Unusual diamagnetic response in PrAlNiCuFe metallic glass

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An unusual diamagnetic response is observed in a $Pr_{60}Al_{10}Ni_{10}Cu_{16}Fe_4$ bulk metallic glass (BMG) during the zero-field-cooled (ZFC) magnetization measurement. The ZFC magnetization of the BMG is found to reverse its direction at low temperature and becomes diamagnetic, whereas the field-cooled branch remains positive. This apparent diamagnetism is ascribed to the specific couple between the ferromagnetic nanoparticles and the amorphous matrix in low fields. Besides, in superconductors, a giant diamagnetic response is unusual in magnetic materials. Therefore, it may simulate scientific and technological interest.
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Diamagnetism has been known as an ubiquitous property of matter that consists in the partial screening of an external magnetic field by the electronics of the matter. However, only in superconductors where electron pairs are unconstrained to react to external fields, can the perfect screening be obtained, resulting in a giant negative magnetic susceptibility.[1] In this work, an unusual diamagnetic behavior in the zero-field cooled (ZFC) branch is observed in the recently developed magnetic $Pr_{60}Al_{10}Ni_{10}Cu_{16}Fe_4$ bulk metallic glass (BMG). The magnetic anomaly is manifested as a negative magnetization at low temperature under ZFC, but the field-cooled (FC) branch remains positive. No superconductivity can be detected down to 2 K in the BMG. Even compared with the general superconductors,[1] the order of the diamagnetic susceptibility (-10^{-2} emu g^{-1} Oe^{-1}) is quite considerable. The analyses show that the magnetic coupling engenders the BMG with anomalous diamagnetic behavior. The BMG with controllable magnetization reversal may simulate scientific and technological interest and give facilities in fields such as ultrahigh-density recording and medicine.[2]

The cylindrical $Pr_{60}Al_{10}Ni_{10}Cu_{16}Fe_4$BMG rod 5 mm in diameter was prepared by arc melting of pure Pr, Al, Ni, Cu, and Fe and sucked into a Cu mold under argon atmosphere.[3,4] Its amorphous nature was ascertained by x-ray diffraction and other methods.[3,4] The magnetism and electrical resistance measurements were performed using a PPMS 6000 of Quantum Design Company. The domain structure was studied using a Digital Instruments NanoScope IIIa D-3000 magnetic force microscopy (MFM).

The ZFC and FC magnetization curves of the BMG measured in 50 and 100 Oe are shown in Fig.1, and the inset shows the enlarged details. The ZFC branch was measured on warming after initially cooling from 300 to 2 K in zero field, in fact, there exits a residual field of about −5 Oe in the magnetometer, which is opposite in direction to the applied field. The FC branch was measured after initially cooling to 2 K in the measuring field. Low field ZFC and FC curves exhibit a typical blocking process at the blocking temperature T_B=14 K (indicated in the inset of Fig.1), which corresponds to the onset of the ZFC curve peak. It is worth noting thatT_B decreases with increasing fields, which is reminiscent of the behavior of spin glass.[5] Above T_B, the FC and ZFC curves coincide as the paramagnetism sets in; whereas below T_B, they split, and the FC branch increases to a plateau while the ZFC branch decreases rapidly, which is consistent with the onset and development of a collective frozen magnetic state with magnetic moments randomly oriented. However, an anomalous diamagnetic response in the ZFC curve is observed at the measuring field H=100 Oe. The ZFC branch apparently becomes negative below 12 K and the magnitude (-10^{-2} emu g^{-1} Oe^{-1}) is considerable. When reducing the H to 50 Oe, the negative magnetization is still obtained, but the value diminishes. Surprisingly, the ZFC curve is similar to that observed in superconducting materials displaying a diamagnetic signal (Meissner effect). These facts also distinguish the BMG from a conventional spin-glass system where the ZFC magnetization departs from the FC magnetization at T_B and just shows positive sign below T_B.[6]

When we focus on the temperature dynamics below T_B and employ the ZFC magnetization measurement at 2 K as a function of H, the ZFC branch becomes negative when $H<1000$ Oe and reaches a minimum around 300 Oe (see Fig. 2), which provides further evidence that confirms the

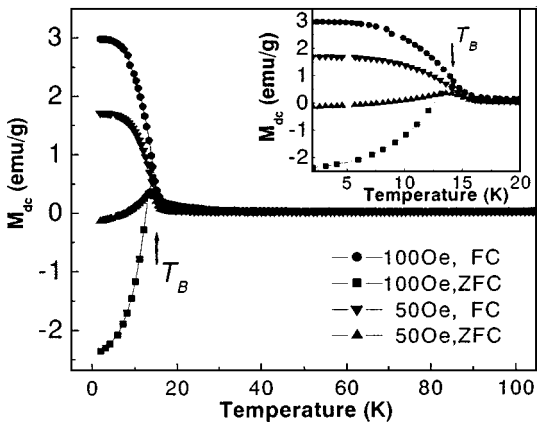

FIG. 1. The dc magnetization as a function of temperature, measured after ZFC and FC. The inset shows the enlarged details.

