Staggered chemical potential and the surface bands of $Sr_2 RuO_4$

Anirudh Chandrasekaran

Loughborough University

14 June 2024

In this notebook we will examine the effect of a staggered chemical potential on the dispersion of the surface bands of $Sr_2 RuO_4$. We will focus in particular on the M point. The staggered chemical potential breaks the lattice reflection symmetry but preserves the fourfold rotation symmetry about each Ru atom. As we will see below, this allows the two d_{xy} bands at the M point to decouple and become individually four-fold symmetric. This allows the bands to host X_9 singularities under tuning.

Loading the Library

We first load the library by executing the following:

```
In[∘]:= AppendTo[$Path, StringDelete[NotebookDirectory[], "/Sr2RuO4"] <> "Package"];
<< BandUtilities`
```

Interpolating between the Wannier90 files

Ingredients needed for symmetrization

```
In[445]:= ρ = {{0, -1, 0}, {1, 0, 0}, {0, 0, 1}};
    (*The matrix part of the C₄ rotation about one of the Ru atoms.*)
    Uρ = {{0, -1, 0}, {1, 0, 0}, {0, 0, -1}};
    (*Representation of C₄ rotation in the d-orbital space.*)

    rxy = {{0, 1, 0}, {1, 0, 0}, {0, 0, 1}}; (*x↔y reflection.*)
    Urxy = {{-1, 0, 0}, {0, 1, 0}, {0, 0, -1}};
    (*Representation of the x↔y reflection.*)

    c0 = {1/4, 3/4, 0}; (*Location of one of the Ru atoms,

    which we shall use as the centre of the C₄ rotation.*)

    AtomLocations =
      {{3/4, 1/4, 0}, {3/4, 1/4, 0}, {3/4, 1/4, 0}, {1/4, 3/4, 0}, {1/4, 3/4, 0}, {1/4, 3/4, 0}};
    (*Location of the basis atoms in the fractional
     coordinates within the unit cell.*)

    SymmetryList = {}; (*This is a multidimensional list which we shall
     construct below. Each element is a sublist corresponding to a particular
     symmetry. First element of the sublist is real space symmetry's matrix part,
    second element is the translation part and the third element is
     its representation in the Hilbert space. Our point group is D₄,
    or D₈ in some pepole's notation.*)
    Do[
      AppendTo[SymmetryList,
        {MatrixPower[ρ, i], (c0 - MatrixPower[ρ, i].c0) // Simplify,
         ArrayFlatten[IdentityMatrix[2]⊗MatrixPower[Uρ, i]]}];

      AppendTo[SymmetryList,
        {MatrixPower[ρ, i].rxy, (c0 - MatrixPower[ρ, i].c0) // Simplify,
         ArrayFlatten[IdentityMatrix[2]⊗MatrixPower[Uρ, i]].
           ArrayFlatten[{{0, 1}, {1, 0}}⊗Urxy]}];
      , {i, 1, 4}];
```

Interpolation with symmetrization

We now interpolate between the Wannier90 data for 13 different rotations of the RuO octahedra: 0°
through 12° to generate a continuously θ dependent Hamiltonian, whilst ensuring that the interpolated model has the right symmetry (for which we are feeding the list SymmetryList):

In[456]:= `FileLocn = NotebookDirectory[] <> "unrenormalized2/";`

`W90Data = LoadInterpolateTBMs[{FileLocn <> "Sr2RuO4_0deg.dat",`
`    FileLocn <> "Sr2RuO4_1deg.dat", FileLocn <> "Sr2RuO4_2deg.dat",`
`    FileLocn <> "Sr2RuO4_3deg.dat", FileLocn <> "Sr2RuO4_4deg.dat",`
`    FileLocn <> "Sr2RuO4_5deg.dat", FileLocn <> "Sr2RuO4_6deg.dat",`
`    FileLocn <> "Sr2RuO4_7deg.dat", FileLocn <> "Sr2RuO4_8deg.dat",`
`    FileLocn <> "Sr2RuO4_9deg.dat", FileLocn <> "Sr2RuO4_10deg.dat",`
`    FileLocn <> "Sr2RuO4_11deg.dat", FileLocn <> "Sr2RuO4_12deg.dat"},`
`   {0, 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12}, "Symmetries" → SymmetryList,`
`   "OrbitalCentres" → AtomLocations, "ParameterSymbol" → "θ"];`

Invalid or no lattice vectors provided! Using unit vectors.

