Observable Topological Effects of Möbius Molecular Devices

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We study the topological properties of quantum states for the spinless particle hopping in a Möbius ladder. This system can be regarded as a molecular device possibly engineered from the aromatic Möbius annulenes, which enjoys a pseudo-spin orbital interaction described by a non-Abelian gauge structure. It results from the nontrivial topology of configuration space, and results in various observable effects, such as optical spectral splitting. The transmission spectrum through the Möbius molecular device is calculated to demonstrate a topological effect as a destructive interferences in the conduction band. The induced interaction also leads to an entanglement between the transverse and longitudinal modes for any locally factorized state.

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Introduction.- With various potential applications, the molecular based devices have motivated extensive experimental and theoretical investigations (see Ref.[1]). Similar to the semiconductor based artificial nanostructures, the engineered molecular architectures also displays different novel quantum effects. The concept of Möbius aromaticity was firstly proposed in 1964[2], but various kinds of Möbius molecule were not designed or claimed to be synthesized[3, 4] until recent years. With these significant developments of engineering topologically nontrivial chemical structures, the topological properties of quantum systems become more and more important. Thus, it is natural to consider the various quantum effects induced by the nontrivial topological configurations[2, 3, 5, 6, 7, 8, 9, 10], such as the Möbius molecule.

For the topological effects in quantum mechanics, it is a fundamental principle (assumption) that the wave function must be single-valued in a topologically trivial configuration. In this sense, various topological effects such as Aharonov-Bohm (AB) effect can be rationally explained without the introduction of any extra assumption in quantum mechanics[11]. In some configuration with nontrivial topological structure, though the motion of particle requires some complex boundary conditions, such topological non-trivialness of the configuration space can be canceled by introducing a singular gauge field. A typical example is the phenomena of persistent currents in a mesoscopic or a superconducting ring threaded by a magnetic flux. Here, the $U(1)$ gauge field can be introduced to cancel the seemingly multi-value of boundary condition with a non-integrable AB phase factor. Another illustration is the fractional statistics of anyons, which describes the effective quantum excitations of the hardcore particles confined strictly in two dimension with the braiding homotopy of the many-body configuration[12].

In this Letter, we show a topology induced quantum effect with a non-Abelian gauge structure, which emerges from the twisted boundary condition in a Möbius ladder. The twisted boundary condition results in a local interaction between transverse and longitudinal modes. And, in the continuous limit, the spinless particle moving in the Möbius ladder is mapped to a pseudo-spin coupled to the orbit corresponding to the longitudinal mode. When we apply a transverse field, the pseudo-spin seems to be confined in a one-dimensional ring and subject to a texture-like effective magnetic field, together with an effective magnetic flux threading the ring(see Fig.1). Different from the existing setups of mesoscopic ring for persistent currents[13], the effective magnetic flux in our molecular device depends on the pseudo-spin state, namely, there exists a non-Abelian gauge field induced by the Möbius topology.

Compared with the topologically trivial configurations, i.e. an ordinary tight binding ladder, the quantum state of the Möbius molecule is strongly affected by its configuration topology. Here, we predict three quantum interference effects, which are distinguished from the ordinary cases obviously: (i) the Stark effect in the electric field cause more optical spectral splitting, due to the effective Zeeman effect by the induce non-Abelian gauge field; (ii) the transmission spectrum through the Möbius molecule is significantly modified due to the destructive interference caused by the non-Abelian flux; (iii) from the view of quantum information, the entanglement is emerged from the locally factorized state in the Möbius molecule.

Induced non-Abelian gauge structure.- Let us con-
sider the hopping of an electron on the tight-binding lattice with a Möbius ladder configuration illustrated in Fig.2(a). Since there is no spin flip, the electron is regarded as a spinless particle. There are \(2N\) lattice sites, located at the two edges of the ladder, whose coordinates are \(r_{j\pm} = (\cos \varphi_j (R \pm w \sin[\varphi_j/2]), \sin \varphi_j (R \pm w \sin[\varphi_j/2]), \pm w \cos[\varphi_j/2])\), with half-width \(w\) and “radius” \(R\), and \(\varphi_j = 2\pi j/N\) being the polar coordinate. We introduce operators \(a_j^\dagger (a_j)\) and \(b_j^\dagger (b_j)\) to denote the creating (annihilating) a particle on \(j\)-th site of each edges respectively. The rungs represent the coupling between \(a\)-chain and \(b\)-chain. We assume the hopping strength along the ladder is homogeneous. Then the Hamiltonian reads

