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Exchange and Anisotropy Noise in Position Entropy Solids*

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Site ordered crystalline materials (zero position entropy) show, below a magnetic ordering temperature, the phenomenon of a magnetic phase state, that is to say, phase relationships exist between the magnetic interactions as well as the relative directions of ordered magnetic moments. On introduction of positional entropy (site disorder, defect, or amorphous materials), fluctuations from site to site of magnetic interactions are created (interaction fluctuation or noise state). In this paper the phenomenology of noise states is classified according to the relative importance of fluctuations from site to site of anisotropy (anisotropy noise) and exchange (exchange noise) interactions, focusing on crystalline site disordered intermetallics of relatively high moment concentration. Perhaps the best telltale sign for the dominance of anisotropy or exchange noise is the nature of the magnetization-field behavior which tends to be inelastic in the former (anisotropy noise, creating magnetic hardness type behavior) and elastic in the latter (exchange noise, reminiscent of metamagnetism). Extreme cases with respect to the relative importance of anisotropy and exchange noise are exemplified and discussed on representative examples. Also comparisons are drawn with "magnetic order" in amorphous intermetallics. A distinction between anisotropy and exchange noise allows for a better understanding of the complex behavior of such systems as $Y_6Mn_{23-x}Fe_x$ or $PrMn_{2-x}Al_x$, for which new information is given, as well as for several related cases, reported earlier in the literature.

Introduction

A new area of materials research has recently come into its own with the charting of the magnetic behavior of materials exhibiting partly randomized high local anisotropy. This randomization is achieved either with site disordered crystalline or with amorphous materials. The relevant phenomenology gains uniqueness when it is seen in broader context. A generalized approach also allows better understanding of complex new phenomenology as presented in this paper.

General

The conventional types of magnetic order such as ferromagnetism, or types of antiferromagnetism (ferrimagnetism, spiral structures, etc.) have in common a periodic arrangement of magnetic moments (magnetic structure) (*1*). This state of magnetic order can be termed "phase order" (*2*, *3*) in the sense that moments, even if modulated over considerable spatial regions, remain periodic and allow for phase relationships. Over the last two decades, however, a healthy awareness of the influence on magnetic order of local environment arose. Whenever magnetic moments are not securely embedded in a well-defined crystallographic environment, such as would be the case only in the limiting situation of well-defined site ordered crystalline compounds (zero positional entropy), magnetic interactions (exchange or crystal field interactions) fluctuate from site to site. Such a material, exhibiting position entropy, may therefore freeze into a magnetic state which will not exhibit phase relationships

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in the magnetic interactions. This state has been termed NOISE (not orderly interacting spin ensemble) state (*3*). The local fluctuations in direction and magnitude or magnetic interactions can lead to a magnetic structure with randomized direction (and magnitude) of local moments. Magnetic moments can be totally scattered in direction such that no macroscopic moment obtains (total scatter state) or else shows partial scatter leading to a resultant moment (resultant scatter state) (*1*). Such scatter magnets will not exhibit the phase relationships of moment directions indicated above. (A tuning of noise into phase order would involve the removal of crystallographic irregularities.)

Historically, the first investigations of scatter magnets involved cases of predominantly low local anisotropy. Terms and subjects such as exchange anisotropy (*4*), mictomagnetism (*5*), mixomagnetism (*6*), or spin glass (*7*) were born spanning the space from concentrated to dilute moment systems. More recently strong anisotropy has been introduced in site disordered (crystalline) as well as amorphous (position disordered) materials of high moment concentration. Perhaps the most striking and novel feature characterizing these high anisotropy randomized distribution (HARD) materials is the occurrence of the often giant intrinsic hardness effects reviewed elsewhere (*8*). A fruitful cross-fertilization among these various fields (high anisotropy–low anisotropy; site disorder–position disorder) is in the process of taking place at this time. This survey of some of the relevant phenomenologies is intended to adumbrate the possibilities for such cross-fertilization. Also the possibility for a more uniform taxonomy as sketched out earlier (*2*, *3*) is introduced. As the field of spin glass (*7*) or mictomagnetism (*9*) and amorphous magnetism (*10*) is relatively well reviewed, this paper will concentrate on NOISE states in site disordered intermetallic compounds with emphasis on systems with relatively high moment concentrations. For this purpose the phenomenology of site disordered pseudobinary compound series will be contrasted with the one of comparable compounds of purely amorphous character. Extreme cases of either high and low local anisotropy will be taken from the literature and discussed. Within this frame it will be possible to gain a clearer understanding on one hand of the complex phenomenology of systems such as $Y_6Mn_{23-x}Fe_x$ or $PrMn_{2-x}Al_x$, for which new information will be presented, and on the other for the behavior of similar compound series reported previously in the literature. In addition, some intriguing possiblities for the creation of unusual magnetic properties will be discussed.

Influence on Magnetic Structure of Exchange and Anisotropy

In the case of magnetically isotropic materials (absence of crystal field anisotropy) on the basis of rare earths the fundamental mechanism responsible for the drop into a magnetically ordered state involves an indirect coupling of magnetic moments over conduction electrons. The type of magnetic structure (mode of moment alignment) is then determined by a minimization of energy constraints arising from polarizations of conduction electrons at moment location within a three dimensional magnetic lattice (*11*). In well-defined structures this leads to a characteristic progression of the type of magnetic structure as a function of the electron concentration whereby regions of ferromagnetic exchange alternate with regions of antiferromagnetism as the number of carrier electrons is altered.

In case of a strong additional energy constraint arising from interactions of the magnetic ground state wavefunctions with the charges of neighboring atoms (crystal field interaction) the influence from the indirect exchange interaction is modified. The anisotropy resulting from crystal field interaction in many cases competes with the one from indirect exchange and as a rule a compromise

is reached which entails a magnetic structure with canted directions of moments (*12*). Several of the complex magnetic types harboring highly anisotropic rare earths have been discussed along these lines especially in the pioneering efforts from the French school originating from Bertaut's work. Regrettably, no survey summarizing success in this direction has apparently been published.

It is obvious that similar constraints concerning exchange or anisotropy interactions in a site or position disordered material can yield a magnetic structure with varying degrees of randomly oriented magnetic moments. While the situation in magnetic materials on the basis of transition metals is different from the one of rare earths in many respects (stronger direct exchange contributions, weaker crystal field anisotropies) the gist of the arguments concerning the stability of magnetic types remains rather similar to the one given for rare earths.

In the following, extreme cases of noise states shall be outlined and a relevant language for site disordered crystalline materials will be introduced and compared with the one for amorphous (position disordered) materials.

