The response of superconductors to controlled introduction of point-like disorder is an important tool to probe their microscopic electronic collective behavior. In the case of iron-based superconductors, magnetic fluctuations presumably play an important role in inducing high-temperature superconductivity. In some cases, these two seemingly incompatible orders coexist microscopically. Therefore, understanding how this unique coexistence state is affected by disorder can provide important information about the microscopic mechanisms involved. In one of the most studied pnictide family, hole-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ (BaK122), this coexistence occurs over a wide range of doping levels, 0.16 ≤ x ≤ 0.25. We used relativistic 2.5 MeV electrons to induce vacancy-interstitial (Frenkel) pairs that act as efficient point-like scattering centers. Upon increasing dose of irradiation, the superconducting transition temperature $T_c$ decreases dramatically. In the absence of nodes in the order parameter this provides a strong support for a sign-changing $s_\pm$ pairing. Simultaneously, in the normal state, there is a strong violation of the Matthiessen’s rule and a decrease (surprisingly, at the same rate as $T_c$) of the magnetic transition temperature $T_{sm}$, which indicates the itinerant nature of the long-range magnetic order. Comparison of the hole-doped BaK122 with electron-doped Ba(Fe$_{0.5}$Co$_{1.5}$)$_2$As$_2$ (FeCo122) with similar $T_{sm}$ ~ 110 K, x = 0.02, reveals significant differences in the normal states, with no apparent Matthiessen’s rule violation above $T_{sm}$ on the electron-doped side. We interpret these results in terms of the distinct impact of impurity scattering on the competing itinerant antiferromagnetic and $s_\pm$ superconducting orders.

**Article**

**Interplay between superconductivity and itinerant magnetism in underdoped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($x = 0.2$) probed by the response to controlled point-like disorder**

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The response of superconductors to controlled introduction of point-like disorder is an important tool to probe their microscopic electronic collective behavior. In the case of iron-based superconductors, magnetic fluctuations presumably play an important role in inducing high-temperature superconductivity. In some cases, these two seemingly incompatible orders coexist microscopically. Therefore, understanding how this unique coexistence state is affected by disorder can provide important information about the microscopic mechanisms involved. In one of the most studied pnictide family, hole-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ (BaK122), this coexistence occurs over a wide range of doping levels, 0.16 ≤ x ≤ 0.25. We used relativistic 2.5 MeV electrons to induce vacancy-interstitial (Frenkel) pairs that act as efficient point-like scattering centers. Upon increasing dose of irradiation, the superconducting transition temperature $T_c$ decreases dramatically. In the absence of nodes in the order parameter this provides a strong support for a sign-changing $s_\pm$ pairing. Simultaneously, in the normal state, there is a strong violation of the Matthiessen’s rule and a decrease (surprisingly, at the same rate as $T_c$) of the magnetic transition temperature $T_{sm}$, which indicates the itinerant nature of the long-range magnetic order. Comparison of the hole-doped BaK122 with electron-doped Ba(Fe$_{0.5}$Co$_{1.5}$)$_2$As$_2$ (FeCo122) with similar $T_{sm}$ ~ 110 K, x = 0.02, reveals significant differences in the normal states, with no apparent Matthiessen’s rule violation above $T_{sm}$ on the electron-doped side. We interpret these results in terms of the distinct impact of impurity scattering on the competing itinerant antiferromagnetic and $s_\pm$ superconducting orders.

