Electronic correlations, spectral and magnetic properties of ZrZn$_2$

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(Dated: March 9, 2020)

We present results of a theoretical study of a prototypical weak ferromagnet ZrZn$_2$. We use the density-functional theory (DFT)+dynamical mean-field theory (DMFT) method to study the electronic and local magnetic properties. The obtained DFT+DMFT electronic self-energies are Fermi-liquid like, indicating a small effective mass enhancement of the Zr 4d states $m^*/m \sim 1.1 - 1.3$ accompanied by partly formed local moments within the electronic states of $t_{2g}$ symmetry. The effect of electronic interaction is shown to be essential for determining the correct topology of some of the Fermi surface sheets, which is changed due to a correlation-induced shift of the peak of the density of states. To study in detail the pressure dependence of the Curie temperature $T_C$ and corresponding pressure-induced quantum phase transition, we consider an effective single-band model, constructed using the $Zr$ 4d contribution to the total density of states. The model is studied within static and dynamic mean-field theory, as well as spin-fermion approach. We show that the spin-fermion approach yields the pressure dependence $T_C(p)$ comparable well with the experimental data, including a first-order quantum phase transition at $p \approx 1.7$ GPa.

PACS numbers: 71.27.+a, 71.10.-w, 79.60.-i

I. INTRODUCTION

ZrZn$_2$ is a well-known weak ferromagnet. Despite its magnetic properties have been studied since the 1950s [1], their peculiarities are still actively debated. At ambient pressure this compound is ferromagnetic below the Curie temperature $T_C \approx 30$ K. The Curie temperature is, however, suppressed by pressure [2] and vanishes at $p \approx 1.65$ GPa [2,3]. It was argued in Ref. [3] that the quantum phase transition in ZrZn$_2$ under external pressure is in fact of the first order; at finite magnetic field the corresponding metamagnetic behavior is observed [3,4]. Near the quantum phase transition in zero magnetic field the exponent of the resistivity $\rho \propto T^\alpha$ changes [1] from the value $\alpha = 5/3$, which is characteristic for systems with ferromagnetic correlations [3,4], to $\alpha = 3/2$, characteristic for the antiferromagnetic correlations.

Density-functional theory (DFT) band structure calculations [2,3] of ZrZn$_2$ revealed an extended van Hove singularity due to a flat part of dispersion near the L point of the Brillouin zone, similar to that in nickel [11]. This flat part yields a peak of the density of states near the Fermi level [10,12], characteristic to many ferromagnetic materials. This peak on one hand promotes ferromagnetism, cf. Ref. [13], but on the other hand, it makes the Stoner theory even qualitatively inapplicable at finite temperatures, since competing channels of electron scattering become important in this situation, as it was studied actively in two dimensional systems with van Hove singularities [14]. In general, Stoner theory predicts transition temperatures much larger than the corresponding experimental data and does not explain the linear temperature dependence of inverse susceptibility [5]. Therefore, correlation effects become especially im-
ties based on a combination of density functional theory and dynamical mean-field theory of correlated electrons (DFT+DMFT) \cite{22} have shown to be a powerful theoretical tool for studying the physics of real materials \cite{22}. Second aspect is the applicability of the effective single-band models with realistic densities of states, and a possibility to use them to study magnetic properties of weak itinerant magnets.

In this paper, by employing the DFT+DMFT method we explore the effect of local Coulomb correlations on the electronic structure and magnetic properties of ZrZn$_2$ (space group Fd$ar{3}$m). We interpret the results of multi-orbital DFT+DMFT calculation within the effective one-band model constructed using the realistic density of states of ZrZn$_2$ and solved by DMFT. To understand details of the paramagnet to ferromagnet transition the one-band model is studied within static mean field and spin-fermion model approaches.

