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# Two-channel Kondo physics due to As vacancies in the layered compound $\text{ZrAs}_{1.58}\text{Se}_{0.39}$

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We address the origin of the magnetic-field independent  $-|A|T^{1/2}$  term observed in the low-temperature resistivity of several As-based metallic systems of the PbFCl structure type. For the layered compound  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$ , we show that vacancies in the square nets of As give rise to the low-temperature transport anomaly over a wide temperature regime of almost two decades in temperature. This low-temperature behavior is in line with the non-magnetic version of the two-channel Kondo effect, whose origin we ascribe to a dynamic Jahn-Teller effect operating at the vacancy-carrying As layer with a  $C_4$  symmetry. The pair-breaking nature of the dynamical defects in the square nets of As explains the low superconducting transition temperature  $T_c \approx 0.14$  K of  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$ , as compared to the free-of-vacancies homologue  $\text{ZrP}_{1.54}\text{S}_{0.46}$  ( $T_c \approx 3.7$  K). Our findings should be relevant to a wide class of metals with disordered pnictogen layers.

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In the last years, several exciting phenomena have been discovered in pnictogen-containing materials. This holds particularly true for high-temperature superconductivity in a class of materials based on iron [1]. Another example is the family of filled skutterudites with pnictogen atoms in the cage [2, 3]. Topologically nontrivial phases of certain 3D insulators and Weyl semimetals have been first observed in pnictogen-based systems, such as  $\text{Bi}_{1-x}\text{Sb}_x$  [4],  $\text{Cd}_3\text{As}_2$  [5], and  $\text{TaAs}$  [6]. Here, we address the origin of the magnetic-field independent  $-|A|T^{1/2}$  term observed in the low-temperature resistivity of several As-based metallic systems. We show that As vacancies in the layered compound  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  give rise to an orbital two-channel Kondo effect (2CK) that is symmetry-protected against level splitting [7, 8]

The standard theory of metals states that at sufficiently low temperatures ( $T$ ), the electrical resistivity  $\rho(T)$  is expected to vary quadratically with  $T$  due to the electron-electron interaction (EEI) [9]. Deviations from this behavior can occur, but generally  $\rho(T)$  continues to decrease upon cooling. Only a few mechanisms are known to produce a low- $T$  resistivity minimum. The paradigmatic example is the Kondo effect where spin-flip scattering of conduction electrons off dynamic centers associated with the local magnetic moments gives rise to

a logarithmic increase of the resistivity upon cooling [9]. Theoretical studies have revealed that an even more exotic 2CK effect can occur if two degenerate channels of the conduction electrons exist, which independently scatter off centers with a local quantum degrees of freedom [10]. The low-energy physics in this case is governed by a non-Fermi liquid fixed point and results in a resistivity minimum followed at lower temperature by a  $\sqrt{T}$  increase of  $\rho(T)$ . However, in a non-magnetic realization of the Kondo effect this would require the exact symmetry between orbital channels that cannot be obtained in any real system. Here, the orbital degree of freedom plays the role of the (pseudo-)spin and the electron spin represents the degenerate channel index. Such a scenario was proposed for metals with dynamic structural defects modeled by double-well potentials [11–13]. However, in spite of considerable interest [14–18], this type of 2CK physics has never been conclusively demonstrated in any bulk metallic system. This appears to be in line with further theoretical efforts that established two-level systems as an unlikely source of the non-Fermi behavior [7, 19–21]. Additional complications arise from the fact that the disorder-enhanced EEI in three-dimensional metals can also give rise to a  $\sqrt{T}$  term in the resistivity [22, 23]. Thus, the clearest evidence to date for the 2CK effect

has come from an artificial nanostructure that can be judiciously tuned to show a  $T^{-1/2}$  term in the differential conductance for about a decade in  $T$  [24].

