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Spin glass behavior of intermetallic compounds after mechanical milling

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Abstract

We investigated the magnetic properties of amorphous Co_2Ge and atomically disordered crystalline GdAl_2 obtained by mechanical milling. Both ac and dc magnetic susceptibility measurements show a single transition below 41 K (T_f) for Co_2Ge and 65 K for GdAl_2 from the paramagnetic to the spin glass state. Thermal hysteresis of the magnetization below T_f , characteristic of a spin glass, is found.

Recently ball milling has attracted much interest since it can be used as a non-equilibrium processing tool. It has been realized that ball milling of crystalline intermetallic compounds (mechanical milling) or of crystalline elemental powder mixtures (mechanical alloying) can be used to synthesize various metastable structures. Examples of these structures are amorphous materials [1], crystalline phases existing in the phase diagram at high temperatures [2], extended solid solutions [3] and solid solutions of immiscible systems [4], nanocrystalline materials [5] and quasicrystalline materials [6]. Since the ball milled materials are far from equilibrium, their properties can be quite different from the equilibrium stable phases. The materials obtained by ball milling can even have unusual and interesting properties because of the special configuration of atomic arrangement. This was indeed observed in e.g. Co_2Ge , where the amorphous state takes a spin glass state at low temperatures whereas crystalline Co_2Ge is a ferromagnet [7,8]. Amorphous Co_2Ge cannot be obtained by traditional melt-spinning. Recently we prepared atomically disordered crystalline GdAl_2 (which we call mm- GdAl_2) by ball milling of the ordered crystalline compound. The magnetic properties of this metastable phase differ markedly from the originally ordered compound. In this paper, we summarize our results on both alloy systems.

For the experimental procedures the reader is referred to Refs. [7,8].

We show first the results for amorphous Co_2Ge in Fig. 1 where the ac magnetic susceptibility (χ_{ac}) vs. temperature is plotted. At low fields (< 2 Oe) a sharp asymmetric cusp occurs at $T_f = 41$ K which is defined as the spin glass transition temperature. When the field is raised the cusp

disappears and a broad maximum is found as illustrated for an applied field of 30 Oe in Fig. 1. At still higher fields (90 Oe) the maximum in χ_{ac} becomes a plateau. These results are further confirmed by static magnetic susceptibility measurements and the observation of thermal hysteresis in low magnetic fields below T_f [7].

Fig. 2 shows the M/B_a vs. T curves of mm- GdAl_2 in different external dc fields. After cooling the sample down to 4.2 K in zero field (ZFC), measurements were successively made during heating. The M/B_a value gives the initial dc susceptibility, χ_{dc} , when the applied fields are small so that magnetization is proportional to susceptibility. Applying a field of 40 Oe a sharp maximum is observed. This maximum becomes a rounded transition and shifts to lower temperature with increasing field. The value of M/B_a decreases with increasing field. A similar

Fig. 1. Temperature dependence of the ac susceptibility χ_{ac} of amorphous Co_2Ge at different amplitudes of the external ac field. A frequency of 109 Hz was used.

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Fig. 2. M/B_0 vs. T curves of atomically disordered crystalline $GdAl_2$ in various dc magnetic fields after zero-field cooling.

behavior is also observed in the ac magnetic susceptibility vs. temperature curves [9].

These phenomena are very similar to those reported for the prototype spin glasses e.g. FeAu and MnCu [10,11]. Thus the magnetic properties of amorphous Co_2Ge and disordered $GdAl_2$ are characteristic of spin glasses. The temperature (T) vs. magnetic field (B) phase diagrams can be established in terms of the change of the peak temperature with external field. As an example, the T vs. B phase diagram for mm- $GdAl_2$ is given in Fig. 3. It is obvious that the decrease of T_f is faster in the low field region and is slower in fields higher than about 200 Oe. This emphasizes the peculiar sensitivity of T_f to low fields in mm- $GdAl_2$.

It is known that both the originally ordered Co_2Ge and $GdAl_2$ are ferromagnets with Curie temperatures of about 47 K [8] and 175 K [9], respectively. After milling both materials become spin glasses as a consequence of disorder on an atomic level. Thus the magnetic properties of the metastable phases obtained by ball milling are markedly different from the stable forms. The spin glass state which represents a metastable magnetic configuration of frozen-in

Fig. 3. The T_f vs. B_{ext} phase diagram for mm- $GdAl_2$ (SG: spin glass region; PM: paramagnetic region); (O) peak temperatures in ac susceptibility, (●) data from dc magnetization measurements.

disorder of the spin system adds another degree of disorder to a system which is already a disordered atomic array. The spin glass behavior of Co_2Ge is observed only after the disappearance of the periodic lattice. This is not the case for $GdAl_2$ where the periodic lattice remains. This difference is most probably due to the fact that the fraction of magnetic element in Co_2Ge is much higher than in $GdAl_2$. Moreover, in Co_2Ge the magnetic contribution is from 3d-Co atoms whereas in $GdAl_2$ it is from the 4f-Gd rare-earth element. Thus the underlying mechanisms can be different. It is also known that an amorphous $Gd_{0.37}Al_{0.63}$ film obtained by sputtering, which composition is not far from $GdAl_2$, shows spin glass behavior with a T_f of about 16 K (a- $GdAl_2$) [12]. However, mm- $GdAl_2$ has a completely different magnetic-ordering temperature ($T_f = 65$ K), which means that mm- $GdAl_2$ represents a new class of spin glass materials because it is neither a dilute solution nor amorphous. It is a crystalline intermediate phase, but atomically disordered and so metastable. Apparently, ball milling is a unique technique to create this spin glass phase in $GdAl_2$. The value of T_f of mm- $GdAl_2$ is much higher than that of a- $GdAl_2$. This could mean that the cluster sizes in mm- $GdAl_2$ are on average larger than those in a- $GdAl_2$ (see Ref. [9] for details).

In conclusion, both amorphous Co_2Ge and atomically disordered crystalline $GdAl_2$ obtained by mechanical milling in a high-energy ball mill exhibit a single transition from the paramagnetic to the spin glass state below 41 and 65 K, respectively. The spin glass transition temperature is lowered with increasing external field. These results demonstrate the use of mechanical milling as a novel technique to synthesize spin glasses and open the way to search for novel spin glass materials.

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