Electron localization/delocalization in incommensurate helical magnets

Shu Tanaka, Hosho Katsura and Naoto Nagaosa

1Department of Physics, The University of Tokyo, 7-3-1, Hongo, Bunkyo-ku, Tokyo 113-0033, Japan
2Department of Applied Physics, The University of Tokyo, 7-3-1, Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
3Correlated Electron Research Center (CERC), National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba Central 4, Tsukuba 305-8562, Japan
4CREST, Japan Science and Technology Agency (JST), Japan

The electronic states in incommensurate (IC) helical magnets are studied theoretically from the viewpoint of the localization/delocalization. It is found that in the multi-band system with relativistic spin-orbit interaction, the electronic wavefunctions show both the extended and localized natures along the helical axis depending on the orbital, helical wavenumber, and the direction of the plane on which spins rotate. Possible realization of this localization is discussed.

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The helical magnets have been studied for a long term since its first discovery by Yoshimori. Their ground states are determined by the (frustrated) exchange interactions and their Fourier transformation $J(q)$. Various properties including the spin wave excitations are analyzed theoretically for many materials. (See [2] for an early review.) Helical spin structure is now attracting recent revived interests from the viewpoints of both dielectric and transport properties. One example is the ferroelectricity induced by the helical magnetic order. Theoretically, the spin current associated with the non-collinear spin configuration is proposed to induce the electric polarization $\mathbf{P}$. Experimentally, it is now found that this mechanism is at work in $\beta$-MnO$_2$ and in other materials. Another new aspect is the anomalous transport properties associated with the spin chirality and in other materials. These developments urge the microscopic theory of electronic states to understand the physical properties associated with the helical spins.

In the absence of the spin-orbit interaction (SOI), one can rotate spin frame so that the $z$-axis is parallel to the direction of the local spin. In this rotated frame, the spins are aligned ferromagnetically and the original spin structure is reflected in the magnitude and phase of the effective transfer integrals. This leads to the double exchange interaction and various phenomena related to the spin chirality, respectively. When we consider the state of single helical wavevector $q$, the relative angle between the neighboring spins does not break the original translational symmetry. Furthermore, there is no spin chirality, i.e., no fictitious magnetic field induced by the solid angle subtended by the spins. Therefore the Hamiltonian in the rotated spin frame preserves the periodicity of the original lattice, and hence one can define the Bloch wavefunction.

This situation is modified in an essential way when the SOI is taken into account. In this case, one cannot rotate the spin frame with the orbitals being intact, and the transfer integrals forming a matrix between ions are transformed in a nontrivial way. Therefore, in general, we expect the incommensurate (IC) modulation of the transfer integrals and even of the site energies in the effective Hamiltonian in the rotated frame.

Localization/delocalization of electronic state in an IC potential is an old issue. Unlike in the case of commensurate periodic potentials, the eigen states are not the extended Bloch states in the case of IC potentials. Therefore the band structures would be unusual, i.e., highly fragmented, in those IC potentials. The central issue is whether electronic states are extended or localized in such kind of potentials, namely, Metal-Insulator Transition (MIT). Aubry and Andre (A-A) have shown that in a simple 1D model MIT occurs simultaneously for all energies when the strength of the IC potential $V_0$ is equal to the transfer integral $t$, i.e., if $V_0 > t$, the electronic states localize. We can also regard A-A model as a two dimensional tight binding model with IC magnetic flux. Actually, well-known Hofstadter butterfly is closely related to this model. Using the trace map technique, Kohmoto et al. has exactly studied the scaling properties of the Fibonacci lattice system which can be regarded as the A-A model with IC modulation $Qa/2\pi$ being the inverse Golden Mean. Similar problems with IC transfer integral are also investigated by Kohmoto et al.

In this paper, we investigate the localization/delocalization of electronic states in IC helical magnets. First we study a model of $d$-orbitals in cubic symmetry taking into account the SOI. We found that as SOI increases, the localization caused by IC starts from the specific $t_{2g}$ wavefunctions at around $q \sim \pi/a$ ($a$: lattice constant). In order to scrutinize this localization, we construct an effective single-band model for $t_{2g}$ bands. With this effective model, the
localization lengths are studied in more detail including its dependence on the angle $\phi$ between the spin rotation angle and the helical wavevector.

