\text{Ni}_{49}\text{P}_{14}\text{B}_6\text{Si}_{12}$.

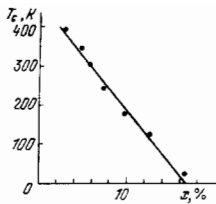


FIG. 18. Curie temperature of the amorphous ferromagnet Y_xNi_{1-x} as a function of the yttrium content x .

magnetic moment results from the noncollinear arrangement of magnetic moments of the $4f$ ions.

A local magnetic anisotropy has important effects on the coercive force of amorphous magnetic materials. Alben *et al.*¹² have shown that the behavior of an amorphous ferromagnet changes quite sharply from magnetically soft to magnetically hard when the energy of the single-ion local anisotropy becomes comparable to the exchange-interaction energy (Fig. 19). The coercive force H_c reaches a level amounting to about 20% of the effective exchange-interaction field, in accordance with the experimental results. In the case of soft amorphous alloys, the local anisotropy leads to values $H_c \sim 10^{-6}$ Oe, which are three orders of magnitude lower than the experimental values. The apparent reason for the discrepancy lies in technological factors, so that we can expect a further increase in the softness of amorphous alloys of the ($3d$ metal)—metalloid type. Figure 20 shows experimental hysteresis curves for three amorphous materials, along with curves for two crystalline analogs for comparison.^{134,135} In the case of the Fe-P-C alloy, the anisotropy is averaged out, and the alloy is very soft. The Co-P alloy is softer in the amorphous state than in the crystalline state, because of a macroscopic anisotropy of technological origin. Finally, for the alloy Tb-Fe, we see that the random anisotropy is not averaged out at 4.2 K.

Real amorphous materials are not ideally isotropic on the macroscopic scale because of various, primarily technological, factors. As a result, a macroscopic magnetic anisotropy appears. Studies have shown¹⁸ that this "technological" anisotropy may be caused by oriented fluctuations of the composition and stress and also by magnetic annealing. Although the technological anisotropy is important from the standpoint, for example, of developing amorphous magnetic-bubble mater-

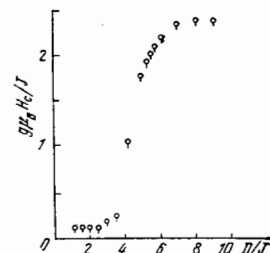


FIG. 19. Dependence of the coercive force H_c on the single-ion anisotropy parameter D (J is the exchange integral).

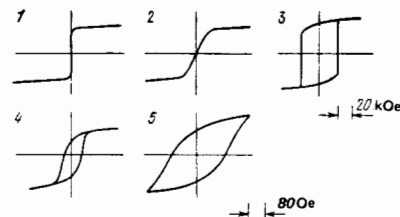


FIG. 20. 1-3—Hysteresis curves of amorphous alloys; 4, 5—of corresponding crystalline alloys. 1, 4) $Fe_{75}P_{15}C_{10}$; 2, 5) $Co_{78}P_{22}$; 3) $Tb_{33}Fe_{67}$.

ials,¹³⁶⁻¹⁴⁰ a theoretical generalization of this phenomenon is still in a beginning stage.

b) Magnetoelastic coupling

Linear magnetostriction is the best-studied manifestation of magnetoelastic coupling. It follows from general considerations that the magnetostriction of an amorphous ferromagnet should be isotropic. With regard to its magnitude, there is no reason for a fundamental difference between the magnetostriction of an amorphous magnetic material and that of a crystal. Figure 21 shows some typical experimental results¹⁷ for the quasiternary system $Fe_{80}B_{20}-Co_{80}B_{20}-Ni_{80}B_{20}$. The solid curve is the line of zero magnetostriction. Table IV shows measurements of the magnetostriction of certain amorphous materials. O'Handley and Chou¹⁷ have carried out a detailed study of the temperature dependence of the magnetostriction constants of alloys of the $Fe_{80}B_{20}$ type with the goal of determining the most important mechanisms for the magnetoelastic coupling. Figure 22 shows the results for $Fe_{80}B_{20}$, $Ni_{50}Fe_{30}B_{20}$, and $Co_{40}Ni_{40}B_{20}$ along with theoretical results on the magnetostriction constants according to calculations by Callen's single-ion theory¹⁵⁰ and in the approximation of a dipole-dipole mechanism. We see that the single-ion mechanism is predominant. A similar analysis for $Co_{80}B_{20}$ has shown that the two magnetostriction mechanisms are of equal importance.

A well-known manifestation of magnetoelastic coupling in magnetic materials is a dependence of the sound velocity on the magnetic field. Tsuya *et al.*¹⁵¹ have found that this dependence is anomalously strong (the "giant ΔE effect," where E is the Young's modulus) in magnetically soft amorphous alloys with a relatively large magnetostriction. Tsuya and Arai¹⁵² have derived a phenomenological theory for this effect. The giant ΔE effect in amorphous ferromagnets may prove useful for developing controllable delay lines.