The paper is organized as follows. In Sec. II A we describe the technical details of the DFT+DMFT approach. The corresponding results for the spectral properties, local spin susceptibility and temperature dependence of the uniform spin susceptibility of ZrZn$_2$ are discussed in Sec. II B. The effective single-band model is considered in Sect. III within DMFT (Sect. III A), mean-field approach (Sect. III B) and spin-fermion model (Sect. III C). Finally, the results are briefly summarized in Sec. IV. In Appendix we provide details on the equations of the spin-fermion model used to account the effect of spin fluctuations.

II. DFT+DMFT STUDY

A. Method

To study the effect of electronic correlations on the electronic structure and magnetic properties of ZrZn$_2$ we have employed the DFT+DMFT method implemented within the plane-wave pseudopotential approach with generalized gradient approximation in DFT \cite{22}. We use a basis set of Wannier functions constructed by means of the projection procedure \cite{23,24} for an energy window spanning occupied Zn $3d$ and partially filled Zr $4d$ bands. The DFT+DMFT realistic many-body problem is solved by the continuous-time hybridization-expansion quantum Monte-Carlo method (segment algorithm) \cite{22}. In these calculations we neglect effects of spin-orbit coupling. The Coulomb interaction term has been treated in the density-density approximation with the average Hubbard interaction $U = 2.5$ eV and Hund’s exchange $J = 0.3$ eV for the Zr $4d$ orbitals \cite{22}. To account for Coulomb interaction energy already described by DFT we employ the fully-localized double-counting correction $V_{dc} = U(N_d - 0.5) - 0.5J(N_d - 1)$ self-consistently evaluated from local occupations $N_d$. Spectral functions and orbital-dependent band mass renormalizations were computed using the real-axis self-energy $\Sigma(\nu)$ obtained from the Padé analytical approximation of the DFT+DMFT imaginary-axis self-energy $\Sigma(i\nu)$ \cite{30}. In order to determine the lattice parameter of cubic ZrZn$_2$ under pressure we compute the total energy as a function of volume using the GGA energy functional and shift the experimental lattice constant according to the third-order Birch-Murnaghan equation of state \cite{31}. For simplicity, only hydrostatic contraction and expansion of the cubic unit cell are considered in these calculations.

Within DFT+DMFT we compute the uniform spin susceptibility as a derivative of the field-induced magnetization $M(T)$ with respect to the energy correction $E_h$ corresponding to the applied field:

$$\chi(T) = \frac{\partial M(T)}{\partial E_h} = \frac{\partial [n_\uparrow(T) - n_\downarrow(T)]}{\partial E_h},$$

where $n_\sigma(T)$ is the occupation of the spin-$\sigma$ at a temperature $T$. In these calculations we check an absence of polarization in the zero field and the linear character of $M$ as a function of $E_h$.

B. Results

1. Uniform spin susceptibility

In Fig. I the inverse uniform spin susceptibility $\chi^{-1}(T)$ of ZrZn$_2$ computed by DFT+DMFT is shown in comparison to experimental data of Shimizu et al. \cite{32}. In the temperature range from $\sim 400$ K to $\sim 1200$ K the calculated temperature dependence of $\chi^{-1}$ to a good accuracy is found to be linear, indicative of the Curie-Weiss-like character of the high-temperature susceptibility. The obtained slope of high-temperature part $\chi^{-1}(T)$ is in agreement with the experimental data, although with some shift of the inverse susceptibility, which origin is to be clarified by future investigations. To highlight the linear region we show a least square fit to the last five computed points. Orbitally-resolved contributions $\chi_\alpha (\alpha = t_{2g}, e_g)$ to the total susceptibility show a strong orbital-selective behavior (Fig. I inset). In particular, we observe that the temperature evolution of $\chi(T)$ curve in a wide temperature range mostly originates from the $t_{2g}$ orbitals. The contribution of the $e_g$ states is an order of magnitude smaller and becomes notable only in the low-temperature region.