Tight-binding data files loaded.

Space dimension: 3

Number of orbitals: 6

Number of supercells: 49

Number of models/data-files to interpolate: 13

Number of lines in the data files:
 {741, 853, 853, 853, 845, 853, 853, 853, 853, 853, 853, 853, 853}

Interpolating between the Hamiltonians.

Valid symmetrization scheme provided! We will symmetrize the loaded Hamiltonian.

Number of supercells after symmetrization: 79

Since this is largely an exploratory analysis, we will not include the overall band renormalization or the spin orbit term which have minimal impact on the M point. We begin by adding a staggered chemical potential Δ

In[459]:= `Hamltn1[k1_, k2_, θ_, Δ_] = W90Data[[1]] + DiagonalMatrix[{Δ, Δ, Δ, -Δ, -Δ, -Δ}];`

Tuning to a HOS with non - zero Δ

We begin by defining a version of the Hamiltonian suitable for series expansion (once again in the bulk Brillouin zone basis for the sake of consistency)

In[460]:= `Ham[k_] := Hamltn1[k[[1]] - k[[2]], k[[1]] + k[[2]], k[[3]], k[[4]]]`

To compute the Hessian determinant with $\delta\theta$ and $\delta\Delta$ corrections, we need to series expand the dispersion to quadratic order with corrections. To find the bands closest to Fermi level to series expand, we first calculate the eigenvalues at the M point for a small $\Delta=0.005$ (that is 5 meV).

In[523]:= `Sort[Eigenvalues[Ham[{0, π // N, 8, 0.005}]]]`

Out[523]= `{-0.555813, -0.555813, -0.076319, -0.066319, 0.286847, 0.286847}`

It is clear that bands 3 and 4 are the bands of interest closest to the Fermi level. We will plot these bands with orbital projection once the critical θ has been figured out.

Band 3

In[480]:= `TaylA = Chop[TaylorExpandBand[Ham,`
    `{0, π // N, 8, 0.005}, 3, 2, "Dimension" → 2, "TuningDegree" → 2,`
    `"TuningSymbol" → {"δθ", "δΔ"}, "Messages" → False][[1]], 10⁻⁸];`

In[481]:= `Total[TaylA] /. {p1 → kₓ, p2 → k_y}`

Out[481]= $-0.076319 - 1.\, \delta\Delta + (-0.0205955 + 0.00119685\, \delta\theta)\, \delta\theta +$
$\left(-0.014985 + 8.34065 \times 10^{-8}\, \delta\Delta^2 + (0.0320869 - 0.000243806\, \delta\theta)\, \delta\theta\right) k_x^2 +$
$\left(-0.014985 - 8.34065 \times 10^{-8}\, \delta\Delta^2 + (0.0320869 - 0.000243806\, \delta\theta)\, \delta\theta\right) k_y^2$

We note that the quadratic term contains a very small $\delta\Delta$ correction. This indicates that the critical θ does not depend strongly on Δ (as long as it is non-zero and small. For large Δ, this analysis based on series expansion to quadratic order may not hold).

We can compute the Hessian determinant and solve for $\delta\theta$ by setting it to zero:

In[482]:= `HesDet = ((D[TaylA[[3]], {p1, 2}] × D[TaylA[[3]], {p2, 2}]) - (D[TaylA[[3]], p1, p2])²) /.`
    `{p1 → 0, p2 → 0, δλ → 0, δΔ → 0} // Simplify`
    `Soln1 = Solve[HesDet == 0, δθ]`

Out[482]= $0.000898201 - 0.00384658\, \delta\theta + 0.00414751\, \delta\theta^2 - 0.000062584\, \delta\theta^3 + 2.37766 \times 10^{-7}\, \delta\theta^4$

Out[483]= $\{\{\delta\theta \to 0.468682\}, \{\delta\theta \to 0.468682\}, \{\delta\theta \to 131.14\}, \{\delta\theta \to 131.14\}\}$