\[
H = \sum_{j=0}^{N-1} \mathbf{A}_j^\dagger \mathbf{M}_j \mathbf{A}_j - \xi \sum_{j=0}^{N-1} \left( \mathbf{A}_j^\dagger \mathbf{A}_{j+1} + \text{h.c.} \right), \tag{1}
\]

where \(\mathbf{A}_j = (a_j, b_j)^T\), and the matrix \(\mathbf{M}_j = \varepsilon_j \sigma_z - V_j \sigma_x\), for \(\sigma_{x,y,z}\) being the Pauli matrices, \(2\varepsilon\) describing the on-site energy difference between \(a\)- and \(b\)-particles, and \(V_j\) representing their coupling strength.

The Möbius boundary conditions are \(a_N = b_0\) and \(b_N = a_0\), or equivalently, \(\mathbf{A}_N = \sigma_z \mathbf{A}_0\). It is this boundary condition that results in the interesting topological properties of the quantum state. In terms of the operator-valued vector \(\mathbf{B}_j \equiv (\epsilon_j, \epsilon_j, \varepsilon_j \sin[\varphi_j/2], V_j)^T\) is the unitary transformation of \(\mathbf{A}_j\), the Hamiltonian is rewritten as

\[
H = \sum_{j=0}^{N-1} \mathbf{B}_j^\dagger (\mathbf{\Omega}_j \cdot \mathbf{\sigma}) \mathbf{B}_j - \xi \sum_{j=0}^{N-1} \left( \mathbf{B}_j^\dagger \mathbf{Q} \mathbf{B}_{j+1} + \text{h.c.} \right), \tag{2}
\]

where \(\mathbf{\Omega}_j \equiv (\varepsilon_j \cos[\varphi_j/2], \varepsilon_j \sin[\varphi_j/2], V_j)^T\) is a direction vector, and \(\mathbf{Q} \equiv \text{diag}([\exp(i\pi/N), 1])\). It should be emphasized that the operator \(\mathbf{B}_j\) only requires the ordinary periodic boundary condition, i.e., \(\mathbf{B}_N = \mathbf{B}_0\). So far, we have shown that the nontrivial Möbius boundary condition is canceled by the unitary transformation, accompanied by an induced non-Abelian gauge field associated with \(\mathbf{\Omega}_j\) and \(\mathbf{Q}\). This point will be seen more clearly in the continuous limit below.

In the continuous limit (i.e. \(N \to \infty\) and \(\varphi_j \to \varphi \in [0, 2\pi]\)), the particle hopping on a Möbius ladder is described by the two-component Hamiltonian \(\mathcal{H}\), which can be mapped to the continuous Hamiltonian

\[
\mathcal{H} = \left( -i \frac{\partial}{\partial \varphi} - \hat{\phi} \right)^2 + \mathbf{\Omega}(\varphi) \cdot \mathbf{\sigma}. \tag{3}
\]

This Hamiltonian describes a pseudo-spin moving in a one-dimension ring subject to non-Abelian gauge field including a spin dependent flux \(\hat{\phi} = (\sigma_z + 1)/4\) and an inhomogeneous magnetic field \(\mathbf{\Omega}(\varphi)\). Here, the natural unit is chosen. In this sense, the induced magnetic flux \(\hat{\phi}\) is an operator, which does not commute with the Zeeman term \(\mathbf{\Omega} \cdot \mathbf{\sigma}\), and the gauge field is called non-Abelian.

**Topological Stark shift and spectral splitting.** Now, we consider the Möbius molecule subject to a uniform electric field \(\mathbf{E} = E_z \hat{\mathbf{e}}_z\) along \(z\) direction. The electric field induced on-site energy difference is \(2\varepsilon_j = \mathbf{E} \cdot (\mathbf{r}_j+\mathbf{r}_{j-1}) \equiv 2\varepsilon \cos(\varphi_j/2)\). By assuming the homogeneous coupling \(V_j \equiv V\), the effective magnetic field

\[
\mathbf{\Omega}_j = \left( \frac{\varepsilon_j}{2} \hat{\sigma}_x, \frac{\varepsilon_j}{2} \sin \varphi_j, V \right) \tag{4}
\]

possesses a texture-like distribution [Fig.2(b)] with spatially varying amplitude and direction.