Basic Considerations Concerning the Origin of Noise Structures

The basic premise for the notion of anisotropy and exchange noise is that these interactions will vary as a function of local environment. As a thought experiment this is made plausible for, say, a hexagonal compound *RPA*, the parent compounds of which are the fictitious compounds RP_2 (strong plane anisotropy in a compound between metal *P* and rare earth *R*) and RA_2 (strong axis anisotropy compound). For ease of presentation, a shorthand shall be employed in the following in a site disordered crystalline material a line will be drawn over that part of its chemical formula which is characterized by random site occupation (for example $Tb\overline{FeAl}$), and an amorphous material will be designated by underlining (for example, $\underline{TbFe_2}$). Assuming weak but uniform ferromagnetic exchange,

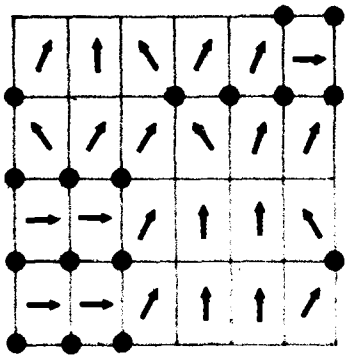

FIG. 1. Schematic representation of a resultant scatter state originating from anisotropy noise (exchange is weak and remains ferromagnetic) of a compound $R\overline{PA}$ as described in the text. Crossing lines indicate atoms *A*, circles indicate atoms *P*. Arrows represent magnetic moments of *R*. Plane clusters and axis clusters are indicated.

compound $R\overline{PA}$ will be in a resultant scatter state as shown schematically in Fig. 1. A thought experiment similar to that leading to anisotropy noise can make plausible the concept of exchange noise in a compound of weak anisotropy $R\overline{FA}$ where RF_2 is ferromagnetic and RA_2 is antiferromagnetic as shown in Fig. 2.

The probability of finding a certain magnitude of moment projected along a preferred direction (M_z) is given in Fig. 3 for various theoretically possible types. It is conceivable that a state of total moment scatter as

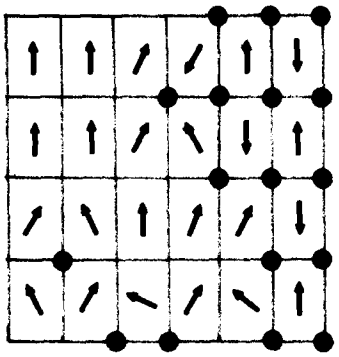

FIG. 2. Schematic representation of a scatter state originating predominantly from exchange noise (anisotropy is weak) of a compound $R\overline{FA}$ as described in the text.

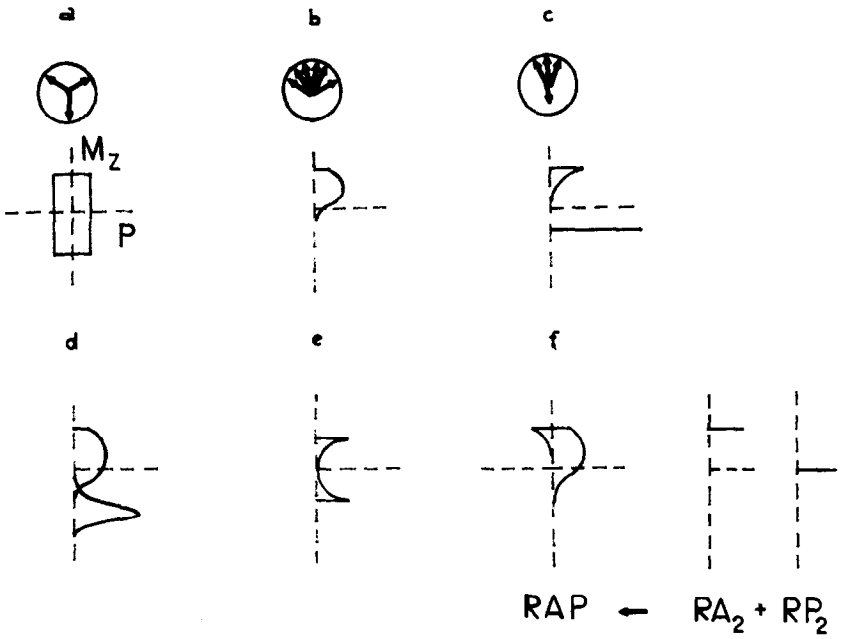

FIG. 3. Schematic representation of total scatter, (a) resultant scatter and (b) complex scatter states (*c–f*). Probability (*P*) of finding a certain magnitude of magnetic moment (M_z), projected along a preferred direction (*z*), is presented as given in a circle cut parallel to that direction. Representations in (a) and (f) include this probability along the positive and negative *x* direction. Total scatter (a) corresponds to speromagnetism of amorphous materials. In (c) one sublattice moment (of different magnitude to the other) does not scatter, presumably due to strong exchange by comparison, say, with anisotropy noise. This condition is relaxed in (d) where both sublattices scatter and the larger moment species exhibits a range in moment magnitudes as well. (e) represents an antiferromagnetic scatter state. (h) indicates a tendentious or skewed resultant scatter type. Symmetrical resultant scatter presumably would be observed by a new choice of *z* axis. This state can be considered to result in a compound $\overline{RAP}$ as described in the text and schematically indicated in the figure. Indications for such a state has been found via a careful reexamination of earlier neutron diffraction patterns on hexagonal $MgZn_2$ type materials in $ErCo_{2-x}Al_x$.

indicated in Fig. 3a will be obtainable in site disordered materials of high magnetic moment concentrations, especially when strong contributions from exchange noise are considered. Among other ways, this will manifest itself in the total loss of intensity to be sampled into magnetic reflections of a neutron diffraction pattern below a moment freezing temperature. Several experimental indications (*5*, *9*, *13*), say in the class of mictomagnets, hint at the aspects of this extreme case which in many ways is related to the concept of a speromagnet (*10*) as defined for amorphous materials.

As the origin of noise structures is connected with fluctuations within atomic sites of exchange interactions and anisotropy interactions the respective phenomenology will depend on the relative importance and type of these interaction fluctuations. Moreover, concentration will play the role of a third variable, allowing yet for another degree of freedom in the development of a characteristic phenomenology. One can then in principle distinguish between the following four extreme cases:

(1) Relative importance of exchange fluctuations and high moment concentration. Massive exchange noise.

(2) Relative importance of exchange fluctuations and low moment concentration. Dilute exchange noise.

(3) Relative importance of anisotropy and high moment concentration. Massive anisotropy noise.

(4) Relative importance of anisotropy and low moment concentration. Dilute anisotropy noise.

It should be noted that the fourth pole in this schematic representation (case 4, dilute systems with weak exchange and relatively strong anisotropy) appears not to be fully realizable, as anisotropy is itself a function of exchange and will become rather small at high moment dilution.

Experimental Evidence

Some of the more or less established types of noise structures are indicated in the following schematic presentation (with representative examples), which spans a space in four poles as it is given on the basis of the relative importance of exchange and anisotropy interactions as well as concentration (concentration increases towards the top). Some of the relevant magnetization field behavior, which is described more fully below, is indicated. All examples show time dependence effects of magnetization.

Relative importance of exchange

Massive exchange noise: ($\overline{\text{Mn–Cu}}$) (displaced "hysteresis loops")

Mictomagnetism ($\overline{\text{Cu–Mn}}$) (displaced "hysteresis loop," elastic, "rebound" hardness)

Dilute exchange noise: spin glass ($\overline{\text{Cu–Mn}}$)

Relative importance of anisotropy

Massive anisotropy noise: ($\text{Tb}\overline{\text{Fe}_{2-x}\text{Al}_x}$) (giant intrinsic magnetic hardness, inelastic type hardness)

Dilute anisotropy noise: (not explored)

In the following some of the more or less well developed cases will be discussed.