Below 400 K theoretical $\chi^{-1}(T)$ shows an upturn which is not observed in experimental data. This upturn is found in DFT+DMFT studies of many other systems with van Hove singularity, slightly shifted off the Fermi level \cite{33,34}, and physically implies a tendency towards non-ferromagnetic (in particular, incommensurate) order, overestimated in DMFT for ZrZn$_2$. This stresses the importance of non-local correlations in the considered multi-band model. On the other hand, this observation agrees with the jump of the exponent of resistivity near the quantum phase transition from the value $5/3$, characteristic for ferromagnetic correlations to the value...
obtained by DFT+DMFT approach. The deviation from the linear dependence is not as strong as the experimental data at ambient pressure, although the temperature dependence of the orbitally-resolved uniform susceptibility $\chi$ at ambient pressure.

FIG. 1: (Color online). Temperature dependence of the inverse uniform spin susceptibility $\chi^{-1}(T)$ of paramagnetic ZrZn$_2$ computed by DFT+DMFT at ambient pressure (a.p.) (black squares) and 2 GPa (red rhombs) in comparison with experiment at ambient pressure (broken line) [32]. The straight solid lines represent least-square fits to the computed points in the temperature range [400, 1200] K. The inset shows temperature dependence of the orbitally-resolved uniform susceptibility $\chi_i(T)$ at ambient pressure.

3/2, characteristic for spin density wave correlations. We note that the fection of temperature dependence of the inverse susceptibility $\chi^{-1}(T)$ at low $T$ is also present in the experimental data at ambient pressure, although the deviation from the linear dependence is not as strong as obtained by DFT+DMFT approach.

To extract a characteristic temperature $T_C^*$ of the onset of strong ferromagnetic correlations (which is approximately identified with the Curie temperature $T_C$) we extrapolate the dependence of susceptibility at high temperatures according to the Curie-Weiss law, $\chi^{-1} \propto T - T_C^*$. The extrapolation of the fit to $\chi^{-1} = 0$ at ambient pressure gives the temperature $T_C^* \sim 32$ K, close to the value reported in Ref. [32].

Upon increasing pressure we observe a growth of $\chi^{-1}(T)$ in the temperature range $T < 1200$ K. The effect of pressure is accompanied by a monotonic reduction of $T_C^*$. In particular, at $p = 1$ GPa we obtain a drop of $T_C^*$ to $\sim 14$ K which further develops to $\sim 4$ K at $p = 2$ GPa. This behavior is in overall qualitative agreement with experimental tendency of decreasing the Curie temperature under compression of the lattice [3–6]. The upturn of $\chi^{-1}(T)$ at low temperatures and slower pressure dependence of $T_C^*$ compared to that of the Curie temperature in the experiment, emphasizes the necessity of considering non-local correlation effects and the role of structural relaxation with account of local Coulomb interaction to determine the structural parameters of ZrZn$_2$ under pressure. The latter is in line with a slight underestimation of the equilibrium volume predicted by the local density approximation [12, 36].

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FIG. 2: (Color online). Upper panel: Temperature dependence of the inverse orbitally-resolved local spin susceptibility $\chi^{-1}_{loc}$ of paramagnetic ZrZn$_2$ obtained with DFT+DMFT. The inset shows a zoom for total $\chi^{-1}_{loc}$ and the corresponding $t_{2g}$ orbitals contribution. Lower panel: Local spin correlation function $\chi(\tau) = \langle \hat{m}_z(\tau)\hat{m}_z(0) \rangle$ of paramagnetic ZrZn$_2$ as computed by DFT+DMFT at $T = 464$ K. Orbitally-resolved Fourier transform of $\chi(\tau)$ as a function of the real energy is shown in the inset.

