Supplementary Information: Electronic anisotropies revealed by detwinned ARPES measurements of FeSe

Matthew D. Watson,1,* Amir A. Haghighirad,2 Luke C. Rhodes,1,3 Moritz Hoesch,1 and Timur K. Kim1

1Diamond Light Source, Harwell Campus, Didcot, OX11 0DE, United Kingdom
2Clarendon Laboratory, Department of Physics, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom
3Department of Physics, Royal Holloway, University of London, Egham, Surrey, TW20 0EX, United Kingdom

Determination of degree of detwinning from constrained fits to MDCs

FIG. 1. a,b) High symmetry cuts in the Z-A_b and Z-A_a directions, reproduced from Fig. 1 of main text. c,d) Peak fitting of the MDC at the Fermi level with two pairs of Lorentzians (and a centered Lorentzian background function.)

TABLE I. Fermi surface size, velocities and width (FWHM) of the hole pocket of FeSe at Z (23 eV) and electron pockets at A (56 eV), extracted from constrained Lorentzian fits to the detwinned data.

|      | k_F (Å⁻¹) | v_F (eV ˚A) | Γ_k (Å⁻¹) |
|------|-----------|-------------|-----------|
| Z, a | 0.097(5)  | 0.40        | 0.068     |
| Z, b | 0.145(5)  | 0.50        | 0.038     |
| A, inner | 0.03(1)   | -           | -         |
| A, outer | 0.19(1)   | 0.66        | 0.04(1)   |

In the main text, we compared ARPES data obtained in the same measurement geometry, but with the sample rotated by 90 degrees. These data generally showed a clear disparity between strong and weak intensities on the dispersions from the primary and minority domains respectively, indicating that the sample had a good but not perfect degree of detwinning. Here we present a method to estimate the degree of detwinning from quantitative analysis of the MDCs for the hole pocket, and justify the figure of 80%-20% domain population used in the main text.
Consider the Outer and Inner bands found in the MDCs, i.e. the long and short axes of the single elliptical hole pocket in one domain. Let them each have an intrinsic amplitude $\tilde{O}$, $\tilde{I}$ - i.e. the intensity that would be seen in a perfectly detwinned sample, and observed amplitudes $O$ and $I$ in each sample orientation $(a,b)$. Let $p$ be the population of the primary domain, and $(1 - p)$ the minority domain population. Then we have

\begin{align*}
O_b &= p\tilde{O} \\
I_b &= (1 - p)\tilde{I} \\
O_a &= (1 - p)\tilde{O} \\
I_a &= p\tilde{I}
\end{align*}

In order to extract $O, I$ we measure the sample in both orientations with otherwise equivalent measurement conditions, and fit the MDCs along $a$ and $b$. We use pairs of Lorentzian peaks to fit the data, and both the widths and peak positions are fixed between the two orientations (Fig.1). This gives more confidence in the values of the amplitudes (i.e. $O_a$ etc) obtained.

We define a ratio $r$, obtained experimentally from the peak amplitudes, as

\[ r = \frac{O_b}{I_b} = \frac{O_a}{I_a}, \quad r > 1 \]

which is a function of $p$ only,

\[ r = \frac{p^2}{(1 - p)^2} \]

Solving the quadratic equation for $p$ gives

\[ p = \frac{r - \sqrt{r}}{r - 1} \]

which has the intuitive limits of $p = 0.5$ for $r = 1$ (i.e. both directions are equivalent for a completely twinned sample) and $p \to 1$ as $r \to \infty$, which would correspond to a perfectly detwinned sample. In our case we obtain $r = 16.5$, giving $p = 0.8$. We show some fitting parameters in Table I.

In our experience, the estimated 80%-20% domain population balance is likely to be close to the maximum detwinning effect possible with ARPES measurements of FeSe, since we found that further strain tends to induce cracks and inhomogeneity to the sample surface. However we note that the detwinned data in the main text is obtained without significant compromise to the resolution of features in the data compared to twinned samples. We further note that for the “accidentally detwinned sample”, no quantitative estimate of the degree of detwinning is possible as this analysis was not performed, but qualitatively it appears to have a similar degree of detwinning to the mechanically detwinned sample in the main text, and is qualitatively different from the twinned Fermi surface shown in Fig. 3.
FIG. 2. Fermi surface maps obtained on a different partially detwinned sample. The analysis of MDCs for this sample (not shown) indicates that the domain populations are approximately 64%-36%. a) 23 eV maps of the Z point, in different sample orientations. Note that due to the unequal intrinsic peak widths and amplitudes of the longer and shorter sections of the elliptical hole pocket, the visual impression of the degree of detwinning varies with the measurement geometry; for this reason it is important to measure the sample in both orientations and to perform the numerical analysis. b) Temperature-dependence of the Fermi surface, measured at 56 eV. While the 50 K data also shows a detwinning effect, the 100 K data resembles a normal unstrained sample (as in Fig. 3(c) of the main text), e.g. the spectral weight around the Γ point shows no anisotropy relative to the tensile stress direction.

Partially detwinned samples

In Fig. 2 we show data on a different partially-detwinned sample, which gives consistent results although with a weaker degree of detwinning.

One important challenge is to identify if the degree of detwinning is homogeneous over a big enough area to allow for reliable rotations of the sample and Fermi surface mapping. Additionally, it is possible for the sample surface to buckle or crack if the strain is large, which can also occur in normal samples sometimes after cleavage. This is a more significant problem for FeSe compared to other Fe-based superconductors due to the structural weakness of the samples. We checked for this on each sample by measuring spectra as a function of sample position, to test for either any significant change in the apparent angular offset of the spectra (which might indicate a buckling of the surface) or for any change in the relative amplitudes of peaks. In the main sample used in the text neither of these was a significant issue but in two other samples these issues were observed.
In Fig. 3 we show a Fermi surface measurement of a twinned sample, covering an extremely large region of momentum space. The parity-switching behavior can be easily identified for both the electron and hole pockets: every time you move by $(\pi, \pi)$ on the map, the opposite selection rules apply. Note that when the measurement is well away from normal emission, selection rules tend to be less strict, for example some $d_{XZ}$ spectral weight is observed at the 3rd $\Gamma$ point, whereas this spectral weight is strictly absent at normal emission (1st $\Gamma$). Nevertheless the parity switching is still clear, since the $d_{YZ}$ weight dominates at the 3rd $\Gamma$ point, while being strongly suppressed at the 2nd and 4th $\Gamma$ points.