New Model for Amorphous Magnetism*

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(Received 9 April 1973)

We propose a new model for magnetism in an amorphous material. The model is particularly appropriate for rare-earth compounds such as $\mathrm{TbF_2}$. It is a Heisenberg model in which each ionic spin is subjected to a local anisotropy field of random orientation. We discuss the magnetic properties of two simple ionic configurations, and show that the model is in qualitative agreement with the experimental data.

In a recent publication Rhyne, Pickart, and Alperin[1] examined the properties of a ferromagnetic $\mathrm{TbFe_2}$ amorphous alloy. They showed that both the Curie temperature T_c and the spontaneous magnetization $M(T)$ of this alloy are substantially lower than those of the corresponding crystalline Laves phase alloy. Their work is particularly significant because they are able to compare directly the properties of crystalline and amorphous phases of the same alloy, in contrast to previous experimental work where the crystalline phases are not accessible to experiment for materials of the same percentage composition.[2]

Previous theoretical work, based on Gubanov's ideas,[3] uses a phenomenological fluctuation of the Heisenberg coupling constant $\mathcal{J}$. However, in order to describe a reduction in T_c from 710 to 388°K or a reduction in $M(0)$ from 4.7 to 2.8 $\mu\mathrm{B}$/atom, as in the case for $\mathrm{TbFe_2}$, it would be necessary to choose very large values of the fluctuation in $\mathcal{J}$, fluctuations of the same order as $\mathcal{J}$ itself. Although this might be possible in insulating systems where the coupling may be very sensitive to the overlap of wave functions, it is less probable in a rare-earth transition-metal alloy where the coupling is likely to be of Ruderman-Kittel-Kasuya-Yosida type.

We propose a model for magnetism in amorphous rare-earth transition-metal alloys which allows a direct comparison with the corresponding crystalline phases. It is a Heisenberg model in which each spin has the same magnitude but is subjected to a local anisotropy field of random orientation. We expect that the anisotropy fields will be important in amorphous $\mathrm{TbFe_2}$ and similar alloys, because the corresponding crystalline materials have huge magnetocrystalline anisotropy.[4] We assume that the randomness of the anisotropy field is the most important characteristic of the amorphous state, and that it is a result of the topological disorder of a random close-packing structure[5] which is characteristic of amorphous metals.[6] In principle, therefore, the local anisotropy can be calculated from a detailed structural model.

The Hamiltonian for the present model is written

$$H=\sum_i V_i - \tfrac{1}{2}\mathcal{J}\sum_{i\delta}\vec{J}(i)\cdot\vec{J}(i+\delta), \tag{1}$$

where V_i is a local single-ion anisotropy field at the site i,[7] $J(i)$ is the angular-momentum operator for the magnetic ion on site i, and $\mathcal{J}$ is the nearest-neighbor coupling constant of the Heisenberg model. We consider two of the simplest configurations for the magnetic ions: (a) ions with $J=1$, 2, or 3 in a uniaxial anisotropy field, and (b) ions with two low-lying singlet states, such as Pr^{3+} in a locally hexagonal field.[8] Although we believe that case (a) describes the basic properties of amorphous materials such as $\mathrm{TbFe_2}$, we intend at this time only to establish guide lines for a future application to this material. Case (b) enables us to make predictions about the amorphous phase of a different class of materials, those which seem likely to possess induced moments. After development of the Hamiltonian (1) for the two simple ionic configurations we write down expressions for T_c and $M(T)$ in the two cases. Finally, we discuss ways in which the model can be extended and refined to bring it closer to describing $\mathrm{TbFe_2}$, and make some suggestions for further experiments.

Case (a), uniaxial anisotropy with $J=1$.—In this case $V_i=-D[J_{z_i}(i)]^2$, where $\hat{z}_i$ refers to the local direction of anisotropy at site i. We analyze the Hamiltonian (1) in the Hartree-Fock approximation (HFA), with a molecular field λ, proportional to the bulk magnetization $M(T)$, in a unique $\hat{z}$ direction. Equation (1) then becomes

$$H_F=-\sum_i D[J_{z_i}(i)]^2-\lambda\sum_i J_z(i), \tag{2}$$

where $\lambda=v\mathcal{J}\langle J_z(i)\rangle$. Here v is the number of nearest neighbors, or the coordination number of the structure, which is assumed to be the same as in the crystalline phase.

