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To cite this article: Xing Guo *et al* 2018 *New J. Phys.* **20** 053032

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OPEN ACCESS

RECEIVED

26 February 2018

REVISED

7 April 2018

ACCEPTED FOR PUBLICATION

27 April 2018

PUBLISHED

11 May 2018

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Xing Guo¹, Yaojin Li¹ and Chenglong Jia^{1,2,3}¹ Key Laboratory for Magnetism and Magnetic Materials of the Ministry of Education, Lanzhou University, Lanzhou 730000, People's Republic of China² Institut für Physik, Martin-Luther Universität Halle-Wittenberg, Halle (Saale) D-06099, Germany³ Author to whom any correspondence should be addressed.E-mail: cljia@lzu.edu.cn**Keywords:** phase transition, spin–phonon interaction, vector-spin-chirality, bound state**Abstract**

We illustrate analytically the formation of vector-spin-chirality bound state due to spin–phonon interaction conditioned by the inverse Dzyaloshinskii–Moriya mechanism. The non-equilibrium dynamics of spin-chirality is effectively mapped into the spin-boson model. For spin-1/2 systems, our study suggests an existence of a gapless first-order phase transition from incoherent to coherent spin fluctuations, which is quantified to an emergence of spin-chirality bound state. The critical strength of spin–phonon interaction is found to be determined by the ratio between the amplitude of spin fluctuations and the Debye frequency of system.

Introduction

The magnetic properties of physical systems dramatically depend on the dimensionality, the frustration, and the thermal/quantum spin fluctuations of the system [1]. One-dimensional (1D) frustrated quantum spin-1/2 chain with competing nearest-neighboring ferromagnetic ($J_1 < 0$) and next-nearest-neighboring antiferromagnetic ($J_2 > 0$) exchange coupling, despite its simple structure, offers a good playground to look for exotic quantum phases in both experiment and theory [2]. In the classical spin approximation, it is well known that the ground state of the J_1 – J_2 spin chain possesses a helimagnetic state with an incommensurate pitch angle $Q = \arccos(-J_1/4J_2)$ in the range of $0 > J_1/J_2 > -4$. For the SU(2)-invariant quantum case, such a long-range helical ordering is destroyed by strong quantum/thermal spin fluctuations, whereas the system possesses types of *hidden* multiple-spin ordering, such as magnetic multipolar phase or spin-nematic state in which magnon bound states are formed from the subtle competition between geometrical balance of ferromagnetic and antiferromagnetic correlations among spins [3–10].

The J_1 – J_2 spin-1/2 chain also provides a minimal model for understanding the multiferroic behavior of the quasi-1D edge-sharing cuprates, such as LiCu_2O_2 [11, 12], LiCuVO_4 [13, 14], CuCl_2 [15] and $\text{PbCuSO}_4(\text{OH})_2$ [16, 17], in which the ferroelectricity is found to be of spin origin and inherently related to a vector spin chirality, $(\mathbf{S}_i \times \mathbf{S}_j)$ of nearby spin \mathbf{S}_i [18]. A spin excitation gap is therefore believed to be required for protecting the vector spin chirality ordered state without a magnetic spiral long-range order in multiferroic cuprates. By introducing a small bond alternation in the nearest-neighboring ferromagnetic exchange and an easy-plane anisotropy into the isotropic J_1 – J_2 Hamiltonian, [19] proposed a gapped vector-chiral dimer state in the absence of magnetic field based on density-matrix renormalization-group calculations. Such gapped vector-chiral phase was also obtained for the J_1 – J_2 Heisenberg model with added uniform Dzyaloshinskii–Moriya (DM) interaction by using the numerical Lanczos diagonalization [20, 21]. However, it is difficult to handle the DM interaction analytically [22], and the role of DM interaction in the vector-chiral state is needed to be further clarified. In the present study, we revisit a vector spin chiral bond that is coupled with phonons by the spin–phonon interaction of DM type. We reveal analytically that a novel first-order phase transition is induced by the formation of a vector-spin-chirality bound state. This phase transition is *gapless* in energy but causes a strongly dynamical suppression of

decoherence in the quantum and thermal spin fluctuations as the strength of spin–phonon interaction increasing.