We start with the following electronic model:

\[
H = H_U + H_{SO} + H_d + H_t,
\]

\[
H_U = -U \sum_j \mathbf{e}_j \cdot \mathbf{S}_j,
\]

\[
H_{SO} = -\lambda \sum_j \mathbf{L}_j \cdot \mathbf{S}_j,
\]

\[
H_d = \sum_j \epsilon_{\alpha}(d_{j\alpha}^2|d_{j\alpha}^2|),
\]

\[
H_t = t_j^\alpha|d_{j\alpha}^0|/d_{j+1\sigma}^\alpha|.
\]

In the octahedral ligand field, the $d$ orbitals are split into $e_g$ and $t_{2g}$-orbitals [23]. The $t_{2g}$-orbitals, i.e., $d^{xy}, d^{yz}$ and $d^{zx}$, have energies lower than $e_g$-orbitals, i.e., $d^{x^2-y^2}$, and $d^{3z^2-r^2}$ by $10Dq$, but the order is reversed as we take the hole picture in the followings, i.e., $\epsilon_{t_{2g}} - \epsilon_{e_g} = 10Dq$.

The on-site SOI is considered, the matrix elements of which are calculated by $L \cdot S$ with $L (S)$ being the orbital (spin) angular momentum. It is noted that $L$ has no matrix elements within the $e_g$ sector, while nonzero coupling occurs within $t_{2g}$ sector and between $e_g$ and $t_{2g}$ sectors.

Considering the hopping between $d$-orbitals and oxygen orbitals [24], we derive the effective transfer integrals $t_{ij}^{\alpha}$ between $d^\alpha$-orbitals at neighboring magnetic ions $i$ and $j$. We took the values $t^{yz} = t^{zx} = 0.1$, $t^{3z^2-r^2} = 0.3$ and $t^{xy} = t^{x^2-y^2} = 0$. In $H_U$ of Eq. (1), the magnetic moment at site $j$ is described by the unit vector $\mathbf{e}_j \equiv (\cos \phi_j \sin \theta_j, \sin \phi_j \sin \theta_j, \cos \theta_j)$ and $\mathbf{S}_j$ denotes the electronic spin operator at site $j$. We assume the IC helical magnetic structure for $\mathbf{S}_j$ along $z$-axis, which is on the spin $(z, x)$-plane, realized as a result of the frustrated spin exchange interaction. We focus on the ordered ground state properties, and hence the mean field treatment gives a good description of the system. We assume the ferromagnetic spin configuration perpendicular to the helical wavevector $\mathbf{q}$, and hence $k_x$, $k_y$ are good quantum numbers, i.e., the electronic wavefunctions are plane waves along $x$- and $y$-directions. We fix $k_x = k_y = 0$ hereafter, and consider the one-dimensional (1D) model only along $z$-direction. Fig. 1 shows the calculated density of states as a function of the helical wavenumber $q$ with the color specifying the localization length $\xi$. We note here that the sample size is a prime number 199, and helical wavenumbers $q$’s are taken to be proximate to the IC values. All the bands states from the $e_g$-orbitals are extended due to the weak SOI, and hence are omitted in Fig. 1. The green region is the extended states while the blue one is strongly localized within the scale of lattice constant. We took the values 10$Dq = 3$, $U = 1.4$, and $\lambda = 1.0$.

The density of states are understood as follows. The largest splitting between $e_g$ and $t_{2g}$ occurs due to the lig- and field 10$Dq$ in Eq. (1). Then the $t_{2g}$ bands are further split into bands of $\Gamma_7$ and $\Gamma_8$ origin, the latter of which is upper in energy since we take the hole picture. Then both the bands are further split by the spin exchange field $U$.