2. Local susceptibility

To complement the analysis of magnetic properties we consider the local spin susceptibility $\chi(\tau) = \langle \hat{m}_z(\tau)\hat{m}_z(0) \rangle$ (where $\hat{m}_z(\tau)$ is the instantaneous magnetization of the impurity site at imaginary time $\tau$ and $\langle \ldots \rangle$ denotes the thermal average computed by CT-QMC) and its Fourier transform $\chi_{loc}(\lambda) = \int d\tau e^{i\lambda \tau} \chi(\tau)$, $\omega_n$ being bosonic Matsubara frequencies. We first discuss the temperature dependence of the static local spin susceptibility $\chi_{loc} = \chi_{loc}(0)$. Our results for the temperature dependence of $\chi_{loc}$ are shown in Fig. 2 (upper panel). Calculated $\chi_{loc}$ to a good approximation is a linear function of temperature. From the relation $\chi_{loc} \propto T + \sqrt{2}k_B T_K$ [37, 38] we find a rather large Kondo temperature $T_K \approx 1000$ K for the $t_{2g}$ states.
and much larger value for the $e_g$ states, which shows that local moments are absent (fully screened) at low temperatures. The presence of flat part of time dependence of $\chi(\tau)$ (see lower panel of Fig. 2) shows that in the $t_{2g}$ band there are short lived local moments with the inverse lifetime, determined by the half width of the peak of the analytical continuation of $\chi(\omega)$ at the half height $2\hbar\omega_F = 0.16\ eV$. This corresponds to the lifetime 25 fs, which is approximately twice longer than that discussed previously for the iron pnictides [40] and $\epsilon$-iron [34], but much shorter than the lifetime of local moments in such strong magnet as $\alpha$-iron [11]. At the same time, almost no local moments are formed in the $e_g$ states. This implies a different degree of electronic coherence for different orbitals of the Zr $4d$ shell and is indicative for orbital-selective local moments.

Next, we discuss the effect of electronic correlations on the spectral properties of ZrZn$_2$. The spectral functions of ZrZn$_2$ calculated by DFT and DFT+DMFT for the experimental crystal structure are presented in Fig. 3. In agreement with previous theoretical investigations [12, 30] our results (both DFT and DFT+DMFT) show that the spectral weight in the vicinity of the Fermi energy ($E_F$) is due to the Zr $4d$ orbitals and mostly originates from the $t_{2g}$ states. These orbitals form a narrow band located in the interval $(-0.5, 0.5)$ eV with a sharp peak above $E_F$. We note however that within DFT the $e_g$ spectral function also shows a peaked feature above the Fermi level, but its amplitude is almost five times smaller than that due to the $t_{2g}$ states. In addition, we observe that the Zn-$3d$ band is located well below $E_F$. The Zr-$4d$ and Zn-$3d$ states are weakly hybridized and the latter provide no contribution close to the Fermi level.

The Zr-$4d$ spectral functions computed by DFT+DMFT share a common shape with those obtained within DFT. Correlation effects only lead to a shift and renormalization of the quasiparticle bands near $E_F$ and do not induce a significant transfer of the spectral weight. In particular, we observe that the sharp peak originating from the $t_{2g}$ states emerges at $\sim 0.025\ eV$, almost twice closer to $E_F$ compared to its position in DFT ($\sim 0.05\ eV$). Such an effect of shifting the peak of the density of states towards Fermi level is common for other correlated metallic systems. For example, it occurs in $\alpha$- [41], $\gamma$- [35] and $\epsilon$-iron [34] and some two-dimensional systems (see, e.g., Refs. [12, 14]). The $e_g$ spectral function shows a similar transformation. However, the $e_g$-derived peak is pushed from above to below the Fermi energy indicating a possible topological transformation of the Fermi surface due to electronic correlations at ambient pressure, as discussed below. The obtained shape of the spectral functions is preserved in the whole pressure range $p < 2\ GPa$ with all the features shifted to higher energies with increasing pressure. In particular, at $p = 2\ GPa$ the peak of the $e_g$ states reaches the Fermi level.

