Supplementary material

“Warping a single Mn acceptor wave function by straining the GaAs host”

I. OVERVIEW

The local density of states of individual Mn acceptors near InAs quantum dots in a GaAs host are mapped at room temperature by scanning tunneling microscopy. Dramatic distortions and a reduction in the symmetry of the wave function of the hole bound to the Mn acceptor are observed originating from local strain induced by quantum dots. Calculations of the acceptor state wave function in the presence of strain, within a tight-binding model and within an envelope-function effective mass model, agree with the experimentally-observed shape. The magnetic easy axes of strained lightly-doped Ga$_{1-x}$Mn$_x$As can be explained based on the observed local density of states for the single Mn spin. The following supplementary section expands on the information provided in the Article providing additional examples of observed strained distorted Mn, details of the high-pass filter used in Fig. 1 of the Article, and additional details of the tight-binding model used to calculate the wave function shape and the magnetic anisotropy energy.
II. SYMMETRY OF THE GAAS (110) SURFACE AND UNSTRAINED MN ACCEPTOR STATE

Shown below is the symmetry of the (110) GaAs surface and the Mn wave function. In (a) red are gallium sites and grey are arsenic sites; the larger circles are in even layers below the surface and the smaller circles are in the odd layers below the surface. The shaded triangles show schematically the unstrained acceptor wave function when the center gallium site is occupied by a Mn atom. The appearance of the Mn wavefunction with respect to the Ga and As sublattices is discussed in detail in the Ref. [1]. The unstrained Mn acceptor wave function shape shown in (b) is from Ref. 12 of the Article, and corresponds to a Mn atom located in the fifth layer below the surface. The acceptor wave function has reflection symmetry along the [110] direction and approximate reflection symmetry along the [001] direction. These symmetries are preserved unless the strain is not collinear to both the [001] and [110] directions.

FIG. 1: (a) Symmetry of the (110) GaAs surface and the (b) Mn wave function. The wave function can be deformed only if the strain axis is not collinear to both [001] or [110] directions.
III. SYMMETRY OF MN ACCEPTOR STATE IN STRAINED GAAS

In Fig. 2 four cases of Mn near InAs QDs are shown. They show Mn atoms located in each of the four quadrants around an InAs QD (white arrows) as well as in regions where the strain is weak or does not break the symmetry of the Mn atoms (purple arrows). The Mn atoms in (c) and (d) are shown in Fig. 1 of the Article.

FIG. 2: Four different cases of relative arrangement of QD and Mn when Mn wave-function deformation occurs. Deformed Mn wave functions are indicated by white arrows and some isolated undeformed Mn wave functions are indicated by purple arrows. The color scale of each panel is adjusted for the best view.
IV. HIGH-PASS FILTER

In figures 3 and 4 we demonstrate the details of the sequential filtering procedure. The Fourier spectrum of the topographical STM image of a single undisturbed Mn wavefunction is shown in the Fig. 3(a). The central part of the spectrum shows the spatial harmonics related to Mn wavefunction. The remote satellite features marked by $a_0$ and $b_0$ correspond to the lattice parameters of the (110) surface 0.565 $nm$ and 0.4 $nm$ respectively. In the figures 3(b) and 4(b) the main features of the spectrum are separated from the high frequency noise by applying the band pass filter. No modification of the Mn wavefunction envelope is introduced at this stage. In the figures 4(c) and 3(c) the isotropic part of the Mn wavefunction is removed by applying high pass filter with the typical radius of 8-10 lattice constants $a_0$. An identical filtering sequence was applied to the images with quantum dots to obtain figures 1(aa) and 1(bb) of the Article.

![Images](attachment:image.png)

FIG. 3: Different stages of sequential Fourier filtering performed for Mn wavefunction in the strain free GaAs ($a$) → ($b$) → ($c$).

Below, Fig. 4 shows the intermediate results of the filtering procedure for one QD-Mn pair. This example shows that the Mn wavefunction deformation is present at any stage of the filtering.
FIG. 4: Different stages of sequential filtering (a) → (b) → (c) → (d). Figures (c) and (d) differ by the size of the high pass filter.
V. CORRESPONDENCE OF MN WAVE FUNCTION SHAPE WITH QD STRAIN

FIG. 5: Symmetry of the ellipsoidal QD. Stress is outwardly directed from the dot, leading to stress along the [\bar{1}11] direction in the blue regions and along the [1\bar{1}1] direction in the red regions. The wave functions in the red and blue regions are mirrored images of each other.

