Robust flat bands in $RCo_5$ compounds

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The mechanism to realize the peculiar flat bands generally existing in $RCo_5$ ($R$=rare earth) compounds is clarified by analyzing the first-principles band structures and the tight-binding model. These flat bands are constructed from the localized eigenstates, the existence of which is guaranteed by the partial cancelation between the inter-site hopping amplitudes among the Co-3$d$ states at the Kagomé sites and those between the Kagomé and honeycomb sites. Their relative positions to other bands can be controlled by varying the lattice parameters keeping their dispersion almost flat, which suggests the possibility of the flat-band engineering.

The nature of flat bands has been attracting much attention from a variety of view points. Ferromagnetism originating from the flat band has been intensively studied in some model systems [1-3]. The nearly flat band with a non-zero Chern number offers a unique playground of the fractional quantum Hall effect [7-10]. An extremely large effective mass for the flat band affects the transport properties of solids and results in various unconventional phenomena such as the inverse Anderson transition [11]. A sharp peak of the density of states (DOS) owing to partially flat dispersion is preferred for the thermoelectric devices to enhance their thermoelectric performance [12-15].

Owing to these various intriguing aspects, seeking for the flat bands in real materials is of significant importance for materials design. One example of experimental realization of the flat band was reported for Cu$(1,3$-tetracyanoquinodimethane) [16], which can be described by a single-orbital model on the Kagomé lattice. Another example is the tetragonal cuprate La$_5$Ba$_2$Cu$_{10}$O$_{16}$ [3, 18, 19], in which the electronic structure for the flat band plays a central role for determining its magnetic order [20]. Also other materials have been reported to possess the flat bands [21-23].

Recently, the intermetallic ferromagnet YCo$_5$ (Fig. 1(a)-(b)), which is known for its large magnetic anisotropy, was pointed out to have peculiar flat bands [24, 25]. It was found that the system experiences the first-order Lifshitz transition [28] and exhibits an unusual isomorphic lattice collapse with a sudden change of the magnetic moment when the Fermi level crosses the flat band (along the $\Gamma$-M-K-$\Gamma$ line in Fig. 1(c)) by applying the pressure. This flat band is observed in the whole $k_z = 0$ plane and consists of $3d_{xz}$ and $3d_{yz}$ states of Co atoms [27]. Similar flat bands were found in other $RCo_5$ ($R$=rare earth) compounds such as LaCo$_5$ [27], SmCo$_5$ [29], and also CePt$_5$ [30], which has the same CaCu$_5$-type structure. However, the mechanism to realize such ubiquitous flat bands has not been clarified yet.

In this Letter, we revealed the origin of the flat bands in $RCo_5$ compounds and attributed their existence to the localized eigenstates realized by the destructive interference among Co-3$d$ orbitals on the Kagomé-honeycomb stacked structure (see Fig. 1(a)-(b)). These flat bands are affected very little by varying the lattice parameters while their relative positions to other bands are altered. Such robustness and controllability of the flat bands are crucial for the system to exhibit the first-order Lifshitz transition where the flat dispersion should be retained under the pressure, and also pave the way for the flat-band engineering. Our mechanism can be applied also to other CaCu$_5$-type structures with $d$-orbitals on Cu sites. In this sense, this study demonstrates a new general mechanism to realize the flat dispersion for a wide range of materials.

For $ab$ initio band structure calculations, we used the Perdew-Wang local-spin-density approximation [31] and the full-potential linearized augmented plane-wave method as implemented in WIEN2K code [32]. For simplicity, we do not include the spin-orbit coupling throughout this Letter since it affects very little to the flat bands. Experimental lattice parameters ($a = 9.313$ a.u. and $c/a = 0.806$) are taken from Ref. [33]. The muffin-tin radii for Co and Y atoms, $r_{Co}$ and $r_Y$, were set to 2.34 and 2.07 a.u., respectively. The maximum modulus for the reciprocal lattice vectors $K_{max}$ was chosen so that $r_Y \times K_{max} = 9.00$.

As was pointed out in Ref. [27], the flat bands in YCo$_5$ exist (i) on the $k_z = k_y = 0$ line and (ii) in the $k_z = 0$ plane (see the flat bands along the $\Gamma$-A and the $\Gamma$-M-K-$\Gamma$ lines, respectively, in Fig. 1(c)-(e)). At a glance these two flat bands have similar eigenvalues, but have different origins as shown later in this Letter. In addition, we shall see that their eigenvalues become different when we change the lattice parameters. Therefore, we shall
individually analyze their origins.

