

Controlling the Curie temperature in (Ga,Mn)As through location of the Fermi level within the impurity band

S.1. Spin dependent density of states, magnetic circular dichroism, and depletion of the valence band

One should note that Figure 3 in the main text, which is a schematic representation of the mechanism of magnetic circular dichroism (MCD) in (Ga,Mn)As, is drawn with exaggeration for pedagogical reasons. If we had plotted this figure to scale, it would be very difficult to see any effects arising from the spin-dependent depletion of the valence band that causes MCD. Specifically, as can be seen in Figure 5 in the main text, the value of MCD at its maximum does not exceed 3%. Since the MCD signal is proportional to the density of states (DOS) pulled from the valence band into the impurity band¹, we can conclude that not more than 3% of the states are missing from the valence band. This estimate also agrees with our assertion that the number of states in the impurity band (IB), and therefore the depletion of one of the spin projections in the VB, is of the order of x_{eff} . Thus, one should not expect any significant changes in the shape of the valence band.

It is worth mentioning that the data in Fig. 2D of Richardella et al.² supports such depletion of the VB density of states with increasing x_{eff} . Indeed, the differential conductance dI/dV plotted in that figure is believed to be proportional to the DOS. Their curves clearly show that the DOS well inside the VB (indicated by the y-axis intercept of the plot) decreases with increasing x_{tot} . Since the entire VB is not shown in the above figure, we cannot estimate what percentage of the DOS is moved into the IB according to their measurements; but the trend is certainly consistent with our findings.

Results reported in the work of Ohya et al.³ are also consistent with our findings. If the Mn ions bind the impurity states in their immediate vicinity, it follows that the (somewhat fewer) states left in the VB will have little overlap with the Mn sites, because the VB states are orthogonal to the impurity states. This also follows from the completeness of the basis of eigenstates. Since the impurity states make a large contribution to the density of states near the Mn ion, the VB states must contribute little in those regions. Because of this very small overlap with the Mn ion, the VB states are quite insensitive to the magnetic properties of Mn, thus resulting in very small p - d exchange. In fact, apart from some depletion in their number and some redistribution of their wavefunctions so as to avoid the regions occupied by impurity states, the VB states are similar to those in a non-magnetic material. This is one of the main observations of the paper by Ohya et al.³, indicating that the VB is very insensitive to any magnetic properties of the material, including T_C . This also explains why the energies of the discrete VB levels trapped in the very thin (Ga,Mn)As films used by those authors are well described by the k -dot- p parameters of the non-magnetic GaAs host.

S.2. Negative contribution to MCD signal below the energy gap

One of the important aspects of the MCD signal in (Ga,Mn)As is that it is comprised of both positive and negative contributions. Earlier studies of the MCD in (Ga,Mn)As with low Mn concentration showed a negative contribution to the MCD signal in some of the samples. In Ref. 1 it was argued that this negative below-gap contribution arises due to compensation. As shown in Figure 4 of that reference, samples A, B and D studied in that paper have the same Mn concentration $x = 0.014$, but were grown under different conditions, resulting in different levels of compensation. In that series, sample D was most highly compensated, and the negative contribution to MCD in that sample was therefore very pronounced; while Sample A had the lowest degree of compensation, and the negative contribution to MCD signal was completely absent for that sample. In this context, it is interesting to compare the MCD spectra of samples A and B in Ref. [1] below the energy gap. Sample B has somewhat higher degree of compensation than sample A, and one can see in Fig. 4 of that paper that, even though the MCD signal in sample B never reaches the “negative territory”, the contribution of compensation to the spectrum is clearly seen in the form of a downward dip in the spectrum of Sample B.

In the present paper a negative contribution to MCD signal is not expected because most of the samples used have a low compensation level, as indicated by their filling factors listed in Table I. The only sample with relatively high compensation level is sample F, for which $f \approx 0.3$, indicating that 70% of the states in the impurity band are occupied by electrons. Figure 5 of the present manuscript compares MCD spectra on samples F (as-grown) and F* (annealed). As one can see, and in agreement with Fig. 4 of Ref. 1 discussed in the preceding paragraph, the negative contribution below the gap is clearly evident (again in the form of a downward dip), even though the signal never actually drops below zero. It is difficult to say whether there is any negative contribution to MCD below the gap in sample F* ($f = 0.7$) because of the presence of a positive below-gap background. However, based on our understanding, such negative contribution (if it exists) would in this case be even weaker than in sample F.

References:

- ¹ Berciu, M. et al. Origin of magnetic circular dichroism in (Ga,Mn)As: giant Zeeman splitting vs. spin dependent density of states. *Phys. Rev. Lett.* **102**, 247202 (2009).
- ² Richardella, A. et al. Visualizing critical correlations near the metal-insulator transition in $\text{Ga}_{1-x}\text{Mn}_x\text{As}$. *Science* **327**, 665-669 (2010).
- ³ Ohya, S., Takata, K. & Tanaka, M. Nearly non-magnetic valence band of the Ferromagnetic semiconductor (Ga,Mn)As. *Nature Physics* **7**, 342-347 (2011).