

## Distinctive exchange bias and unusual memory effects in magnetically compensated

### $\text{Pr}_{0.75}\text{Gd}_{0.25}\text{ScGe}$

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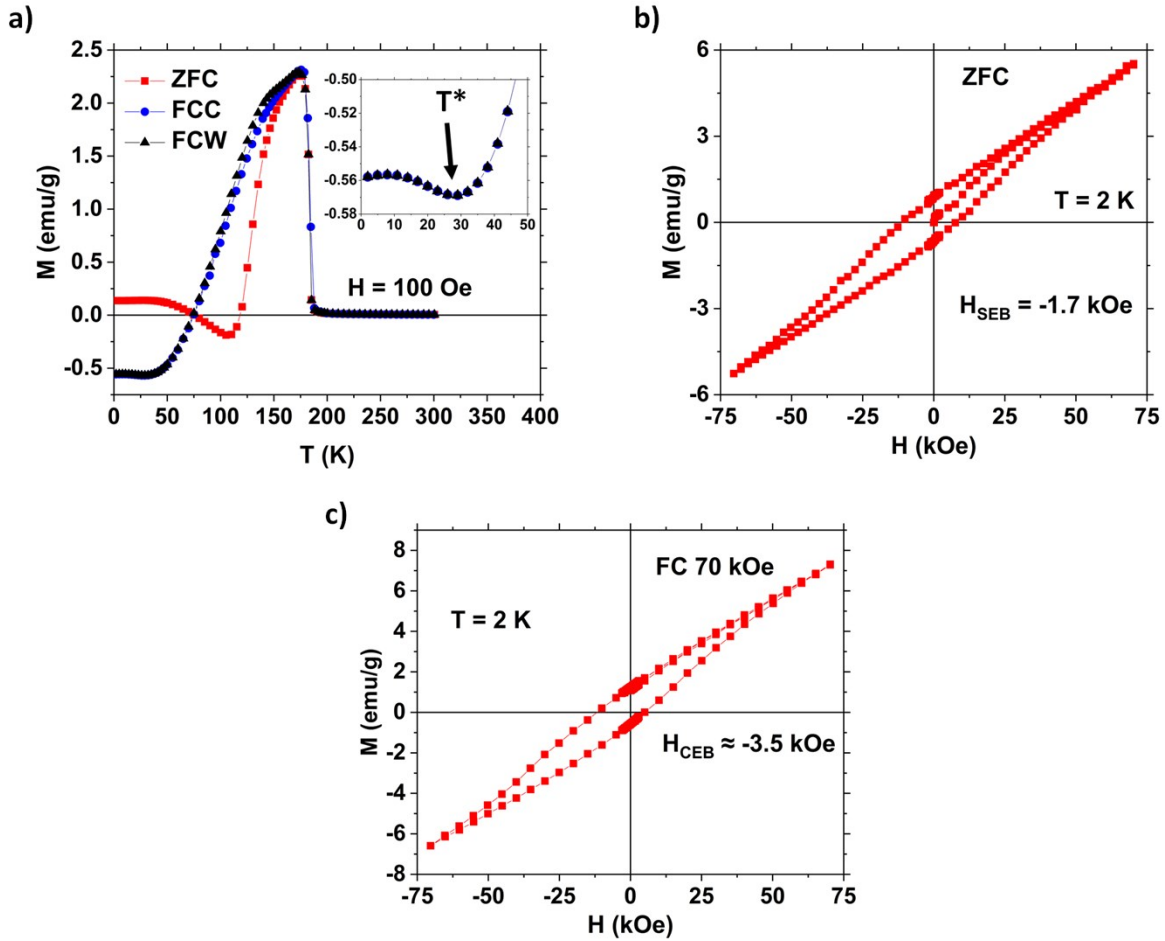
**Table S1:** List of the major impurities (over 0.001 wt.%, later converted to at.%) in the gadolinium metal used to prepare samples 1 and 2. Significant differences appear in bold font. The analysis was performed by inductively coupled plasma mass spectroscopy (ICP-MS).

