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# Charge doping-induced quasiparticle scattering in iron-pnictide superconductors as probed by vortex pinning

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Charge doping of iron-pnictide superconductors leads to collective pinning of flux vortices, whereas isovalent doping does not. Moreover, flux pinning in the charge-doped compounds is consistently described by the mean-free path fluctuations introduced by the dopant atoms, allowing for the extraction of the elastic quasiparticle scattering rate. The absence of scattering by dopant atoms in isovalently doped  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  is consistent with the observation of a linear temperature dependence of the low-temperature penetration depth in this material.

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The suggestion of exotic multi-band superconductivity in the iron-pnictide superconductors [1–3], has raised questions as to the relevance of pair-breaking by atomic-scale disorder in these materials [4–13]. In particular, the proposed fully-gapped  $s_{\pm}$  state, characterized by a sign-reversal of the order parameter phase on different Fermi surface sheets, should be strongly affected by weak interband scattering [5–7, 13]. A number of authors argue that, in the case of the fully gapped  $s_{\pm}$ -state, a moderate impurity density leads to the appearance of gapless superconductivity [8, 13], a situation that may explain the temperature dependence [5] of the *ab*-plane penetration depth observed in Ref. [9]. In the case of nodal  $s_{\pm}$  superconductivity [2, 3], the presence of disorder would be responsible for the lifting of nodes [7]. This scenario is possibly relevant for the observation of fully gapped superconductivity in  $(\text{Ba,K})\text{Fe}_2\text{As}_2$  [14]. Alternatively, the latter experimental result could be the hallmark of emerging competing  $s_{++}$  superconductivity [8], or of  $s_{\pm}$  superconductivity with strong pair-breaking [5, 13]. Finally, the observation of gap nodes in  $\text{LaFePO}$  [15–17] and  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  [18–20] could either be the manifestation of a nodal  $s_{\pm}$  state, or the result of weak pair-breaking disorder [8]. In order to settle these controversies, it is necessary to characterize the scattering rates of impurities and other types of disorder in these materials by an independent means.

Here, we argue that pinning of the vortex lattice, sensitive not only to extrinsic but also to intrinsic disorder in superconductors, can be used as a probe for the microscopic quasi-particle scattering mechanism. In particular, in electron-doped  $\text{PrFeAsO}_{1-y}$ ,  $\text{NdFeAsO}_{1-x}\text{F}_x$ , and  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ , the critical current density  $j_c$  is consistently described in terms of collective pinning mediated by spatial fluctuations of the quasi-particle mean free path [21, 22]. The density of point pins responsible for pinning closely corresponds to the dopant atom

concentration. On the other hand, isovalently doped  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  is characterized by a very small  $j_c$  as a result of negligible vortex pinning by atomic-scale point defects. From this, we conclude that charged point defects are responsible for quasiparticle scattering and collective pinning, but neutral atomic-scale point defects are not. Analysis of the critical current allows for the estimate of the scattering cross-section and scattering phase angle  $\delta_0$  of the defects, which turns out to be best described by the Born limit. Hence, the absence of quasiparticle scattering in the  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  material allows for superconductivity characterized by a gap with line nodes to survive, whereas, in charge-doped iron pnictides, gap nodes may be lifted [7], or putative  $s_{\pm}$  superconductivity compromised [8].

Critical current densities of single crystalline  $\text{PrFeAsO}_{1-y}$  (with  $y \sim 0.1$ ,  $T_c \sim 35$  K) [23–26],  $\text{NdFeAsO}_{0.9}\text{F}_{0.1}$  ( $T_c \sim 36$  K) [26–29],  $\text{Ba}_{0.45}\text{K}_{0.55}\text{Fe}_2\text{As}_2$  ( $T_c \sim 34$  K) [14, 24], and  $\text{BaFe}_2(\text{As}_{0.67}\text{P}_{0.33})_2$  ( $T_c = 29$  K) [18, 19] were obtained from local measurements of the magnetic flux density perpendicular to the crystal surface,  $B_{\perp}$ , and the flux density gradient  $dB_{\perp}/dx$ . Previous work has shown the critical current of superconducting iron pnictide crystals to be spatially inhomogeneous [26]. While a *global* measurement of the *average* flux density over the crystal surface, or of the magnetic moment of the entire crystal, may result in a spurious temperature dependence  $j_c(T)$ , local measurements do not have this shortcoming. Local  $j_c$  values in applied fields up to 50 mT were obtained from magneto-optical imaging (MOI) of the flux density [26, 30]. Measurements in fields up to 2 T were performed using micron-sized Hall probe arrays, tailored in a pseudomorphic GaAlAs/GaAs heterostructure [25]. The 10 Hall sensors of the array had an active area of  $3 \times 3 \mu\text{m}^2$ , while an 11th sensor was used for the measurement of the applied field.