On the other hand, there are a number of experimental results which point to unconventional scattering mechanisms whose precise nature has remained enigmatic [25]. In particular, several As-based metallic systems with the layered PbFCl structure display a low- $T$  resistivity minimum. Investigations on e.g. diamagnetic ThAsSe (nominal composition) single crystals revealed a magnetic-field- ( $B$ )-independent  $-|A|T^{1/2}$  term in the low- $T$  resistivity [26]. Furthermore, the thermal conductivity and specific heat of ThAsSe showed glass-type temperature dependences which support the presence of structural defects with internal degrees of freedom [26]. Investigations of the closely related Zr- and Hf-based arsenide selenides have pointed to a  $B$ -independent  $-|A|T^{1/2}$  contribution to  $\rho(T)$  as a generic feature of metallic arsenide selenides crystallizing in the PbFCl structure [27]. To address the physical mechanism leading to the  $B$ -independent  $-|A|T^{1/2}$  term in the low- $T$   $\rho(T)$  we have identified two homologues,  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  and  $\text{ZrP}_{1.54}\text{S}_{0.46}$ , that allow us to identify the microscopic origin of the anomalous behavior. By combining precise physical property measurements with chemical and structural investigations performed on the same single crystals, we show that the only viable explanation for the observed transport anomalies of  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  is in terms of a 2CK. We argue that 2CK physics is possible due to vacancies in the square nets of As atoms.

The ternary pnictide-chalcogenides ( $Pn$ - $Ch$ ) contain intermediate phases which crystallize in the tetragonal ( $P4/nmm$ ) PbFCl structure, a substitution variant of the  $\text{Fe}_2\text{As}$  type. This crystal structure consists of square-planar  $4^4$  nets stacked along the  $[001]$  direction. For compounds with exact chemical composition  $M:Pn:Ch=1:1:1$ , each of the  $4^4$  nets is exclusively occupied by one element resulting in a layer-sequence  $\dots Pn-M-Ch-Ch-M-Pn$ , and forming a puckered double-layer  $M_2Ch_2$  with an ordered distribution of  $M$  and  $Ch$ .

Phases with  $M$ , being a metal with the fixed +4 oxidation state such as Zr, Hf, and Th, are characterized by significant excess of pnictogen atoms (see Fig. 1). In case that the homogeneity range is restricted to the tie-line between  $MPn_2$  and  $MCh_2$  the chemical formula corresponds to  $MPn_xCh_y$  with  $x+y=2$ . For  $M=\text{Zr}$ , it has been shown that only the  $2c$  site is randomly occupied by  $Pn$  and  $Ch$ . Exceptionally for the arsenide selenide, however, the homogeneity range is located right next to the tie-line  $\text{ZrAs}_2$ - $\text{ZrSe}_2$  on the As depleted side and hence  $1.90 \leq x+y \leq 1.99$  [28] and Fig. S4 of [29]. As a result, vacancies are *solely* present within the As layers ( $2a$  site). For details on the crystal growth, chemical characterization, and physical measurements of two homologues  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  (3% vacancies at the  $2a$  site)

and  $\text{ZrP}_{1.54}\text{S}_{0.46}$  (full occupation), see [29].

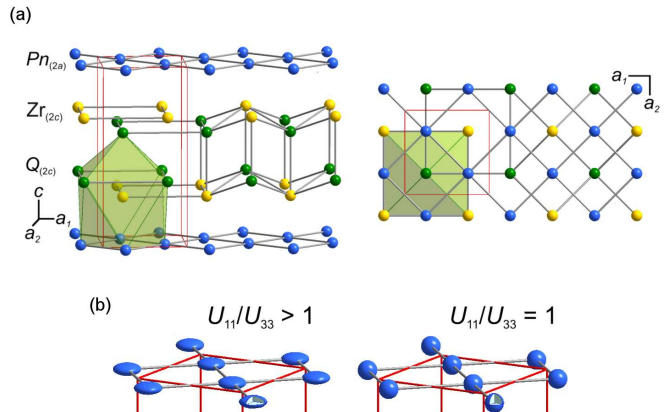


Figure 1. Structural disorder in zirconium pnictide chalcogenides. (a) The PbFCl structure type with the fully occupied  $Q(2c)$  site (green) by  $Ch$  together with  $Pn$ . The  $Pn(2a)$  site (blue) arranged within planar layers is only occupied to 97% by As in  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$ , but fully occupied by P in  $\text{ZrP}_{1.54}\text{S}_{0.46}$ . (b) The vacancies in  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  (left) manifest random displacements of As within the layer due to homoatomic covalent bond formation. This is indicated by flattened displacement ellipsoids in the structure refinements, while the refined displacement ellipsoids in  $\text{ZrP}_{1.54}\text{S}_{0.46}$  (right) are nearly spherical.