The critical θ to the first approximation is then given by

In[484]:= `TaylB = Chop[TaylorExpandBand[Ham, {0, π // N, 8 + δθ /. Soln1[[1]], 0.005},`
    `3, 2, "Dimension" → 2, "TuningDegree" → 2,`
    `"TuningSymbol" → {"δθ", "δΔ"}, "Messages" → False][[1]], 10⁻⁸];`

In[485]:= `Total[TaylB] /. {p1 → kₓ, p2 → k_y}`

Out[485]= $-0.0856538 - 1.\, \delta\Delta + (-0.0191726 + 0.00162093\, \delta\theta)\, \delta\theta +$
$\left(0.0000166482 - 3.94981 \times 10^{-8}\, \delta\Delta^2 + (0.0319471 - 0.000137449\, \delta\theta)\, \delta\theta\right) k_x^2 +$
$\left(0.0000166482 + 3.94981 \times 10^{-8}\, \delta\Delta^2 + (0.0319471 - 0.000137449\, \delta\theta)\, \delta\theta\right) k_y^2$

Indeed the coefficients of the quadratic terms have become quite small. We can repeat the process once again to obtain better convergence

In[486]:= `HesDet = ((D[TaylB[[3]], {p1, 2}] × D[TaylB[[3]], {p2, 2}]) - (D[TaylB[[3]], p1, p2])²) /.`
    `{p1 → 0, p2 → 0, δλ → 0, δΔ → 0} // Simplify`
    `Soln2 = Solve[HesDet == 0, δθ]`

Out[486]= $1.10864 \times 10^{-9} + 4.25488 \times 10^{-6}\, \delta\theta + 0.00408246\, \delta\theta^2 - 0.0000351289\, \delta\theta^3 + 7.55692 \times 10^{-8}\, \delta\theta^4$

Out[487]= $\{\{\delta\theta \to -0.000521115\}, \{\delta\theta \to -0.000521115\}, \{\delta\theta \to 232.429\}, \{\delta\theta \to 232.429\}\}$

Thus, the critical theta is given by

In[488]:= `θcrit = 8 + (δθ /. Soln1[[1]]) + (δθ /. Soln2[[1]])`

Out[488]= `8.46816`

Using this, we go ahead and define the Hamiltonian at the critical θ:

In[490]:= `HamCrit[k_] := Hamltn1[k[[1]] - k[[2]], k[[1]] + k[[2]], θcrit, 0.005]`

In[491]:= `TaylC =`
`    Chop[TaylorExpandBand[HamCrit, {0, π // N}, 3, 4, "Messages" → False][[1]], 10^-8];`

In[492]:= `Total[TaylC] /. {p1 → k_x, p2 → k_y}`

Out[492]= $-0.0856438 - 0.630949 k_x^4 + 0.102619 k_x^3 k_y + 0.860439 k_x^2 k_y^2 - 0.102619 k_x k_y^3 - 0.630949 k_y^4$

This is obviously a singularity belonging to the X_9 class. We can confirm this explicitly

In[493]:= `DiagnoseHOS[Total[TaylC] /. {p1 → k_x, p2 → k_y}]`

Polynomial has corank 2. It is 5-determinate. Its determinacy is either 5 or 4
  and its codimension is 8. Comparing with the known list of singularities:

Polynomial has a catastrophe belonging to the X_9 singularity class.

Out[493]= {2, 8, 5}

We can also explicitly check that this satisfies the four fold rotation symmetry $(k_x, k_y) \rightarrow (k_y, -k_x)$ but not the mirror symmetry $(k_x, k_y) \rightarrow (k_y, k_x)$ since the latter is broken while the former is preserved under the staggered chemical potential Δ.