By using the iterative method developed by MacKinnon [25], we can calculate the Green’s function $G_{1,N}^{(N)} \equiv \langle 1| (E - H)^{-1} |N \rangle$, which connects both ends of the long chain. $G_{1,N}^{(N)}$ is still a $10 \times 10$ matrix and the Lyapnov exponent, i.e., the inverse of the localization length $\xi$ is obtained as $\frac{\xi}{N} \equiv -\lim_{N \to \infty} \frac{1}{2N} \ln \text{Tr}|G_{1,N}^{(N)}|$. The blue color at around $q \sim \pi/a$ means the strong localization along the helical axis. When we change $\lambda$, we still observe the localization down to $\lambda \sim 0.2$. Therefore we conclude that the localization starts in some part of the electronic spectrum at around $q = \pi/a$ as one increases the SOI. The most remarkable point we can grasp from the above figure is the energy dependence of localization/delocalization nature, namely that there are both localized and extended states at different energies for the same $q$. This is in sharp contrast to the case of $A-A$ model where all the states are either extended or localized depending only on the ratio $V_0/t$ as mentioned above.
In order to study this localization in more depth, we now derive the effective model for a limiting case, i.e., $10Dq \gg 3\lambda/2 \gg U \gg t$. Even though this is not necessarily a suitable limit for realisite systems, it clarifies why $\xi$ depends on the orbitals. By taking into account the spin degree of freedom, there is six-fold degeneracy of the $t_{2g}$ energy levels. Because of the on-site SOI, this degeneracy is lifted and we have two groups of spin-orbit coupled states, labeled $\Gamma_7$ and $\Gamma_8$. The two-fold degenerate states, $\Gamma_7$, and the four-fold degenerate one, $\Gamma_8$, are given by $|3^+\rangle = (|d_{1}^{xy}\rangle + |d_{1}^{yz}\rangle + i|d_{1}^{zx}\rangle)/\sqrt{3}$, $|3^-\rangle = (|d_{1}^{xy}\rangle - |d_{1}^{yz}\rangle + i|d_{1}^{zx}\rangle)/\sqrt{3}$, $|1^+\rangle = (|d_{2}^{yz}\rangle + i|d_{2}^{zx}\rangle)/\sqrt{2}$, $|1^-\rangle = (|d_{2}^{yz}\rangle - i|d_{2}^{zx}\rangle)/\sqrt{2}$, $|2^+\rangle = (2|d_{3}^{yz}\rangle - |d_{3}^{zx}\rangle - i|d_{3}^{xy}\rangle)/\sqrt{6}$, respectively, where the quantization axis of spin is taken to be the $z$ axis. Henceforth, we assume that the spin-orbit coupling in our system is sufficiently large and focus only on the case where the two multiplets, i.e., $\Gamma_7$ and $\Gamma_8$, do not hybridize with each other.

Now, we construct the normalized state $|g_j\rangle$ to minimize $|g_j\rangle - U\vec{e}_j \cdot \vec{S}_j |g_j\rangle$ in the Hilbert space spanned by the states in $\Gamma_7$ or $\Gamma_8$. The desired states whose spins are parallel to the unit vector $\vec{e}_j$ are explicitly given for $\Gamma_7$ and $\Gamma_8$ by

$$|g_j^7\rangle = \sin \frac{\theta_j}{2} |3^+\rangle + e^{i\phi_j} \cos \frac{\theta_j}{2} |3^-\rangle$$

and

$$|g_j^8\rangle = e^{-i\frac{\theta_j}{2} \cos \frac{\theta_j}{2} \cos \phi_j} |1^+\rangle + e^{i\frac{\theta_j}{2} \sin \frac{\theta_j}{2} \sin \phi_j} |1^-\rangle - \sqrt{3} e^{-i\frac{\theta_j}{2} \cos \frac{\theta_j}{2} \sin \phi_j} |2^+\rangle - \sqrt{3} e^{i\frac{\theta_j}{2} \sin \frac{\theta_j}{2} \cos \phi_j} |2^-\rangle,$$

respectively. Here, subscript $j$ denotes the site number and subscripts 7 and 8 correspond to $\Gamma_7$ and $\Gamma_8$ respectively. Using these states, we can derive the effective Hamiltonian $H = \sum_n T_n c_n^\dagger c_{n+1} + h.c. + V_n c_n^\dagger c_n$, where $c_n^\dagger$ denotes the renormalized annihilation/creation operator and the effective transfer integral $T_n$ and site energy $V_n$ are given as:

$$T_n = \frac{2t}{3} \left( \sin \frac{\theta_n}{2} \sin \frac{\theta_{n+1}}{2} + e^{-i\Delta\phi} \cos \frac{\theta_n}{2} \cos \frac{\theta_{n+1}}{2} \right),$$

$$V_n = -4t/3,$$

and $\Delta\phi = \phi_n - \phi_{n+1}$ for $\Gamma_7$, and

$$T_n = t(e^{i\Delta\phi/2} \cos \frac{\theta_n}{2} \cos \frac{\theta_{n+1}}{2} + e^{-i\Delta\phi/2} \sin \frac{\theta_n}{2} \sin \frac{\theta_{n+1}}{2}) \times (e^{i\Delta\phi} \cos \frac{\theta_n}{2} \cos \frac{\theta_{n+1}}{2} + e^{-i\Delta\phi} \sin \frac{\theta_n}{2} \sin \frac{\theta_{n+1}}{2}),$$

$$V_n = -t(1 + \cos^2 \theta_n),$$

for $\Gamma_8$.

As for the $\Gamma_7$ case, we can write down $T_n$ as $\frac{2t}{3} e^{i\theta_{n,n+1} \cos \frac{\theta_{n,n+1}}{2}}$, where $\theta_{n,n+1}$ is the angle between the two spins $\vec{S}_n$ and $\vec{S}_{n+1}$. The phase $\alpha_{n,n+1}$ is the vector potential generated by the noncollinear spin configuration, but we can eliminate it by appropriate gauge transformation. Then we can conclude that we have no incommensurability in our 1D $\Gamma_7$ model.