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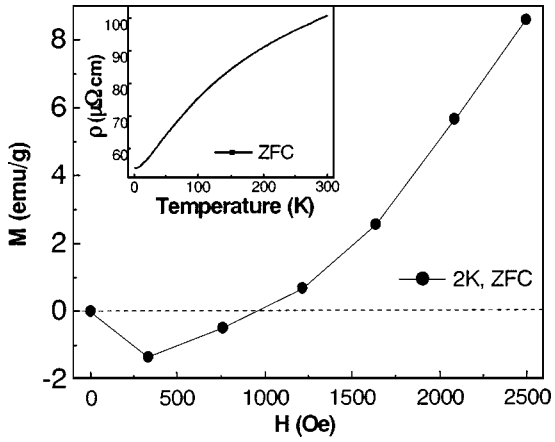

FIG. 2. Magnetization vs the applied field at 2 K after ZFC. The inset shows the electrical resistivity as a function of temperature under ZFC condition.

diamagnetic behavior in the BMG. The inset in Fig. 2 displays temperature-dependent electrical resistivity of the alloy under ZFC. Unlike other metallic glasses,[7] the BMG exhibits a positive temperature coefficient of resistivity, and no superconductivity is detected until 2 K.

To account for the observed giant diamagnetic behavior, the $M-H$ hysteresis loops and magnetic domain structure of the alloy were studied. Figure 3 presents the $M-H$ hysteresis loops at 300 and 2 K. It reveals that the BMG exhibits a hard magnetic property, and the coercivity (146 kA m^{-1}) is almost the same at 300 and 2 K, but the M_r/M_s value decreases from 0.59 at 300 K to 0.33 at 2 K. The typical (10×10 μm) MFM image of the sample is presented in Fig. 4. Some regions with obvious magnetic contrast in large gray (nonmagnetic) areas are visible, and the average domain width is about 180 nm, which is similar to that of other hard magnetic BMGs.[4,8] The results indicate that ferromagnetic Pr–Fe clusters coexist with amorphous matrix in the BMG. However, the BMG exhibits a typical amorphous XRD pattern, which gives an upper limit of a few nanometers for the cluster size. The average domain width is much larger than the cluster size because of the existence of exchange-coupling interaction in the alloy,[8] i.e., the domain is actually a collection of a group of the Pr–Fe nanoclusters with similar magnetic orientation aligned by exchange coupling. The nanoclusters with size of nearly 2 nm in other rare earth

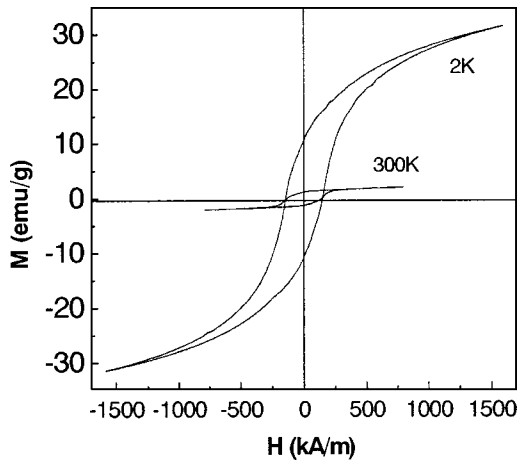

FIG. 3. The $M-H$ hysteresis loops of the as-cast BMG at 300 K and 2 K (ZFC).

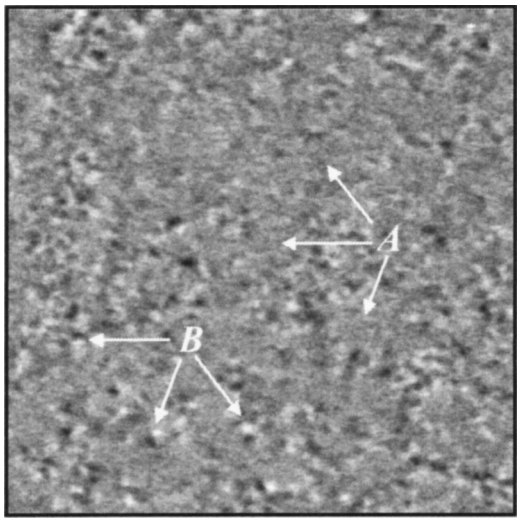

FIG. 4. (Color online) MFM image with scan size 10×10 μm for the as-cast BMG. The large gray areas (nonmagnetic) are indicated by marker *A*; the regions marked by *B* denote the magnetic domain structures with random distribution.

