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Effects of iso-valent substitution and pressure on the inter-plane resistivity of single crystal $Ba(Fe_{1-x}Ru_x)_2As_2$

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Temperature-dependent inter-plane resistivity, $\rho_c(T)$, was measured in iso-valent substituted ironarsenide compound Ba(Fe_{1-x}Ru_x)₂As₂ over a substitution range from parent compound to slightly below optimal doping, x=0.29. The feature of interest in the $\rho_c(T)$, a broad resistivity crossover maximum found in parent compound at $T_{max} \approx 200$ K, shifts to higher temperatures with Ru substitution, ~340 K for x=0.161 and goes out of the 400 K range for x=0.29. Nearly Tlinear dependence of inter-plane resistivity is found at the highest substitution level x=0.29. This temperature-dependent ρ_c and its evolution with substitution bear close similarity to another type of iso-valent substituted system, BaFe₂(As_{1-x}P_x)₂. Similarly to the iso-valent substitutions, the measurements of inter-plane resistivity in the parent BaFe₂As₂ compound under pressures up to 20 kbar also revealed a rapid rise in T_{max} .

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I. INTRODUCTION

Magnetically mediated mechanism of superconductivity in iron-based materials is discussed in relation to the observation of a quantum critical point in the phase diagram. In BaFe₂As₂ based superconductors, in which superconductivity can be induced by different types of substitutions and pressure, in all cases the maximum T_c is not far from a point where magnetism vanishes as a function of the tuning parameter¹⁻⁵. The existence of a quantum critical point governs systematic evolution of all electronic properties, in particular of the electrical resistivity.

The most clear evolution is found in the iso-valent substituted material, $BaFe_2(As_{1-x}P_x)_2^6$. Here resistivity for both in-plane, ρ_a , and inter-plane, ρ_c , current directions reveals extended range of T-linear dependence at optimal doping 6,7 , and signatures of the quantum critical point are found in both normal and superconducting state properties^{6,8,9}. Much more complex evolutions of in-plane and inter-plane resistivity are found in electron- doped $Ba(Fe_{1-x}Co_x)_2As_2^{10-12}$ and hole-doped $Ba_{1-x}K_xFe_2As_2^{13}$. Here dominant feature of $\rho_c(T)$ is a broad cross-over maximum, which in the BaFe₂As₂ parent compound takes place at $T_{max} \approx 200$ K. This maximum shifts to lower temperatures with Co-doping and does not change position with K-doping up to $x=0.34^{13}$. The maximum in $\rho_c(T)$ for the hole-doped materials correlates well with a slope-change feature in the in-plane $transport^{13,14}$.

We correlated the maximum in $\rho_c(T)$ with an anomaly in the temperature dependent magnetic susceptibility¹¹, as found most clearly in the temperature-dependent NMR Knight shift measurements^{15,16}. This interpretation suggests that the maximum is caused by the onset of activation of carriers over a minimum of the pseudogap, while the pseudogap maximum and restoration of normal metallic properties correspond to significantly higher temperatures and becomes visible only at very high electron dopings $x > 0.16^{11}$. The existence of pseudogap in iron based superconductors was later confirmed with spectroscopic^{17–19} and ARPES^{20,21} techniques. Some recent advanced dynamical mean-field theory (DMFT) band structure calculations^{22–24} also predict the existence of the pseudogap with strong orbital selectivity. The prediction is that the effect would be the strongest in the d_{z^2} orbital²³, which would naturally lead to a much stronger effect in the inter-plane transport.

This interpretation of the resistivity maximum is not unique though. The characteristic energy scale T_{max} is significantly smaller than the energies found in band structure calculations $^{22-24}$. An alternative interpretation is the loss of spin-disorder scattering at low temperatures, though this effect alone cannot explain resistivity decrease with temperature above T_{max} . Clearly, additional studies are required to get a new insight into an anomalous resistivity behavior at high temperatures. It is of particular interest, if the unique evolution of resistivity observed in the phosphorus doped materials can be found in other systems. Iso-valent substitution of Fe by Ru in BaFe₂As₂ also suppresses magnetism and brings about superconductivity. Importantly, Ru substitution does not lead to changes of the Fermi surface beyond the suppression of the folding effects of the magnetic wavevector²⁵. Ru substitution was also shown to act similar to application of pressure $^{26-28}$. Study of the interplane resistivity using pressure as a tuning parameter provides an additional possibility to tune system without introducing substitution disorder, inevitable for all types of dopings and particularly strong when substitutions are made in Fe site. With this motivation in mind here we perform systematic study of the inter-plane transport in BaFe₂As₂ compound using iso-valent Ru substitution and pressure as tuning parameters. We find that the evolution of the resistivity in samples with Ru-substitution