To quantify electronic correlations in ZrZn$_2$ we analyze the local DFT+DMFT self-energy $\Sigma(i\nu_n)$ and the effective band mass enhancement $m^*/m = 1 - \partial \text{Im} \Sigma(i\nu)/\partial(i\nu)|_{i\nu \to 0}$. Here $i\nu_n$ is the fermionic Matsubara frequency and the derivative $\partial \text{Im} \Sigma(i\nu)/\partial(i\nu)$ is computed using Padé extrapolation of $\Sigma(i\nu)$ to $i\nu = 0$. Our results for the frequency dependence of orbitally-resolved self-energies $\text{Im} \Sigma_{m}(i\nu_n)$ ($m = t_{2g}$, $e_g$) at an electronic temperature $T = 464\ K$ are presented in Fig. 4. We observe that the $e_g$ self-energy exhibits a Fermi liquid-like behavior with insignificant damping of quasiparticles ($\text{Im} \Sigma_{e_g}(0) \sim 0.002\ eV$) and yields the mass enhancement $m^*/m = 1.16$. The $t_{2g}$ states are less coherent and show a stronger renormalization with $\text{Im} \hat{\Sigma}_{t_{2g}}(0) \sim 0.015\ eV$ and $m^*/m = 1.34$. These results are in line with or-

|FIG. 3: (Color online). Orbitally-resolved spectral functions of paramagnetic ZrZn$_2$ computed by DFT+DMFT ($T = 464\ K$) at ambient pressure (a.p.) and $p = 2\ GPa$. The DFT results are shown in the inset. The Fermi energy is set to 0 eV.|

|FIG. 4: (Color online). Orbitally-resolved imaginary parts of the local self-energy of paramagnetic ZrZn$_2$ at ambient pressure on the Matsubara mesh obtained with DFT+DMFT ($T = 464\ K$).|
bital selectivity of local moments traced from the local susceptibility.

We proceed further with investigation of the effect of electronic correlations on the Fermi surface (FS) of ZrZn$_2$. To compute the FS within DFT+DMFT we locate poles of the lattice Green function $G(k,0)$, diagonalized in the orbital space. In Fig. 5 we compare our results for the FS computed by DFT and that obtained by DFT+DMFT at ambient pressure. Specifically, we observe that upon inclusion of electronic correlations the topology of one of the FS sheets changes near the $L$ point of the Brillouin zone, such that DFT+DMFT Fermi surfaces are in agreement with the experimental data [45]. This transformation occurs due to above discussed shift of the $e_g$-derived peak of the density of states from its position above the Fermi level in DFT to the energies below $E_F$ in DFT+DMFT. Therefore, correlations are crucially important to obtain the correct topology of the Fermi surface of ZrZn$_2$ (see also discussion in Ref. [45]).

At high pressures $p > 2$ GPa the neck encircling the $L$ point disappears, but the resulting sheets are nearly nested. This may explain antiferromagnetic correlations, which appear in the paramagnetic phase and yield experimentally observed $T^{3/2}$ dependence of the resistivity in the paramagnetic state under pressure [3].

III. EFFECTIVE SINGLE-BAND MODEL

A. The model and the dynamical mean-field theory

To study in more detail the phase transition to ferromagnetic phase, we construct an effective single band model. To construct this model we use a cut of the realistic total density of states of ZrZn$_2$ computed by DFT in an energy window chosen to span the full width of Zr-$4d$ band for each considered pressure. The normalized density of states defines a single-band model which is solved by dynamical and static mean-field theory, as well as investigated within the spin-fermion model approach.