based magnetic BMGs have been directly observed.[9–11] The hysteresis loops and MFM image shed light on the microstructure of the alloy, which consists of amorphous matrix and ferromagnetic Pr–Fe nanoclusters with small fraction. According to the strong pinning model,[12] the Pr–Fe nanoclusters may act as pinning sites for the domain walls of the amorphous matrix, and the constant coercivity suggests that these particles keep stable in low temperature, the significant H_S enhancement is in good accordance with the blocking process.

We ascribe the anomalous diamagnetic response in the alloy to its inhomogeneous microstructure and the neglected residual field in which the ZFC branch is cooled. Figure 5 shows the enlarged ZFC magnetization curve under 100 Oe, which displays a weak paramagnetic-to-ferromagnetic transition around $T_C = 25$ K. It also shows an antiferromagnetic ordering temperature T_N at 20 K, which has been observed in other metallic glasses.[12] Because the ZFC branch is actually cooled in −5 Oe, the ferromagnetic Pr–Fe clusters will create a negative dipolar field in the sample interior during initially cooling process. This strong polarization even remains after the amorphous matrix loses its ferromagnetic order, causing the slight negative susceptibility in the ZFC branch;

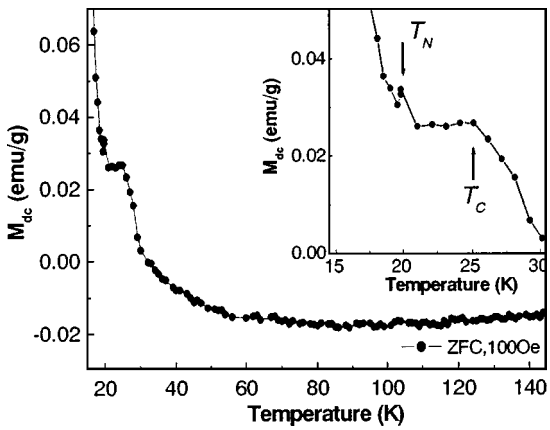

FIG. 5. The temperature dependence of the dc magnetization under ZFC condition with more enlarged details, the measuring field is 100 Oe.

the magnetic stability of these Pr–Fe nanoclusters also can be evidenced by the coercivity (see Fig.3). As the paramagnetic amorphous matrix becomes ferromagnetic below the Curie temperature, the spontaneous ferromagnetic order starts. With further cooling below T_B, the long-range ferromagnetic order in the amorphous matrix will be unstable, the competition between the FM and AFM interactions causes the frustration leading to the blocking process.[13] However, due to the strong negative dipolar field induced by the nanoclusters, a large number of magnetic moments of the amorphous matrix will eventually freeze in opposite direction to the measuring field. With increasing field from 50 to 100 Oe, the antiferromagnetic order is depressed by the applied field,[14] and the smaller frustration makes more magnetic moments tend to align in the dipolar field, leading to a large negative value. Similarly, a diamagnetic effect in a weakly ferromagnetic Pd0.5at.%Fe alloy has also been recently found when presenting FC magnetization measurement in very low field (<25 mOe), which arises from an inhomogeneity induced interplay of demagnetization fields and domain-wall pinning.[15]

Consequently, the magnetic sign of the Pr-based BMG can be easily tuned according to the different magnetization processes. This kind of system with multistate (non, positive, and negative) magnetic phases provides a flexible alternative for the magnetic nanostructure with controllable magnetization reversal, which is highly desirable in ultrahigh density magnetic recordings and new submicron magneticelectronics devices.[16,17] The inhomogeneous microstructure can be easily fabricated by introducing magnetic elements like Fe in various rare earth based BMGs.[4] Very recently, with an appropriate choice of magnetic matrix, an increase in T_B from 10 to 290 K was reported.[18] Therefore, it may pave the way for this diamagnetic response in the BMG being applied near room temperature.

In conclusion, a diamagnetic response in the $Pr_{60}Al_{10}Ni_{10}Cu_{16}Fe_4$ BMG during the ZFC magnetization measurement is observed. This unusual behavior is ascribed to the specific ferromagnetic particles couple to the amorphous matrix in low fields. Although it is clear that the material is not suitable in itself for engineering applications, the approach developed should in principle apply to nanoparticles deposited on an amorphous layer, a way suitable for use as patterned magnetic nanostructure with controllable magnetization reversal, which has both fundamental and applied significances.

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