We would like to discuss in greater detail than is possible in our main article the differences between our results and others where sign changes in exchange bias have been observed just below the blocking temperature. This effect has been seen in Co/CoO bilayers by Gredig et al. and Radu et al., and in by Shi et al. in Fe\textsubscript{x}Zn\textsubscript{1-x}F\textsubscript{2}/Co bilayers. Let us make some general remarks and then take each of these cases in turn.

In all these papers a plot of bias field against temperature is given that appears similar to the data that we show in Figure 3(a) of our article – that is a hysteresis loop measured well below the blocking temperature of the system shows an offset in field that is opposite in sign to the cooling field that was applied to set the bias, but in addition there is an interval of temperatures below the blocking temperature where an offset with the same sign as the cooling field is also observed. This positive exchange bias is unusual, and the change in sign remarkably so. Various qualitative explanations of the sign change effect have been offered in these papers, but as yet quantitative modelling of their ideas has not been attempted by any of these groups.

In the past a positive exchange bias has been convincingly explained as a result of a very high cooling field overcoming an antiferromagnetic interaction at the ferromagnet/antiferromagnet interface, which then leads to a reversal in direction of the ferromagnet when this large field is removed. In a pair of papers Shi et al. report on their experiments with Fe\textsubscript{x}Zn\textsubscript{1-x}F\textsubscript{2}/Co bilayers. These are closely related to the archetypal FeF\textsubscript{2}/Fe system in which positive exchange bias was first discovered. To date it has only been observed for a rather small range of related materials systems and requires cooling fields much larger than those usually used in exchange bias experiments. At first the system diluted with Zn appears very similar to the pure system – the usual negative bias is found for small cooling fields, whilst positive bias appears for very large cooling fields, indicating an antiferromagnetic exchange coupling between the interfacial moments in the fluoride layer and those in the ferromagnet. However, around the range of cooling fields where the change in sign is observed, an intermediate behaviour is observed, where negative bias is obtained at low temperatures, switching to positive bias for temperatures higher than about half the blocking temperature. Hence in this materials system the change of sign in bias with temperature is observed for a small window of cooling fields, whilst a positive bias is observed for all temperatures when a high cooling fields. We observed neither feature, even though the highest cooling field we applied (90 kOe) was three times larger than the highest reported in this pair of papers (30 kOe), and our blocking temperatures and bias fields—which should provide a measure of the cooling field scale required—are much lower. Hence it is clear that the underlying mechanism of our sign reversal is different to that controlling the properties of these fluoride-based samples.

Gredig et al. observed a small positive exchange bias in an interval about 15 K wide below the blocking temperature (260 K) of their Co/CoO bilayer. By measuring the left and right coercivities of their sample they showed that this was related to an anomalous increase in the right coercivity (that for a field sweeping into the cooling field direction), which showed a marked peak in this region. Hence they dubbed their effect a unidirectional coercivity enhancement, rather than a positive exchange bias.
We have also plotted the left and right coercivities of our 154 Å thick Cu$_{94}$Mn$_6$ sample, shown in Figure 1 below: this sample shows one of the most pronounced bias inversions that we have observed. Our sign convention is opposite to that of Gredig et al.: we cooled in a negative field. When sweeping the field into the field cooling direction—our left hand coercivity $L_{HC}$—we observe a peak similar to that of Gredig et al. In fact this is very common as it is this effect that gives rise to the coercivity enhancements that are always seen around the blocking temperature$^{6,7,8}$, although Ref 1 is the only instance we are aware of where this has given rise to an apparently positive bias. In Figure 1 we also show our data on a sample of the conventional IrMn/Co system where the form of the curves is exactly like that of Gredig et al., with the exception that the minimum in the left coercivity is not quite large enough to produce positive bias. However, when sweeping the field against the cooling direction in the spin glass sample we see a small dip centred around ~11 K. This non-monotonic feature was definitely not present in the data of Gredig et al., whose equivalent curve was completely smooth, and we do not observe it in the IrMn/Co data either. This is a new feature that requires a new explanation, as the ideas about disordered interfaces due to reversed AF grains cannot account for this. Hence it is necessary to develop a new model on another physical basis, as we have done.

Figure 1 - (Upper panels) The left (L $H_C$) and right (R $H_C$) hand coercive fields for a Co layer exchange biased by a 154 Å thick Cu$_{94}$Mn$_6$ film (left graph) and an Ir$_{20}$Mn$_{80}$ film (right graph) as a function of temperature. The measurements were made after cooling to base temperature in a field of -70 Oe. The small dip in R $H_C$ at T ~ 11 K for the CuMn sample is what gives rise to the inversion of the exchange bias field, and is absent from the data for the IrMn sample, which displays a monotonic decline with T. (Lower panels) The exchange bias ($H_e$) and coercive ($H_C$) fields derived from these data as a function of temperature.

Radu et al. also studied a CoO/Co bilayer, although their material was prepared on a single crystal substrate and was approaching epitaxial crystallographic quality. These authors also show the left and right coercive fields in their article, which they refer to as $H_{e1}$ and $H_{e2}$. In their case both curves appear monotonic and the small bias inversion they see is due to a weak inflexion in the curve for $H_{e2}$. Again, they do not see any non-monotonic feature, as we do in Figure 1. They offer a qualitative model based on the idea of antiferromagnetic superexchange across a layer of oxygen atoms. Although this is known to be the mechanism causing antiferromagnetism in CoO, that fact that they always observe the same effect regardless of cooling field, unlike the results of Shi et al., suggest that alternative explanations could be considered.
Hence, although examples of an inversion of bias below the blocking temperature have been seen before, and tentative proposals for the underlying mechanisms given, we would claim to be the first group—with the usual caveats—to have put forward a quantitative model for how this effect can come about in a spin glass based system. Moreover the mechanism in our model is different, as it relies on the RKKY interaction that gives rise to the glassy behaviour itself. This model predicts that if the spin glass needs to be dilute enough that the RKKY interaction can dominate over nearest neighbours, confirmed by our experiments using CuMn layers with much higher Mn concentrations and some associated computational modelling. Hence a bias inversion due this effect has not been previously observed in any of the antiferromagnetic materials mentioned here. Moreover, there are qualitative features in our data that differ from all the other previous observations when examined in detail.

References

3 Shi H., Lederman D., Dilley N. R., Black R. C., Diedrichs J., Jensen K., and Simmonds M. B., Temperature-induced sign change of the exchange bias in Fe$_{0.82}$Zn$_{0.18}$F$_2$/Co bilayers, J. Appl. Phys. 93, 8600-8602 (2003).