The average $\langle J_z(i)\rangle$ is both a thermal average, as in standard mean-field theory, and also an average over the random directions $\hat{z}_i$ of the local anisotropy axis. The latter average can be performed using various probability distributions $P(\mu)d\mu$ for the directions $\hat{z}_i$, where $\mu = \hat{z}\cdot\hat{z}_i$. The crystalline case corresponds to $P(\mu)=\delta(1-\mu)$. In the amorphous case we consider at present only a completely isotropic distribution, with $P(\mu)=1$, $0\leq\mu\leq1$, postponing discussion of more realistic distribution based on random close packing[5] until a later publication.

An exact expression for T_c is obtained by linearizing the self-consistent equations for $M(T)$ near $M(T)=0$. For $J=1$ the result is

$$KT_c/v\mathcal{J} = T_0 = 2\int_0^1 P(\mu)\{\mu^2\exp(\alpha/T_0) + (T_0/\alpha)(1-\mu^2)[\exp(\alpha/T_0)-1]\}[2\exp(\alpha/T_0)+1]^{-1}d\mu, \tag{3}$$

where $\alpha = D/v\mathcal{J}$. In Fig. 1 we plot the ratio of amorphous to crystalline critical temperatures as functions of α for $J=1$, 2, and 3. The expressions corresponding to (3) for $J=2$ and 3 are tediously long and will not be quoted in this paper, but we see that the curves for $J=2$ and 3 essentially coincide. However, the behavior of the spontaneous magnetization at zero temperature, $M(0)$, as a function of α is much more strongly dependent on the size of J. The appropriate functions are also plotted in Fig. 1, and we see that when $J=3$ the ratio of amorphous to crystalline $M(0)$ is already comparable to the ratio of critical temperature as is experimentally found in TbFe$_2$.[1]

In Fig. 2 we show the temperature dependence of the magnetization $M(T)$ for $J=1$ and several values of α. For comparison we also plot $M(T)$ for a crystalline ferromagnet with $J=1$ and $\alpha=0.4$. Curve E is the crystalline function for $\alpha=1.6$ scaled down to illustrate the point that $M(T)$ in the amorphous material decreases faster than $M(T)$ in the crystalline case. This seems to be a characteristic feature of all amorphous ferromagnets so far studied.[2]

Case (b), the singlet-singlet model.—In this case the level structure of the magnetic ions due to the crystalline field is composed of two low-lying singlet states, separated by energy 2Δ, and $2J-1$ excited states of higher energy. When $\mathcal{J}$ is of the order of 2Δ, the low-lying collective excitations in the crystalline material are no longer spin waves, but magnetic excitons, and the character of the ferromagnetic state changes. In such circumstances the singlet-singlet model represents an "induced-moment system," which has a soft mode in the exciton spectrum at the Curie temperature T_c.[8]

The Hamiltonian for a crystalline singlet-singlet induced-moment system can be derived from (1) by introducing pseudospin operators for spin $\frac{1}{2}$.[8] In an amorphous material the same operators can be introduced, but as in case (a), we take explicit account of the random orientation of the local axes of anisotropy $\hat{z}_i$. The Hamiltonian

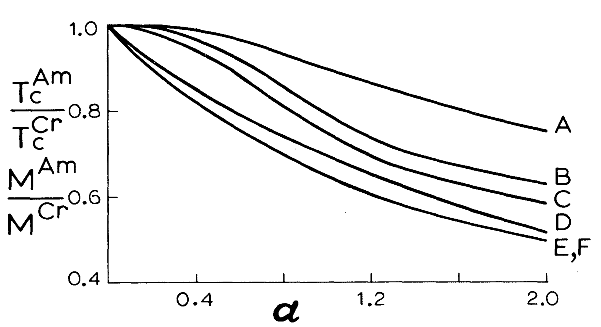

FIG. 1. Ratio of zero-temperature amorphous and crystalline magnetizations, M^{Am}/M^{Cr}, as a function of $\alpha = D/v\mathcal{J}$. Curve A, $J=1$; B, $J=2$; C, $J=3$. Also plotted is the ratio of critical temperatures T_c^{Am}/T_c^{Cr}. Curve D, $J=1$; E, $J=2$; F, $J=3$.