Replacement of As and Se by P and S, respectively, does not distinctly alter basic physical properties, such as electrical resistivity and specific heat of the tetragonal  $\text{Zr-Pn-Ch}$  phases. In fact, differences in the metallic behavior of  $\rho(T)$  are of minor significance only, as depicted in Fig. 2(a). For both homologues  $\text{ZrPn}_x\text{Ch}_y$ , a drop of  $\rho(T)$  signals the onset of a superconducting phase transition at  $T_c \approx 0.14$  K ( $\text{ZrAs}_{1.58}\text{Se}_{0.39}$ ) and 3.9 K ( $\text{ZrP}_{1.54}\text{S}_{0.46}$ ), respectively. However,  $\rho = 0$  is found only substantially below the onset temperature [see Fig. 2(b)]. The complex behavior of  $\rho(T)$  in the vicinity of  $T_c$  is ascribed to a likely delicate variation of chemical composition, i.e., below 0.5 wt.% which is the resolution limit of our analysis. The normal-state specific heats  $C(T)$  for both  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  and  $\text{ZrP}_{1.54}\text{S}_{0.46}$  are very similar, leading to virtually the same value of the Sommerfeld coefficient of the electronic specific heat  $\gamma = 1.7(\pm 0.1)$  mJK $^{-2}$ mol $^{-1}$  [see Fig. 2(c)].

Summing up, the main physical properties of  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  and  $\text{ZrP}_{1.54}\text{S}_{0.46}$  demonstrate a far-reaching similarity between both homologues. Therefore, the observed difference in their  $T_c$ 's by a factor  $\approx 30$  is an unexpected result, especially in view of the tiny concentration of magnetic impurities in the arsenide selenide crystals, i.e. less than 0.10 wt.% [27]). Note that the As vacancies themselves, as (static) potential scatterers, cannot be the source of strong pair-breaking.

A hallmark of the 2CK effect is the  $\sqrt{T}$  dependence of the low- $T$  resistivity. In the case of dynamic structural

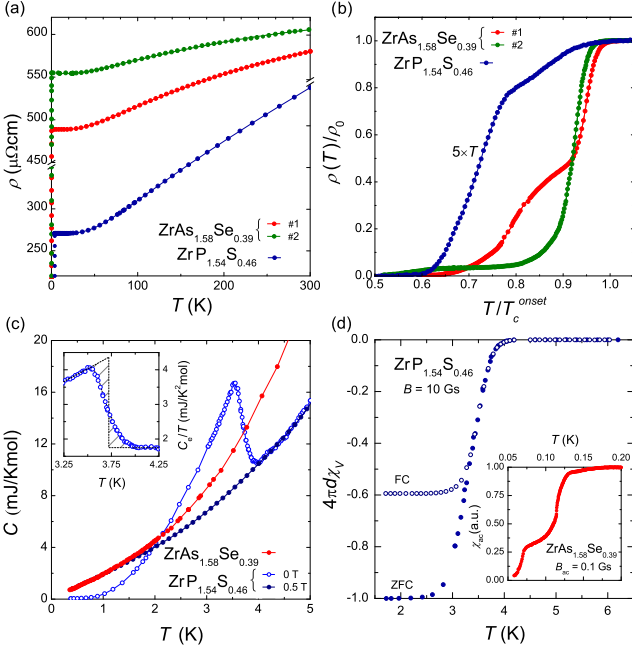


Figure 2. Characteristics of the pnictide chalcogenide superconductors  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  and  $\text{ZrP}_{1.54}\text{S}_{0.46}$ . (a) Normal-state electrical resistivity along the  $c$  axis of  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  and  $\text{ZrP}_{1.54}\text{S}_{0.46}$  (the two specimens of the former material were cut off from the same single crystal). (b) The same results, but with focus on the vicinity of the superconducting transition, as  $\rho/\rho_0$  vs.  $T/T_c^{\text{onset}}$ . For clarity, the temperature scale of the  $\rho(T)$  data for the P-based system was multiplied by a factor of 5. (c) Low-temperature specific heat for  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  and  $\text{ZrP}_{1.54}\text{S}_{0.46}$ . Inset: Electronic specific heat of  $\text{ZrP}_{1.54}\text{S}_{0.46}$  in the vicinity of the superconducting phase transition, as  $C_e/T$  vs temperature.  $C_e = C - C_{\text{ph}}$ , where  $C_{\text{ph}}$  is the phonon contribution, estimated from the normal-state  $B = 0.5$  T data. (d) Zero-field-cooled (ZFC) and field-cooled (FC) dc magnetic susceptibility as a function of temperature for  $\text{ZrP}_{1.54}\text{S}_{0.46}$ . Inset: Evidence for large shielding in  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  is provided by the sizable diamagnetic signal of the ac magnetic susceptibility below about  $T = 0.125$  K.

