Zhou and Bakker's reply to Klein's comment on 'Spin-glass behaviour of mechanically milled crystalline GdAl2'

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Zhou and Bakker Reply: Klein argues [1] that our results [2,3] can also be explained as the behavior of fine ferromagnetic grains, i.e., as the blocking process of such grains, because the freezing of a spin glass is very similar to blocking. He suggests that further local-probe experiments are desired to distinguish between an intrinsic spin glass and a granular ferromagnet. It must be pointed out that Klein's Comment is not relevant to amorphous Co_2Ge [2], because there are no crystalline grains in amorphous Co_2Ge. Thus, we focus on $GdAl_2$. Following Klein's suggestion further experiments on both materials are planned.

We now argue that the results in [3] are conclusive for a phase transition from a ferromagnet to a pure spin glass. As described in [3], from the χ_{ac} vs T curves (Fig. 1. [3]) it is clear that upon milling the paraferromagnetic transition becomes weaker and disappears completely after milling for long periods. The spin-glass phase starts to appear after 60 h of milling as indicated by the peaklike transition at 45 K. Upon further milling, the fraction of spin-glass phase increases, while the amount of ferromagnetic phase decreases as indicated by the relative change of the transition intensities. After 600 h of milling, the transformation from ferromagnet to spin glass is complete. The transition shape and temperature of the newly formed spin glass still change upon further milling until 1000 h. Let us compare the changes of intrinsic parameters, e.g., lattice strain and magnetic ordering temperature (T_f), which directly reflect the atomic arrangement in the lattice, with the change of crystallite (grain) size (Fig. 1). The crystallite size and the relative strain were obtained by x-ray diffraction using the Scherrer equation (see [4]). The peak temperatures in the χ_{ac} vs T curves were taken as T_f. The crystallite size decreases abruptly up to 120 h and then becomes constant at about 21 nm. Both strain and T_f increase continuously up to 1000 h due to increasing atomic disorder. This disorder is further evidenced by the disappearance of a few x-ray reflections [4]. The χ_{ac} vs T curves for milling between 60 and 120 h show two transitions, i.e., the original paraferromagnetic transition and a new peaklike transition at lower temperature. Here the original ferromagnet and a new spin glass coexist. Both transitions might also be explained by fine ferromagnetic grains as proposed by Klein. To distinguish between both explanations, it is very important that the crystallite size becomes constant after 120 h (Fig. 1). This means that milling longer than 120 h would not change magnetic properties if only the refinement of crystallites would be responsible for such change. However, both lattice strain and T_f change significantly between 120 and 1000 h, i.e., atomic rearrangement continues in these periods. Furthermore the coexistence of the two transitions in the χ_{ac} vs T curves is still observed for milling between 120 and 600 h, but the intensity of the paraferromagnetic transition decreases, whereas that of the peaklike transition increases. This strongly indicates that the transition is intrinsic which is claimed by us to be a paramagnetic–spin-glass transition. Apparently, continuous disordering results in an increase of the spin-glass phase and

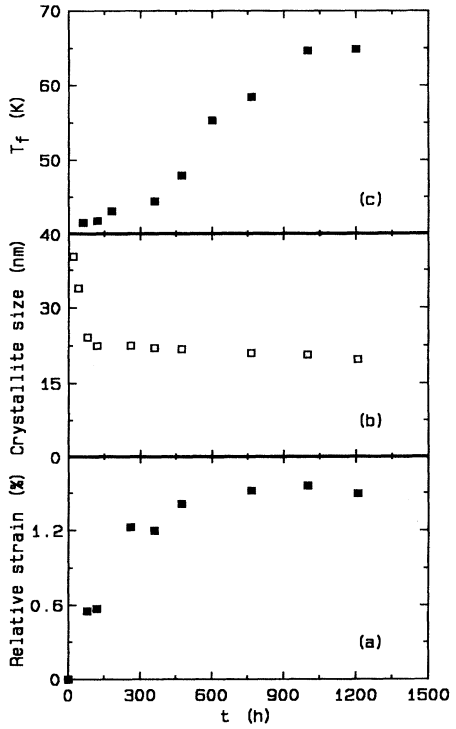

FIG. 1. The relative strain (a), the average crystallite size (b), and the freezing temperature T_f (c) of $GdAl_2$ as a function of milling time.

a decrease of the ferromagnetic phase until complete disappearance of ferromagnetic material. Therefore, atomic disorder induces an intrinsic transformation from the ferromagnetic to the nanocrystalline spin-glass phase. All intrinsic parameters become constant after milling for 1000 h or longer. Then a stationary state of disorder is established. It is impossible to understand the change of various intrinsic parameters, e.g., lattice strain and T_f, and the relative change of the intensities of the two transitions in the χ_{ac} vs T curves during the intermediate stage of milling (longer than 120 h) by the picture suggested by Klein. The grains are not ferromagnetic for milling periods of 600 h and longer; in contrast, the nanocrystals have a spin-glass character.

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