Spin–phonon interaction

For convenience's sake, let us define a dimensionless vector spin chirality along the chain as,

$$\chi_i = \frac{\mathbf{S}_i \times \mathbf{S}_{i+1}}{|\mathbf{S}_i \times \mathbf{S}_{i+1}|}. \quad (1)$$

Under the uniform DM interaction, the $SU(2)$ spin symmetry is broken down to $SO(2) \times \mathbb{Z}_2$. χ_i corresponds to a spontaneous break of the discrete \mathbb{Z}_2 symmetry. Along the two-fold symmetry axis, χ_i^z has only two eigenvalues, $+1$ and -1 , but its direction remains unfixed because of strong thermal/quantum fluctuation of spins. Particularly for spin-1/2 systems, given that the spin fluctuations does spontaneously reverse the local spin (i.e., $\mathbf{S}_i \rightarrow -\mathbf{S}_i$), any incoherent spin fluctuation of two neighboring spins would result in χ_i -flip. In other word, the vector spin chirality χ_i of spin-1/2 systems behaviors as a two-level system with strong fluctuations. In the present study, we can take χ_i as the (pseudo-)Pauli operators. The associated local (ferro-)electric polarization based on the general spin-current model [23–27] reads $\mathbf{P}_i = e\mathbf{u}_i \sim \hat{\mathbf{e}}_{ij} \times \chi_i$, where $\hat{\mathbf{e}}_{ij}$ is the unit vector connecting two spins, \mathbf{S}_i and \mathbf{S}_j . This polarization corresponds to the transverse optical phonons in relevant cuprates. Accordingly, we have a linear coupling between the (transverse) phonon mode and the spin chirality,

$$H_{\text{DM}} = \frac{\lambda}{2} \mathbf{u}_i \cdot \chi_i, \quad (2)$$

where λ stems from the spin–orbit interaction [23, 25]. \mathbf{u}_i is the transverse atomic displacement (here the redefined $\mathbf{u}_i = \mathbf{u}_i \times \hat{\mathbf{e}}_{ij}$ has been exploited for a condensed form with no loss of physical properties), satisfying the (optical) phonon model

$$H_p = \frac{\kappa}{2} \mathbf{u}_i^2 + \frac{1}{2M} \mathcal{P}_{\text{ui}}^2 \quad (3)$$

with κ and M being the spring constant and the effective mass of \mathbf{u}_i , respectively. \mathcal{P}_{ui} is the momentum operator. Minimizing the $(H_{\text{DM}} + H_p)$ yields the local condition, $\mathbf{u}_i = -\frac{\lambda}{2\kappa} \chi_i$. However, it should be noted that the displacement \mathbf{u}_i is not essential to the spin-driven ferroelectricity. \mathbf{u}_i represents the contribution of the displacement of electronic cloud as well. H_{DM} corresponds to the DM interaction once the static displacement (\mathbf{u}_i) is nonzero and breaks the space inversion symmetry.

On the other hand, considering that the exchange interaction J falls off as a power law with the separation of the magnetic ions,

$$J(|\mathbf{r}_i - \mathbf{r}_j|) = J(|\mathbf{R}_i + \mathbf{u}_i - (\mathbf{R}_j + \mathbf{u}_j)|)^{-\gamma} \quad (4)$$

with γ in the range 6–14 [28]. \mathbf{R}_i is the bare value of the position of the magnetic atom at site i , and $|\mathbf{R}_i - \mathbf{R}_j|$ determines the lattice constant (set here to 1). The dynamical exchange striction of transverse displacement [29],

$$J(|\mathbf{r}_i - \mathbf{r}_j|) \approx J \left[1 - \frac{\gamma}{2} (\mathbf{u}_i - \mathbf{u}_j)^2 \right] \quad (5)$$

intrinsically generates a quadratic coupling, $\sim \mathbf{u}_i \cdot \mathbf{u}_j$ between the neighboring transverse displacements, which gives rise to an effective coupling between the neighboring spin chirality χ_i and χ_j under the inverse DM mechanism in the J_1 – J_2 spin chain.