defects, the resulting non-Fermi liquid properties are not expected to depend on an applied magnetic field as long as the Zeeman splitting does not cause a difference in the conduction-electron DOS at the Fermi level between up and down subbands. The only other mechanism that can in principle result in a  $B$ -independent  $-|A|T^{1/2}$  correction to the resistivity is the electron-electron interaction (EEI) in a three-dimensional, disordered metal [22, 23]. This could happen if electron screening is strongly reduced, yielding the unique case of a screening factor,  $\tilde{F}_\sigma$ , very close or even equal to zero [22, 23]. To analyze possible corrections due to the EEI in  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  and  $\text{ZrP}_{1.54}\text{S}_{0.46}$ , we thus have plotted in Fig. 3(a) the relative change of the resistivity normalized to the minimum value,  $(\rho - \rho_{\text{min}})/\rho_{\text{min}}$ , as a function of  $T^{1/2}$ . Below

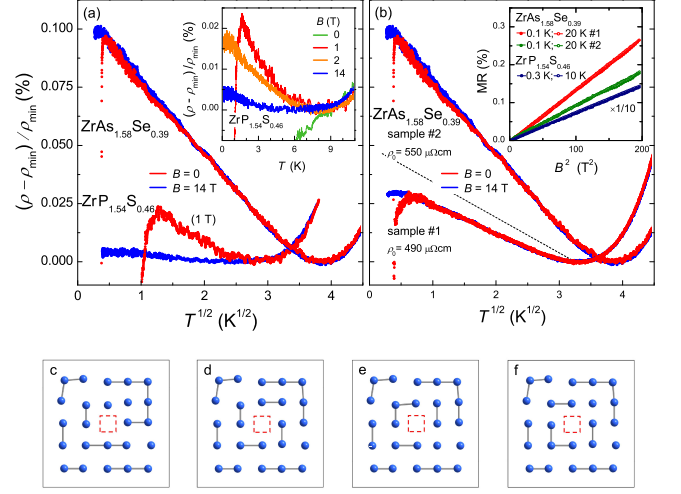


Figure 3. Evidence for the 2CK effect due to As vacancies. (a) Despite very similar basic low-temperature properties of  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  and  $\text{ZrP}_{1.54}\text{S}_{0.46}$ , these sister compounds display qualitatively different responses of  $(\rho - \rho_{\text{min}})/\rho_{\text{min}}$  vs.  $T^{1/2}$ . Note the magnetic-field-independent  $-|A|T^{1/2}$  contribution to  $\rho(T)$  for  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$ , i.e., for a system with vacancies in the pnictogen layer. Inset: Low- $T$  resistivity of  $\text{ZrP}_{1.54}\text{S}_{0.46}$  measured in zero and varying magnetic fields up to 14 T. For  $B = 0$ ,  $\rho_{\text{min}} = \rho(9\text{ K})$  was taken. (b) Remarkable differences of the  $B$ -independent low- $T$   $\rho(T)$  as  $(\rho - \rho_{\text{min}})/\rho_{\text{min}}$  vs.  $T^{1/2}$  for two single-crystalline  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  specimens with similar residual resistivities. Dashed line shows the magnitude of a hypothetical  $-|A|T^{1/2}$  correction to  $\rho(T)$  for sample #2 due to 3D EEI assuming  $\tilde{F}_\sigma = 0$ . Inset: Magnetic field dependence of  $\rho(T)$  of  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  and  $\text{ZrP}_{1.54}\text{S}_{0.46}$  single crystals. Note that the data for  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  are divided by a factor of 10. (c)-(f) Dynamic structural scattering centers in metallic arsenide selenides. Shown are different possible arrangements of dimers or oligomers within the As layer triggered by the vacancy (red square). Arsenic atoms may for example arrange as dimers, trimers, oligomers of several As atoms, or infinite chains with zigzag or sawtooth configuration. (f) is obtained from (e) by a  $\pi/2$  rotation of the arrangement of (e) indicating how the  $C_4$  symmetry is dynamically restored.

