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Research articles An extension of the Handrich model for ferrimagnetic amorphous

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ABSTRACT

In this paper we propose an extension to the Handrich model considering the fluctuations in exchange energies between ions located in different magnetic sublattices. Thus, each magnetic sublattice has an amorphization parameter that changes with temperature. A systematic study of the magnetization as a function of temperature is performed, which allows to observe significant differences when such fluctuations are not considered.

#### 1. Introduction

Magnetic amorphous alloys are very interesting systems. Usually they are part of a class of soft magnetic materials with attractive magnetic and mechanical properties. We can highlight some of them: large critical shear stress [1], high corrosion resistance [2], low eddy current loss resulting from a large electrical resistivity, negligible magnetic hysteresis (desirable for minimizing longitudinal thermal conduction) [3], and others. There are many magnetic applications for amorphous alloys, from the use of small micro-sensors such as large electrical power devices [4]. In addition, with a number of new technologies emerging, the nature and the scope of the existing applications are changing rapidly. One of these promising technologies is magnetic refrigeration. Theoretical and experimental investigations on the application of amorphous alloys in this type of refrigeration has grown considerably in recent years.

Amorphous systems have collinear and non-collinear magnetic structures. In the case of collinear, is observed the ferromagnetic configuration for amorphous alloys for many materials as, e.g., Gd<sub>1-x</sub>M alloys with M = Ag, Au, Al or Cu and transition-metal-metalloid alloys like  $TM_{1-r}M_r$  with TM = Fe, Co or Ni and M = B, P or C [5]. The collinear ferrimagnetic is observed, such as, for  $Gd_{1-x}T_x$  alloys with T = Fe, Co or Ni [6] and thus represents a two sublattices configuration containing two distinguishable groups of atoms.

Many amorphous alloys have the magnetic matrices and their moments very similar to the related crystalline compounds, suggesting similar local environments. Indeed, Corb and coworkers [7] presented a quite successful model of local order and magnetism in amorphous alloys that describes well the moment variation with metalloid content in several alloys based on Co, Ni, and Fe by supposing a local coordination in the amorphous phase like that in a related crystalline compound.

Amorphous materials have no long-range order (periodicity) like crystals. By the way, amorphous ferromagnets have the magnetic moments oriented in a specific direction, but the spatial distribution of these is not regular. In order to describe the behavior of ferromagnets with this irregularity, Handrich and Kaneyoshi proposed a simple model whithin a mean field theory which a localized spin interacts with your neighbors, but fluctuations in exchange energies are present [8]. These fluctuations are characterized by a parameter ( $\delta$ ) which modifies the Brillouin function turning possible to fit experimental data with good agreement. Many works were reported using this model to describe some amorphous ferromagnets properties like magnetic, caloric and so on [9-11]. However, in some cases, the model fail to describe the magnetization behavior. Therefore, Bhatnagar et. al. proposed an empiric expression for  $\delta$  in order to fit the experimental data considering the temperature dependence [12]. The new parameter was  $\delta = \delta_0 (1-t^2)$  with  $t = T/T_C$  and  $T_C$  the Curie temperature. Later, Gallagher and coworkers proposed an extension to the Handrich's model introducing an asymmetrical distribution of the exchange interactions based on empirical knowledge of the Bethe-Slater curve [13]. The fluctuations suggested by Gallagher and coworkers was defined like  $\delta^{\pm} = \delta_0^{\pm} + \delta_1^{\pm} (1-t^2)$  to consider the influence of temperature.

In this paper, we suggest a theoretical model to reach the dependence of temperature on exchange energies fluctuations of ferrimagnetic systems with two magnetic sublattices based on a mean field theory. An expression for delta is obtained allowing a systematic study about the behavior of the magnetization as well as  $\delta$  for differents values of molecular fields.