For this purpose, we constructed a tight-binding model consisting of Co-3\textit{d} states with all possible hopping paths taken into account [35]. We neglected Co-4\textit{s} and Y states because they were found not to contribute to these flat bands in our first-principles band structure calculations.

To construct the tight-binding model, we employed spin-unpolarized calculations because the flat bands also appear there and our story does not depend on whether the spin is polarized or unpolarized. In Fig. (1c), we can see that our tight-binding model reproduces the first-principles band structure well.

To obtain the flat band by the destructive interference [2,6], the corresponding Bloch states $\phi_k(\mathbf{r})$ for the crystal wave vectors $\mathbf{k}$ are constructed as follows:

$$
\phi_k(\mathbf{r}) = \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot \mathbf{R}} \varphi(\mathbf{r} - \mathbf{R}),
$$

where $\varphi(\mathbf{r})$ is the localized eigenstate that is confined in a finite region and $\mathbf{R}$ runs all the lattice vectors. We can easily see that the eigenvalues for $\phi_k$ are independent on $\mathbf{k}$. We made two extensions for this theory. First, the range of $\mathbf{R}$ in this summation is restricted to its subspace represented as $(0,0, R_z)$ and $(R_x, R_y, 0)$ for the $k_x = k_y = 0$ and $k_z = 0$ flat bands, respectively. By this extension, we can deal with not only the flat band in the whole Brillouin zone but also that in some subspace. Second, the localized eigenstate can have a damping tail and is not necessarily confined in a finite region as long as the summation in this equation is well-defined, i.e., a weight of the Bloch wave function on every site does not diverge [38]. In this study, we obtained the localized eigenstates (in an extended meaning as mentioned above) by Fourier transform of the Bloch states of the corresponding flat bands in our tight-binding model.

\begin{enumerate}
\item \textit{The flat band on the $k_x = k_y = 0$ line}
\end{enumerate}

Fig. 2(a) presents a schematic picture of the localized eigenstate for the $k_x = k_y = 0$ flat band in our tight-binding model. This state infinitely extends to $x$ and $y$ directions, but is confined in a Kagomé plane and does not have any weight in the honeycomb planes, which allows an infinite summation over $R_z$ in Eq. (1). This state consists of $3d_{x^2-y^2}$ and $3d_{xy}$ states of Co atoms in the Kagomé plane.

We can easily see that this localized state $\varphi(\mathbf{r})$ is the exact eigenstate of our tight-binding Hamiltonian $\mathcal{H}$ by considering the hopping amplitudes between atomic orbitals $\varphi_{\text{atom}}(\mathbf{r})$ and the localized state, i.e., $\langle \varphi_{\text{atom}} | \mathcal{H} | \varphi \rangle$, as follows. It is obvious that the hopping amplitudes between the localized state and any other Co-3\textit{d} states in the Kagomé plane are zero. In addition, those for any Co states in the honeycomb planes also disappears by cancelation of transfers thanks to two symmetry operations depicted in Fig. 2(b). Thus, any atomic orbitals other than those depicted in Fig. 2(a) do not appear in $\mathcal{H}\varphi$. Along with this fact, the equivalence between all the atomic orbitals depicted in Fig. 2(a) results in their same weights also in $\mathcal{H}\varphi$, so that this localized state is the exact eigenstate of our tight-binding Hamiltonian with all possible hopping paths taken into account. Also there are no hopping amplitudes between the localized eigenstate and the Co-4\textit{s} or Y states.

Realization of this localized eigenstate is not affected by any lattice parameter such as the Kagomé-honeycomb distance, so that the resulting flat band is ubiquitous in many materials with the same structures having $d$ orbitals on the Kagomé sites [39, 40]. The same story also holds when $p$ orbitals exist on the honeycomb sites. For example, YCr$_6$Ge$_6$, where Cr and Ge atoms constitute the Kagomé and honeycomb lattices respectively, has the same flat band [40].

\begin{enumerate}
\item \textit{The flat band in the $k_z = 0$ plane}
\end{enumerate}