With the aforementioned properties in hand, after applying the molecular mean-field approximation to the spin-chirality interactions, we have an effective spin–phonon model that describes the coupling between the local spin-chirality χ_i and the transverse phonon modes

$$H = \sum_i H_p + \sum_i H_{\text{DM}} + \sum_i \frac{\mathcal{J}_i^z}{2} \chi_i^z - \sum_i \frac{\mathcal{J}_x}{2} \chi_i^x, \quad (6)$$

where \mathcal{J}_i^z is an effective local Zeeman energy given by the interactions with the neighboring χ_j ($j \neq i$) after taking into account of the above dynamic exchange-striction effect and other inversion-symmetry-broken effects including applied external electric field. Uniform \mathcal{J}_x is introduced to value the strength of thermal/quantum fluctuations that gives rise to a transition between the two basis eigenstates at given temperature (i.e., $|\uparrow\rangle$ and $|\downarrow\rangle$) of χ_i^z . Here the z -axis is determined by the direction of the non-vanishing ferroelectric polarization, $\mathbf{P} \sim \frac{1}{N} \sum_i \langle \mathbf{u}_i \rangle$ with N being the number of spins. Equation (6) describes the ‘free’ spin-chiralities that couple separately with the phonons via the DM interaction. Furthermore, as demonstrated below, one can see it is the fluctuation \mathcal{J}_x that dominates the non-equilibrium dynamics of χ_i , which can be effectively described by the spin-boson model, consisting in a single two-level χ_i interacting with the transverse phonon bath.

Spin-chirality bound state

In the presence of the spin–phonon coupling, complete information about the interaction effects (H_{DM}) can be further encapsulated in the spectral function, $g(\omega) = \pi \sum_{\mathbf{k}} |\lambda_{\mathbf{k}}|^2 \delta(\omega - \omega_{\mathbf{k}})$ in the momentum \mathbf{k} -representation of discrete phonon modes after thermal equilibrium average [30]. Here $\lambda_{\mathbf{k}}$ denotes the effective coupling strength between the spin chirality and the k th mode of transverse phonons with frequency $\omega_{\mathbf{k}}$. The Hamiltonian H of a single vector-spin-chirality can then be rewritten as the spin-boson model

$$H = \sum_{\mathbf{k}} \omega_{\mathbf{k}} a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} + \sum_{\mathbf{k}} \frac{\lambda_{\mathbf{k}}}{2} \chi_i^z (a_{\mathbf{k}}^{\dagger} + a_{\mathbf{k}}) + \frac{\mathcal{J}_z}{2} \chi_i^z - \frac{\mathcal{J}_x}{2} \chi_i^x \quad (7)$$

with $a_{\mathbf{k}}^{\dagger}$ ($a_{\mathbf{k}}$) being the phonon creation (annihilation) operator. In the Debye model, there are no phonon modes with the frequency above the Debye frequency ω_{D} . Such an abrupt cutoff can result in a simple form of the spectral density $g(\omega)$ in the continuum limit [30],

$$g(\omega) = 2\pi\alpha\omega_{\text{D}}^{1-s} \omega^s \Theta(\omega_{\text{D}} - \omega), \quad (8)$$

where α is a dimensionless coupling constant and $\Theta(x)$ is the usual step function. For the case of (three-dimensional) phonon-related interaction in a solid, s can be 3 or 5 for frequencies well below the Debye frequency [31, 32]. However, in general, the frequency behavior of $g(\omega)$ is complicated, especially in the low dimensional systems. In the following discussion, we treat s as a free parameter with no *qualitative* change in conclusion of the present study.

Under the unitary transformation $U = \exp(-i\pi\chi_i^y/4)$ and the successive the rotating-wave approximation (RWA) [33], we have then a transformed integrable Hamiltonian \tilde{H} (hereafter, transformed quantities are marked by a tilde),

$$\tilde{H} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} + \sum_{\mathbf{k}} \frac{\lambda_{\mathbf{k}}}{2} (\tilde{\chi}_i^{\dagger} a_{\mathbf{k}} + \tilde{\chi}_i^{-} a_{\mathbf{k}}^{\dagger}) + \frac{\mathcal{J}_x}{2} \tilde{\chi}_i^z. \quad (9)$$

Noted that $\mathcal{J}_z = 0$ is assumed in \tilde{H} at the first step in analytical discussions for emphasizing the effect of spin fluctuations. As for the case of $\mathcal{J}_z \neq 0$, we find that \mathcal{J}_z does modify slightly the tunneling parameter [34], but has no qualitative influence on the non-equilibrium dynamics of spin-chirality discussed in the following section. We have now $[\hat{N}, \tilde{H}] = 0$ with $\hat{N} = \sum_{\mathbf{k}} a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} + \tilde{\chi}_i^{\dagger} \tilde{\chi}_i^{-}$ being the total excitation number operator, and the eigenstate of \tilde{H} is given by the direct sum of the subspace with definite quantum number N .