VI. PARAMETERS FOR THE EFFECTIVE SPIN HAMILTONIAN FROM THE TIGHT-BINDING MODEL

The first-order modification of the energies of the valence band edge and of the acceptor level due to the strain tensor can be described, by symmetry, with the effective Hamiltonian

\[ H = a (\epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz}) + b \left[ (J_x^2 - J^2/3) \epsilon_{xx} + c.p. \right] + \frac{d}{\sqrt{3}} \left[ (J_x J_y + J_y J_x) \epsilon_{xy} + c.p. \right] \]  

(1)

where \( c.p. \) stands for cyclic permutation. For the valence band edge states \( J = 3/2 \) and for the acceptor level \( J = 1 \). The sign convention for \( a, b, \) and \( d \) is set so the Hamiltonian describes hole energies.

Shown below are the values for the coefficients in Eq. (1) for the valence band edge (hole convention), calculated for several values of the strain along the stress direction. The results for the hydrostatic deformation potential \( a \) show greater variability, but those for the shear...
deformation potentials show at most 3% variability or nonlinearity. Thus Eq. (1) appears accurate for strains in this regime.

<table>
<thead>
<tr>
<th>[001]</th>
<th>-0.1%</th>
<th>-0.05%</th>
<th>0.05%</th>
<th>0.1%</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>-1.1</td>
<td>-1.2</td>
<td>-1.3</td>
<td>-1.4</td>
</tr>
<tr>
<td>b</td>
<td>-3.6</td>
<td>-3.6</td>
<td>-3.5</td>
<td>-3.5</td>
</tr>
</tbody>
</table>

TABLE I: Values of deformation potentials (in eV) for the valence band edge states for use in Eq. (1) extracted from the application of stress along the [001] direction, producing a strain of −0.1%, −0.05%, 0.05%, or 0.1% along the [001] direction.

<table>
<thead>
<tr>
<th>[111]</th>
<th>-0.1%</th>
<th>-0.05%</th>
<th>0.05%</th>
<th>0.1%</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>-1.2</td>
<td>-1.3</td>
<td>-1.3</td>
<td>-1.3</td>
</tr>
<tr>
<td>d</td>
<td>-3.3</td>
<td>-3.3</td>
<td>-3.3</td>
<td>-3.3</td>
</tr>
</tbody>
</table>

TABLE II: Same as Table I but for stress along the [111] direction.

The value of \(a\) is for the shift of the valence band edge, not the gap of the semiconductor, and hence high quality experimental values for this are not available. The experimental values for \(b\) and \(d\) are within a factor of 2 of these calculated ones. Deformation potentials can be produced which are closer to the experimental values from tight-binding calculations which include more states, such as \(s^*\) or \(d\) states, or which alter the distance scaling of the tight-binding matrix elements from the \(\ell^{-2}\) scaling we have assumed. As these models introduce many additional band-structure and strain parameters we have retained a smaller basis set and simpler scaling rules. As the deformation potentials we calculate are within a factor of 2 of the experimental ones we do not expect the use of more complex tight-binding models (larger basis sets and less transparent scaling rules) to significantly modify the appearance of the Mn wave function in the strained crystal or the magnetic anisotropy we calculate from this model.
VII. ACCEPTOR LEVEL ENERGY SHIFTS AND SPLITTINGS FOR ARBITRARY STRESS DIRECTIONS

The splittings of the acceptor energy levels with strain can be described for arbitrary stress direction with Eq. (1) with deformation potentials determined for the acceptor state. Shown below are the values for the $a$, $b$, and $d$ coefficients for the acceptor level in Eq. (1) calculated for several values of the strain along the stress direction. Slightly larger variability or nonlinearity is found for the acceptor level than for the valence band energy shifts. Still, Eq. (1) appears accurate for strains in this regime. Strains approaching 1%, however, produce clearly nonlinear effects.

\[
\begin{array}{cccccc}
[001] & -0.1\% & -0.05\% & 0.05\% & 0.1\% \\
 a & -0.9 & -1.3 & -1.1 & -1.4 \\
 b & -6.9 & -6.8 & -6.3 & -6.3 \\
\end{array}
\]

TABLE III: Values of deformation potentials (in eV) for the acceptor level for use in Eq. (1) extracted from the application of stress along the [001] direction, producing a strain of $-0.1\%$, $-0.05\%$, $0.05\%$, or $0.1\%$ along the [001] direction.

\[
\begin{array}{cccccc}
[111] & -0.1\% & -0.05\% & 0.05\% & 0.1\% \\
 a & -0.9 & -0.9 & -1.2 & -1.3 \\
 d & -8.1 & -8.2 & -8.3 & -8.3 \\
\end{array}
\]

TABLE IV: Same as Table III but for stress along the [111] direction.

These deformation potentials and Eq. (1) can be used to determine the magnetic anisotropy energy for a Mn with a hole in the acceptor state under stress along an arbitrary direction.