**Introduction**

The use of controlled disorder is a powerful phase-sensitive way to study the nature of the superconducting state without affecting the chemical composition. According to Anderson’s theorem, conventional isotropic $s$–wave superconductors are not affected by the scalar potential (i.e., non spin-flip) scattering, but are sensitive to spin-flip scattering due to magnetic impurities (for recent theoretical results on the impact of impurities on $T_c$, see for example refs. 11–13). In single-band high superconducting transition temperature (high-$T_c$) cuprates, both magnetic and non-magnetic impurities cause a rapid suppression of $T_c$, consistent with the nodal $d$–wave pairing. In multi-band iron-based superconductors (IBS), a sign-changing order parameter between the electron-like and hole-like Fermi sheets, $s_{sr}$, is the most plausible pairing state. Although its response to non-magnetic scattering depends sensitively on the multi-band structure of the pairing interaction, on the chemical potential, and on the gap anisotropy, it is generally expected that intraband scattering is much less efficient in causing pair-breaking than interband scattering. Additionally, the orbital content of the bands can also affect the suppression of $T_c$. We note that the multi-band character of the superconducting state alone is not sufficient to have $T_c$ suppression. For instance, in the known two-gap $s_{++}$ superconductor, MgB$_2$, where the gap does not change sign, electron irradiation resulted only in a small change due to gap magnitude difference between two bands. While the effect of scattering induced by various means from chemical substitution to irradiation with various particles on $T_c$, has been studied in many IBS, there is limited experimental information on the effects of point-like disorder simultaneously on superconducting and magnetic transitions in the regime where superconductivity and antiferromagnetism coexist. The expected physics, however, is very intriguing. Assuming an itinerant nature for long-range magnetic order (LRMO), it has been shown that $T_c$ may actually increase upon the introduction of disorder due to the stronger effect on magnetism quantified via the suppression of the magnetic transition temperature, $T_{sm}$. (Here “sm” is used to indicate simultaneous structural and magnetic transitions in underdoped BaK122). However, this is not a universal trend, as it depends on the relative ratio of the magnetic and superconducting state energies and on the relative strength of the intraband and interband scattering rates.

Irradiation of relatively thin crystals (~20 μm in our case) with 2.5 MeV relativistic electrons is known to produce vacancy—interstitial Frenkel pairs, which act as efficient point-like scattering centers. In the high-$T_c$ cuprates these defects are known to be strong unitary scatterers causing significant suppression of $T_c$. There is a growing number of studies of the effects of electron

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irradiation not only on \( T_c \) (see ref. 8 and references therein), but on other properties, such as vortex pinning and creep 38 and London penetration depth.39–42 In a previous study of electron irradiated \( \text{Ba}_1-\delta\text{K}_\delta\text{Fe}_2\text{As}_2 \) we focused on the evolution of the superconducting gap structure with the potassium concentration and found noticeable changes in the behavior, such as increasing gap anisotropy.40,41,43

In this paper, we focus on the effects of electron irradiation simultaneously on \( T_p \), \( \tau_{sm} \) and normal state resistivity in an underdoped composition of \( \text{Ba}_1-\delta\text{K}_\delta\text{Fe}_2\text{As}_2, \ x=0.2 \), in which LRMO coexists with superconductivity. In the normal state, we find strong violation of the Matthiessen’s rule below \( T_{sm} \) which is expected due to a change of the band structure and thus effective carrier density in the magnetically ordered state, and above \( T_{sm} \) in the broad temperature range, which is unexpected at least in the simple picture. Moreover, this behavior is in a stark contrast with the electron-doped \( \text{Ba}(\text{Fe}_1-x\text{Co})_x\text{As}_2 \) \( (x=0.02) \) with similar \( T_{sm} \) in which the Matthiessen’s rule is expectedly violated below \( T_{sm} \) but obeyed above \( T_{sm} \). At a first sight this could be understood that in this case additional disorder is not so effective, because despite notably lower substitution level, \( x \), required to suppress magnetism, doping into Fe-As planes introduces much higher scattering rates as evidenced by notably higher residual resistivity values. This argument, however, does not hold, because (1) the magnetic transition temperature, \( T_{sm} \), changes by a similar amount as in \( \text{BaK}_{1.2} \) and (2) the same compliance with the Matthiessen’s rule above \( T_{sm} \) is observed in isovolumetric electron-replaced \( \text{BaFe}_{1-x}\text{Ru}_x\text{As}_2 \) 45 and very clean \( \text{BaFe}_2\text{As}_2(\text{Fe}_{0.8}\text{Ru}_{0.2})_2 \). Therefore, the difference is likely in the electronic structure of \( \text{BaK}_{1.2} \) and specifics of its inter- and intra-band interactions and scattering channels.26