Fig. 2(a) presents a schematic picture of the localized eigenstate for the $k_z = 0$ flat band in our tight-binding model. This state consists of Co-3$d_{xz}$ and 3$d_{yz}$ states [27] both on Kagomé and honeycomb planes. Since we consider the $k_z = 0$ state here, each atomic orbital is defined as an infinite sum over the $z$ direction with the same phase, and the localized eigenstate also infinitely extends to the $z$ direction. It is important that atomic orbitals except $3d_{xz}$ and $3d_{yz}$ states are not involved owing to the mirror symmetry with respect to the Kagomé plane, i.e., only $3d_{xz}$ and $3d_{yz}$ states have odd parity for this reflection and other orbitals have even parity. Atomic orbitals
FIG. 2: (color online). (a) Schematic picture of the localized eigenstate in a Kagomé plane for the \( k_x = k_y = 0 \) flat band. (b) Two symmetry operations of this state: reflection and \( C_3 \) rotation.

on the honeycomb planes are also depicted in this figure, where the signs of all the orbitals at the middle between the Kagomé and the honeycomb planes are shown. We observed an interesting step-by-step damping of the orbital weights; the relative weights of the large, middle, and small orbitals depicted in Fig. 3(a) are about 31 : 4 : 1 in our calculation. Note that the sizes of these orbitals are depicted just schematically and the same sizes only mean the similar weights.

Two important cancelations to realize the localization of the eigenstate are shown in Fig. 3(b)-(c). In Fig. 3(b), the hopping amplitudes among the Kagomé sites are partially canceled with that between the Kagomé and honeycomb sites. By this partial cancelation, the step-by-step damping of the localized state is realized. The other cancelation is shown in Fig. 3(c), where the hopping amplitudes to an atomic orbital depicted in the center of this figure are canceled, and then this orbital is not involved with the localized eigenstate. Without this cancelation, a large number of the orbitals are involved with this eigenstate, which will prevent its localization. Though, in fact, we observed some tilting of the outer orbitals in Fig. 3(a), which might be due to distant transfers other than the nearest-neighbor Co(3g)-Co(3g) and Co(3g)-Co(2c) hoppings.

Whereas our tight-binding model takes all possible hopping processes into consideration, to obtain clear understanding of our mechanism, let us first focus on the nearest-neighbor Co(3g)-Co(3g) and Co(3g)-Co(2c) hoppings. Then the state in Fig. 3(a) can be the eigenstate, and in addition, our localized eigenstate still leads to the flat dispersion even if one takes into account the nearest-neighbor Co(2c)-Co(2c) hopping except the \( \delta \)-bonding contribution, i.e., \((dd\delta)\) in the Slater-Koster parameterization [11]. This is because the weights of the orbitals on the honeycomb planes for our localized state can be constructed as the linear combination of the localized eigenstates in the honeycomb lattice, which is realized when \((dd\delta)\) (or \((pp\pi)\)) if \( p \) orbitals are considered is neglected [22], as shown in Fig. 3(d). Owing to this fact, the Co(2c)-Co(2c) hopping except \((dd\delta)\) only shifts the on-site energy of the honeycomb sites. The hopping processes neglected in this discussion can only slightly change the localized eigenstate depicted in Fig. 3(a) because their amplitudes are relatively small.