is similar with that observed on iso-valent substitution of As with P. A broad range of *T*-linear dependence is observed in both in-plane and inter-plane transport in samples with close to optimal substitution level. The general trend of shifting the maximum in $\rho_c(T)$ for parent BaFe₂As₂ to higher temperatures with pressure, is similar to the effects of two iso-valent substitutions, P for As and Ru for Fe.

However, using the temperature of the structural/magnetic transition as a reference, the rate at which T_{max} increases is notably higher for pressure than for the iso-electron substitutions.

II. EXPERIMENTAL

A. Sample preparation

Single crystals of Ba(Fe_{1-x}Ru_x)₂As₂ (BaRu122) were grown using high temperature FeAs flux technique²⁶. The samples from the same batches have some distribution of T_c . As a first step of sample selection for our study, we cleaved thin slabs typically of 20 μ m thickness with two clean cleavage surfaces from the inner part of the crystals. Numerous smaller pieces with sides along (100) directions for resistivity measurements were further cleaved using a razor blade. The samples for interplane resistivity measurements typically had dimensions of $0.5 \times 0.5 \times 0.02 \text{ mm}^3$ size $(a \times b \times c)$. Elongated samples for in-plane resistivity measurements typically were of $1 \times 0.2 \times 0.02$ mm³ size. All samples were prescreened using magnetooptical imaging^{29–31} and dipper version of the tunnel diode resonator (TDR) technique^{32,33}. These measurements allowed us to exclude samples with macroscopic inhomogeneity and possible inclusions with lower T_c .

Both in-plane and inter-plane resistivity were measured on big sets of crystals coming from the same slab. Conventional four-probe measurements were performed on samples with ultra-low contact resistance (typically 10 $\mu\Omega$) soldered contacts^{31,34}. For inter-plane resistivity measurements we relied on negligible contact resistance and used two-probe technique. Top and bottom surfaces of the samples were covered with Sn solder and 50 $\mu {\rm m}$ diameter silver wires were attached to enable measurements in four-probe configuration, which was used down to the sample to measure series connected sample, R_s , and contact, R_c resistance. Taking into account that typical sample resistance R_s is 1 m Ω , and contact resistance $R_c \sim 10 \ \mu\Omega$, the contact resistance represents a minor correction of the order of 1 to 5%. The validity of the assumption $R_s \gg R_c$ can be directly seen for our samples for temperatures below the superconducting T_c , where $R_s = 0$ and the measured resistance represents $R_c^{(10,29,31)}$. The details of the measurement procedure can be found in Refs. 10, 11, and 35. Measurements on samples with $c \ll a$ are very sensitive to any inhomogeneity in the contact resistance or internal sample connectivity, which tend to mix the in-plane component ρ_a due to redistribution of the current. Measurements on a large number of samples are necessary in order to select the ones with minimal intra-plane meandering. Typically this screening process involved at least 5 samples from each batch yielding the same dipper TDR T_c . In all cases we obtained qualitatively similar temperature dependencies of the electrical resistivity. The resistivity value at room temperature, $\rho_c(300K)$ was approximately in the range 1000 to 1500 μ Ω cm. The resistivity value for inplane resistivity, $\rho_a(300K)$, was in 300 ± 50 μ $\Omega {\rm cm}$ range and did not reveal any evolution with x beyond error bars, contrary to previous reports suggesting significant decrease^{36,37}