The Stark effect, i.e. the energy shift under the weak electric field, is calculated by the perturbation approach. For simplicity of notation, we present the result in the continuous limit. By taking the unperturbed Hamiltonian \(\mathcal{H}_0 = (-i \frac{\partial}{\partial \varphi} - \hat{\phi})^2 + \mathbf{\Omega}_0 \cdot \mathbf{\sigma}_z\), the zero-th order eigenenergy are \(E_{n_1} = (n - 1/2)^2 + V\) and \(E_{n_1} = n^2 - V\) respectively, corresponding to eigen-states denoted as \(|n, \chi\rangle = |n\rangle |\chi\rangle\), for \(\chi = \uparrow, \downarrow\) and \(|\phi | n\rangle = \exp(in\varphi)/\sqrt{2\pi}\). The energy spectra of these unperturbed states, illustrated in Fig.2 in comparison with the ordinary ring, shows the obvious spectral splitting.

The perturbation \(\mathcal{H}' = \mathbf{\Omega}_0 \sigma_x + \Omega_0 (\varphi) \sigma_y\) results in the superposition of the zero-th order states with different pseudo-spin components. The Stark shifts \(\delta E_{n_1\chi}\). 

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**FIG. 2:** Energy levels (the solid lines with dash marks) for the ordinary ring (a) and the Möbius ladder (b). The dotted lines with empty square marks are the energy levels in the continuous limit. The allowed transitions induced by a electric field in the \(z\) direction are depicted by the arrows. (c) The optical spectra of the Möbius ladder corresponding to the transitions shown in (b). The gray peak centered at the origin is the spectra for the ordinary ring. The Lorentz profile are assumed for each peaks with phenomenological broadening. Parameters used in calculation are \(N = 12\) and \(V = 50\).

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[sources and references mentioned]
under the static electric field are calculated as

$$
\delta E_{n1} = \frac{\varepsilon^2 (V - n - 1/8)}{3n^2 - 8Vn + (4V^2 - V - 3/16)}, \quad (5a)
$$

$$
\delta E_{n1} = \frac{-\varepsilon^2 (V - n + 5/8)}{3n^2 - (8V + 3)n + (4V^2 + 5V + 9/16)}, \quad (5b)
$$

Together with the non-vanishing transition matrix elements \(\langle n \uparrow | H' | n + 1 \downarrow \rangle = \langle n \uparrow | H' | n \downarrow \rangle = \varepsilon/2\), this energy shifts due to the twisted boundary condition are regarded as an observable quantum effect of the induced non-Abelian gauge field.

In view of the future experiments, the topology state of the Möbius molecule may be detected by the splitting of the optical spectra. We consider the transitions of the electron in the Möbius molecule under an optical excitation. We assume that the molecules are subject to the linearly polarized light, whose electric field component \(\mathbf{E}(t) = E_z(t)\mathbf{e}_z\) oscillates in the \(z\) direction. The time dependent Hamiltonian is \(H(t) = H_0 + H'(t)\), where the perturbation \(H'(t) = \mathbf{E}(t) \cdot \mathbf{r} = H' \cos \omega t\), for \(\omega\) being the frequency of the pumping light. The transition selection rule shown in Fig.2 follows from the above mentioned transition matrix elements as (i) \(|n \uparrow\rangle = |n \downarrow\rangle\) and (ii) \(|n \downarrow\rangle = |n + 1 \uparrow\rangle\). The Fermi’s golden rule is applied to calculate the spectra with various excitation energy. In comparison with the case of ordinary ring, where only one peak is located at frequency \(\hbar \omega = 2V\) since the only transition (i) is allowed in this case, the optical spectra of Möbius molecule show clear splitting due to the non-trivial topology.

**Transmission through a Möbius ring.**—Besides the topology induced Stark shift and the spectral splitting, the transmission through the Möbius ring exhibits obvious differences from the ordinary ring.\(^{10}\)

We consider the molecule is connected to two leads as shown in Fig.3(c). The leads are modeled by two semi-infinite chains. The electron can hop along the chains and tunnel between the lead and the Möbius ring at the junctions. By assuming that the leads are connected with the ring at \(a_0\) and \(a_{N/2}\) sites, the Hamiltonians of the electron moving in the leads and its tunneling to the Möbius ring are written as \(H_{\text{lead}} = t_1 \sum_{k=1}^{\infty} c_{i,k}^\dagger c_{i,k+1} + \text{h.c.}\) and \(H_{\text{tun}} = t_1 (c_{L,1}^\dagger a_0 + c_{R,1} a_{N/2}^\dagger) + \text{h.c.}\), respectively, where \(c_{i,k}\) for \(i = L, R\) are the annihilation operators of the electron in the leads, and \(t_1\) describes the electron hopping amplitude.