(*a*) *Exchange Fluctuations Predominant*

Exchange scatter state. Exchange fluctuations include tendencies for parallel and antiparallel moment alignment. This results in an apparent resultant scatter state, that is to say, magnetic moments will be scattered in direction but show a resultant magnetization say for unique conditions. As concentration of moments is increased, various more or less distinct states of short range "order" are reached. A textbook example is the system Cu–Mn which, at low concentration in Mn, is described as a spin glass (*7*), then changes, on increasing Mn concentration, to a mictomagnetic state (*5*) with clusters of ordered moment in a glass matrix, and finally exhibits massive short-range "order" (*5*, *14*) before exhibiting signs of long-range antiferromagnetic order (*14*) (which plausibly should still retain some aspects of a resultant scatter state).

In a *spin glass* each moment is in its own random field, H_i, but unlike a simple paramagnet, a spin glass exhibits, as $T \to 0$, a certain rigidity in an external magnetic field, obviously as a result of disorderly weak exchange (and possibly anisotropic) interactions (garbled low anisotropy spins—GLAS). Some of the most important questions concerning the physical picture of a spin glass appear to be still largely unsettled (*7*). Such questions are: Is there a phase transition at T_F; are there clusters and domain walls; and perhaps most important for the aim of the present study, is additional anisotropy essential? The relevant phenomenology of spin glasses shows a gently sloping section in χ^{-1} vs T, with $(\chi^{-1})_{sg} > (\chi^{-1})_p$ and a paramagnetic Curie point of $\theta \cong 0$. The magnetization, in low fields, peaks around a freezing temperature. In cases where no major importance of anisotropy is suggested there exist no hysteresis effects in magnetization vs field, but displaced curves are obtained when the material has been cooled in a field. Beyond these general remarks the spin glass phenomenology is of considerable diversity as witnessed by the various terms endemic to various laboratories (infinite cluster, cluster glass, etc.). As this article searches primarily for common denominators in the phenom-

enology of relatively dense moment systems these points shall not be further amplified.

The paramount importance of (increasing) moment concentration is brought out in the evolution from the spin glass to the *micto-magnetic state* say in Mn–Cu. Mictomagnets are, at high temperatures, superparamagnetic but moment orientations freeze on decreasing temperature into a state without long-range order. Magnetic clusters, in sizes considerably larger than would correspond to chemical clustering, are then embedded in a glassy matrix. Thermomagnetic history and displaced "hysteresis loops" are among the fingerprints of this complex magnetic state. Present-day understanding of mictomagnetism rests on a model in which an intricate mixture of very small ferromagnetic and antiferromagnetic regions is formed, say in Mn alloys such as Cu–Mn, through local composition fluctuations (*9*). A more involved version of this model envisages domain ensembles incorporating interacting pairs of ferro- and antiferromagnetic regions which exhibit some local anisotropy (*9*). In a recent paper (*13*) neutron diffraction has been employed to elucidate the magnetic states of $Fe_{0.7}Al_{0.3}$. This material is unusual in that it is ferromagnetic below T_c = 400°K, paramagnetic below an inverse Curie temperature T_i = 170°K, and mictomagnetic below the freezing temperature T_f = 92°K. Low-angle neutron diffraction shows that this material exhibits ferromagnetic clusters (in size larger than would correspond to chemical clusters) through all of these regions. The size of these clusters is smoothly decreasing with decreasing temperature. It was postulated, therefore, that the ordering transitions result from the varying mode of coupling between these clusters. It was furthermore suggested that an antiferromagnetic exchange interaction increases towards lower temperatures resulting, first in the loss of magnetic order and subsequently in the freezing into a cluster glass type state.

In a critical review several difficulties with the models presented so far have been indicated (*9*). Several mictomagnets exist, which do not show straightforward indications for antiferromagnetic interactions (Fe–Au, Fe–Rh). Perhaps especially interesting is the fact that field cooling produces remanence proportional to the strength of the field but the field at which reversal of magnetization is achieved (displacement without hysteresis effects) is indirectly proportional to the magnitude of external field. Also in cases where the field is applied below the freezing temperature, at progressively decreasing temperatures, the remanence at 4.2°K becomes correspondingly smaller while the displacement of the "hysteresis curve" increases in proportion, such that the product of remanence and displacement remains constant.

Finally, materials involving *massive exchange fluctuations* (Mn–Cu, $Mn_{3-x}Fe_xSn$ (*4*), and Mn–Ni–Fe (*6*)), show negative deviations from Curie–Weiss behavior as well as displaced "hysteresis loops." Rather large (pseudo) coercive forces with reversible magnetization are observed in materials cooled in a field. This rebound magnetic hardness obviously originates from a process analogous to the metamagnetic one in antiferromagnets and can probably still be further maximized.

Complex cases. Besides an apparent scatter in moment direction due to exchange noise, fluctuations in magnitude of magnetic moment without scatter of moment direction have to be considered. $YCo_{5-x}Ni_x$, where a binomial distribution of nearest neighbors dictates the magnitude of moment on a local moment model, is an example (*15*) in the limited sense of exhibiting exchange fluctuations within (presumably) complete ferromagnetism. However, the high local anisotropy and the concomitant strong hardness effects classify this material predominantly as a representative for the case of anisotropy noise. Similarly, rare earth moments are subject to pronounced local moment fluctuations in cases where the influence of the crystalline electric field is strong. This is plausibly the case in $PrAl_2$ with partial disordered Cu or Ag substitution for

Al. Here, variations in local magnitude of exchange field will have a pronounced influence on the local magnitude of moment. In fact, neutron diffraction on $PrCu_{0.5}Al_{1.5}$ (*16*) ($MgCu_2$ type) has shown that the ordered moment on Pr is further reduced beyond the one in $PrAl_2$ without a sizable concomitant increase in the diffuse scatter. This crystal field reduction in magnetic moment then is assumed to be locally fluctuating. In fact, for $PrAl_{2-x}Ag_x$, the loss of signs of magnetic order has been explained (*17*) on the basis of the creation of a subthreshold situation concerning exchange by variation of the local crystal field parameters (partial singlet creation due to randomization).

In cases where anisotropy is very strong and remains nonfluctuating (say uniaxial), exchange fluctuations may remain cryptic, that is to say, magnetic moments will remain in a state of phase order. Nevertheless, exchange fluctuations will manifest themselves in an indirect way, say via their effects on domain wall thickness and propagation.