The measurements of electrical resistivity under pressure were carried out with a piston-cylinder Be-Cu pressure cell, with a core of tungsten carbide. The sample, manganin, and Pb manometers were mounted on a feed-through, which was inserted into a Teflon capsule filled with a 60:40 mixture of n-pentane: light mineral oil, which served as the pressure transmitting medium. Pressure was generated at ambient temperature with a hydraulic press, using manganin as a reference manometer. The pressure was locked in, and the cell was then loaded into a Quantum Design Physical Property Measurement System (PPMS-9), which provided the temperature environment for the measurements, as well as the dc measurements of resistance for the sample, manometers, and a Cernox temperature sensor attached to the body of the cell. The pressure at low temperatures was determined from the superconducting transition temperature of the Pb manometer. The cooling and warming rates were kept below 0.35 K/min, which maintained the T-lag between the Cernox sensor and the sample well bellow 0.5 K throughout the whole temperature range. In light of the approximately linear variation of pressure from ambient temperature to ~ 90 K in piston-cylinder cells³⁸, the pressure values at temperatures between these limits were estimated from linear interpolation.

III. RESULTS

A. Inter-plane resistivity in samples with iso-valent Ru substitution

Figure 1 shows the evolution of the in-plane (top panel) and inter-plane resistivity (bottom panel) of samples of iso-valent substituted Ba(Fe_{1-x}Ru_x)₂As₂. The in-plane resistivity data are very similar to previous reports on the same material^{26,36,37}. The main feature of $\rho_a(T)$ is gradual drop of the temperature of coinciding structural/magnetic transition $T_{sm}^{26,39,40}$, which goes away at $x \approx 0.29$. For all $x \leq 0.24$ we still see the presence of slight $\rho_a(T)$ up-turn on cooling above T_c , which is completely suppressed for x=0.29. The data for sample x=0.29 reveal very close to T-liner dependence over a broad range from above T_c to almost 400 K, with only



(Color online) (Top panel) Doping evolu-FIG. 1. tion of the temperature-dependent in-plane resistivity of $Ba(Fe_{1-x}Ru_x)_2As_2$. The curves are presented using normalized $\rho/\rho(300K)$ plots and offset for clarity. Top to bottom x=0, 0.073, 0.126, 0.21, 0.24 and 0.29. (Bottom panel) Temperature dependent inter-plane resistivity of $Ba(Fe_{1-x}Ru_x)_2As_2$, top to bottom x=0, 0.073, 0.126, 0.161, 0.21, 0.24 and 0.29. Down triangles show positions of the coupled structural/magnetic transition, open circles show positions of T_{max} . For the highest substitutions x=0.24 and 0.29no maximum is observed in the range, but a slope change crossover is still clearly visible, as indicated by the square symbols.

a mild slope change at around 200 K. The temperature of the slope change in $\rho_a(T)$ is reminiscent of the much more pronounced feature in the $\rho_a(T)$ of hole-doped materials, $(Ba_{1-x}K_x)Fe_2As_2^{13,14,41}$. A previous study on a higher doped materials³⁷ suggested that eventually, at $x \approx 0.35$, $\rho_a(T)$ becomes *T*-linear over the whole temperature range from T_c to 300 K.



FIG. 2. (Color online) Left panel shows the compositional phase diagram of $Ba(Fe_{1-x}Ru_x)_2As_2$ as determined from inter-plane resistivity measurements in this study (down triangles, circles and open squares, see Fig. 1 for definition) in comparison with magnetization measurements (up-triangles and pentagons) of Thaler *et al.*, Ref. 26. Right panel compares temperature - composition diagrams for two iso-valent substitutions, of Fe with Ru (blue lines and symbols) and of As with P (red lines and symbols)^{6,7}.

Evolution of the inter-plane transport in iso-valent substituted Ba(Fe_{1-x}Ru_x)₂As₂ is distinctly different from both electron-doped BaCo122¹¹ and hole- doped BaK122¹³ materials. The inter-plane resistivity $\rho_c(T)$ reveals a broad cross-over maximum at T_{max} , which, with Ru-substitution, moves from approximately 200 K to above 300 K in samples with x=0.21. The $\rho_c(T)$ data for $x \ge 0.24$ suggest that the substitution drives T_{max} past 400 K, the highest temperature of our measurements. However, the leftover of the maximum can be found in a mild slope change in samples with x=0.24 and x=0.29, marked with squares in the bottom panel of Fig. 1. This evolution trend suggests that in samples with higher substitution level both in-plane and inter-plane resistivity would become *T*-linear, similar to BaP122 materials.