To analyze the transmission for a given injection energy \(E\), we calculate the self-energies \(\Sigma_{L,R}\) to determine the Green’s function of the Möbius ring \(G(E) = [E - H - \Sigma_L - \Sigma_R]^{-1}\) by taking account of the influence of the semi-infinite leads.\(^{14}\) The self-energies \(\Sigma_{L,R}\) can be obtained numerically, and they give the level broadenings \(\Gamma_{L,R} = -2 \text{Im} \Sigma_{L,R}\). The transmission coefficient \(T(E)\) is obtained by the relation \(T(E) = \text{Tr}[\Gamma_R G L G^\dagger]\).\(^{14}\)

The transmission spectrum of the Möbius ring is shown in Fig.3(b), comparing with that of the ordinary mesoscopic ring with periodic boundary conditions Fig.3(a). For simplicity, \(\varepsilon_j = 0\) and \(V_j = V\) are assumed. Through the unitary transformation from \(A_j\) to \(B_j\), the Möbius ladder is decomposed into two independent rings (channels), see Fig.3(d). In the strong coupling limit, i.e. \(V \gg \xi\), the energy spectrum of the channels are split into two bands. The energy gap between them is determined by the coupling strength \(V\). Below the energy gap (or in the “valence band”), the transmission behaviors are locally similar in both cases. It is not affected by the topology of configuration space, since, as discussed before, the induced gauge field does not present in the valence band channels. Above the energy gap (or in the “conduction band”), the transmission coefficient is completely suppressed in the Möbius ring due to the induced gauge field, which equals to a half magnetic flux quanta. The particle could not transmit through the Möbius ring at such energies, due to the destructive interference between the two arms of the ring.

**Decoherence from induced Stern-Gerlach effect of pseudo-spin.**—The third topological phenomenon is the quantum decoherence of the pseudo-spin caused by the Stern-Gerlach effect of the induced gauge field.\(^{15}\) Actually, through the spin-orbit coupling, quantum entanglement between different spin states is created so that a quantum measurement can be realized. Similar to the Stern-Gerlach experiment, the spatial degrees of freedom interacts with spin in a non-demolition fashion, and thus measures the spin states. Here, we point out that, the situation may be different from the topologically nontrivial case without obvious local coupling. We have shown that the gauge field can be induced by the Möbius boundary condition, and the effective pseudo-spin orbital interaction further arises from this gauge field. Thus, the entanglement could be created by the topology induced effect in the absence of any real local interactions.

In order to emphasize the main physical mechanism in our argument, we assume the homogeneous conditions...
\[ \varepsilon_j = 0 \quad \text{and} \quad V_j = V . \] Thus, only is the \( \sigma_z \) component retained, and thus Hamiltonian (2) is of a block-diagonal form, i.e., \( H = \text{diag}[H_\uparrow, H_\downarrow] \) with the conditional Hamiltonians
\[ H_x = \pm V \sum_{j=0}^{N-1} c_{jX}^\dagger c_{jX} - \sum_{j=0}^{N-1} \left( \xi_j c_{jX}^\dagger c_{j+iX} + \text{h.c.} \right), \tag{6} \]
where \( \chi \in \{ \uparrow, \downarrow \} \), \( \xi \downarrow = \xi \), and \( \xi \uparrow = \xi \exp(i\pi/N) \).