(*b*) *Anisotropy Fluctuations Predominant*

Anisotropy scatter state. Historically speaking, the diagnosis of an anisotropy scatter state grew out of some of the pioneering efforts connected with the study of the influence of the electron concentration (ec) on the type of magnetic order in the middle sixties [details of what will be said is summarized in a recent review paper (*8*)]. However, instead of finding changes within phase order states, such as ferromagnetism developing into modulated types of antiferromagnetism as the ec was altered, a new type of magnetic order was fingerprinted altogether. Among the hallmarks in the phenomenology of such systems as $TbCo_{2-x}Al_x$, exhibiting high local anisotropy and partial crystallographic disorder, were signs for random scatter in moment direction and the development of strong intrinsic magnetic hardness effects. Hardness effects, however, are viscive (inelastic) in nature, that is to say, magnetization does not recover in an elastic manner as, say, in mictomagnets. The, often giant, hardness effects are a result of interaction fluctuations in an otherwise homogeneous material, at least speaking in the conventional sense.

The importance of domain walls modified by interaction fluctuations is clearly borne out by the dramatic effects minor impurities can have on intrinsic hardness. An example is $SmNi_5$ ($T_c = 41°K$) which in well-defined form exhibits a negligible coercive force (at 4.2°K) while $SmNi_{4.8}Fe_{0.2}$ ($T_c = 103°K$) has $H_c = 166$ kOe at 4.2°K. While intrinsic hardness in phase order compounds is of minor importance, due to the near equivalence of domain wall location at possible loci in the crystal, this energy degeneracy is lifted on introduction of interaction fluctuations (noise state) which both narrows the dimension of the wall and simultaneously pins it. It is therefore gratifying to observe that a relation exists between the magnitude of coercive force and the number of weakly coupled magnetic constituents (binomial distribution function for T metals–maximum density of scatter centers for R metals). The propagation of these interaction fluctuation domain walls involves thermal and field activation over energy barriers. As a result a characteristic temperature and time dependence of magnetic hardness are observed. A similar situation appears to pertain in amorphous magnetic materials on the basis of highly anisotropic rare earths.

Quite generally speaking it should be pointed out that anisotropy fluctuations create exchange fluctuations in their wake due to the dependence of exchange on the angle of interacting moments. Moreover, there exists a relationship between the strength of exchange and the development of anisotropy such that a situation of byzantine complexity results. This was hinted at in Fig. 3f. The relative influence of exchange and anisotropy fluctuations on intrinsic hardness has been exemplified among others in $SmCo_{5-x}Ni_x$ (*15*) and $SmNi_{5-x}Cu_x$ (*18*).

In order to be able to discuss, on a comparative basis, phenomenology of site and position disordered materials some aspects of the latter shall be briefly introduced. Details are given in a recent review (*8*).

Magnetic Taxonomy of Amorphous Materials

The question of what happens to magnetic structure and concomitant magnetic phenomenology in amorphous materials introduces only minor variations to the theme presented so far for site disordered materials. However, as an impressive library of experimental data is now available for these metastable relatives of crystalline position entropy materials, two of the terms used for magnetic structures in amorphous materials shall be briefly introduced. Accordingly a state in which total scatter in moment direction is observed is termed speromagnetism (scatter state analog for site disorder). An asperomagnet differs from a speromagnet in that some moment directions are more likely than others (this corresponds to the resultant scatter magnet defined for site disorder materials). An asperomagnet, therefore, has a resultant magnetization.

Some recently discovered examples where either anisotropy or exchange noise is predominant shall be discussed in the following section by way of comparing phenomenology in site and position disordered materials.

Comparison of Site and Position Disordered Materials

A direct comparison of site disordered (crystalline) with position disordered (amorphous) materials of identical compositions is so far not possible, as most information on amorphous materials is available for binary systems which exist as well-defined compounds in their crystalline state. As an example the celebrated amorphous $TbFe_2$ exists in the crystalline state as a well defined $MgCu_2$ type structure (zero position entropy). Partial substitution for Fe by, say Al, however, develops a phenomenology largely comparable with the one of amorphous $TbFe_2$.

Comparisons shall now be made concerning systems on the basis of rare earths and high and of low anisotropy, respectively, and of one system with a magnetic transition metal only.

(*a*) *$TbFe_{2-x}Al_x$ vs $TbFe_2$.* Tb, as a rare earth with large orbital contribution, exhibits the potential for high magnetic anisotropy which in turn dominates the respective phenomenologies in site and position disordered materials. $TbFe_{2-x}Al_x$ and the related $TbCo_{2-x}Al_x$ were among the first systems on which the fingerprints of anisotropy noise, namely scatter order (*2*) and strong intrinsic hardness, were recognized (*8*). A similar phenomenology was subsequently also diagnosed in $TbFe_2$ (*19*). Lack of saturation and strong intrinsic magnetic hardness are characteristic for both position and site disordered materials. Site disordered materials display this behavior over practically the entire homogeneous regions even at Al rich compositions where the transition metal has become nonmagnetic. A characteristic concentration, temperature, and time dependence of intrinsic hardness is observed (*8*). Details of demagnetization behavior and its temperature dependence are practically identical in site and position disordered compounds. Neutron diffraction (*19*) on $TbCo_{0.5}Al_{1.5}$ has shown that only part of the total moment freezes in a manner to be sampled as magnetic reflections. The balance is found in the disorder background scattering. Both $TbFe_2$ and $TbFe_{1.5}Al_{0.5}$ exhibit strong low-angle neutron scattering at 4.2°K indicating the presence of magnetic clusters (*20*, *21*). A preliminary Mössbauer experiment (*22*) on $ErCo_{0.5}Al_{1.5}$ appeared to indicate that moments identified as disordered (neutron disorder background scattering) are frozen in nature rather than paramagnetic.

(*b*) *$GdCo_{2-x}Al$ vs $GdAl_2$.* By contrast with the similarities in the magnetization behavior of site and position disordered materials with a high anisotropy constituent, the magnetization behavior of site and position disordered materials on the basis of Gd (spin only system) are quite different for the special

cases of $GdAl_2$ and $GdCo_{2-x}Al_x$. While $GdAl_2$ shows lack of complete saturation in laboratory fields (*22*), the site disordered materials derived from $GdAl_2$ by initial Co substitution for Al ($GdCo_{2-x}Al_x$, Co nonmagnetic) saturate readily, exhibiting saturation moments of order of the tripositive rare earth moment (*24*). This difference can be explained in terms of asperomagnetism due to local sign reversals of exchange interactions in $GdAl_2$ while materials in $GdCo_{2-x}Al_x$, at least in moderate fields, remain in the state of phase order (ferromagnetism) presumably as a result of the absence of sign reversals in exhange on Gd sites. No data of temperature dependence of magnetization in very low fields (below 100 Oe) are available. Such measurements may indicate peaking magnetizations in analogy to the situation in $La_{0.88}Gd_{0.12}Al_2$ which was taken as evidence for a spin glass type state (*25*). It will have to be ascertained, however, that this behavior is not a result of hardness effects.