Element	GdM-010 (at.%) (Sample 1)	GdM-011 (at.%) (Sample 2)
<b>Al</b>	<b>0.522</b>	<b>0.038</b>
Si	0.001	0.002
Fe	< 0.001	0.003
C	0.038	0.017
<b>O</b>	<b>0.293</b>	<b>0.559</b>
Gd	99.144	99.380

Most impurities listed are of similar amounts, except for Al and O. These impurities are expected to affect the structure differently: O will likely precipitate out to the grain boundaries in the form of oxides, while Al will either do the same in the form of  $\text{GdAl}_2$ , or other Gd-Al compounds, or substitute Ge within the bulk (Gd and Pr atomic radii are too large compared to Ge and Al). Despite these different impurity amounts, the crystal structure and magnetic properties

remain fundamentally unaffected, however there are some minute differences. It is worth noting that even with the higher oxygen content in sample 2, we believe that most of the differences come from the elevated Al content in sample 1.

Both samples maintain a CeScSi-type structure with slightly different lattice parameters:  $a = b = 4.31794(5) \text{ \AA}$  and  $4.3146(3) \text{ \AA}$ ,  $c = 15.8339(3) \text{ \AA}$  and  $15.8243(3) \text{ \AA}$ , resulting in cell volumes of  $295.217(7) \text{ \AA}^3$  and  $294.58(4) \text{ \AA}^3$  for samples 1 and 2, respectively. This correlates to 0.08%, 0.06%, and 0.2% differences for  $a$ ,  $c$ , and volume, respectively; lattice parameters of both samples are slightly larger than those predicted by Vegard's Law,  $a=b=4.313 \text{ \AA}$  and  $c=15.817 \text{ \AA}$ , which is not uncommon for intra rare earth alloys <sup>1</sup>.