$T_{\text{min}} \approx 15.0$  K, the zero-field  $\rho(T)$  data for  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  depend strictly linearly on  $T^{1/2}$  over almost two decades in temperature. A magnetic field of  $B = 14$  T, being the largest field accessible in our experiment, does not alter the  $-|A|T^{1/2}$  dependence for the system containing vacancies in the pnictogen layers.

For the free-of-vacancies system, the low- $T$  upturn displays a completely different response to an applied field. (In the absence of a magnetic field, superconducting fluctuations dominate the transport properties of  $\text{ZrP}_{1.54}\text{S}_{0.46}$  leading to a strong decrease of  $\rho(T)$  in a temperature region which significantly exceeds  $T_c \approx 3.7$  K.) Although at  $B = 1$  T a small fraction of the  $\text{ZrP}_{1.54}\text{S}_{0.46}$  sample still displays surface superconduc-

tivity below 1 K, at elevated temperatures, we observe an upturn in  $\rho(T)$  with a rather complex  $T$  behavior. More remarkably, however, the  $(\rho - \rho_{\min})/\rho_{\min}$  correction is distinctly smaller at intermediate fields of about 2 T [cf. the inset of Fig. 3(a)]. For 14 T, the low- $T$  upturn is strongly reduced, and its magnitude is about 7 times smaller than for  $B = 1$  T. Furthermore, the  $\rho(T)$  rise is restricted to a rather narrow temperature window, i.e. from above 4.2 K to around 1 K. At  $T < 1$  K, the resistivity tends to saturate. These findings point to a negligible EEI in single crystals of  $\text{ZrPn}_x\text{Ch}_y$  and yield striking evidence for an entirely different origin of the  $-|A|T^{1/2}$  term in the low- $T$   $\rho(T)$  occurring in the material without ( $\text{ZrAs}_{1.58}\text{Se}_{0.39}$ ) and with ( $\text{ZrP}_{1.54}\text{S}_{0.46}$ ) a full occupancy of the square-planar pnictogen layers. For an in-depth analysis, see [29]. In the latter case, structural disorder (driven from the mixed occupied  $2c$  sites only) leads to weak localization. Its negative contribution to the magnetoresistance  $\text{MR} = [(\rho(B) - \rho(0))/\rho(0)]$  increases upon cooling and amounts to about 0.02% at  $T = 2$  K. The weak-localization contribution is nearly two orders of magnitude smaller than the classical MR [cf. the inset of Fig. 3(b)]. Therefore, the total MR of  $\text{ZrP}_{1.54}\text{S}_{0.46}$  is essentially temperature independent at  $T \leq 10$  K and approaches 1.4% at  $B = 14$  T.

Figure 3(b) shows the temperature dependence of  $(\rho - \rho_{\min})/\rho_{\min}$  for two  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  specimens with similar elastic relaxation times. In spite of this moderate variation, the size of the magnetic-field-insensitive  $-|A|T^{1/2}$  term between these samples differs by more than a factor of 3 and hence the  $A$  coefficient amounts to 0.038 and 0.167  $\Omega\text{cm}/\text{K}^{1/2}$  for samples #1 and #2, respectively. Similarly to the afore mentioned results, the experimental observations shown in Fig. 3(b) are at strong variance to the expectation based on enhanced EEI in 3D specimens of the same disordered metal [22, 23]. Indeed, a nearly identical screening factor  $\tilde{F}_\sigma$ , see Ref. [29], implies that the magnitude of a hypothetical  $(\rho - \rho_{\min})/\rho_{\min}$  anomaly would only vary with  $\rho D^{-1/2} \propto \rho^{3/2}$ . This, however, is not observed in  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$ . In fact, a supposed  $-|A|T^{1/2}$  correction, calculated in respect to the  $|A|$ -coefficient value of sample #1 and schematically sketched by a dashed line in Fig. 3(b), would be substantially smaller than what is experimentally found for sample #2. (Since  $A$  is field independent,  $\tilde{F}_\sigma = 0$  was assumed in our calculations [29]).