In[498]:= `Chop[((TaylC /. {p1 → k_x, p2 → k_y}) - (TaylC /. {p1 → k_y, p2 → -k_x})) // FullSimplify,`
`    10^-6] // Total`
`Chop[((TaylC /. {p1 → k_x, p2 → k_y}) - (TaylC /. {p1 → k_y, p2 → k_x})) // FullSimplify,`
`    10^-6] // Total`

Out[498]= `0`
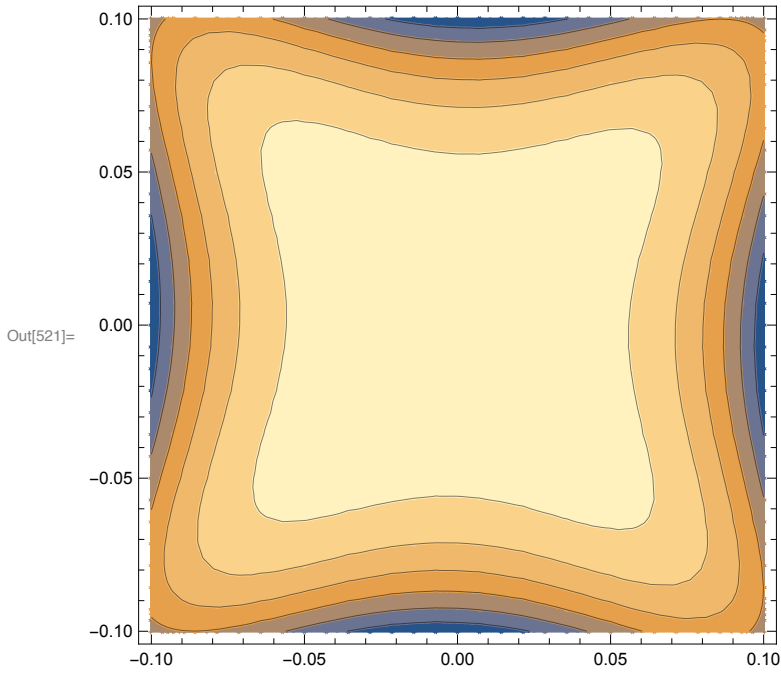
Out[499]= $0.205239 k_x^3 k_y - 0.205239 k_x k_y^3$

The 3D and surface plot of the dispersion

In[522]:= `Plot3D[Total[TaylC], {p1, -1/4, 1/4}, {p2, -1/4, 1/4}]`

Out[522]=
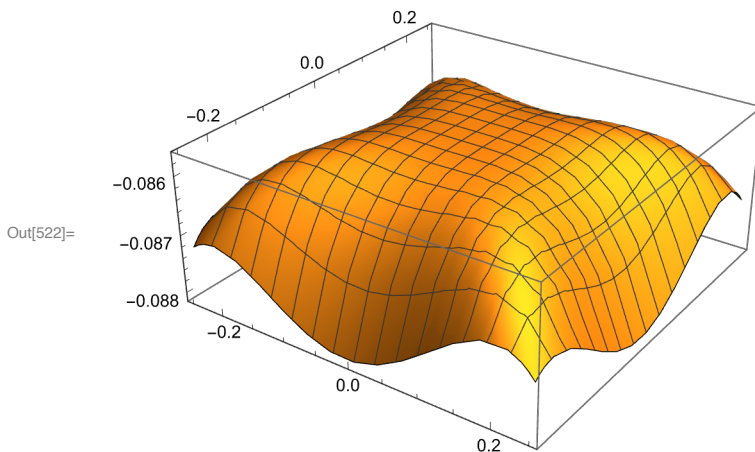
In[521]:= `ContourPlot[Total[TaylC], {p1, `$\frac{-1}{10}$`, `$\frac{1}{10}$`}, {p2, `$\frac{-1}{10}$`, `$\frac{1}{10}$`}]`

Out[521]=

These indicate that this is a four fold maximum rather than a saddle.

Band 4

In[500]:= `TaylA2 = Chop[TaylorExpandBand[Ham,`
`    {0, π // N, 8, 0.005}, 4, 2, "Dimension" → 2, "TuningDegree" → 2,`
`    "TuningSymbol" → {"δθ", "δΔ"}, "Messages" → False][[1]], 10`$^{-8}$`];`

In[501]:= `Total[TaylA2] /. {p1 → k`$_x$`, p2 → k`$_y$`}`

Out[501]= $-0.066319 + 1. \, \delta\Delta + (-0.0205955 + 0.00119685 \, \delta\theta) \, \delta\theta +$
$(-0.014985 - 8.34065 \times 10^{-8} \, \delta\Delta^2 + (0.0320869 - 0.000243806 \, \delta\theta) \, \delta\theta) \, k_x^2 +$
$(-0.014985 + 8.34065 \times 10^{-8} \, \delta\Delta^2 + (0.0320869 - 0.000243806 \, \delta\theta) \, \delta\theta) \, k_y^2$

We note that the quadratic term contains a very small $\delta\Delta$ correction. This indicates that the critical θ does not depend strongly on Δ (as long as it is non-zero and small. For large Δ, this analysis based on series expansion to quadratic order may not hold).