Based on these observation, we shall show the robust existence of the localized eigenstate as depicted in Fig. 3(a) on the Kagomé-honeycomb stacked structure of Co atoms in RC05 compounds, both by the analysis of the tight-binding model and by the observation of the first-principles band structures. For the former purpose, we considered a simplified situation as shown in Fig. 4(a) only with the nearest-neighbor Co(3g)-Co(3g), Co(3g)-Co(2c), and partial Co(2c)-Co(2c) hoppings (i.e., the hopping process between the sites labeled \( x_2 \) and \( x_4 \) in Fig. 4(b) is taken into account but that between the sites labeled \( x_2 \) and \( \alpha^{-1}x_4 \) or between the sites labeled \( x_4 \) and \( \alpha x_2 \) is not). The weight of each atomic orbital in Fig. 4(a) is shown in Fig. 4(b), where inequivalent
sites are named 1, 2, 3, and 4 that have weights $x_1$, $x_2$, $x_3$, and $x_4$. We assumed the exponential decay of the weight on each site with a damping factor $\alpha$ as presented in Fig. 4(b), which is fast enough to obtain the well-defined Bloch states in Eq. (1). In addition, the boundaries among the regions weighted as ..., $\alpha^{-1}$, $\alpha^0$, $\alpha$, ..., are parallel in Fig. 4(a) and not concentric as in Fig. 3(a) to avoid complex mathematical treatment. The region far from the center of the localized eigenstate in Fig. 3(a) is expected to be well described with our simplified picture. There are two conditions to be satisfied: (I) the exponentially decaying state shown in Fig. 4(a) and (b) is an eigenstate and (II) the cancelation as shown in Fig. 3(d) occurs at the site 3 to have the orbital depicted in the center of Fig. 3(d) not involved with our eigenstate. We do not require $|\alpha| < 1$ because even if we obtain a solution with $|\alpha| > 1$, a damping factor $1/\alpha$ obviously yields another solution. Using the hopping amplitudes defined in Fig. 4(c) along with $t_5$, $\epsilon_h$, $\epsilon_{k1}$, and $\epsilon_{k3}$ that denote the hopping amplitude between the sites 2 and 4 (Co(2c)-Co(2c)), the on-site energies of the honeycomb site, sites 1 and 3 of the Kagomé lattice, respectively, we can derive the condition [43]: If we can choose $\alpha$ to have the eigenvector $(x_1, x_2, x_3, x_4)$ of the matrix
\[
\begin{pmatrix}
\epsilon_{k1} & t_3 & 2(1 + \alpha^{-1})t_4 & \alpha^{-1}t_3 \\
t_3 & \epsilon_h + 2t_5 & 2t_2 & 0 \\
2(1 + \alpha)t_4 & 2t_2 & 2\epsilon_{k3} + 4t_1 & 2t_2 \\
\alpha t_3 & 0 & 2t_2 & \epsilon_h + 2t_5
\end{pmatrix}
\]
(2)
satisfies
\[-(1 + \alpha)t_1x_1 + \frac{t_3}{2}(x_2 + x_4) + 2t_4x_3 = 0,
\]
(3)
then the exponentially decaying state as shown in Fig. 4(a) exists. We can prove that this condition is always satisfied for arbitrary real values of the tight-binding parameters [43]. Though an effect of the long-ranged hopping amplitudes we neglected here is non-trivial and it is difficult to provide a rigorous relationship between the exponentially decaying eigenstate shown in Fig. 4(a) and the localized eigenstate depicted in Fig. 3(a), the above analysis supports the robustness of the flat band.

Let us now move on to the first-principles calculations to demonstrate that these two flat bands are affected very little by varying the lattice parameters. Fig. 5 shows the band structures of YCo$_5$ with lattice parameters (a) increased or (b) decreased by 10% isotropically, and those with only the $c$ axis (c) increased or (d) decreased by 10%. Though the flat dispersion was verified to be remained by a small variation of the lattice parameters such as one for $c/a$ lesser than 1% in previous studies [28, 29], it is surprising that it is almost completely retained by such large variation of the lattice parameters whereas other band dispersions are affected. The relative positions of the flat bands to other bands and the Fermi level are different among these figures. Such controllability suggests the possibility of the flat-band engineering. In Fig. 5(c)-(d), the $k_z = 0$ flat bands acquire slight dispersion, which might be due to the issues mentioned in the last of the previous paragraph. Related to the robustness of the flat bands, as mentioned in the introduction of this Letter, other $R$Co$_5$ compounds [27, 29] and also CePt$_5$ [30], which has the same CaCu$_5$-type structure, are reported to have the same flat bands. This is because our mechanism can be obviously applied to all CaCu$_5$-type structures with $d$-orbitals on Cu sites.

To conclude, we found that the realization of the flat bands in $R$Co$_5$ compounds can be attributed to the existence of the localized eigenstate defined in particular directions. Their flat dispersion is very robust against the variation of the lattice parameters whereas their position can be controlled by it, which offers the possibility of the flat-band engineering.

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SUPPLEMENTAL MATERIAL

DERIVATION OF EQS. (2) AND (3) IN THE MAIN TEXT

The eigenvalue equation for the Hamiltonian of the system presented in Fig. 4(a) in the main text is

\[
\begin{pmatrix}
\ddots & 2\epsilon_k + 4t_1 & 2t_2 & 2t_4 \\
2t_2 & \epsilon_h + 2t_5 & t_3 & \ddots \\
t_4 & 2t_4 & \epsilon_{k1} & t_3 \\
\end{pmatrix}
\begin{pmatrix}
\alpha^{-1}x_3 \\
x_1 \\
x_2 \\
x_3 \\
x_4 \\
\end{pmatrix}

= \lambda
\begin{pmatrix}
\ddots \\
\alpha^{-1}x_3 \\
\alpha^{-1}x_4 \\
x_1 \\
x_2 \\
x_3 \\
\alpha x_1 \\
\alpha x_2 \\
\ddots \\
\end{pmatrix}
\]  

(4)

where \( \lambda \) is the eigenvalue and the eigenvector is assumed to be the above form as presented in Fig. 4(b) in the main text. Then we can immediately obtain Eq. (2) in the main text.