We assume one electron is initially located at the \( a_0 \) site, i.e., \( |\psi(0)\rangle = a_0^j(\text{vac}) = |j = 0 \rangle \otimes (|\uparrow \rangle + |\downarrow \rangle) / \sqrt{2} \). In the pseudo-spin representation, this initial state stands for a pseudo-spin pointing in the \( x \) direction at the \( j = 0 \) site. Obviously, it is a locally factorized state. At time \( t \), the wave function evolves into a superposition \( |\psi(t)\rangle = (|\psi_+(t)| \uparrow \rangle + |\psi_-(t)| \downarrow \rangle) / \sqrt{2} \) where the \( |\psi_\pm(t)\rangle = \exp(-iH_x t)|j = 0\rangle \) can be understood as the detector states measuring the pseudo-spin states. As the so-called decoherence factor, the overlap \( D(t) = \langle \psi_-(t)|\psi_+(t)\rangle = \sum_{j=0}^{N-1} G_\uparrow(j, t)G_\downarrow(j, t)/2 \) of \( |\psi_\pm(t)\rangle \) characterizes the quantum coherence of pseudo-spin states. Here,
\[ G_\pm(j, t) \equiv \langle j|\psi_\pm(t)\rangle = \sum_{k=-\infty}^{+\infty} e^{i\varphi_\pm(k)} J_k(2\xi t), \tag{7} \]
are the propagators of the two spin components, where \( J_n(x) \) is the \( n \)-th order Bessel function. Here, \( j_k = j + kN \) represents the \( j \)-th site with winding number \( k \) with respect to the two decomposed rings, and the two pseudo-spin dependent phases \( \varphi_\pm(j_k) = j_k\pi/2 \) and \( \varphi_\mp(j_k) = \varphi_\pm(j_k) + j_k\pi/N \) accompany the longitudinal motions in the two rings. It is the induced phase shift \( j_k\pi/N \) between \( \varphi_\pm(j_k) \) that gives rise to the decoherence of pseudo-spin. Straightforwardly, the decoherence factor is calculated as
\[ D(t) = \frac{1}{2} e^{2\xi'Vt} \sum_{\delta = -\infty}^{\infty} i^\delta J_{\delta N}(2\xi' t) \tag{8} \]
where \( \xi' = \xi \sqrt{2 - 2 \cos(\pi/N)} \). It is clear that, in the limit with large \( N \), the short time behaviors of the decoherence factor is dominated by only one term with \( \delta = 0 \), thus \( |D(t)| \approx |J_0(2\xi' t)| \) with an time dependent envelop \( \sqrt{N/(2\pi\xi t)} \) decaying as inverse square-root of time.

**Conclusion.**- Taking the Möbius ladder as an illustration, we have explored the role of topological structure of the configuration space on the quantum states of the particles confined in a topologically-nontrivial manifold. The global properties of the topological system can be locally described by a non-Abelian gauge structure, which can result in some observable effects in the aspects of spectroscopy, such as the topological induced Zeeman splits and the higher energy band suppression of the transmission of the Möbius molecule. We also show the quantum decoherence of the pseudo-spin and the entanglement due to the pseudo-spin orbital interaction. On the other hand, from the view of chemistry, these observable effects can be regarded as the physical signals of the successful synthesis of some Möbius aromaticity molecule. These methods may be used to distinguish the topologically nontrivial molecules from the ordinary ones.

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[1] V. Balzani, A. Credi, and M. Venturi, *Molecular Devices and Machines: A Journey Into the Nanoworld* (Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim, 2003).
[2] E. Heilbronner, Tetrahedron Lett. **29**, 1923 (1964).
[3] H. S. Rzepa, Chem. Rev. **105**, 3697 (2005); M. K. Cyrański, Chem. Rev. **105**, 3773 (2005); R. Herses, Chem. Rev. **106**, 4820 (2005).
[4] S. Tanda, et. al., Nature **417**, 397 (2002).
[5] J. Gravenes and M. Willatzen, Phys. Rev. A **72**, 032108 (2005).
[6] S. Nakamura, et. al., Physica E **22**, 684 (2004).
[7] F. Mila, et. al. Phys. Rev. B **57**, 1457 (1998).
[8] K. Yakubo, et. al., Phys. Rev. B **67**, 125319 (2003).
[9] M. Hayashi and H. Ebisawa, Phys. Rev. B **72**, 024505 (2005).
[10] S. K. Maiti, Solid State Commun. **142**, 398 (2007).
[11] N. Byers and C. N. Yang, Phys. Rev. Lett. **7**, 46 (1961); C. N. Yang, in *Proc. Int. Symp. Foundations of Quantum Mechanics*, Tokyo, 1983, pp.5-9.
[12] F. Wilczek, Phys. Rev. Lett. **48**, 1144 (1982); Y. S. Wu, Phys. Rev. Lett. **52**, 2103 (1984).
[13] D. Loss, et. al., Phys. Rev. Lett. **65**, 1655 (1990).
[14] S. Datta, *Quantum transport: Atom to transistor* (Cambridge University Press, Cambridge, 2005).
[15] Y. Li, et. al., Phys. Rev. Lett. **99**, 130403 (2007).