We can compute the Hessian determinant and solve for $\delta\theta$ by setting it to zero:

In[502]:= `HesDet =`
`    ((D[TaylA2[[3]], {p1, 2}] × D[TaylA2[[3]], {p2, 2}]) - (D[TaylA2[[3]], p1, p2])`^{2}`) /.`
`    {p1 → 0, p2 → 0, δλ → 0, δΔ → 0} // Simplify`
`Soln12 = Solve[HesDet == 0, δθ]`

Out[502]= $0.000898201 - 0.00384658 \, \delta\theta + 0.00414751 \, \delta\theta^2 - 0.000062584 \, \delta\theta^3 + 2.37766 \times 10^{-7} \, \delta\theta^4$

Out[503]= $\{\{\delta\theta \to 0.468682\}, \{\delta\theta \to 0.468682\}, \{\delta\theta \to 131.14\}, \{\delta\theta \to 131.14\}\}$

The critical θ to the first approximation is then given by

In[504]:= `TaylB2 = Chop[TaylorExpandBand[Ham, {0, π // N, 8 + δθ /. Soln12〚1〛, 0.005},`
        `4, 2, "Dimension" → 2, "TuningDegree" → 2,`
        `"TuningSymbol" → {"δθ", "δΔ"}, "Messages" → False]〚1〛, 10⁻⁸];`

In[505]:= `Total[TaylB2] /. {p1 → k_x, p2 → k_y}`

Out[505]= $-0.0756538 + 1. \, \delta\Delta + (-0.0191726 + 0.00162093 \, \delta\theta) \, \delta\theta +$
$(0.0000166482 - 9.02946 \times 10^{-8} \, \delta\Delta^2 + (0.0319471 - 0.000137449 \, \delta\theta) \, \delta\theta) \, k_x^2 +$
$(0.0000166482 + 9.02946 \times 10^{-8} \, \delta\Delta^2 + (0.0319471 - 0.000137449 \, \delta\theta) \, \delta\theta) \, k_y^2$

Indeed the coefficients of the quadratic terms have become quite small. We can repeat the process once again to obtain better convergence

In[506]:= `HesDet =`
        `((D[TaylB2〚3〛, {p1, 2}] × D[TaylB2〚3〛, {p2, 2}]) - (D[TaylB2〚3〛, p1, p2])²) /.`
        `{p1 → 0, p2 → 0, δλ → 0, δΔ → 0} // Simplify`
        `Soln22 = Solve[HesDet == 0, δθ]`

Out[506]= $1.10864 \times 10^{-9} + 4.25488 \times 10^{-6} \, \delta\theta + 0.00408246 \, \delta\theta^2 - 0.0000351289 \, \delta\theta^3 + 7.55692 \times 10^{-8} \, \delta\theta^4$

Out[507]= $\{\{\delta\theta \to -0.000521115\}, \{\delta\theta \to -0.000521115\}, \{\delta\theta \to 232.429\}, \{\delta\theta \to 232.429\}\}$

Thus, the critical theta is given by

In[508]:= `θcrit2 = 8 + (δθ /. Soln12〚1〛) + (δθ /. Soln22〚1〛)`

Out[508]= `8.46816`

Using this, we go ahead and define the Hamiltonian at the critical θ:

In[510]:= `HamCrit2[k_] := Hamltn1[k〚1〛 - k〚2〛, k〚1〛 + k〚2〛, θcrit2, 0.005]`

In[511]:= `TaylC2 =`
        `Chop[TaylorExpandBand[HamCrit2, {0, π // N}, 4, 4, "Messages" → False]〚1〛, 10⁻⁸];`

In[512]:= `Total[TaylC2] /. {p1 → k_x, p2 → k_y}`

Out[512]= $-0.0756438 + 0.201354 \, k_x^4 - 0.102619 \, k_x^3 \, k_y - 0.804168 \, k_x^2 \, k_y^2 + 0.102619 \, k_x \, k_y^3 + 0.201354 \, k_y^4$