RESULTS AND DISCUSSION

The panel (b) in Fig. 1 shows the evolution of the temperature-dependent resistivity of under-doped \( \text{Ba}_1-\delta\text{K}_\delta\text{Fe}_2\text{As}_2, \ x=0.20 \), with increase of irradiation dose/disorder. We zoom on the low-temperature range revealing features in \( \rho(T) \) curves at the structural/magnetic, \( T_{sm} \) and superconducting, \( T_c \) transitions. The resistivity of the samples at temperatures just above \( T_c \) follows well a \( \rho(T)+AT^2 \) dependence, similar to previous reports, 46 allowing easy extrapolation of \( \rho(T) \) and tracking its evolution with disorder. The residual resistivity of the pristine samples was about 40 \( \mu \Omega \cdot \text{cm} \), and residual resistivity ratio \( \rho(300 \text{~K})/\rho(0)>7 \). Residual resistivity increased up to approximately 120 \( \mu \Omega \cdot \text{cm} \) at the highest dose of 6 \( \mu \text{C/cm}^2 \). On structural/magnetic ordering, resistivity of the sample shows small down-turn on cooling, due to a loss of spin-disorder scattering. The structural transition temperature was determined using temperature-dependent resistivity derivative and peak position as a criterion, as shown in Fig. 1c. The \( T_{sm} \) is monotonically suppressed with increase of sample residual resistivity as shown in Fig. 1 panel (d), right scale. The superconducting transition temperature was determined at crossing points of linear extrapolations of the sharp resistivity drop at the transition and smooth \( T \) extrapolations of the curves in the normal state. The irradiation does not change the sharpness of the resistive transition, so the use of the alternative criterion (midpoint or zero resistance cross-point) for \( T_c \) determination does not alter any of our conclusions. The \( T_c \) shows monotonic decrease with irradiation from above 16 to 9 K Fig. 1d. Interestingly, the decrease of both temperatures in absolute numbers is almost the same and the two are linearly proportional to each other, see Fig. 1 panel (e).

The top panel (a) of Fig. 2 shows 4-probe resistivity measured in the sample \( \text{Ba}_1-\delta\text{K}_\delta\text{Fe}_2\text{As}_2, \ x=0.20 \), in pristine state and after electron irradiation with the dose of 0.8 \( \mu \text{C/cm}^2 \). Even at this relatively small dose, a clearly non-parallel shift of the curves indicates significant violation of the Matthiessen’s rule in the paramagnetic state. In Drude model, the electrical conductivity of metal \( \sigma=en\tau/m^* \), here \( n \) is the carrier density, \( \tau \) is the scattering time, \( e \) is electron charge and \( m^* \) is the effective bare mass of the carriers. The Matthiessen’s rule states that the total scattering rate \( \tau^{-1}=\tau_s^{-1}+\tau_d^{-1} \), which leads to a parallel shift of \( \rho(T) \) curve with the increase of residual \( \rho(0) \) with disorder. The Matthiessen’s rule can be violated by Fermi surface reconstruction, which takes place below \( T_{sm} \) or even above \( T_{sm} \) as a consequence of the anisotropic character of the magnetic fluctuations.43 Close to room temperature the Matthiessen’s rule is valid, with the violation being closely linked with a crossover feature in the temperature-dependent resistivity at around 200 K. For comparison in inset in panel (a), Fig. 2, we show temperature-dependent resistivity of slightly electron-doped \( \text{Ba(Fe}_1-x\text{Co})_x\text{As}_2, \ x=0.02, \) in pristine state and after 3.4 \( \mu \text{C/cm}^2 \) electron irradiation. Notably, electron irradiation leads to a comparable increase of the room temperature resistivity, \( \rho(300 \text{~K}) \), in both electron and hole-doped compositions, around 3–4 \( \mu \Omega \cdot \text{cm} \) per C/cm\(^2 \), respectively. However, the increase remains practically constant above \( T_{sm} \) in electron-doped composition, similar to the behavior of hole-doped composition above the crossover feature and to phosphorus-substituted samples.40 Also, the resistivity change at \( T_{sm} \) upon cooling is quite different between hole and electron-doped compounds, showing only a slight downturn in the former, but a pronounced jump in the latter.

To understand the difference in behavior, it is important to note that orbitals of iron and arsenic in FeAs layer are contributing the most to the density of states at the Fermi level in \( \text{BaFe}_2\text{As}_2 \) based materials. Therefore disorder introduced by random positions of substitutional Co atoms in the FeAs layer, affects electron scattering significantly stronger than substitutional disorder of K on Ba site. This can be directly seen in notably lower residual resistivity in \( \text{Ba}_1-\delta\text{K}_\delta\text{Fe}_2\text{As}_2, \ x=0.20, \rho(0)\sim 40 \mu \Omega \cdot \text{cm} \) than in \( \text{Ba(Fe}_1-x\text{Co})_x\text{As}_2, \ x=0.02, \rho(0)\sim 170 \mu \Omega \cdot \text{cm} \), despite five times smaller level of substitution in the latter case. Because of this high level of substitutional disorder in Co-doped material, additional disorder introduced by electron irradiation plays relatively smaller role than in K-doped compound. This different level of background disorder leads to different resistivity behavior on passing \( T_{sm} \) in pristine samples. Loss of the carrier density below \( T_{sm} \) due to partial gap opening in conditions when carrier mean free path is controlled by disorder and is essentially temperature-independent, gives resistivity increase in disordered Co-doped material. Some loss is compensated by notable increase of mean free path due to the loss of spin-disorder scattering in hole-doped compositions. These considerations were directly illustrated recently in irradiation study on \( \text{BaFe}_2\text{As}_2 \) with P substitution.10 Note, however, that these considerations do not explain the violation of the Matthiessen rule above \( T_{sm} \) in hole-doped as opposed to its validity in electron-doped compositions. The difference is not related to the level of substitutional disorder in two cases, since both absolute increase of resistivity above \( T_{sm} \) and suppression rate of \( T_{sm} \) with disorder are very similar on both sides.