As $\lambda_{\mathbf{k}} = 0$, the spin chirality decouples completely from the phonons. The ground state is given by a tensor product of the zero-excitation modes of two subsystems, i.e., $|\tilde{\psi}_0\rangle = |\tilde{\downarrow}\rangle \otimes |0_{\mathbf{k}}\rangle$ with $\tilde{\chi}_i^{-} |\tilde{\downarrow}\rangle = 0$, $a_{\mathbf{k}} |0_{\mathbf{k}}\rangle = 0$, and $E_0 = \langle \tilde{\psi}_0 | \tilde{H} | \tilde{\psi}_0 \rangle = -\mathcal{J}_x/2$. The expected value of spin chirality in the original space reads then

$$\langle \chi_i^z \rangle_0 = \langle \tilde{\psi}_0 | U \chi_i^z U^{\dagger} | \tilde{\psi}_0 \rangle = 0.$$

Consequently, we have zero value of ferroelectric signal, and the long-range order of spin chirality is broken down by the spin fluctuations even down to zero temperature. However, $|\tilde{\psi}_0\rangle$ is not always the ground state of the whole system when the spin–phonon coupling is turned on, as shown below.

Considering the single-excitation state with $N = 1$, $|\tilde{\psi}_1\rangle = c_0 |\tilde{\uparrow}\rangle \otimes |0_{\mathbf{k}}\rangle + \sum_{\mathbf{k}} \alpha_{\mathbf{k}} |\tilde{\downarrow}\rangle \otimes |1_{\mathbf{k}}\rangle$, its eigen-energy E_1 is determined by the following transcendental eigenequation [33],

$$\mathcal{F}(E_1) \equiv \frac{\mathcal{J}_x}{2} - \frac{1}{4\pi} \int_0^{\infty} \frac{g(\omega)}{\omega - (E_1 + \frac{\mathcal{J}_x}{2})} d\omega = E_1. \quad (10)$$

The analysis for algebraic relationship of above equation reveals two inequality constrains,

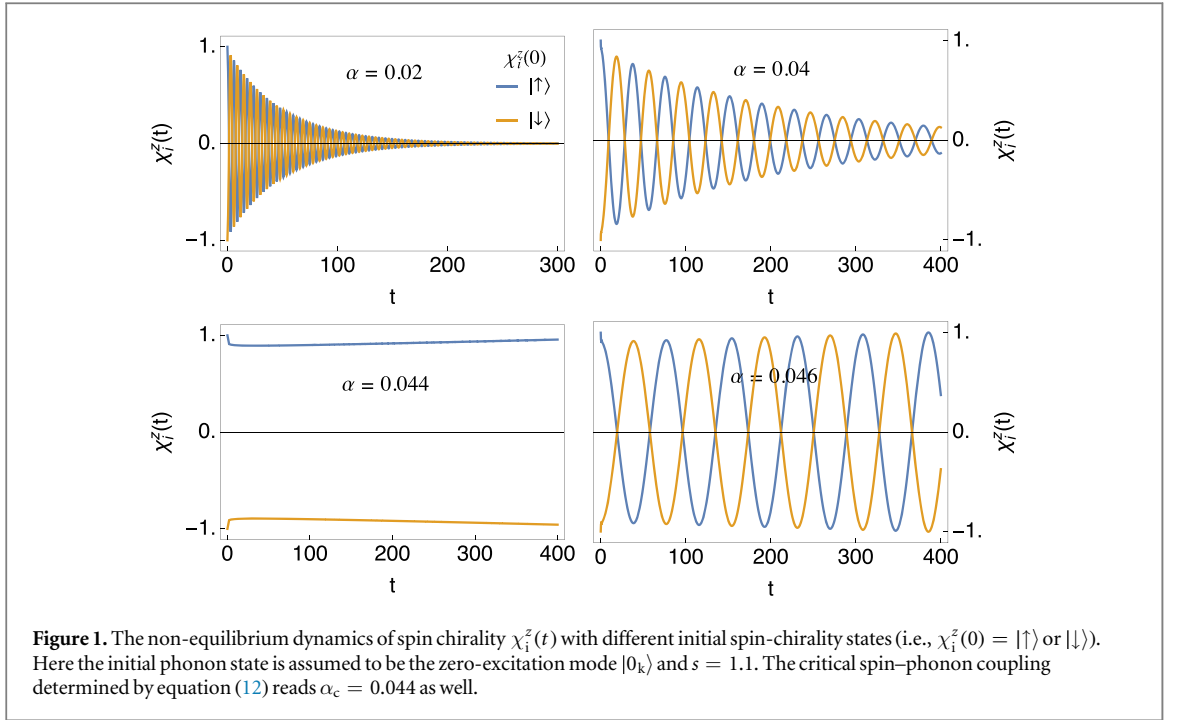
$$E_1 < -\mathcal{J}_x/2 \quad \text{and} \quad \mathcal{F}(-\mathcal{J}_x/2) \leq -\mathcal{J}_x/2. \quad (11)$$