This is obviously a singularity belonging to the X_9 class. We can confirm this explicitly

In[513]:= `DiagnoseHOS[Total[TaylC2] /. {p1 → k_x, p2 → k_y}]`

```
Polynomial has corank 2. It is 5-determinate. Its determinacy is either 5 or 4
  and its codimension is 8. Comparing with the known list of singularities:
Polynomial has a catastrophe belonging to the X₉ singularity class.
```

Out[513]= `{2, 8, 5}`

We can also explicitly check that this satisfies the four fold rotation symmetry $(k_x, k_y) \to (k_y, -k_x)$ but not the mirror symmetry $(k_x, k_y) \to (k_y, k_x)$ since the latter is broken while the former is preserved under the staggered chemical potential Δ.

In[514]:= `Chop[((TaylC2 /. {p1 → k_x, p2 → k_y}) - (TaylC2 /. {p1 → k_y, p2 → -k_x})) //`
`    FullSimplify, 10^{-6}] // Total`
`Chop[((TaylC2 /. {p1 → k_x, p2 → k_y}) - (TaylC2 /. {p1 → k_y, p2 → k_x})) // FullSimplify,`
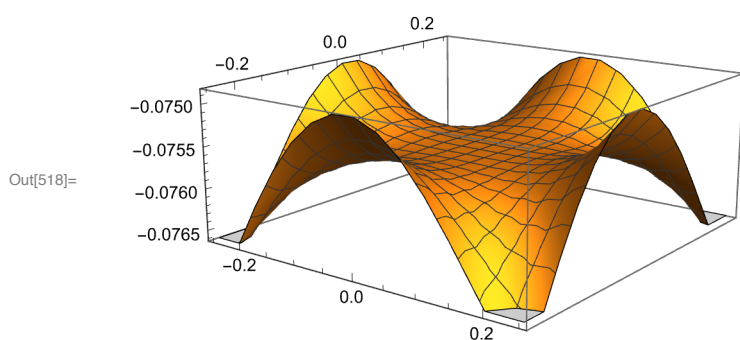`    10^{-6}] // Total`

Out[514]= `0`

Out[515]= $-0.205239\ k_x^3\ k_y + 0.205239\ k_x\ k_y^3$

The 3D and surface plot of the dispersion

In[518]:= `Plot3D[Total[TaylC2], {p1, -1/4, 1/4}, {p2, -1/4, 1/4}]`

Out[518]=

In[520]:= `ContourPlot[Total[TaylC2], {p1, -1/10, 1/10}, {p2, -1/10, 1/10}]`

Out[520]=
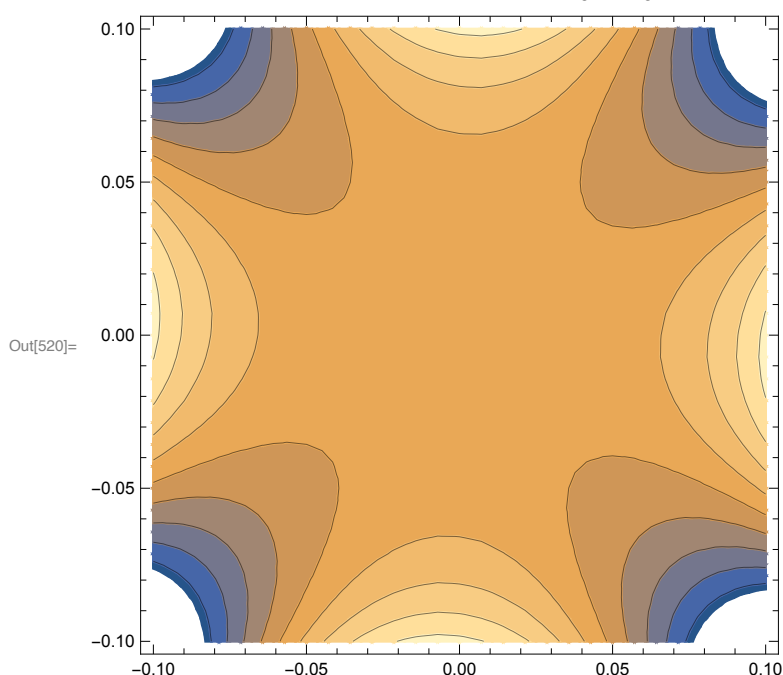
These confirm that this is a genuine four fold saddle.