The left panel of Fig. 2 summarizes the evolution of the main features of the temperature-dependent inter-plane resistivity with Ru substitution. For reference we show the temperatures of the coupled magnetic/structural transition and bulk superconducting T_c as determined from magnetization measurements²⁶. The temperature of the inter-plane resistivity maximum T_{max} moves up very rapidly with x, similar to the behavior in iso-valent substituted $BaFe_2(As_{1-x}P_x)_2$. In the right panel we directly compare the phase diagrams of the two iso-valent substitutions in BaFe₂As₂, which indeed reveal clear similarity. Keeping in mind the uncertainties in compositional determination x, the potential effect of disorder on the phase diagram, 4^{42} and the uncertainty in the determination of the maximum position due to possible admixture of the $\rho_a(T)$ component, the similarity of the two phase diagrams is just remarkable.



FIG. 3. (Color online) Comparison of the temperaturedependent inter-plane resistivity of under-doped samples of BaFe₂As₂ based superconductors, for the compositions selected to have comparable T_{sm} of about 100 K. Top left panel (a) shows $\rho_c(T)$ for hole-doped (Ba_{1-x}K_x)Fe₂As₂¹³, top right panel (b) for electron doped Ba(Fe_{1-x}Co_x)₂As₂¹¹, bottom left panel (c) for iso-valent substituted BaFe₂(As_{1-x}P_x)₂⁷, bottom right panel (d) for isovalent substituted Ba(Fe_{1-x}Ru_x)₂As₂ (this study). Note the very different evolution of the position of maximum of interplane resistivity T_{max} , shown with open circles.

B. Comparison of different types of substitutions

To put our findings in a broader perspective, in Fig. 3 we directly compare the temperature dependent interplane resistivity of BaFe₂As2 derived compositions with four different types of substitutions in the "under-doped" regime. For the sake of comparison we selected compositions with similar $T_{sm} \sim 100$ K. This selection criterion in fact does not correlate well with the superconductivity; samples with T_{sm} in this range have significantly different T_c , and reveal just traces of superconductivity in BaP122 and BaRu122. Nevertheless, this comparison reveals interesting features. Samples with substitutions into the Fe site, Co and Ru, have significantly higher normalized residual resistivity, $\rho(0)/\rho(300K)=1.1$ and 0.8, respectively, as compared to approximately 0.3 in BaP122 and BaK122. The values of $\rho_c(300K)$ in all cases are the same



FIG. 4. (Color online) Comparison of the temperaturedependent in-plane (blue lines) and inter-plane (red lines) resistivity of close to optimally doped samples of BaFe₂As₂ based superconductors. Top left panel (a) shows data for hole-doped (Ba_{1-x}K_x)Fe₂As₂¹³, top right panel (b)for electron doped Ba(Fe_{1-x}Co_x)₂As₂¹¹, bottom left panel (c) for isovalent substituted BaFe₂(As_{1-x}P_x)₂^{6,7}, bottom right panel (d) for iso-valent substituted Ba(Fe_{1-x}Ru_x)₂As₂ (this study). Note that Ru-doped samples are actually under-doped, which is the most likely reason that the evolution towards *T*-linear dependence is incomplete.

within error bars and are in the range 1000 to 1500 $\mu\Omega$ cm, so that the difference is in fact observed between actual resistivity values. This finding is a natural consequence of the substitution disorder introduced right into orbitals forming the states at the Fermi energy. Secondly, there is a robust upturn in $\rho_c(T)$ just below T_{sm} in samples with more disorder, while the increase is not as pronounced in samples with substitution away from Fe-site.

In Fig. 4 we compare temperature-dependent in-plane (blue lines) and inter-plane (red lines) resistivity of samples of BaFe₂As₂ derived superconductors with four different types of substitutions at close to optimal level. As suggested by the T - x phase diagrams, Fig. 2, two different types of iso-valent substitutions yield quite similar temperature-dependent resistivity, as indicated in the two bottom panels in Fig. 4. Neither $\rho_a(T)$ nor $\rho_c(T)$ in BaP122 and BaRu122 show any evident slope-change features for optimal substitution level, which is in stark contrast with hole-doped BaK122 revealing maximum in $\rho_c(T)$ and slop-change in $\rho_a(T)$ at around 200 K^{13,14}, and with electron-doped BaCo122^{11,12}, showing cross-over only in the inter-plane transport.



