Comment on "Spin-Glass Behavior of Mechanically Milled Crystalline $GdAl_2$"

In a recent Letter [1] Zhou and Bakker (ZB) describe a technique in which they mechanically mill ferromagnetic crystalline $GdAl_2$, until the crystallite size is found to be about 21 nm. ZB argue that the milling induces "atomic (chemical) disorder in the $GdAl_2$ lattice" which yields "a real magnetic phase transition from a ferromagnet to a spin glass." The supporting experimental findings for this claim are (a) the cusp in the ac susceptibility which shifts to lower temperatures as the field is increased, and the (b) the irreversibility of the dc magnetization which is consistent with the ac measurements. ZB emphasize that "the observed phenomena are bulk phenomena and the contribution of the grain boundaries is very small." The purpose of this Comment is to point out that the experimental data are, in fact, consistent with the anticipated behavior of fine *ferromagnetic* grains and that ZB's claim, that an intrinsic magnetic transition to a "pure" spin glass has been induced, is dubious.

Fine ferromagnetic grains are known to have particular magnetic properties which are distinct from those of the bulk material. When the grains are less than a critical size (of the order of 30 nm in conventional magnets), it is energetically unfavorable to have domain walls, and thus the magnetic response of these grains is described in terms of a coherent rotation of a single magnetic domain [2]. In some temperature range below the Curie temperature, the magnetic behavior is described as superparamagnetism, which is characterized by (large) reversible magnetization without hysteresis, due to free coherent rotation of the spins in each grain. However, as soon as the temperature drops below some "blocking temperature" (which is related to the energy barriers induced by the shape and crystalline anisotropy) the free coherent rotation is blocked [2]. The blocking temperature decreases as a field is applied, since the field reduces the energy barriers. The observation of blocking depends on the time scale of the experiment, and thus the blocking temperature decreases as the frequency decreases.

The ac susceptibility exhibits a cusp, since the blocking of the grains suppresses their response. Furthermore, the field and frequency dependence of the cusp reflects the dependence of the blocking temperature [3,4]. The dc magnetization reflects the blocking in the difference between the zero-field-cooled magnetization (where the sample is cooled in zero field below the blocking temperature, and then a field is applied and the magnetization is measured as a function of increasing temperature) and the field-cooled magnetization (where the magnetization is measured upon cooling in a field). It is clear that the zero-field magnetization should fall below the field-cooled magnetization as long as the temperature is below the blocking temperature for the appropriate field.

The process of blocking the coherent rotation resembles the freezing process which describes the spin-glass transition. In fact, if one interchanges the terms blocking temperature and "freezing temperature", it is very difficult to distinguish between the behavior of fine ferromagnetic grains and canonical spin-glass systems like CuMn or AgMn [3]. In some cases only a detailed comparison of the frequency dependence of the blocking temperature and the freezing temperature could reveal that the two systems are not in the same universality class [4].

Due to the crystallite size of the mechanically milled $GdAl_2$, it is reasonable to assume that the grains consist of a single magnetic domain. Since the behavior of such ferromagnetic particles is known to be qualitatively similar to that of a spin-glass, only experiments which are sensitive to the *local* magnetic properties can distinguish between an intrinsic spin-glass and a granular ferromagnet. The muon-spin-depolarization and the neutron diffraction are sensitive to local interactions. Local scanning using a Hall probe or a magneto-optical probe can detect if there are ferromagnetic domains in comparable sizes to that of the grains. The heat capacity is also different for the two cases, as the entropy is very distinct. Without affirmative results from these (or other) local-probe-experiments, one cannot rule out the possibility that no intrinsic (atomic scale) ferromagnet to spin-glass transition has occurred, and that all the observed phenomena stem from the granularity of the material.

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