It is of interest to note that the magnetic entropy up to the freezing temperature (T_F) in $GdAl_2$ (*25*) is 9.2 J/mole°K or 55% of R ln 8. By contrast for DyCu (*10*) the entropy up to T_F is only 5.8 J/mole°K or 25% of R ln 16, reflecting the strong influence of random crystal fields. (The situation would appear to be more complex than discussed so far. A random freezing of well-developed moments will only remove part of the paramagnetic entropy in analogy with the spin glass problem. In addition crystal field effects, resulting in a lowest lying doublet for Dy can act to lower magnetic entropy both above and below the magnetic ordering temperature.) Peaking magnetizations with temperature in both cases are related to the spin glass type behavior but, again, could also be described in the context of domain wall effects (intrinsic hardness).

(*c*) *$YFe_{2-x}Al_x$ vs YFe_2.* Site disorder introduced into YFe_2 by partial substitution for Fe by Al establishes a situation (*27*) in several respects rather similar to the one of YFe_2 (*28*). Both site and position disordered materials exhibit lack of complete saturation and intrinsic magnetic hardness effects. Mössbauer spectroscopy (*29*) on $YFe_{2-x}Al_x$ indicates that all magnetic moments are frozen, in large part in a scatter state, involving fluctuations in direction as well as magnitude of Fe moments (absence of remaining paramagnetism). The spectrum of hyperfine fields, as observed, indicates the dependence of Fe moments on the number of Fe nearest neighbors. $TbFe_2$ appears to exhibit scatter in moment direction of both Tb and Fe (*30*, *31*). Tb moments likely are relatively uniform in magnitude due to large exchange. Fe moments by contrast may show a spread of magnitude besides a spread of orientation.

While materials in $Y_{1-x}Fe_x$ have noncollinear structures, it should be mentioned that $Y_{1-x}Co_x$ (*32*, *33*) and $Y_{1-x}Ni_x$ (*34*) are ferromagnets. Similar behavior may then be retained in $YFe_{2-x}Al_x$ on the one hand and, say, $LaCo_{5-x}Al_x$ on the other (*15*) a contention which would yet have to be verified. The relative influence of cluster formation and of scatter order contributions (*27*), as defined above, is not clear at present for the site disordered materials.

Complex Cases (Site Disordered Systems)

In the following, some new information shall be given for further cases where no obvious predominance of exchange or anisotropy noise exists.

$Y_6Mn_{23-x}Fe_x$, $PrMn_{2-x}Al_x$ and related systems. The essential features of magnetization behavior of $Y_6(Mn_{0.75}Fe_{0.25})_{23}$ at 4.2°K are given in Fig. 4 for a Th_6Mn_{23} type material as obtained by quenching from the melt. [Details of experimental procedure and further results are to be given elsewhere (*35*).] Clearly the material shows both strong intrinsic hardness as well as an asymmetric hysteresis loop. According to what has been said above, one has to consider both exchange and anisotropy noise in this case. Anisotropy noise is predominantly responsible for the intrinsic hardness while asymmetry effects reminding of mictomagnetism appear related to exchange

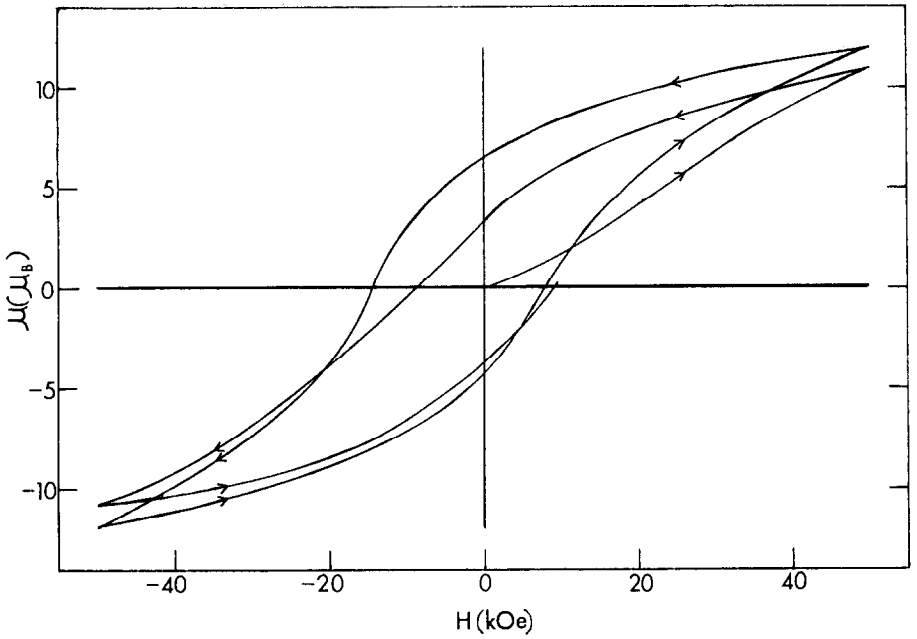

FIG. 4. Magnetization (μ_B/mole) versus applied magnetic field for $Y_6(Mn_{0.75}Fe_{0.25})_{23}$. The curve reading higher magnetization has been obtained for a specimen cooled in a field of 50 kOe while the other curve was obtained for a material cooled in a field of ~4 Oe.

noise. A schematic representation of the superposition of these two effects is given in Fig. 5. It is of interest to note in this connection that the temperature dependence of the part denoted as mictomagnetic is faster than the one characterized as being due to intrinsic hardness.

Another representative example for complex magnetic scatter types, in terms of magnetization vs temperature behavior, is shown in Fig. 6 for a material in $Pr\overline{Mn_{2-x}Al_x}$ of $MgZn_2$ type ($Pr\overline{Mn_{1.6}Al_{0.4}}$). The peaking magnetizations with temperature hint at the presence of a scatter state originating from a mixture of anisotropy noise on Pr and exchange noise as well as anisotropy noise on Mn. This is corroborated by asymmetry found in the "hysteresis" curves. It should be noted that complex magnetization data, reported in the literature on pseudobinary systems, as a rule reflect the presence of exchange or anisotropy noise or a mixture of both. The survival of phase order under such conditions is rather the exception. Indications for noise states include data reported on $Y\overline{Mn_{2-x}Al_x}$ (*36*) or $Er\overline{Mn_{2-x}Al_x}$ (*37*). In the latter case a considerable broadening of magnetic peaks characteristic of the magnetic spiral structure in analogy to $TbMn_2$ is observed in the neutron diffraction pattern. In this connection it is of interest to note that the magnetic neutron diffraction peaks in the ferromagnetic scatter compounds $Er\overline{Co_{2-x}Al_x}$ (*38*, *39*) remain sharp, although sizable uniform diffuse background scattering develops. This indicates that exchange noise in this special case yields a spread in the periodicity of the magnetic structure while anisotropy noise does not.