C. Evolution of the temperature-dependent inter-plane resistivity with pressure

dependent in-plane (top panel) and inter-plane (bottom panel) resistivity with pressure in the parent compound of 122 iron-based superconductors, BaFe₂As₂. Pressure values are changing on cooling, as shown for temperatures of 300 K and 4.2 K. The inset in the top panel shows pressure dependence of T_s as determined from ρ_a (solid down-triangles) and from ρ_c (open down-triangles) measurements. Inset in the bottom panel shows evolution of T_s (down triangles) and of T_{max} (circles) with pressure. Symbols in the main panel show the respective temperatures vs. actual temperaturedependent resistivity curves.

It is remarkable that two different types of iso-valent

substitution give rise to similar effects on both the inplane and inter-plane temperature-dependent electrical resistivity. It was shown previously that the effects of pressure and of the iso-valent Ru substitution on in-plane resistivity are very similar^{27,28}. In a way, pressure provides an independent tuning parameter for quantum criticality, which has a significant advantage: it does not introduce substitutional disorder. At the simplest approximation, pressure tunes the system by changing lattice parameters which in turn tune the bandwidth, so its effect can be significantly different from doping. Here we study evolution of the in-plane and inter-plane resistivity of parent BaFe₂As₂ with pressure.

In Fig. 5 we show evolution of in-plane (top panel) and inter-plane (bottom panel) resistivity of parent BaFe₂As₂ with pressure. Use of the piston-cylinder cell allowed us to reach pressures of about 20 kbar (at 4.2 K), which is not nearly sufficient to suppress magnetism of the parent compound⁴³, but acceptable to suppress T_{sm} to close to 100 K, as was the case for samples shown in Fig. 3. As can be seen from inset in top panel of Fig. 5, the structural/magnetic transition shifts from 134 K to about 115 K at the maximum pressure of our experiment. Very similar shift of T_{sm} is observed from $\rho_c(T)$ measurements, shown for comparison in inset with solid symbols. This comparison suggests that the c-axis resistivity measurements performed on samples with Sn soldered contacts covering the whole sample surface and which are strong enough to prevent samples from detwinning⁴⁴, do not significantly affect resistivity studies as a function of pressure.



FIG. 6. (Color online) Evolution of the position of the maximum of the temperature-dependent inter-plane resistivity, T_{max} as a function of the structural transition temperature T_S for cases of iso-valent Ru (blue circles) and P (red diamonds) substitutions and of application of pressure (black right triangles). Note the significantly faster increase of T_{max} under pressure.

Despite the limited pressure range, the maximum in the inter-plane resistivity at T_{max} shows quite significant shift to higher temperatures. It moves up from $T_{max} \sim 200$ K to almost 300 K in the pressure range studied. In Fig. 6 we compare the behavior of T_{max} and T_{sm} for parent Ba122 under pressure with their evolution upon Ru and P iso-valent substitutions. As $T_{sm} \rightarrow 0$ as the system approaches putative quantum critical point, this dependence reveals evolution of the features on its approach. Note that for BaP122 the structural and magnetic transitions are split with doping, while they remain coincident on Ru substitution^{39,40}. As can be seen from Fig. 6, pressure is moving maximum much faster than iso-valent substitution.

As can be seen from Fig. 5, the residual resistivity of BaFe₂As₂ does not change much with pressure. This reflects the fact that pressure does not change the degree of disorder in the samples. Simultaneously, this feature suggests that the electronic structure in the antiferromagnetic phase below T_{sm} does not change with pressure. Application of pressure reduces both in-plane and inter-plane resistivity in the metallic phase above T_{sm} , suggestive of weakening electronic correlations. The resistivity in the antiferromagnetic phase below T_{sm} shows an increase, indicating an increase of spin scattering below the transition, consistent with increase of spin disorder with pressure. The overall pressure evolution of the temperature-dependent resistivity is suggesting that the T-linear dependence of inter-plane resistivity can be eventually found at higher pressures corresponding to pressure-tuned QCP. Additional studies at higher pressures will be necessary in order to test this trend experimentally.

