Supplement for "Electric field control of phonon angular momentum in perovskite BaTiO$_3$"

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The python code provided with this supplement can be used to construct spring models used in the manuscript. The main function that creates a spring model is called `ph_model`. This function specifies parameters of the model such as number of periodic dimensions in the model (`dim_k`), number of dimensions of atomic displacements (`dim_d`), periodic lattice vectors (`lat`), Cartesian coordinates of atoms (`crt`), and masses of atoms (`mas`). Here is the syntax used to call this function,

```
ph_model(dim_k, dim_d, lat, crt, mas)
```

Once the model has been created with `ph_model` we use another function, called `set_spring` to specify the springs in the model. Here is the syntax of function `set_spring`,

```
set_spring(k_stretch, k_bending, i, j, R)
```

Here `k_stretch` is the K_r parameter from the main paper specifying the stretching strength of the spring. Similarly `k_bending` specifies K_θ parameter. Finally, `i` specifies index of the first atom connected to this spring (atoms are indexed so that the first atom has index `0` not `1`), `j` specifies the second atom, and `R` specifies the unit cell vector of the second atom in terms of the periodic lattice vectors (`lat`). As periodicity is implicitly assumed in the code, it is enough to specify a spring between `i`-th and `j+R`-th atom, as this automatically specifies equivalent springs between `i+P`-th and `j+R+P`-th atom for all lattice vectors `P`. Similarly, by hermiticity specifying spring between `i`-th and `j+R`-th atom also specifies the same spring between `j`-th and `i-R`-th atom.

The following snippets of python code generate the models used in the main manuscript. The complete code that generates these models and creates all figures in this document are provided in the supplementary material.

In all cases we use the following, somewhat arbitrarily selected, numerical parameters in the code,

```
kr  = 1.0
krS = 1.5
krL = 1.0
ktheta  = 0.20
kthetaS = 0.24
kthetaL = 0.18
delta = 0.1
m1 = 2.5
m2 = 1.0
```

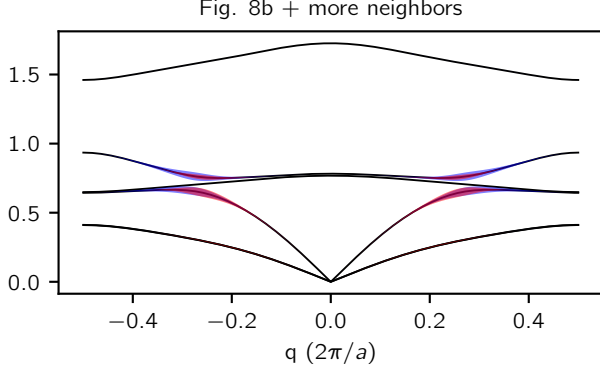## A. Model shown in Fig. 8 (b) in the main text

The model shown in Fig. 8 (b) consists of a chain of two atoms connected with a spring. One of the atoms is displaced perpendicular to the chain direction. This model can be specified with the following python code (see supplementary material for the entire code),

```
model = ph_model(dim_k = 1,
                 dim_d = 3,
                 lat = [[0.0, 0.0, 1.0]],
                 crt = [[0.0, delta*0.5, 0.0],
                        [0.0, 0.0       , 0.5]],
                 mas = [m1, m2])
model.set_spring(kr, ktheta, 0, 1, [ 0])
model.set_spring(kr, ktheta, 0, 1, [-1])
```

Below is the resulting phonon band structure.
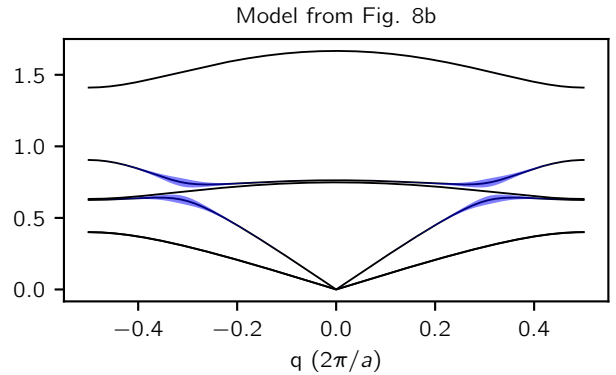
Blue color indicates phonon bands which have non-zero contribution of a single atom to the phonon angular momentum. Thickness of the blue curve is proportional to the contribution to the phonon angular momentum. As discussed in the main text, in this model total phonon angular momentum is zero, as contributions from two atoms in the unit cell cancel each other. This cancellation does not occur in the two-dimensional version of the model (as discussed shortly) or if we include springs between further neighboring atoms, as shown in the following figure.