### 2. Theory

We start this section writing the local field on a *i*-th ion as

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$$h_i^{a(b)} = H + \sum_j J_{ij} \langle S_j^z \rangle_T^{a(b)} + \sum_k J_{ik} \langle S_k^z \rangle_T^{b(a)}$$
(1)

The first term is the external magnetic field whereas the second and third terms are the exchange interaction between spins  $S_j$  on common sublattice (*a*) and exchange between  $S_k$  spins on different sublatice (*b*), respectively. Due to structural disorder, the exchange energies  $J_{mn}$  can be replaced by

$$J_{mn} = \langle J_{mn} \rangle + \Delta J_{mn} \tag{2}$$

The symbol  $\langle ... \rangle$  represents the random average over all possible configurations and  $\Delta J_{mn}$  characterizes the fluctuations of exchange energies due to non-uniforme distribution of the ions. In addition, we define the quantities according to [8]

$$X_i^{a(b)} = h_i^{a(b)} - W_0^{a(b)}$$
(3)

$$W_0^{a(b)} = \langle h_i^{a(b)} \rangle \tag{4}$$

Considering that the expectation value  $\langle S_i^z \rangle_T$  can be replaced by the mean value  $S^{\gamma}\sigma^{\gamma}$  (with  $\sigma^{\gamma} = \frac{M^{\gamma}(T)}{M^{\gamma}(0)}$ ) – i. e. it's independent of random average, the Eq. (1) can be replaced on Eqs. (3) and (4), providing (for H = 0)

$$X_i^a = S^a \sigma^a \sum_j \Delta J_{ij} + S^b \sigma^b \sum_k \Delta J_{ik}$$
<sup>(5)</sup>

and

$$W_0^a = S^a \sigma^a \sum_j \langle J_{ij} \rangle + S^b \sigma^b \sum_k \langle J_{ik} \rangle$$
(6)

where the index b was dropped and we'll consider only the sublattice a without loss of generality. The reduced magnetization can be reached by performing the random average of all magnetic moments

$$\sigma^{a} = \langle \sigma_{i}^{a} \rangle = \langle B_{S}(\beta S^{a}(W_{0}^{a} + X_{i}^{a})) \rangle$$
(7)

 $B_S$  is the Brillouin function,  $\beta = \frac{1}{k_B T}$  and we take, for convenience,  $g\mu_B = 1$  in the argument ( $k_B$ , g and  $\mu_B$  are, respectively, Boltzmann constant, Landè factor and Bohr magneton). Applying the Handrich-Kaneyoshi approximation [8]

$$\sigma^{a} = \frac{1}{2} \sum_{l=1,-1} B_{S}(\beta S^{a}(W_{0}^{a} + l\Delta^{a}))$$
(8)

where

$$(\Delta^a)^2 = \langle (X_i^a)^2 \rangle = (W_0^a \delta^a)^2 \tag{9}$$

and

$$(\delta^{a})^{2} = \frac{\left\langle \left( S^{a} \sigma^{a} \sum_{j} \Delta J_{ij} + S^{b} \sigma^{b} \sum_{k} \Delta J_{ik} \right)^{2} \right\rangle}{\left( S^{a} \sigma^{a} \sum_{j} \langle J_{ij} \rangle + S^{b} \sigma^{b} \sum_{k} \langle J_{ik} \rangle \right)^{2}}$$
(10)

Considering the fluctuations  $\Delta J_{ij}$  and  $\Delta J_{ik}$  independents, i.e.,  $\langle \Delta I_{ij} \Delta J_{ik} \rangle = \langle \Delta J_{ij} \rangle \langle \Delta J_{ik} \rangle$ , we obtain ( $\langle \Delta J_{nm} \rangle = 0$ )

$$(\delta^a)^2 = \frac{\left\langle (S^a \sigma^a \sum_j \Delta J_{ij})^2 \right\rangle + \left\langle (S^b \sigma^b \sum_k \Delta J_{ik})^2 \right\rangle}{\left( S^a \sigma^a \sum_j \left\langle J_{ij} \right\rangle + S^b \sigma^b \sum_k \left\langle J_{ik} \right\rangle \right)^2}$$
(11)