As for the $\Gamma_8$ case, on the other hand, the effective site energy $V_n$ explicitly depends on the local spin angle $\theta_n$. If we have the spin configuration in the plane which is parallel to the $xz$ plane, i.e., $\theta_n = \text{const.}$, and set the pitch $\Delta\phi = \text{const.}$, $V_n$ and $T_n$ are constant. On the other hand, if we have the tilt of the spin rotation plane from the above plane to the other plane, $\theta_n$ is no longer a constant and then $V_n$ would generally be IC. $|T_n|$ also depends on both the angles of $\vec{S}_n$ and $\vec{S}_{n+1}$. Here we can conclude that the case where holes are in $\Gamma_8$, the effective 1D model would generally be IC. This explains why the upper part of the $t_{2g}$ density of states in Fig.
1 are localized more strongly, where the wavefunction is mainly from $\Gamma_8$ components.

Now we focus on the $\Gamma_8$ case and numerically examine whether the localization of the wavefunction occurs in more details. We consider the helical spin configuration $\hat{S}_n = (S \cos(qn), S \cos \varphi \sin(qn), S \sin \varphi \sin(qn))$, where $q$ is helical wavenumber, and $\varphi$ denotes the tilt angle of the spin rotating plane from $xz$-plane (See Fig. 2).

The numerical calculations are performed for systems of size 1000, a large prime number, with nearly incommensurate modulations $q/2\pi = j/1000, \; (j = 1, 2, 3, \ldots)$.

The results are shown in Figs. 3, where the vertical and the horizontal axes represent the energy and the helical wavenumber, respectively. We take the unit where $t = 1$ and $a = 1$. The tilt angles are $\varphi = 0^\circ, 30^\circ, 60^\circ$ and $90^\circ$ for Figs. 3(a),(b),(c), and (d), respectively. The energy spectrum in Fig. 3(d) is almost same as the lowest band of $\Gamma_8$ bands in Fig. 1. In Figs. 3, the localization length increases as the color changes from blue to green. The figures clearly display that there are domains of strong localization $\xi \sim 1$ when we have a finite tilt angle $\varphi$. On the other hand, for $\varphi = 0^\circ$ the transfer integrals $T_n$ and the on-site potentials $V_n$ are constants, and there is no localized states. Even in the most suitable case for localization, i.e., Fig. 3(d), however, the helical wavenumber $q$ should be approximately in the range of $2\pi/3 < q < 4\pi/3$ for the localized states. This is because the long period of the helical structure means the slowly varying and weak perturbations in the rotated frame, and hence does not cause the localization.

Now we discuss the possible realization of the localized states in realistic systems. From the above results, three important conditions for the localization are (i) strong SOI, (ii) short helical period, and (iii) the direction of rotating spin plane. The SOI increases as the mass of the atom get heavier, and hence the present model becomes more relevant from the viewpoint (i). For 3d-orbitals of transition metal oxides, the SOI is typically of the order of 20-30meV, which is an order of magnitude smaller than the transfer integral $t$. Therefore the localization length is expected to be rather large, and hence the disorder effect such as the impurity scattering might hide the IC effect. Therefore, even though $\beta$-MnO$_2$ [1] and SrFeO$_3$ [2,3] show interesting transport properties, it is unlikely that the localization found in this paper is relevant to these materials. 4d or 4f, 5f-orbitals, where SOI is larger than $\sim 0.3eV$, are more promising. Actually there are many rare-earth metals showing the helical spin structure such as Tb, Dy, Ho, Er [4,5]. From the condition (ii), it is rather hard to find the short period helical spin structure. It is typically $4a-5a$ or even larger [6]. This viewpoint, MnO$_2$ [7,11] is an interesting case, but the localization is unlikely as discussed above. As for the condition (iii), we need more study since only the cubic case has been considered. The directional dependence of the spin plane might be useful to control the localization/delocalization by an external magnetic field.

Even though the conditions for localization discussed above are rather stringent, which explains why it has never been observed experimentally thus far, it will play a vital role in the quantum transport properties of the system once realized. One direct consequence is the large anisotropy of the resistivity between parallel and perpendicular to the helical axis, i.e., it should be much more resistive in the parallel direction.

In conclusions, we have studied the localization/delocalization of the electronic states in helical magnets. We found the localized states under the condition of (i) strong spin-orbit interaction, (ii) short helical wavelength, and (iii) proper direction of the plane on which spins rotate. The strong dependence of the localization length $\xi$ on the orbital is also found, which is explained by an effective model for a certain limiting case.

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