We first compare the results of the dynamical mean-field theory for the single-band model, constructed as discussed above, with the results of Sect. IIIB for multi-band model. In DMFT calculations for single-band model we use the local Coulomb interaction $U = 4.5$ eV chosen such that the value of characteristic temperature $T_C^*$ at ambient pressure is close to that obtained for the multi-band model. In Fig. 6 we show the temperature dependence of the inverse spin susceptibility $\chi^{-1}(T)$ computed by DMFT at ambient pressure and $p = 2$ GPa. Similar to the multi-orbital model the computed inverse susceptibility shows an upturn in the low-temperature region, corresponding to the tendency to the incommensurate order. By analogy to the multi-orbital case we compute the characteristic temperature $T_C^*$ of the onset of strong ferromagnetic correlations from a linear fit of the high-temperature part of $\chi^{-1}(T)$. At ambient pressure our calculations yield a $T_C^* \sim 30$ K which drops to $\sim 10$ K at $p = 2$ GPa in qualitative agreement with the realistic DFT+DMFT calculation (Fig. 6 inset). Studying the paramagnetic to ferromagnetic phase transition in details requires an account for non-local correlations and comparison of energies of para- and ferromagnetic state. This is performed in the following subsections on the basis of a simplified mean-field theory based approach.

B. Mean-field approximation

The simplest methods to treat the electron correlation effects is the static mean-field approximation, described by the action
To find energetically preferable solution, we compare the paramagnetic (\(\Omega_{\mu, n} = \langle n | n \rangle \)) and ferromagnetic (\(\Omega_{\mu, s} = \langle s | s \rangle \)) solutions.

We solve the equations (1) for \(\mu\) and \(\langle s \rangle\) for a given electron concentration per orbital \(\langle n \rangle\), obtained in DFT calculations. This yields paramagnetic (\(\mu_{\text{PM}}\), \(\langle s \rangle = 0\)) and ferromagnetic (\(\mu_{\text{FM}}\), \(m_{\text{FM}} = \langle s \rangle \neq 0\)) solutions. To find energetically preferable solution, we compare the values of thermodynamic potential

\[
\Omega(\mu; \langle n \rangle, \langle s \rangle) = U \left( \frac{\langle s \rangle^2}{4} - \frac{\langle n \rangle^2}{4} \right) - \frac{1}{2} \sum_{\sigma} \int f \left( \tilde{\epsilon} + \sigma U \langle s \rangle \right) \rho(\epsilon) d\epsilon.
\]

If \(\Omega(\mu_{\text{PM}}; \langle n \rangle, 0) < \Omega(\mu_{\text{PM}}; n_{\text{FM}}, \langle s \rangle_{\text{FM}})\), where \(n_{\text{PM}}\) and \(\langle s \rangle_{\text{PM}} \neq 0\) fulfill the mean-field equations for \(\mu = \mu_{\text{PM}}\), then the paramagnetic solution is considered energetically preferable, while in case \(\Omega(\mu_{\text{FM}}; \langle n \rangle, m_{\text{FM}}) < \Omega(\mu_{\text{FM}}; n_{\text{FM}}, 0)\), where \(n_{\text{FM}}\) and \(\langle s \rangle = 0\) is the solution of MF equations for \(\mu = \mu_{\text{FM}}\), the ferromagnetic solution dominates.

Solution of the equations (1) and (5) in the ground state for moderate Coulomb interaction shows that the ferromagnetic ground state is energetically preferable for positions of the peak of the density of states close to the Fermi level (sufficiently low pressures), while paramagnetic state is energetically preferable for peak positions far from the Fermi level.

We note that the effective interaction \(U\) in the static mean-field theory is typically smaller than the bare Coulomb interaction of the one-band model, since it accounts for the screening processes, cf. Refs. \[46, 47\]. We choose below the effective Coulomb interaction \(U = 1.8\) eV from the condition that the first-order quantum phase transition occurs at the pressure \(p_c\) close to the experimental value \(p_c = 1.65\) GPa.

The calculated dependence of Curie temperature \(T_C(p)\) in the mean-field theory is presented in Fig. 7. The obtained pressure dependence of \(T_C\) is in qualitative agreement with the experimental data \[5, 6\]. However, the obtained Curie temperatures overestimate experimental values by an order of magnitude, which is usual for mean-field theory of weak magnetic systems \[5\]. In the narrow region of pressures near quantum phase transition, where none of the above criteria of preference of paramagnetic or ferromagnetic solution is fulfilled, the phase separation between para- and ferromagnetic phases occurs. We do not study this region in details since it is rather narrow for ZrZn2.