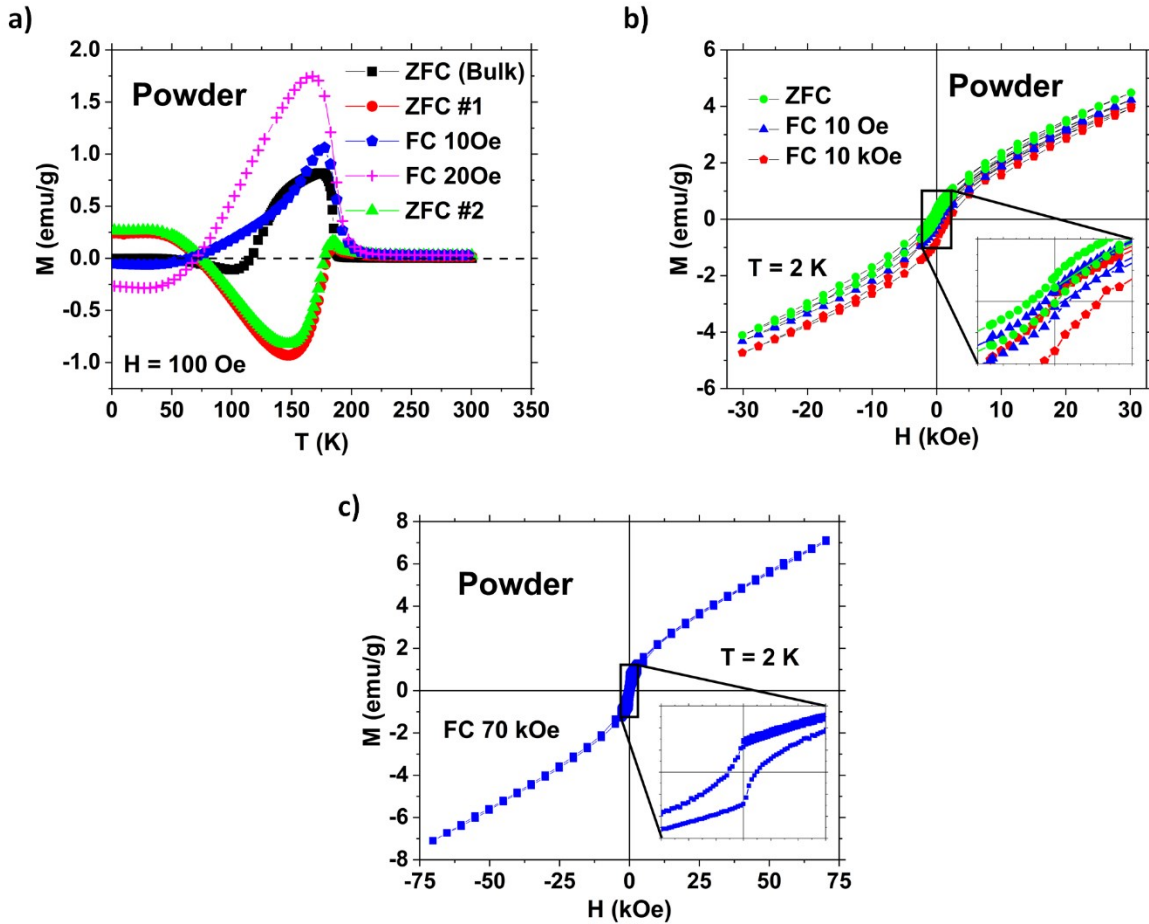


**Figure S1:** Figure 3 (main work) analog for sample 2. (a) ZFC, FCC, and FCW  $M(T)$  measured at 0.1 kOe. Inset shows detail around the  $T^*$  in FCC and FCW. (b) ZFC  $M(H)$  at  $T = 2$  K. (c)  $M(H)$  curve at  $T = 2$  K after cooling in 70 kOe field.

The magnetic properties of bulk sample 2 are presented in figure S1. Compensation is seen in all  $M(T)$  measurements taken at  $H = 0.1$  kOe.  $M(T)$  shows a local minimum in FCC and FCW data and a local maximum in ZFC data at  $T^* = 29$  K similar to the  $T^* = 30$  K of sample 1. Furthermore, the  $T_{Comp}$  for both samples under ZFC conditions agree fairly well, 115 K and 118

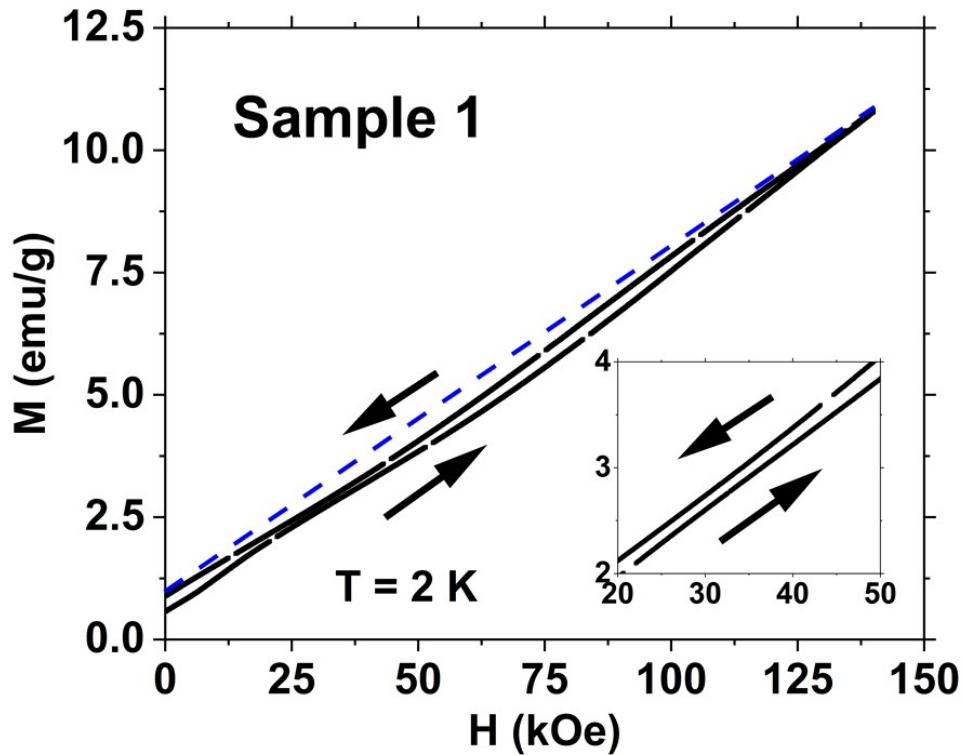
K for samples 1 and 2, respectively. However, sample 2 shows a  $T_{\text{Comp}}$  for FCC and FCW at 76 K as well as a second  $T_{\text{Comp}}$  for ZFC also at 76 K.