Fig. 8b + more neighbors

Here the red region is enhanced about 2000 times relative to the blue region.

B. Model shown in Fig. 8 (c) in the main text

The model from Fig. 8 (c) can be generated by the following code,

```
model = ph_model(dim_k = 1,
                 dim_d = 3,
                 lat = [[0.0, 0.0, 1.0]],
                 crt = [[0.0, 0.0, delta*0.5],
                        [0.0, 0.0,        0.5]],
                 mas = [m1, m2])
model.set_spring(krS, kthetaS, 0, 1, [ 0])
model.set_spring(krL, kthetaL, 0, 1, [-1])
```

The resulting phonon band structure is,

Model from Fig. 8c
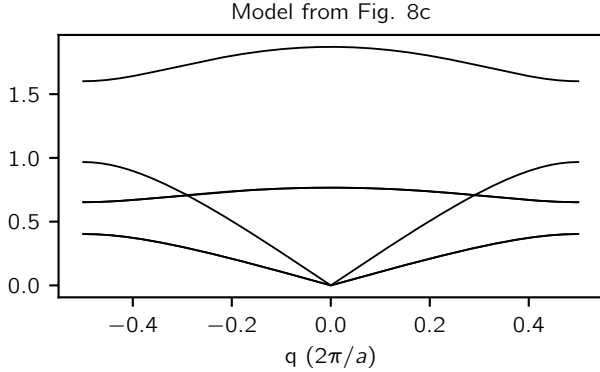
As discussed in the main text, this model has both zero phonon angular momentum and zero contribution to the phonon angular momentum from a single atom in the unit cell. The same is true even if we include springs between further neighboring atoms.

C. Two-dimensional model shown in Fig. 9 in the main text

Finally, the two-dimensional model from Fig. 9 in the main text can be generated with the following code,

```
model = ph_model(dim_k = 2,
                 dim_d = 3,
                 lat = [[1.0, 0.0, 0.0],
                        [0.0, 1.0, 0.0]],
                 crt = [[0.0, 0.0, delta*0.5],
                        [0.5, 0.0,        0.0],
                        [0.0, 0.5,        0.0]],
                 mas = [m1, m2, m2])
model.set_spring(kr, ktheta, 0, 1, [ 0, 0])
model.set_spring(kr, ktheta, 0, 2, [ 0, 0])
model.set_spring(kr, ktheta, 0, 1, [-1, 0])
model.set_spring(kr, ktheta, 0, 2, [ 0,-1])
```

The resulting phonon band structure shows again in blue contribution of individual atom to the phonon angular momentum. On the other hand, red color is used to show the total phonon angular momentum, when summed over all atoms in the unit cell. As discussed in the main text, in this model the total phonon angular momentum is non-zero. The trajectory in the reciprocal space of phonon wavevectors is a low-symmetry line that avoids all high-symmetry points in the Brillouin zone.

Model from Fig. 9
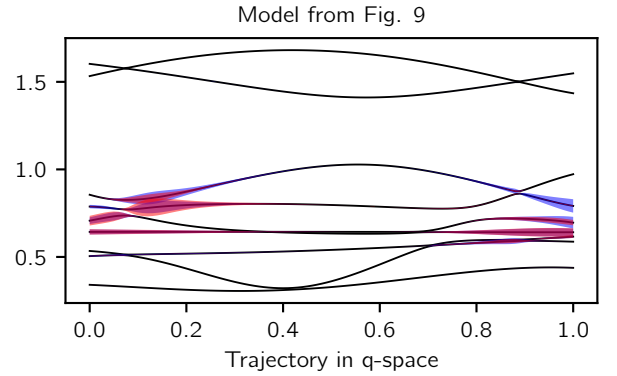
The two-dimensional histogram of the phonon angular moment for this model is shown below.

Model from Fig. 9 (histogram)
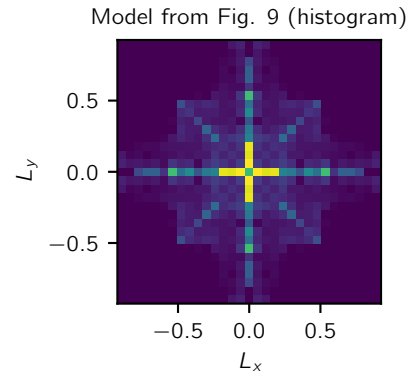
As can be seen from the figure, the qualitative features of the phonon angular momentum in this model are similar to that we found from first-principles, as shown in Fig. 2 in the main text.