C. Calculation of critical temperature from the spin-fermion model

To improve the results of the mean-field approximation, let us take into account the effect of spin fluctuations in the spin-fermion model. To this end, we consider the partition function

\[
Z = \int D[\epsilon] \int d^3 \tilde{S} \exp \left( -\beta S_{\text{eff}} \right)
\]

with the effective action

\[
S_{\text{eff}} = S_{\text{MF}} + 2U s \tilde{S} + D \tilde{S}^2,
\]

containing fluctuating field \(\tilde{S}\); \(s = (1/2) \sum_{k, \sigma, \sigma'} c_{k, \sigma} \sigma \sigma' c_{k, \sigma'}\) corresponds to the spin of itinerant degrees of freedom, \(\sigma = (\sigma_x, \sigma_y, \sigma_z)\) is the vector of Pauli matrices, \(D\) is the strength of spin fluctuations. For simplicity we consider here only the effect of spin fluctuations of static uniform (\(q = 0, \omega_n = 0\)) spin mode, which can be justified at sufficiently large correlation lengths \[16, 19, 48\]. We also note that at finite temperatures the classical fluctuations in dimension \(d = 3\) are relevant from renormalization group point of view, in contrast to quantum fluctuations, having dimension \(d + z\), where \(z = 3\) is the dynamic critical exponent \[13\]. To be consistent with the mean-field action (2), we keep the spin-fermion interaction equal to \(2U\), cf. Ref. \[19\]. This yields the following equations.
In conclusion, by employing a combination of the DFT+DMFT method and analytical model-based techniques we investigated the electronic and magnetic properties of a prototypical itinerant ferromagnet ZrZn$_2$. Our DFT+DMFT results show that electronic correlations in ZrZn$_2$ are rather weak but not negligible as characterized by an effective mass renormalization of the Zr-4$d$ states $m^*/m \sim 1.1 - 1.3$. Most importantly, we demonstrate that the effect of local Coulomb correlations is essential for determining the correct topology of some of the Fermi surface sheets. We show that the topological change of the Fermi surface occurs due to a correlation-induced shift of the peak of the density of states.

Extrapolation of the linear high-temperature part of the inverse uniform susceptibility $\chi^{-1}(T)$ of ZrZn$_2$ calculated by DFT+DMFT yields an estimate of the Curie temperature $T^*_C \sim 32$ K at ambient pressure and $T^*_C \sim 4$ K at $p = 2$ GPa in qualitative agreement with experiment. Nevertheless, at low temperatures the shape of $\chi^{-1}(T)$ shows a significant deviation from the linear behavior (an upturn). This behavior is indicative of an overestimation of the tendency towards the non-ferromagnetic (in particular, incommensurate) order within DMFT.

To study details of the paramagnet-ferromagnet transition, we have constructed an effective single-band model by using the Zr 4$d$ contribution to the total density of states. We have shown that for an appropriate choice of the Coulomb repulsion DMFT analysis of this model yields the results, close to DFT+DMFT analysis of the multi-band model, showing however an upturn of the inverse susceptibility at low temperatures.

The study of the effective single-band model within static mean-field approximation shows a second-order paramagnet-ferromagnet phase transition with decreasing temperature at small pressures, while with increasing pressure the first-order quantum phase transition into paramagnetic phase occurs in agreement with the experimental data. Yet, in contrast to DMFT the static mean-field approximation overestimates the transition temperature by almost one order of magnitude.

To reconcile the results of static and dynamic mean-field approaches to the single-band model, we have also considered a spin-fermion approach to the single-band model. Similarly to DMFT, this approach yields reasonable values of transition temperatures and, at the same time, it preserves the second-order phase transition with changing temperature at small pressures and the first-order quantum phase transition into paramagnetic phase with increasing pressure. The resulting pressure dependence of Curie temperature is in good semi-quantitative agreement with the experimental data.