To derive Eq. (3) in the main text, we consider the cancelation of the hopping amplitudes to the site 3 as presented in Fig. 1 in this supplemental material, where the central orbital is rotated by 90 degrees clockwise from that of Fig. 4(a) in the main text to represent the cancelation shown in Fig. 3(c) in the main text.

FIG. 1: Cancelation of the hopping amplitudes in derivation of Eq. (3) in the main text. This is a part of Fig. 4(a) in the main text except that the central orbital is rotated by 90 degrees clockwise.
SOLVING EQS. (2) AND (3) IN THE MAIN TEXT

Eqs. (2) and (3) in the main text read

\[
\begin{align*}
\epsilon_k x_1 + t_3 x_2 + 2(1 + \alpha^{-1})t_4 x_3 + \alpha^{-1} t_3 x_4 &= \lambda x_1 \\
t_3 x_1 + (\epsilon_h + 2t_5) x_2 + 2t_2 x_3 &= \lambda x_2 \\
2(1 + \alpha) t_4 x_1 + 2t_2 x_2 + (2\epsilon_k + 4t_1) x_3 + 2t_2 x_4 &= \lambda x_3 \\
\alpha t_3 x_1 + 2t_2 x_3 + (\epsilon_h + 2t_5) x_4 &= \lambda x_4,
\end{align*}
\]

and

\[
\alpha = -1 + \frac{1}{t_1 x_1} \left[ t_3 \frac{t_3}{2} (x_2 + x_4) + 2t_4 x_3 \right].
\]

respectively. Here we assume \( x_3 \) to be nonzero and then can take \( x_3 = 1 \) without loss of generality. If one would like to normalize the wave function, \( (x_1, x_2, x_3, x_4) \) should be multiplied by the normalized constant. Eq. (6) reads

\[
x_1 = -\epsilon h/4 - 2t_5 - \lambda t_3 \frac{x_2}{t_3} - 2t_2/ t_3.
\]

By substituting Eq. (9) into Eq. (7), we obtain

\[
x_2 = -x_4 - \frac{(2\epsilon_k + 4t_1 - \lambda) t_1 + 4t_2^2}{2t_1 t_2 + t_3 t_4}.
\]

By substituting Eqs. (9)-(11) into Eq. (8), we obtain

\[
2 \left( 2t_2 + \frac{t_3 t_4}{t_1} \right)^2 - \left( \epsilon_h + \frac{t_3^2}{2t_1} + 2t_5 - \lambda \right) \left( 2\epsilon_k + 4t_1 + 4 \frac{t_2^2}{t_1} \right) = 0.
\]

By solving this equation, we can represent \( \lambda \) with the tight-binding parameters \( t \) and \( \epsilon \). Since \( \lambda \) is a real value, the following equation should be satisfied:

\[
\left[ \left( \epsilon_h + \frac{t_3^2}{2t_1} + 2t_5 \right) + \left( 2\epsilon_k + 4t_1 + \frac{t_3^2}{2t_1} \right) \right]^2 + 4 \left[ \left( 2t_2 + \frac{t_3 t_4}{t_1} \right)^2 - \left( \epsilon_h + \frac{t_3^2}{2t_1} + 2t_5 \right) \left( 2\epsilon_k + 4t_1 + \frac{t_3^2}{2t_1} \right) \right] \geq 0.
\]

We can immediately see that this equation is always satisfied because

\[
\text{LHS of Eq. (13)} = \left[ \left( \epsilon_h + \frac{t_3^2}{2t_1} + 2t_5 \right) - \left( 2\epsilon_k + 4t_1 + \frac{t_3^2}{2t_1} \right) \right]^2 + 8 \left( 2t_2 + \frac{t_3 t_4}{t_1} \right)^2.
\]

By substituting this and Eqs. (9)-(11) into Eq. (5), we can obtain a quadratic equation for \( x_4 \). Because it is a quadratic equation, it always has solution(s). Using obtained \( x_4 \) and Eqs. (9)-(11), now we can represent all variables, \( x_1, x_2, x_3, x_4, \alpha, \) and \( \lambda \) with the tight-binding parameters \( t \) and \( \epsilon \).