Discussion

General Relationships Among the Various Scatter Types

From the juxtaposition of magnetic phenomenologies in a side variety of partly

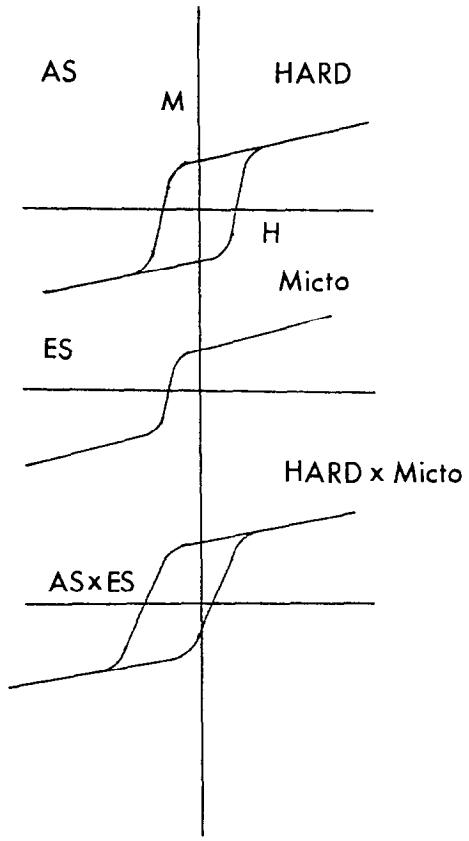

FIG. 5. Schematic representation of the magnetization–field behavior of anisotropy scatter state (AS), exchange scatter state (ES), and an admixture of both, such as found in $Y_6(Mn_{0.75}Fe_{0.25})_{23}$. AS × ES indicates that no straightforward addition of magnetization curves is involved in the mixed state.

disordered magnetic systems it appears that some unifying aspects emerge. Perhaps the best fingerprint for the relative importance of anisotropy versus exchange noise is the mechanism of demagnetization which tends to be inelastic in nature in the former and elastic in the latter case. The identification of elastic type hardness with exchange noise and inelastic hardness with anisotropy noise can be made plausible in the following way. Progress in magnetization can be made against exchange noise by rearranging an elastic medium of exchange coupled moments, a process which reminds of the metamagnetic one. On the other hand magnetization processes involving strong local anisotropy contributions result, even without the implications of domain walls, in the locking in of moments in energy minima given by local anisotropy directions. Connected with this is an avalanching rearrangement of exchange and anisotropic interactions resulting in an irreversible process. Domain wall propagation localizes this phenomenon.

The more complex cases discussed pose perhaps too difficult a problem, at present, to be treated in a way more quantitative than on a descriptive phenomenological level. Nevertheless the variety of phenomena observed is certainly intriguing. Indeed the pioneering experimental and theoretical ventures (*4*, *9*) into the phenomenologies of site disordered intermetallics have reaped a bountiful harvest.

The suggestions of cluster formation, in practically all systems discussed, make it quite possible that even poles in the schematic representation such as, say spin glasses (dilute exchange noise) and massive anisotropy noise materials, are related in the sense that clustering of moments and domain walls (at least in a broader definition) are a common denominator to both classes of materials. In this connection it should be pointed out that domain walls in massive anisotropy fluctuation materials may not be considered to have their classical character, but rather correspond to regions of disoriented spins separating magnetic clusters, a situation reminiscent of the mictomagnetic state. In general one will expect to find, say with a mictomagnet on increasing anisotropy, the nature of the glass matrix to gain gradually the character of interaction fluctuation domain walls while clusters will attain the nature of domains.

Also, many spin glasses (as mictomagnets) may not exhibit the low anisotropies contended. The question of the limits of applicability of, say, the term spin glass may be answered most straightforwardly by reserving this classification for situations where inelastic magnetic hardness (unsymmetrical hysteresis loops) is developing in materials cooled either in or without fields. No such question arises

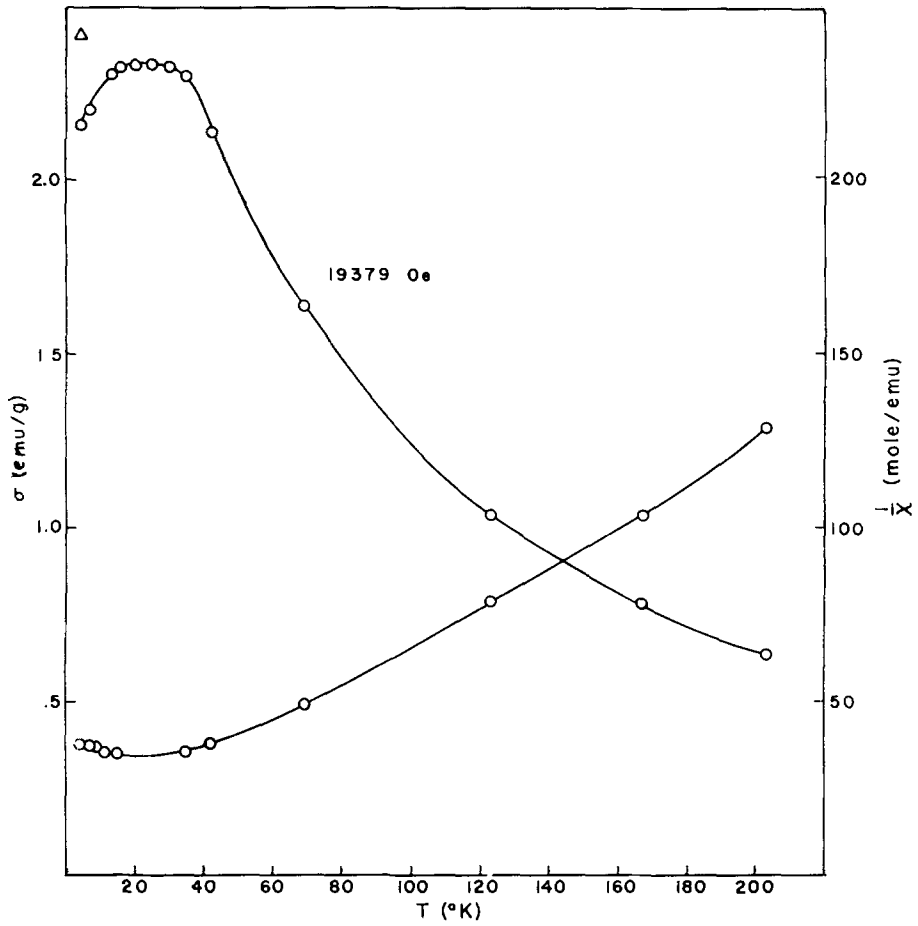

FIG. 6. Magnetization (σ) and reciprocal susceptibility ($1/\chi$) versus temperature (T) for $Pr\overline{Mn_{1.6}Al_{0.4}}$. The material was cooled to 4.1°K in the absence of a field. The triangular point was obtained after field cooling (19379 Oe) from 200 to 4.2°K. Magnetization versus field behavior was measured up to $H = 50$ kOe at 4.2°K and was found to be approximately linear.

when the terminology exchange or anisotropy noise is used. Several dilute magnetic systems show relatively high local anisotropy as they exhibit magnetization-field behavior typical of magnetically hard materials. In these cases it might be preferable to speak of dilute anisotropy noise (or anisotropy glasses?) rather than spin glasses.