The obtained results confirm importance of both, local and non-local correlations for various properties of weak magnets. While the electronic properties are well described by considering only local correlations, for magnetic properties non-local correlations appear to be more important, and only local correlations are insufficient to describe these properties. In this respect, the proposed spin-fermion model approach is perspective to study
magnetic properties of other weak itinerant magnets. On
the other hand, this calls for further development of the
approaches describing both, local and non-local correla-
tions. Although number of such approaches is already
developed (see, e.g., Ref. [49]), their application to the
weak itinerant magnets seems to be interesting.

V. ACKNOWLEDGMENTS

The DFT+DMFT calculations were supported by the
Russian Science Foundation (Project 19-12-00012). The
dynamical mean-field calculations of the one-band model
were performed within the state assignment of Mino-
brauki of Russia (theme Electron No. AAAA-A18-
118020190098-5). The study of the one-band model
within static mean-field theory and spin-fermion app-
roach was supported by RFBR grant 17-02-00942a.

Appendix A: Derivation of the equations of the
spin-fermion model

The action \( \mathcal{W} \) can be written as quadratic form of
fermionic operators as follows:

\[
S_{\text{eff}} = \sum_{k,\sigma,\sigma'} c_{k,\sigma}^\dagger M_{\sigma,\sigma'}(k)c_{k,\sigma'} + D\hat{S}^2 + E_0,
\]

where \( E_0 = U(\langle s_z \rangle^2 - \langle n \rangle^2/4) \).

Therefore, in the presented spin-fermion model, the
average total occupation \( \langle n \rangle \) is given by

\[
\langle n \rangle = -\frac{\partial \Omega}{\partial \mu} = \frac{e^{-\beta E_0}}{Z} \int d^3\hat{S} \left[ \sum_{\sigma} \int f(\tilde{\epsilon} + \sigma \gamma) \rho(\epsilon) d\epsilon \right] \times \exp \left( -\beta D\hat{S}^2 - \sum_{\sigma} \int d\epsilon \rho(\epsilon) \ln (1 - f(\tilde{\epsilon} + \sigma \gamma)) \right)
\]

and the average magnetization \( \langle s_z \rangle \) is

\[
\langle s_z \rangle = -\frac{\partial \Omega}{\partial H} = -\frac{e^{-\beta E_0}}{Z} \int d^3\hat{S} \times \left[ \sum_{\sigma} \int \frac{H_{\text{MF}}}{2\gamma} f(\tilde{\epsilon} + \sigma \gamma) \rho(\epsilon) d\epsilon \right] \times \exp \left( -\beta D\hat{S}^2 - \sum_{\sigma} \int d\epsilon \rho(\epsilon) \ln (1 - f(\tilde{\epsilon} + \sigma \gamma)) \right),
\]

where \( \Omega = -\beta^{-1} \ln Z \) is the thermodynamic potential.

The expansion of logarithmic contribution in the ex-
ponents of Eqs. \((A4)-(A7)\) in powers of \( \gamma \) contains only
even powers and corresponds to weak magnetization of
the spin-fluctuation field (the first order in \( \hat{S} \) term), the
renormalization of the value of \( D \) (for the second-order
term) and multi-paramagnon interactions (higher-order
terms). We neglect the weak magnetic field acting on \( \hat{S} \)
and multi-paramagnon interactions effects and assume
that the value of \( D \) is already renormalized. These
approximations imply replacement of $\gamma$ in these logarithmic contributions by its $\bar{S} = 0$ value. At fixed
$\langle n \rangle = 2 \int_{-\infty}^{\infty} \rho(e)de$, $H = 0$, $T$, $D$, the equations (A3) and (A9) reduce therefore to the equations (8) and (9) of the main text.

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