The lower panel (b) of Fig. 2 shows temperature dependent Hall coefficient, \( R_H \), obtained using van der Pauw technique in the sample \( A \) of \( \text{Ba}_1-\delta\text{K}_\delta\text{Fe}_2\text{As}_2, \ x=0.20 \) before and after irradiation. For reference we show data in other hole-doped samples, \( x=0.3 \) and \( x=0.4 \), in all cases normalizing data at 20 K, the lowest temperature of our Hall effect measurements. (For normalization, the data for \( x=0.4 \) and \( x=0.3 \) were extrapolated to \( T=20 \text{~K} \)). Irradiation does not change either magnitude or temperature dependence of the Hall effect in sample with \( x=0.20 \), despite three-fold variation of sample the resistivity. On the other hand doping clearly changes magnitude and temperature dependence of the Hall effect. These observations clearly show that defects introduced by irradiation are not doping the system. It should be noted that the independence of Hall coefficient on the residual
resistivity is possible only if all types of carriers change their mobility by the same factor, - not so easy condition to meet in multi-band systems. If magnetism was due to localized spins, one would expect that disorder, as introduced in our experiment, would affect $T_{sm}$ primarily via the effect of random dilution. If magnetism however arises from a Fermi surface instability, the change in the lifetime of the electronic states will affect $T_{sm}$. Indeed, ref.26, studying a simplified two-band model for the interplay between superconductivity and magnetism, showed that both intraband and interband impurity scattering suppress $T_{sm}$. This is to be contrasted with the case of $s_{\pm}$ superconductivity, in which $T_c$ is only affected by interband impurity scattering. Because LRMO competes with superconductivity, depending on how strong this competition is, it is possible that the net effect of disorder is to increase $T_c$ in the coexistence region. This seems to be the case in P-doped Ba122, but not in BaK122, where we find $T_c$ to also be suppressed. One possible explanation for this difference would be that the competition between superconductivity and magnetism...
is not as strong in K-doped systems as in P-doped systems, or that the intraband scattering is dominant over the interband scattering in the system studied here.

As for the Matthiessen’s rule, a known scenario in which it is violated is when impurities are added in a system whose main scattering mechanism is strongly anisotropic in momentum space. In BaK122, a natural candidate is the scattering by spin fluctuations, which in this system are strongly peaked at the finite wave-vectors \((n, 0)\) and \((0, n)\). In this case, the violation of the Matthiessen’s rule would imply that the resistivity of the normal state is dominated by magnetic fluctuations. Such a preponderance of magnetic fluctuations could in principle favor a higher \(T_c\) state, if indeed pairing is mediated by spin fluctuations.

It should be noted, however, that the position of hot spots as well as the strength of inelastic scattering are very similar on electron and hole-doped sides, while the effect of disorder (as seen in direct comparison for Fig. 3) is dramatically different. Alternative explanation for the strong violation of the Matthiessen rule was suggested in a recent study of the effect of natural growth disorder on properties of BaFe1−xPdxAs2 with Ru substitution. Here it was assigned to predominant suppression of high mobility carriers with disorder.