The importance of magnetic concentration, as seen for the canonical exchange noise materials, has so far not been sufficiently explored for their anisotropy noise counterparts. However, an interesting point can be made in this connection concerning the behavior of $Y\overline{Co_{5-x}Ni_x}$. The onset of strong intrinsic hardness coincides here with an accelerated saturation moment decay as modeled on a local moment picture. This suggests concomitant increase in cluster formation, a situation in many ways similar to the mictomagnetic case. Therefore, moment concentration and degree of disorder appear to play a similarly important role for magnetic behavior of anisotropy noise materials. Parallels in phenomenology of exchange and anisotropy noise materials can be anticipated to become unraveled in the future.

Fascinating possibilities in the creation of

new material properties should open upon further admixing of the various more or less extreme cases. As an example one can contemplate planting a constituent with potential for high anisotropy (Sm) into, say, a massive exchange fluctuation matrix and so further maximizing magnetic hardness effects.

Applicability of Theoretical Models

Various theoretical treatments intended for one special case or another appear to be flexible enough to be applied for related variations of the common theme. As an example, theories of spin glass behavior are not limited to low concentrations and appear applicable also for situations of massive exchange noise. Likewise models of random local anisotropy, developed for amorphous materials, can be of relevance for site disordered crystalline materials. It has been shown (*40*) for amorphous random dense packed models that second-order crystal field terms, absent, say in well-defined cubic compounds, are dominant in amorphous materials. It was suggested, however, that site disorder, say in cubic compounds, likewise introduces second-order terms (*8*, *17*). A simple Hamiltonian (*41*) has been introduced for this problem, namely

$$H = -\sum_i D_i S_{iz}^2 - \sum_{ij} J(r_{ij}) S_i S_j,$$

where the first term introduces an average anisotropy (D). It would appear that, with certain modifications, a similar Hamiltonian should also be applicable to the situation of site disorder in high anisotropy intermetallics. A review of random anisotropy models is given in Ref. 42.

An interesting result from computer modeling on amorphous magnetic states, arrived at on Monte Carlo simulations of Heisenberg spins (*43*), is that there are to be no completely free paramagnetic spins. This is also of relevance to site disordered analogs such as $\overline{ErCo_{2-x}Al_x}$.

Also the principle of frustration (*44*), which has to do with the incompatibility of exchange interactions, when these interactions exhibit random local variations (exchange noise) is obviously of relevance to both position and site disordered materials. These principles have been discussed in the context of the spin glass problem (*45*) where in practice Gaussian distributions of positive and negative exchange interactions are employed (Edwards–Anderson model).

Relationship between Site and Position Disordered Materials

An interesting question pertains to the uniqueness of magnetic types in amorphous as distinct from crystalline materials. While arguments advanced in favor of this point (*10*) are not immediately convincing, given the thought experiments on compounds $\overline{RAP}$ and $\overline{RAF}$, perhaps a uniqueness by degree emerges. While a scatter state on the basis of exchange noise may be largely indistinguishable to most relevant experimental probing from one of speromagnetism (save for subtle aspects connected with the inherent difference in location of atoms) in the sense that, say neutron diffraction will detect all but noise in both cases, it is not quite clear whether a total scatter state on the basis of anisotropy noise can be established in crystalline materials. Neutron diffraction on $\overline{ErCo_{2-x}Al_x}$ has indicated (*37*, *38*) that, as a limit, so far only about half of the total moment was forced into the diffuse background scattering (accordingly the average scatter angle is of order 60°) by disordered site substitution. While this value may be different in materials with a larger ratio of anisotropy to exchange, it is of interest to note that it is also about half of the magnetization that is fairly easily saturated in $DyNi_3$ (Ni is nonmagnetic) both, according to experiment and according to general theory of high anisotropy amorphous materials (*47*). The degree of scatter reached in $\overline{ErCo_{2-x}Al_x}$ may, therefore, come close to the limit achievable. As in $DyNi_3$, the initial part of the magnetization curve for $\overline{ErCo_{2-x}Al_x}$ corresponds to alignment of domains while the high

field section, above a knee, corresponds to the closing of the scatter cones against local anisotropy. The initial part of the magnetization curve in site disordered materials will then be represented as coherent neutron scattering. Given the limit in achieving disorder neutron scattering, it would appear that there exists indeed an obvious difference to the case of say $DyNi_3$ in the sense that the site disordered material retains, at least in part, its crystallographic heritage (presence of coherent neutron scattering) while the position disordered material, even though also in a resultant scatter state, (asperomagnetic) appears amorphous to neutrons. By contrast, exchange noise is more likely to be capable of producing a total scatter state as exemplified in various cases where neutron diffraction gives evidence only of short range order (absence of coherent magnetic scattering in site disordered materials such as mictomagnets).

The most obvious difference between site and position disordered materials, from a crystallographic point of view, concerns the range of local coordination numbers in amorphous materials compared with uniform coordination numbers in crystalline materials including site disordered ones. However the condition that no two atomic sites are equivalent magnetically in an amorphous compound can be considered to be rather well simulated in its site disordered counterpart, especially when contributions beyond nearest neighbor shells are included. As with position disordered materials, where all macroscopic directions may be equivalent in the bulk, a similar situation should therefore also hold for special site disordered scatter magnets.

The chaos of an amorphous magnetic structure is then in principle also achievable in the relative harmony of a crystalline environment when individual magnetic atoms receive conflicting information from the various constellations in the sky of their nearest or next nearest neighbor shells. As in the theory of magnetic structures which involve Shubnikov groups by allowing for "colored" atoms, that is, "atoms with directions," the various magnetic constellations around individual atoms can be considered to reflect a rather large array of environments with different "color shades" resulting in the diaspora of the magnetic state.

Comparing the potential for realization of various magnetic types, it is, quite generally speaking, of interest to note that various cases of ferromagnetism (a magnetic state usually considered to be intimately connected with crystalline order) have been found in amorphous materials [e.g., $Y_{1-x}Ni_x$ (*33*), Gd_4Au (*48*). By contrast antiferromagnetism appears to be realizable only in the crystalline state (*10*).

Both site disordered and position disordered materials exhibit limitations in the development of theoretically accessible phenomenologies. A deplorable limitation with amorphous materials is the more or less fixed degree of disorder in this class of materials. This limitation is relaxed in site disordered compounds where the degree of disorder can often be systematically varied. From this point of view, site disordered materials would appear to pose the greater challenge for an unfolding of magnetic phenomenology in response to an increase in the degree of disorder. On the other hand a considerably wider variety in stoichiometries, at least around certain compositions, is available with (metastable) amorphous materials than is the case with their stable crystalline counterparts. However, in this respect one will also look with interest to metastable site disordered materials. A further degree of limitations is the predominance of second-order crystal field parameters in amorphous materials compared with the rather more catholic behavior of crystalline compounds.

Relative Temperature Dependence of Exchange and Anisotropy

It is well known that magnetic anisotropy exhibits a temperature dependence, considerably more pronounced than the one corres-

ponding to a Brillouin function (*1*). Several quantum mechanical calculations for crystal field anisotropy of 4*f* systems (*49*) attest to this experimental experience outlining the dependence of crystal field anisotropy on exchange. Position entropy materials near the ordering temperature will then exhibit small random scatter, originating from the low values of randomized local anisotropy. As the temperature decreases, local anisotropy noise is expected to increase. This, in turn, will have a deleterious effect on the magnitude of exchange interaction as scatter in moment direction decreases the average projection of interacting moments. It is, therefore, plausible to assume that position entropy materials might exist where this decrease in exchange interaction would lead to a reexiting of magnetic order as the temperature is decreased.