Once such conditions are satisfied, $\mathcal{F}(E_1)$ always has one and only one *real* root, which is exact the eigen-energy E_1 of the formed bound state $|\tilde{\psi}_1\rangle$ of the system in the presence of the spin–phonon interaction of DM type. For a excited state with $N \geq 2$, its eigenvalue is found to be always greater than E_1 . This suggests that the state with higher-phonon modes cannot be the ground state.

The above inequality constraints, equation (11), yields a critical value of the spin–phonon coupling,

$$\alpha_c = 2s\mathcal{J}_x/\omega_{\text{D}}, \quad (12)$$

which is simply determined by the ratio between the strength of spin fluctuations and the Debye frequency of phonons. In the case of strong spin–phonon interaction, i.e., $\alpha \geq \alpha_c$, the ground state of the system is not the zero-excitation mode $|\tilde{\psi}_0\rangle$ but the single-excitation bound state $|\tilde{\psi}_1\rangle$. Considering that $\langle \tilde{\psi}_1 | \tilde{\psi}_0 \rangle = 0$ and the discontinuity of the first derivative of the ground state energy, one can see that the system undergoes a first-order phase transition, accompanying with a *gapless* change in the ground state from $|\tilde{\psi}_0\rangle$ to $|\tilde{\psi}_1\rangle$ by increasing the spin–phonon interaction.



As a well-established feature of the bound state, the excited state population in $|\tilde{\psi}_1\rangle$ should be constant in time. For a more clearer picture of the decoherence dynamics of the spin chirality, let us investigate in detail the time evolution of the spin chirality under different strength of spin–phonon coupling but with same initial state,

$$|\Psi(0)\rangle = |\uparrow\rangle \otimes |0_k\rangle \text{ or } |\downarrow\rangle \otimes |0_k\rangle. \quad (13)$$

The expected value of the spin chirality at time t reads then

$$\langle \chi_1^z(t) \rangle = \langle \Psi(0) | e^{iHt} \chi_1^z e^{-iHt} | \Psi(0) \rangle = \sqrt{2} \Re[c(t)]. \quad (14)$$

Here the probability amplitude $c(t)$ is given by the Schrödinger equation under the RWA,

$$\dot{c}(t) + i\frac{J_x}{2}c(t) + \frac{1}{4\pi} \int_0^\infty \int_0^t J(\omega) \exp\left[-i\left(\omega - \frac{\omega_0}{2}\right)(t - \tau)\right] c(\tau) d\tau d\omega = 0 \quad (15)$$