We thus conclude that the  $B$ -field independent  $-|A|T^{1/2}$  correction to  $\rho(T)$  in  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  cannot be caused by EEI and can only be explained by the existence of non-magnetic defects with degenerated ground state which, at low temperatures, place the system near the 2CK fixed point. Note that in the dilute limit, where the dynamic scattering centers are independent of each other, the amplitude  $|A|$  is proportional to the concentration of dynamic scattering centers in the 2CK regime which is in general smaller than, and not expected to

scale in a simple fashion with, the concentration of As vacancies.

The chemical composition of  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  implies that the vacancies in the As layer are in the dilute limit. As discussed above, our analysis shows that interstitial As does not occur in the pnictogen layer. Each vacancy thus preserves the  $C_4$  symmetry of the pnictogen layer. As a result, a Jahn-Teller distortion forms in the pnictogen layer concomitant with a formation of As dimers or oligomers. This phenomenon is well known to occur in such square nets of the PbFCl structure-type compounds [44]. The flattened displacement ellipsoids shown in Fig. 1(b) are indicative of the occurrence of the dynamic Jahn-Teller effect in the As ( $2a$ ) layer. As the Jahn-Teller distortion develops, the doublet states split and one of them becomes the new ground state. A finite tunneling rate between the different impurity positions compatible with the overall square symmetry restores the square symmetry through a dynamic Jahn-Teller effect, see Figs. 2(c)-(f). The group  $C_4$  possesses only one-dimensional irreducible representations and one two-dimensional irreducible representation (IRREP). Thus, a non-Kramers doublet associated with the dynamic Jahn-Teller distortion transforms as the two-dimensional irreducible representation of the group  $C_4$ , and allows for a two-channel Kondo fixed point to occur at sufficiently low energies [45–47]. The pseudo-spin index  $\pm$  of the effective 2CK Hamiltonian labels the basis states of this two-dimensional subspace [29]. As a result of the coupling to the conduction electrons the doublet can become the ground state [48]. In fact, the doublet is renormalized below the singlet in a wide parameter regime [49].

Our conclusion that 2CK centers exist in  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  is further corroborated by the low superconducting transition temperature, as compared to  $\text{ZrP}_{1.54}\text{S}_{0.46}$ , which points to the presence of efficient Cooper pair breakers [50]. The 2CK effect in  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  has been argued to arise out of the non-Kramers doublet transforming as the two-dimensional IRREP of  $C_4$ . This representation cannot be transformed into its complex conjugate through the application of a unitary transformation and thus the associated basis states of the two-dimensional IRREP, labeled by  $+$  and  $-$ , are time-reversed partners. Expanding the BCS order parameter around the quantum defect will thus have to involve singlets of  $+$  and  $-$ . The full Hamiltonian including the quantum defect involves scattering from one of the basis states of the two-dimensional IRREP to the other. These scattering processes therefore have to break up Cooper pairs and thus reduces  $T_c$ . For details, see [29]. Thus, our model is in line with all observed properties of  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  and is not susceptible to the difficulties that exist for the 2CK scenario based on two-level systems. The observed suppression of superconductivity in  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  as compared to  $\text{ZrP}_{1.54}\text{S}_{0.46}$  is naturally explained. A

$\log T$ -behavior of  $\rho(T)$  is expected for the  $T$ -behavior right above the  $\sqrt{T}$  regime for a 2CK system, which is indeed observed [29]. Direct evidence of the dynamic scattering centers could come from scanning tunneling spectroscopy apt to zoom into one of these dynamic scattering centers.

In conclusion, we have shown that the  $B$ -independent  $-|A|T^{1/2}$ -term in  $\rho(T)$  of  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  is triggered by non-magnetic centers with a local quantum degree of freedom. Our analysis indicates that a dynamic Jahn-Teller effect concomitant with the formation of As oligomers places the scattering center in the vicinity of the orbital two-channel Kondo fixed point. These quantum impurities act as efficient Cooper pair breakers which explains the suppression of  $T_c$  of  $\text{ZrAs}_{1.58}\text{Se}_{0.39}$  as compared to the homologue  $\text{ZrP}_{1.54}\text{S}_{0.46}$ . We expect that similar dynamic scattering centers occur in other materials PbFCl structure type with square nets of pnictogen.

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