Comparing bands 3 and 4

We can finally compare the HOS in band 3 and band 4 (recall that these bands were degenerate before the staggered chemical potential was applied)

In[516]:= `Total[TaylC] /. {p1 → k_x, p2 → k_y}`
`Total[TaylC2] /. {p1 → k_x, p2 → k_y}`

Out[516]= $-0.0856438 - 0.630949 \, k_x^4 + 0.102619 \, k_x^3 \, k_y + 0.860439 \, k_x^2 \, k_y^2 - 0.102619 \, k_x \, k_y^3 - 0.630949 \, k_y^4$

Out[517]= $-0.0756438 + 0.201354 \, k_x^4 - 0.102619 \, k_x^3 \, k_y - 0.804168 \, k_x^2 \, k_y^2 + 0.102619 \, k_x \, k_y^3 + 0.201354 \, k_y^4$

It is clear that while band 3 (with negative k_x^4 and k_y^4 terms) and band 4 (with positive k_x^4 and k_y^4 terms) disperse roughly in the opposite fashion. This is obviously an approximate qualification since the $k_x^2 \, k_y^2$ terms also play a role. We now plot the band structure with orbital projections

In[524]:= `δk = 3.869 × 0.35;`
`HSymmPts = {{{-δk, π - δk}, "← -M_x"}, {{0, π}, "M_y"}, {{0, π + δk + 0.01}, "Γ →"}};`
`Bnds = GenerateBandStructure[HamCrit, HSymmPts,`
    `"OrbitalProjection" → True, "OrbitalGrouping" → {{1, 4}, {2, 5}, {3, 6}}];`

In[536]:= `PlotBandStructure[Bnds, (*{-0.042,0.012}*){-0.2, 0.1}, AspectRatio → 3/4,`
  `"yLabel" → {(*Label*)"E (meV)", (*Font Size*)13, (*Font Color*)`
    `Black, (*Font Family*)"Arial"}, "xLabel" → {13, Black, "Arial"},`
  `"yTicks" → {12, Black, "Helvetica"}, "LineThickness" → 0.008,`
  `"LineColorScheme" → {Red, Brown, Blue}, (*Dividing Lines*)"DividingLines" →`
   `{(*Dashing[..]*)0.01, (*Color*)Gray, (*Thickness*)0.004},`
  `(*,Legend*)"PlotKeyLegend" → {{"d_xz", "d_yz", "d_xy"}, {0.25, 0.85}}]`

Out[536]=
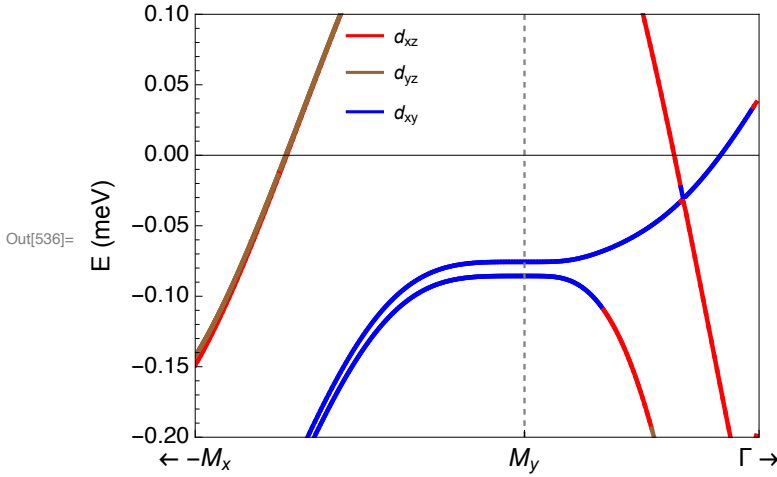
We notice that the M point singularities arise primarily from the d_{xy} orbitals. Further the higher order saddle nature of band 4 and the higher order maximum nature of band 3 are quite apparent.