with the initial condition $c(0) = \frac{1}{\sqrt{2}}$ or $c(0) = -\frac{1}{\sqrt{2}}$, respectively. In figure 1, $\langle \chi_1^z(t) \rangle$ are presented with different α . Regardless of initial spin-chirality state ($|\uparrow\rangle$ or $|\downarrow\rangle$), $\langle \chi_1^z(t) \rangle$ shows an oscillatory decay to zero in the weak coupling regime ($\alpha < \alpha_c$) because of the absence of the bound state. However, for the case of strong spin–phonon interaction ($\alpha > \alpha_c$), the bound state is formed. The $|\Psi(t)\rangle$ component contains $|\tilde{\psi}_0\rangle$ and $|\tilde{\psi}_1\rangle$. We have then a lossless oscillation of $\langle \chi_1^z(t) \rangle$ with the frequency determined by the energy difference of these two eigenstates (i.e., $E_0 - E_1$). At the critical value of $\alpha = \alpha_c$, one has $E_0 = E_1$. The bound state $|\tilde{\psi}_1\rangle$ emerges in time. The quantum coherence from $|\tilde{\psi}_1\rangle$ does not change during the time evolution, which results in a finite asymptotical $\langle \chi_1^z(t) \rangle$. In other words, the decoherence of the spin chirality tends to be inhibited with the strong spin–phonon coupling.

Discussions

In conclusion, our analytical study reveals that the incoherent spin fluctuations can be strongly suppressed due to the formation of spin-chirality bound state driven by the spin–phonon coupling of DM type. The ratio between the spin fluctuations and the Debye frequency of phonons determines the critical point of spin–phonon interaction, beyond which the gapped spin-chiral state is resulted in. The local effective Zeeman field on the vector-spin-chirality χ_1 does not however have influence on the value of critical point. Experimentally, considering that the amplitude of spin fluctuations does depend on the temperature of system, such the first-order phase transition can be realized even in the system with moderate spin–phonon coupling by lowering the temperature to tone down the critical point.

In the absence of the spin–phonon coupling, the spin-chirality decouples completely from the phonon bath, the dynamical behavior of spin-chirality χ_1 becomes analogous to an isolated quantum two-level system. We have then two instantaneous eigenstates $|\pm\rangle$ with the eigen-energy $E_\pm = \pm \frac{1}{2} \sqrt{\mathcal{J}_z^2 + \mathcal{J}_x^2}$. Considering that the energy difference \mathcal{J}_z between the two diabatic (unperturbed) bases of the χ_1^z can be controlled via applying

electric field, the spin-chirality dynamics is described by the well-known Landau–Zener transition [35]. The transition probability reads, $P_{LZ} = \exp\left[-\frac{\pi \mathcal{J}_x^2}{2\hbar\nu}\right]$, where $\nu = |d\mathcal{J}_z/dt|$ is the Landau–Zener speed. P_{LZ} gives the probability of finding the system in the excited state at $t \rightarrow +\infty$ when it is started in the ground state at $t \rightarrow -\infty$. In the weak fluctuation limit ($|\mathcal{J}_z| \gg \mathcal{J}_x$), the Landau–Zener formula gives an excellent approximation to the actual transition probability, $P_{\uparrow} \approx P_{LZ}$. That is to say, when the external electric field changes very fast ($\nu \rightarrow \infty$), the transition probability $P_{\uparrow} \rightarrow 1$, and the spin-chirality will tend to completely jump from low energy level to the high energy level with evolution of times. As a quasi-static processes with $\nu \rightarrow 0$, the system is under the condition of the adiabatic limit, the transition probability $P_{\uparrow} \rightarrow 0$. Tunneling will mostly not happen and the system will always stay in the low energy level. However, when \mathcal{J}_z is given, the transition probability will be a constant value. In other words, if the initial state is $|\downarrow\rangle$ with time evolution we will find the final state $|\uparrow\rangle$ in a fixed probability. On the other hand, as the fluctuation \mathcal{J}_x becomes strong and is comparable to \mathcal{J}_z , P_{\uparrow} is found to oscillate coherently as a function of $E(t \rightarrow +\infty)$ due to the underlying transient dynamics of Landau–Zener transition. The system possesses the coherent Landau–Zener oscillations [36].

Acknowledgments

This work is supported by the National Natural Science Foundation of China (No. 11474138), the German Research Foundation (No. SFB 762), the Program for Changjiang Scholars and Innovative Research Team in University (No. IRT-16R35), and the Fundamental Research Funds for the Central Universities.

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