The most notable difference between samples 1 and 2 is the magnitude of SEB (0.5 kOe and 1.7 kOe, respectively). The 0.5 kOe SEB in sample 1 (figure 3b, main work) is significantly lower than the 1.7 kOe SEB in Sample 2 (Fig. S1b). A possible explanation is Al leaching Gd out of the sample and forming  $\text{Gd}_x\text{Al}_y$  precipitates along the grain boundaries, creating potential AFM-FiM or FM-FiM domain boundaries. However, CEB values are the same for samples 1 and 2 making these domain boundaries sparse or unlikely, pointing towards the likelihood of Al substitution for Ge rather than precipitation of  $\text{Gd}_x\text{Al}_y$  intermetallics. More work needs to be done to better understand the physical roots of the large SEB difference.



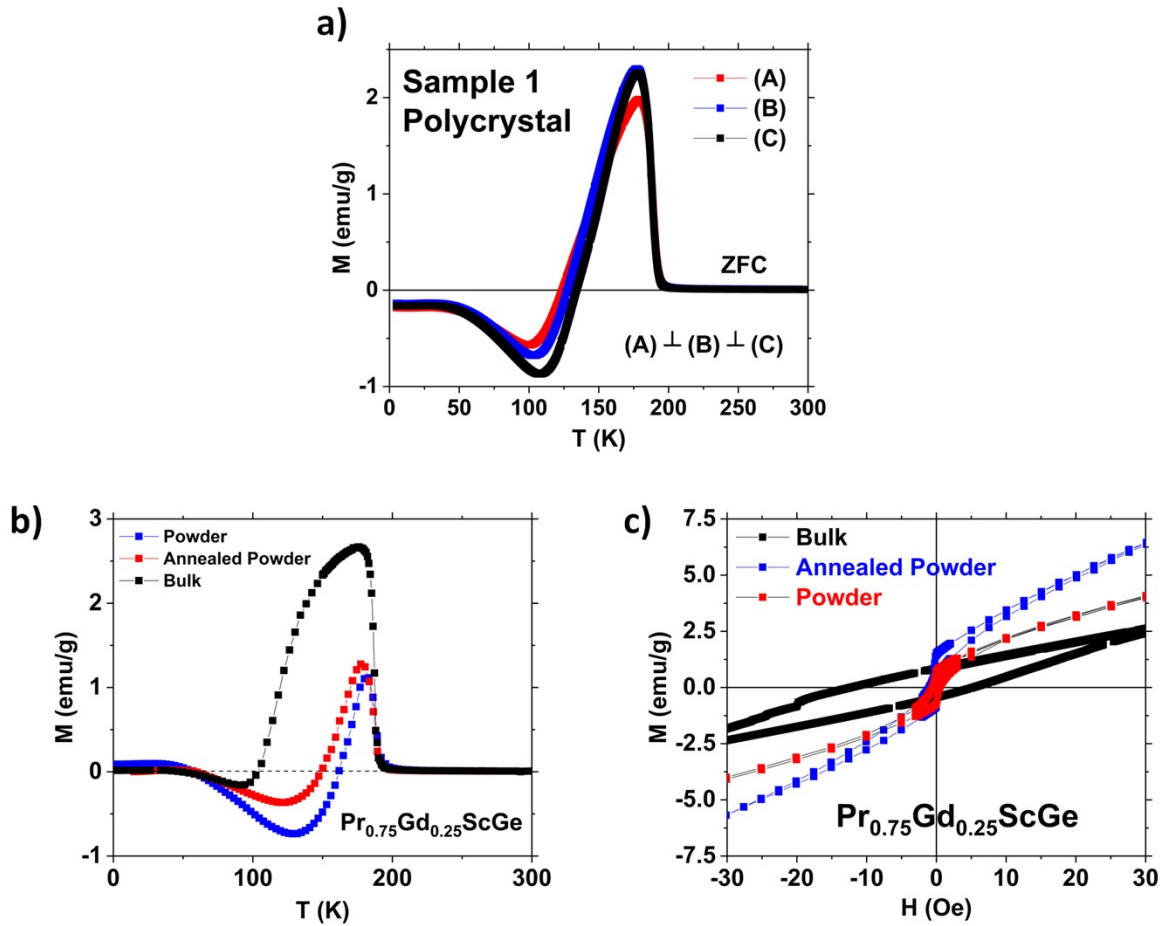
**Figure S2:** Figure 4 (main work) analog for sample 1. (a)  $M(T)$  behavior of powdered sample 1 with the legend arranged in the chronological order of the measurements. (b)  $M(H)$  of powdered sample 1 at  $T = 2$  K with an inset to highlight the SEB. (c) 70 kOe field cooled  $M(H)$  of sample 1 at  $T = 2$  K. Inset highlights the lack of CEB.