An example for this intriguing possibility may, in fact, already have been identified with the mictomagnet $Fe_{0.70}Al_{0.30}$, which does show a certain inelastic part to its magnetic hardness. As outlined earlier the unusual behavior of this compound has been explained by a different temperature dependence for ferro- and antiferromagnetic exchange interactions, without taking into account anisotropy. Even if in this case the actual mechanism for reexiting magnetic order would be clearly shown to be indeed only based on temperature dependence of exchange noise, one can plausibly expect that cases will be identified in the future where a representative of the model world (predicted on the basis of anisotropy noise as above) will enter the real world.

Crystal Chemical Considerations

The cacophony of magnetic scatter states, observed as a rule at relatively low temperatures, obviously is a gratuity of high entropy crystallographic states which reflect equilibrium or nonequilibrium situations characteristic of high temperature (large influence of the entropy), subsequently frozen in by activation energy barriers at lower temperatures. For an artful unfolding of scatter state phenomenology one will therefore have to master the effects of entropy upon crystal structure. For this, crystal chemical knowledge and intuition are of essence.

The field of magnetism is site disordered materials remains, therefore, a challenge to the imagination of the structural chemist to produce a spectrum of various degrees of disorder. The design of interesting new model compounds (especially ones with high anisotropy) will hinge on the judicial choice of such parameters as electrochemical, size, and electronegativity factors. Large homogeneous regions of site disordered structures have to be specially designed as stable or metastable compound series. Variation in heat treatment and mode of preparation will allow for various degrees of disorder in complex systems (e.g., tendencies towards superstructure formation are a common low-temperature answer to the question of free energy minimization in partly disordered structural modifications). Recrystallization of amorphous materials into stable or metastable low-temperature modifications will be of interest. Also hydrides of intermetallic compounds and their desorbed partly amorphous products have opened one more variety in this respect (*50*).

In general, variations in the mode of site occupancy of the various atomic species allow one to follow the relevant magnetic phenomenology as it develops out of a more orderly crystallographic situation. Accordingly, phase order is expected to change into various modes of homogeneous or inhomogeneous (clusters) scatter states. In order to observe the influence of anisotropy, a structure with preferred axis aids in the interpretation of experimental data. Candidates fulfilling these criteria are usually relatively densely packed phases such as the hexagonal Laves phases and their derivatives (Haucke phase related structures) or the AlB_2 type. Other phases allowing for large homogeneous regions are the cubic CsCl or Th_6Mn_{23} type. Complex intermetallic structures (such as σ, R, P phases

and others) exhibit a considerable variety in the mode of site occupancy. Also, the role of magnetic concentration can be probed by selective dilution of moment carrying atoms on crystallographic sites.

In view of the impressive progress of the last few years on magnetic properties of amorphous materials, largely a gift to fundamental science of technological interest in information storage, it can be hoped that progress in related crystalline materials will also participate in an added impetus.

Considerations Concerning Magnetic Entropy in NOISE States

Perhaps contrary to intuition not all noise states can be expected to result in unusually high values of entropy below freezing temperatures or on approaching absolute zero temperature. In fact anisotropy and exchange noise materials, in extreme cases, would appear to present somewhat different thermal characteristics in this respect. In the spin glass problem a high density of closely spaced states in energy is responsible for broad low-temperature specific heats as well as the freezing (say in a magnetic field) into metastable states differing from the ground state by small increments in energy. In fact, it is very unlikely that the actual ground state (with entropy approaching zero) would be reached in a particular cooling experiment close to 0°K. (Finite entropy at 0°K is a result of a nonequilibrium state and does therefore not violate the third law of thermodynamics.) Neglecting the concomitant exchange noise in site disordered materials of high anisotropy, one expects the system of magnetic moments to drop, at 0°K, into one well-defined scatter state (zero entropy), dictated by the given local anisotropy axes. Only in second approximation, that is to say on inclusion of exchange noise, will one anticipate metastable states in analogy to the spin glass problem. The actual magnitude of specific heat effects has yet to be firmly established for representative examples of more of less extreme cases. In this respect it will also be of interest to investigate the potential of noise systems for applications in adiabatic magnetic cooling. Of particular promise is the cooling potential of anisotropy noise materials around the ordering temperature. It should be mentioned that a technological interest exists for adiabatic magnetic cooling in the temperature range for 20–300°K utilizing powerful new superconducting field capabilities.

Scatter States in Systems other than Metallic

It should be noted that scatter state phenomena are not limited to metallic systems. Disordered canting of moments has long been known for several oxidic phases such as $\overline{Zn_{1-x}Co_x}Fe_2O_4$ (*51*, *52*) as a result of competing exchange paths. A wealth of unusual domain wall effects and concomitant magnetic hardness in response to extended defects and site disorder as found in oxide phases, is known (*52*). The general phenomenology, however, is somewhat different as befits a different electronic makeup.

Future Areas of Interest and Probing Techniques

It is clear that an abundance of questions await clearer answers. Among them is the nature of the phase transition in all partly disordered materials discussed, including the question of magnetic phase segregations (broad or 'misorder" transitions), the changing character of domains and domain walls (or cluster and glass), and, quite generally, the limits in achieving special effects. From a practical point of view the use of scatter types is of interest, say for magnetic cooling, permanent magnets, as well as applications in information storage.

Various experimental techniques beyond measurements of bulk magnetization appear to be promising to help further elucidate magnetic states in site and position disordered compounds. The average magnetic moment is accessible via paramagnetic susceptibility or the average hyperfine field (Mössbauer

spectroscopy) and information from neutron diffraction. Mössbauer transition probabilities can yield the angle between moment alignment and the direction of radiation employed in the experiment. Small-angle neutron diffraction and, in some cases, the initial susceptibility give the size of correlated clusters. Specific heat can yield information pertaining to the mode of loss of entropy as well as thermal exitation processes and the nature of the phase transition.

Conclusion

A relatively new area of materials research, namely, one in which the harmony of well-developed magnetic types is partly destroyed through disordered substitutions (a tuning out of phase), has been outlined. It is shown that the diverse field of site disordered magnetic materials can be viewed in unifying manner by taking account of the relative importance of anisotropy, exchange, and magnetic moment concentration, resulting in an enlarged conceptual approach. This sense for the relationship among chaotic magnetic states helps sharpen one's intuition for new adventures in the charting of the phenomenology of relatively dense intermetallic compounds of either high (HARD) or low (GLAS) anisotropy. Within this frame the novel and complex behavior of such materials as $Y_6Mn_{23-x}Fe_x$ can be better understood. Also a rather close relationship between the scatter states of site disordered crystalline materials and their amorphous (position disordered) counterparts is indicated. In fact, the vigorously expanding research on amorphous magnetic materials is now giving impulse to related research in crystalline compounds.

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