In conclusion, irradiation with relativistic 2.5 MeV electrons leads to rapid suppression of both superconducting \(T_c\) and the temperature of concomitant orthorhombic/antiferromagnetic transition \(T_{\text{sm}}\). In the absence of nodes in the superconducting order parameter, observation of rapid suppression of \(T_c\) provides a strong support for a sign-changing \(s_\pi\) pairing. Rapid suppression of \(T_{\text{sm}}\) surprisingly at the same rate as \(T_c\) indicates the itinerant nature of the LRMO. Comparison of the hole-doped \(\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2\) with electron-doped \(\text{BaFe}(\text{Fe}_{1-x}\text{Co}_x)\text{As}_2\) with similar \(T_{\text{sm}}\sim 110 \text{ K}\), \(x = 0.02\), reveals significant differences in the normal states, with no Matthiessen’s rule violation above \(T_{\text{sm}}\) on the electron-doped side and strong violation on the hole-doped side. Our results provide strong evidence of the itinerant nature of the AFM phase and non-trivial influence of non-magnetic disorder on coupled superconductivity and magnetism in iron-based superconductors.

**METHODS**

Single crystalline samples and transport measurements

Single crystals of \(\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2\) were synthesized using high-temperature FeAs flux method. Two samples \(A\) and \(B\) were used. The electrical resistivity and Hall effect of sample \(A\) were measured on \(\text{Si} \times \text{Si}\) sample with four contacts soldered with Sn at the corners in van der Pauw configuration. The electrical resistivity of sample \(B\) was measured in standard 4-probe configuration. The resistivity of both samples at room temperature, \(\rho(300 \text{ K})\), before irradiation was set to 300 \(\mu\Omega\text{cm}\), the value determined from measurements on big arrays of crystals, with actually measured value being within 10% uncertainty of geometric factor determination.

Single crystals of \(\text{BaFe}(\text{Fe}_{1-x}\text{Co}_x)\text{As}_2\) were grown from FeAs/CoAs flux from a starting load of metallic Ba, FeAs, and CoAs, as described in detail elsewhere. The composition of the sample was determined using wavelength dispersive spectroscopy (WDS) version of electron probe microanalysis as \(x = 0.02 \pm 0.002\). The electrical resistivity of the sample \(C\) was measured in four-probe configuration. Similar to hole-doped sample, resistivity of the sample before irradiation was set as 300 \(\mu\Omega\text{cm}\). The samples were mounted on a thin mica plate in a hollow Kyocera chip, so that they could be moved between irradiation chamber and resistivity and Hall effect setups in a different \(^4\text{He}\) cryostat without disturbing the contacts.

**Electron irradiation**

The low-temperature 2.5 MeV electron irradiation was performed at the SIRIUS Pelletron linear accelerator operated by the Laboratoire des Solides Irradiés (LSI) at the Ecole Polytechnique in Palaiseau, France. The Kyocera chip was mounted inside the irradiation chamber and was cooled by a flow of liquid hydrogen to \(T = 22 \text{ K}\) to remove excess heat produced by relativistic electrons upon collision with the ions. The flux of electrons amounted to about 2.7 \(\mu\text{A}\) of electric current through a 5 \(\text{mm}\) diameter diaphragm. This current was measured with the Faraday cage placed behind the hole in the sample stage, so that only transmitted electrons were counted. The irradiation rate was about \(5 \times 10^{-6} \text{ C}/(\text{cm}^2\text{s})\) and large doses were accumulated during several irradiation runs. The resistance of sample \(A\) at 22 K was monitored in situ during irradiation, revealing linear increase with irradiation dose, one segment of the broken line in Fig. 1a. Periodically the sample \(A\) was extracted from irradiation chamber and the effect of irradiation was characterized by ex situ measurements of electrical resistivity as function of temperature, Fig. 1b, and of the Hall effect, Fig. 2b. Warming the sample to room temperature leads to partial defect annealing, as can be seen as the down-steps in the dose dependence of resistivity at 22 K at the start of the next irradiation run. This annealing is incomplete, as evidenced by gradual increase of resistivity for subsequent runs. The resistivity of the sample at room temperature remained stable for a period of at least several months, unless the sample was further warmed above room temperature. For sample \(B\) of K-doped and sample \(C\) of Co-
doped materials the whole dose was applied in one shot without intermediate measurements.

**DATA AVAILABILITY**
The authors declare that all data supporting the findings of this study are available within the article or from the corresponding author upon reasonable request.

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**AUTHOR CONTRIBUTIONS**
R.P. and M.A.T. conceived the project. M.K. and R.P. conducted electron irradiation with the technical support from the SIRIUS team at École Polytechnique. M.K., R.P. and M.A.T. performed transport measurements and data analysis. P.C.C. and H.-H.W. characterized and selected the samples. R.M.F. contributed theoretical analysis. All authors discussed the data, interpreted the results and wrote the manuscript.

**ADDITIONAL INFORMATION**
Competing interests: The authors declare no competing interests.

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