The magnetic properties for powdered sample 1 are presented in Fig. S2. While there are similarities between samples 1 and 2, Fig. 4b clearly shows that negative field cooling results in a positive exchange bias, while Fig. S2b shows that positive fields result in positive exchange bias. This reflects the differences in impurities and cooling fields between the measurements.



**Figure S3:** Magnetization vs magnetic field of sample 1 cooled in a 140 kOe field. The dashed line is a guide to the eye, illustrating deviations from linearity. The inset clarifies behavior between 20 and 50 kOe.

Fig. S3 depicts the high field  $M(H)$  behavior. This sample is close to be nearly fully compensated, displaying an almost antiferromagnetic (AFM) behavior, but still having some hysteresis. This nearly fully compensated behavior gives rise to the large jump in conventional exchange bias (predicted to be about 10 kOe from linear extrapolation, much larger than the 3.5 kOe seen after cooling in a 70 kOe magnetic field).



**Figure S4:** Potential effects of anisotropy and strain on magnetic properties - a) depicts  $M(T)$  of a single polycrystalline bulk piece in 3 distinct orientations, b) and c) depicts how strain affects the  $M(T)$  and  $M(H)$ , respectively. Both  $M(T)$  are measured in a 100 Oe field, and  $M(H)$  is measured at  $T = 2$  K. The data for the bulk in (b) are measured using a polycrystalline sample that was different from the specimen used in (a); both were extracted from the same ingot.

Differences between the bulk and powder measurements are most likely attributed to one (or more) of three factors: surface oxidation, anisotropy in the particles causing them to rotate and repack, and/or internal strain caused by pulverization. Potential oxidation was measured by

placing a powder sample in an oven at 150°C for 1 day and determining the weight gain. There was no measurable change in weight. However, it is worth noting that the new surfaces created by pulverization will oxidize during the grinding, forming a thin protective layer of rare earth oxides. Anisotropy was determined qualitatively using  $M(T)$  in 3 unique orientations using the same irregularly shaped polycrystalline piece (Fig. S4a). The different directions show only minor differences in  $M(T)$  behavior associated with a certain degree of preferred orientation developed during solidification of the arc-melted button. Lastly, strain due to pulverization was analyzed (Fig. S4b and S4c). A powder sample was wrapped in Ta and sealed in a quartz tube under partial helium atmosphere. The tube was purged multiple times and the final purge involved heating Y filings, placed on the opposite side of the tube, in order to remove any residual oxygen. The sealed tube was put in a furnace and temperature quickly ramped to 700 °C where the powder dwelled for 2 hours. The furnace was then turned off and allowed to slowly cool back to room temperature, after which its magnetic properties were re-measured. The reduction of strain plays a larger role than anisotropy and surface oxidation, as the relief of internal strain clearly shows a partial return of the  $M(T)$  of the powder to that of the bulk (Fig. S4b), but the  $M(H)$  shows little to no change (Fig. S4c). Furthermore, 2 hours at 700°C should be enough time to remove all internal strain, and yet there remains a significant difference in the magnetic properties, mainly in CEB and SEB, of the annealed powder and the bulk sample.

Furthermore, there is a noticeable difference between  $M(T)$  of two bulk pieces illustrated in Fig. S4a and S4b despite global similarity. In fact, each one of the  $M(T)$  curves recorded from different portions of the same ingot looks slightly differently. The minor differences in behaviors stem from small variations due to inhomogeneities in the atomic environment, similar to those reported in the past in  $Mn_2PtGa$  and  $Pr_{0.6}Er_{0.4}Al_2$ <sup>2,3</sup>. A good way of thinking about this is that Pr



and Gd's carry different magnetic moments, and the measured values of  $M(T, H)$  are going to be dependent upon the local chemistry. So, a slightly Gd-rich region will behave magnetically different than a slightly Pr-rich region.

## References

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