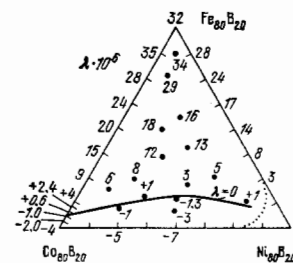


FIG. 21. Linear magnetostriction of $Fe_{80}B_{20}-Co_{80}B_{20}-Ni_{80}B_{20}$ amorphous alloys.

TABLE IV. Magnetostriction of amorphous alloys (300 K).

Composition	$k \cdot 10^6$	Reference	Composition	$k \cdot 10^6$	Reference
Co_{91}P_9	-4.3	141	$\text{Fe}_{88}\text{B}_{12}$	49	147
$\text{Fe}_3\text{Co}_{72}\text{P}_{16}\text{B}_4\text{Al}_3$	~0	142	$(\text{Fe}_{60,93}\text{Co}_{0,05})_{78}\text{Si}_8\text{B}_{14}$	44	147
$\text{Fe}_6\text{Co}_{74}\text{B}_{20}$	0.4	143	$\text{Co}_{78}\text{Si}_8\text{B}_{14}$	-2	148
$\text{Co}_{46}\text{Ni}_{31,1}\text{P}_{22,7}$	-9	144	$\text{Fe}_{78}\text{Si}_8\text{B}_{14}$	35	148
$\text{Fe}_{80}\text{P}_{13}\text{C}_7$	31	145	$\text{Co}_{39}\text{Ni}_{39}\text{Si}_{18}\text{B}_{14}$	1	148
$\text{Co}_{80}\text{B}_{20}$	-4	146	$\text{Gd}_{32}\text{Co}_{78}$	35	149
$\text{Fe}_{40}\text{B}_{20}$	31	146			

c) Electrical resistance

There are two distinctive features in the behavior of the electrical resistance of amorphous metallic magnetic materials: The resistance of amorphous metals at room temperature is 2-4 times that of the corresponding crystalline metals; the temperature dependence of the resistance of amorphous metals (magnetic or not) exhibits a minimum, usually below room temperature. Figure 23 shows results obtained by Rapp *et al.*¹⁵³ on the amorphous ferromagnetic alloys $\text{Fe}_{75}\text{P}_{16}\text{B}_6\text{Al}_3$ and $\text{Fe}_{60}\text{Ni}_{15}\text{P}_{16}\text{B}_3\text{Al}$. Their results can be described well by the theory for the electrical resistance of amorphous alloys derived by Cochran *et al.*¹⁵⁴

Such features of the electrical conductivity of amorphous alloys as the small temperature coefficient of the resistance and the anomalously large Hall effect result from the strong effects of the chemical and configurational disorder on the electron mean free path.

7. CONCLUSION

The rapid progress in research on amorphous magnetism over roughly the past decade has led to an understanding of many important aspects of the physical properties of amorphous materials. We can now see the foundations for a theory for the ground state and elementary excitations of atomically disordered magnetic materials. Important aspects of the magnetic behavior of amorphous systems, in particular, the role played by fluctuations of the parameters of the system, have been discovered. At the same time, several important questions require further study.

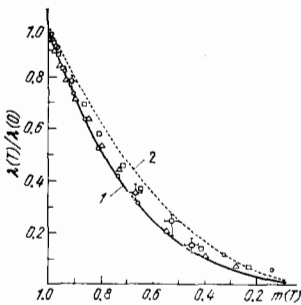


FIG. 22. Temperature dependence of the magnetostriction of several amorphous ferromagnets. \circ — $\text{Fe}_{80}\text{B}_{20}$; Δ — $\text{Ni}_{50}\text{Fe}_{30}\text{B}_{20}$; \square — $\text{Co}_{40}\text{Ni}_{40}\text{B}_{20}$. Curve 1—Single-ion theory of magnetostriction; 2—theory based on the mechanism of dipole-dipole coupling $\lambda \sim m(T)^2$; $m(T)$ —relative magnetization.

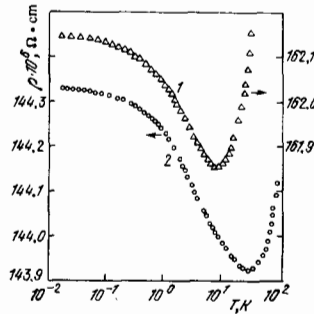


FIG. 23. Temperature dependence of the electrical resistance of amorphous alloys. 1— $\text{Fe}_{75}\text{P}_{16}\text{B}_6\text{Al}_3$; 2— $\text{Fe}_{60}\text{Ni}_{15}\text{P}_{16}\text{B}_3\text{Al}$.

There is the problem of developing a closed theory for the magnetic state and magnetic properties on the basis of the real disordered atomic structure. We lack a general theory of elementary magnetic excitations of various types. There has been essentially no study of critical phenomena in amorphous magnetic material. Research on the dynamics of the spin system is still in the beginning stage.

The work which has been carried out has confirmed the excellent prospects for technological applications of amorphous magnetic materials. Important magnetically soft and magnetically hard materials have already been developed, and certain amorphous alloys look promising for use as heads in magnetic tape recording. Work is being carried out to develop media for magnetic recording of information.

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