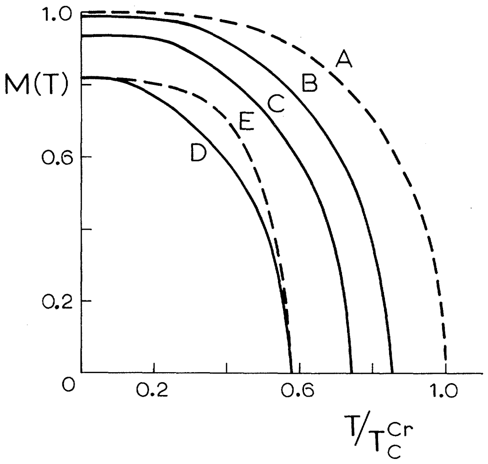

FIG. 2. $M(T)$ for several values of $\alpha = D/v\mathcal{J}$ for $J=1$. Dashed curves, crystalline magnetizations; T_c^{Cr} is the crystalline critical temperature for the particular α under consideration. Curves A, B, $\alpha=0.4$; C, $\alpha=0.8$; D, $\alpha=1.6$; E, scaled crystalline magnetization for $\alpha=1.6$.

becomes

$$H_s = \Delta \sum_i \sigma_{x_i}(i) - \tfrac{1}{2}\alpha^2 \mathcal{J} \sum_{i,\delta} \mu_{i,i+\delta}\sigma_{z_i}(i)\sigma_{z_{i+\delta}}(i+\delta). \quad (4)$$

Here $\alpha = \langle 1i | J_{z_i}(i) | 0i \rangle$, where $|0i\rangle$ and $|1i\rangle$ are the state vectors for the two singlet states at site i, and $\sigma_{z_i}(i)$ and $\sigma_{x_i}(i)$ are Pauli matrices in pseudospin space, relative to the local axes at site i. The angle between local axes on nearest-neighbor sites enters through $\mu_{ij} = \hat{z}_i \cdot \hat{z}_j$.

The Hamiltonian (4) can be treated in the HFA, as before, by introducing a molecular field λ in the $\hat{z}$ direction where $\lambda = v\mathcal{J}\langle J_z(i)\rangle = v\mathcal{J}\alpha\langle\sigma_z(i)\rangle = v\mathcal{J}M(T)/\mu_B$. The self-consistent equation for the bulk magnetization then becomes

$$1 = v\mathcal{J}\alpha \int_0^1 \frac{\mu^2 d\mu\, P(\mu)}{(\Delta^2+\lambda^2)^{1/2}} \tanh\left(\frac{(\Delta^2+\lambda^2)^{1/2}}{KT}\right), \quad (5)$$

where $\mu = \hat{z}\cdot\hat{z}_i$, and the resultant expression for T_c is $KT_c = \Delta/\tanh^{-1}(3\Delta/v\mathcal{J}\alpha)$. In contrast to case (a), the shape of the $M(T)$ curves is essentially the same in amorphous and crystalline materials, although $M(0)$ and T_c for the amorphous phase are even more sharply reduced than in case (a).

However, experiments on amorphous induced-moment systems would be useful from another point of view. Because of the relative simplicity of these systems it is possible to derive equations for their collective excitations (magnetic excitons) whereas the corresponding equations for the spin waves in case (a) are much more complicated. For example, it can be shown that in the random-phase approximation the Hamiltonian gives a soft mode at T_c for any distribution $\rho(\mu)$ of the local axes. Appropriate experiments on amorphous induced-moment systems would therefore be a very direct test of our model. A detailed analysis of this point will be presented in a later publication.

We have presented two forms of a new model for magnetism in amorphous rare-earth transition-metal alloys, and have performed some simple illustrative calculations. To make case (a) more directly applicable to $TbFe_2$ we would have to introduce different mean fields on the Tb and Fe sites and consider more realistic values of J ($J=6$ for Tb and $J=2$ for Fe). These complications will be dealt with in a later publication.

It would be convenient to compare our simple calculations with data on ferromagnetic (as opposed to ferrimagnetic) amorphous alloys such as $TbRh_2$; unfortunately such data do not exist. However, we can predict that when further rare-earth transition-metal alloys can be made amorphous, those containing S-state or nonmagnetic rare-earth atoms, such as Gd, Y, or Lu, will not exhibit a significant decrease in magnetization or Curie temperature compared with their crystalline counterparts. This is certainly consistent with existing data on amorphous[9] and crystalline[10] Gd-Fe alloys.

A suitable choice for further experiments would be YFe_2. Those alloys which contain other rare earths ought to exhibit significant decreases in both $M(T)$ and T_c when they are made amorphous—good candidates might be $TbNi_2$, $TbRh_2$, or $TbIr_2$.

It would also be interesting to look for compounds which are induced-moment systems in the amorphous state. Crystalline $PrIr_2$, $PrRu_2$, and $PrOs_2$ are probably examples of induced-moment systems, and would likely have the same character in the amorphous state. It is also possible that certain compounds which are ordinary ferromagnets in the crystalline state may become induced-moment systems when made amorphous.

We wish to thank R. J. Birgeneau, R. W. Cochrane, and J. O. Ström-Olsen for stimulating discussions.

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*Work supported in part by the National Research Council of Canada.

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