IV. DISCUSSION

The pseudogap or partial gap on the Fermi surface⁴⁵ in the cuprates is well established by a number of spectroscopic techniques^{46,47}, however, its microscopic origin is still debated⁴⁸. Main theories and experiments link it to either two neighboring phases, an antiferromagnetic Mott-insulator, with pseudogap arising due to exotic magnetism⁴⁹, and superconductivity, as an effect of the preformed superconducting pairs⁵⁰, or to a competing charge order phase⁵¹. The pseudogap is universally observed in both hole and electron⁵² doped cuprates, though it is much more pronounced in the former.

Our previous study of electron-doped BaCo122 found a clear correlation between the maximum in the interplane resistivity and anomalies in the temperature-dependent Knight shift and spin relaxation time^{11,16}. This correlation between the pseudogap features in resistivity and NMR measurements is also known in the cuprates⁴⁵. Further similarity between the two families is strong asymmetry of the pseudogap features on electron- and hole-doped sides^{45,52}. There are, however, some significant differences. In the cuprates the pseudogap vanishes close to optimum doping⁴⁷, while in electron- doped iron-based materials the superconducting dome is completely imbed-

ded into the pseudogap range 11,15 .

Because of the correlation between features in magnetic and resistive measurements, it is natural to consider magnetic origin of the pseudogap. Long range magnetic order, developing in the parent BaFe₂As₂ below T_m , leads to two effects. Reconstruction of the Fermi surface 53,54opens a gap on part of the Fermi surface, which is expected to give rise to resistivity increase. Simultaneously long-range magnetic ordering is accompanied by a loss of inelastic spin disorder scattering 55,56, which leads to an increase of the mean-free-path, giving rise to resistivity decrease. The increase of the mean-free path is limited at low temperatures by elastic scattering on residual impurities. In the parent compound the loss of the spin-disorder scattering dominates and the resistivity decreases gradually below T_m , particularly strongly in the cleanest annealed samples⁵⁷. The partial gapping is responsible for an increase of the resistivity below T_m in Ru- and Cosubstituted materials, see Fig. 3, in which contribution of spin-disorder scattering is small compared to impurity scattering, as evidenced by high residual resistivity value. The increase is mild in BaP122 and is virtually absent in BaK122 with significantly smaller residual resistivity.

Study of the in-plane resistivity anisotropy in straindetwinned samples^{44,58,59} found that actually resistivity increase starts at a structural transition at a temperature T_s which is always higher than T_m^{60} . Based on this observation, it was suggested that the tetragonalto-orthorhombic transition is also of magnetic origin and reflects directional nematic correlations between spins, without static long range magnetic order $^{61-64}$. These correlations lead to dramatically different effects for two directions in the plane, with resistivity increase for one direction of the current and the decrease for another 44,65 . Interestingly, strained samples reveal in-plane resistivity anisotropy at temperatures even significantly higher than T_s , showing that magnetic correlations (and thus nematic susceptibility) start significantly higher than actual long range ordering below T_m^{66-68} .

Following the same line of argument, it is natural to assign pseudogap features in the inter-plane resistivity to a build-up of magnetic correlations, reflected in NMR measurements^{15,16,69}. Significant difference, though, is that the directional inter-plane transport would be most sensitive to the inter-plane magnetic correlations, which should be quite strong in proximity to three-dimensional magnetism, as found in BaFe₂As₂. Our observations of the different doping-dependence of the pseudo-gap features for three types of substitutions may be suggestive that evolution of magnetic correlations proceeds significantly different in these cases. This difference may be also responsible for a difference between doping-evolution of the pseudogap features in magnetically two-dimensional cuprates and magnetically threedimensional iron-based materials.

V. CONCLUSIONS

Iso-valent Ru substitution in Ba(Fe_{1-x}Ru_x)₂As₂ leads to a systematic evolution of the temperature-dependent inter-plane resistivity towards a *T*-linear dependence at the optimal doping, in line with expectations for the scenario invoking existence of the quantum critical point in the substitutional phase diagram. The dominant hightemperature feature of the temperature-dependent interplane resistivity, a maximum at a temperature T_{max} , shifts rapidly to higher temperatures with x, revealing a trend similar to another type of iso-valent substitution in BaFe₂(As_{1-x}P_x)₂⁷. Because the two substituseries show different evolution of the Fermi surface^{25,70}, this similarity suggests that the maximum is not related to evolution of the electronic structure.

Our observations suggest that despite very different evolution of T_{max} feature for four different types of

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