The equation above shows that the amorphization parameter  $\delta^a$  is temperature dependent through the magnetizations  $\sigma^a$  and  $\sigma^b$ . With a bit of algebra, we can rewrite the Eq. (11) as

$$(\delta^{a})^{2} = \frac{\frac{\left\langle \left(\Sigma_{j} \Delta J_{ij}\right)^{2} \right\rangle}{\left\langle \left(\Sigma_{j} J_{ij}\right)^{2} \right\rangle} + \frac{\left\langle \left(\Sigma_{k} \Delta J_{ik}\right)^{2} \right\rangle}{\left\langle \left(\Sigma_{j} J_{ij}\right)^{2} \right\rangle} \left(\frac{s^{b_{\sigma}b}}{s^{a_{\sigma}a}}\right)^{2}}{\left(1 + \frac{s^{b_{\sigma}b}}{s^{a_{\sigma}a}} \frac{\Sigma_{k} \langle J_{ik} \rangle}{\Sigma_{j} \langle J_{ij} \rangle}\right)^{2}}$$
(12)

$$(\delta^{a})^{2} = \frac{(\delta^{a}_{0})^{2} + (\delta^{a}_{1})^{2} \left(\frac{s^{b}\sigma^{b}}{s^{a}\sigma^{a}}\right)^{2}}{\left(1 + \frac{s^{b}\sigma^{b}}{s^{a}\sigma^{a}}\frac{\lambda^{ab}}{\lambda^{aa}}\right)^{2}}$$
(13)

Finally, one can note that the amorphization parameter obtained for each lattice is characterized by both energies fluctuations between ions localized in a common lattice ( $\delta_0$ ) and different lattice ( $\delta_1$ ). Furthermore, the parameter is temperature-dependent through the magnetization of each lattice (it is important to highlight that if the second lattice does not exist we recovery the parameter defined by Handrich). Combining Eqs. (8) and (9), we obtain

$$\sigma^{a} = \frac{1}{2} \sum_{l=1,-1} B_{S}[x_{a}(1+l\delta^{a}(T))]; \qquad x_{a} = \beta S^{a} W_{0}^{a}$$
(14)

#### 3. Application

Now we investigate the magnetization of ferrimagnetic amorphous systems taking into account the parameter  $\delta(T)$ . From Eq. (14) the total magnetization can be written as

$$M(T) = \frac{1}{2} \sum_{\gamma=a,b} \sum_{l=1,-1} \mu_B (NgJ)^{\gamma} B_J [x_{\gamma}(1+l\delta^{\gamma}(T))]$$
(15)

where for each lattice  $\gamma$ , *J* is the total angular momentum, *N* the number of magnetic ions, *g* the Landè factor and  $x = g\mu_B J\beta \langle h \rangle$ .

For convenience, the system consists of two lattices with identical ions, i.e.,  $J_a = J_b = \frac{5}{2}$  and  $g_a = g_b = 2$ . The ferrimagnetic state is obtained by considering that a lattice has  $p = \frac{2}{3}$  of the magnetic ions while the other has  $(1-p) = \frac{1}{3}$ . Using the mean field approximation, field  $\langle h^{\gamma} \rangle$  can be written as  $h^{a(b)} = \eta^{aa(bb)}M^{a(b)} + \eta^{ab(ba)}M^{b(a)}$ . Here, the  $\eta$ 's parameters represent the renormalized exchange interactions as defined in [9]. Fig. 1 shows the magnetization profiles for differents  $\eta$ . The choice of the  $\eta$  parameters defines the values for the compensation and magnetic ordering temperature. It is based on the experimental values of these quantities usually observed in ferrimagnetic amorphous alloys [14]. In all situations, we consider  $\eta^{ab} = \eta^{ba} = -100 \frac{T^2}{me^{\gamma}}$ .

Usually, in amorphous ferromagnets, the magnetization vs. temperature curve exhibits a pronounced "depression" when compared to



**Fig. 1.** Magnetization profiles for different combinations of renormalized exchange parameters: solid  $\left(\eta^{aa} = 300\frac{T^2}{mev} \text{ and } \eta^{bb} = 0.001\frac{T^2}{mev}\right)$ ; dashed  $\left(\eta^{aa} = 30\frac{T^2}{mev} \text{ and } \eta^{bb} = 150\frac{T^2}{mev}\right)$ ; dotted  $\left(\eta^{aa} = 10\frac{T^2}{mev} \text{ and } \eta^{bb} = 300\frac{T^2}{mev}\right)$ ; dashed dotted  $\left(\eta^{aa} = 0.001\frac{T^2}{mev} \text{ and } \eta^{bb} = 400\frac{T^2}{mev}\right)$ . In all cases,  $\eta^{ab} = \eta^{ba} = -100\frac{T^2}{mev}$ .

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