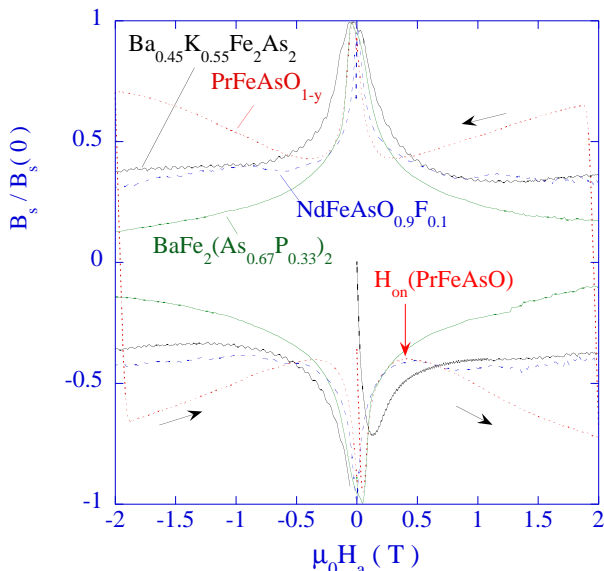


FIG. 1: (Color online) Normalized hysteresis loops of the local “self-field”, measured on the top surfaces of  $\text{PrFeAsO}_{1-y}$ ,  $\text{NdFeAsO}_{0.9}\text{F}_{0.1}$ ,  $\text{Ba}_{0.45}\text{K}_{0.55}\text{Fe}_2\text{As}_2$ , and  $\text{BaFe}_2(\text{As}_{0.67}\text{P}_{0.33})_2$  single crystals, at reduced temperature  $T/T_c = 0.2$ . Arrows indicate the direction in which the cycles are traversed.

Figure 1 shows hysteresis cycles of the local “self-field”  $B_s \equiv B_{\perp} - \mu_0 H_a$  (where  $\mu_0 \equiv 4\pi \times 10^{-7} \text{ Hm}^{-1}$ ) versus  $H_a$  for a variety of iron pnictide superconductors, at the same reduced temperature  $t \equiv T/T_c = 0.2$ . A salient feature of the hysteresis loops is the presence of a pronounced peak at small field. In the  $\text{REFeAsO}$  (1111) family of iron pnictide superconductors (where RE stands for a rare earth ion) [26], as in the  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  [31–33] and  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  [34] “122” superconductors, this peak is superposed on a field-independent contribution. At higher fields, the hysteresis loop width in these compounds increases again, at a field  $H_{on}$ , the result of a structural change of the vortex ensemble [26]. On the contrary, in isovalently doped  $\text{BaFe}_2(\text{As}_{0.67}\text{P}_{0.33})_2$ , the hysteresis loop width shows a monotonous decrease.

Figure 2 shows the critical current density of five iron-pnictide compounds. For the four compounds measured in the present study,  $j_c = 2\mu_0^{-1} dB_{\perp}/dx$  is extracted using the Bean model, [36], while data for  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  and  $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$  are taken from Refs. [32] and [34], respectively. In what follows, we describe  $j_c$  as the superposition of two contributions,  $j_c^{coll}$  and  $j_c^s(B)$ , with the former accounting for the constant hysteresis width at higher fields. It is seen that for all materials, the low-field peak in  $B_s(H_a)$  amounts to a plateau in  $j_c(B)$ ,

$$j_c(0) = j_c^{coll} + j_c^s(0), \quad (1)$$

followed by a power-law decrease, such that

$$j_c(B) = j_c^{coll} + j_c^s(B) \sim j_c^{coll} + AB_{\perp}^{-\beta} \quad (2)$$

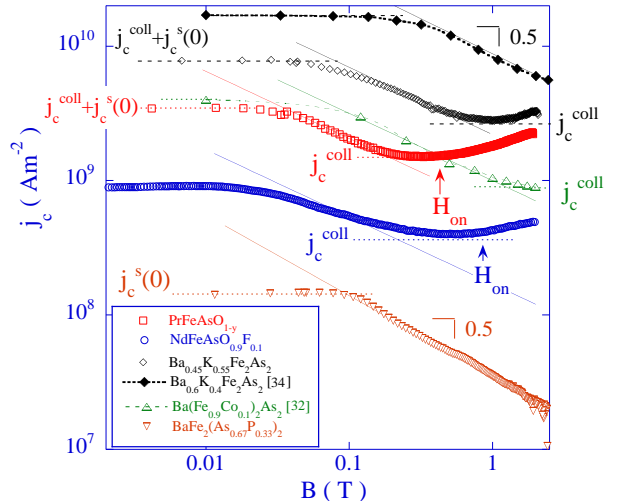


FIG. 2: (Color online) Critical current density as function of magnetic flux density for  $\text{PrFeAsO}_{1-y}$  ( $\square$ ),  $\text{NdFeAsO}_{0.9}\text{F}_{0.1}$  ( $\circ$ ),  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  ( $\triangle$ ) [32],  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  ( $\diamond$ ), and isovalently doped  $\text{BaFe}_2(\text{As}_{0.67}\text{P}_{0.33})_2$  single crystals, at reduced temperature  $t = 0.21$ . Drawn lines indicate the power-law dependence of the strong pinning contribution to the critical current density at intermediate fields.

with  $0.5 < \beta < 0.63$ . The behavior of  $j_c^s$  is precisely that expected for vortex pinning by sparse pointlike defects of radius larger than the coherence length  $\xi$ , which are inevitably present in any real, imperfect crystal [37–39]. An analysis of data on the (1111) iron pnictides has shown that spatial variations of the average dopant atom density on a (large) scale of several dozen nm, leading to concomitant modulations of  $T_c$ , account for the measured magnitude and temperature dependence of  $j_c^s$  [26]. Fig. 3a shows that a similar vortex pinning mechanism is certainly relevant in the “122” family of compounds. In all investigated crystals,  $j_c^s$  depends on the spatial location at which it is measured, but the field-independent  $j_c^{coll}$  is rather homogeneous and will be attributed to atomic scale fluctuations of the dopant atom positions (collective pinning) [21].

The different field dependence of the two critical current contributions (Fig. 2) allows one to extract both as function of temperature. Figure 3a shows the  $T$ -dependence of  $j_c(0) = j_c^{coll} + j_c^s(0)$ , while the lower panel (b) shows only the field-independent contribution  $j_c^{coll}$ . This is found to be non-zero in the charge-doped iron pnictides  $\text{PrFeAsO}_{1-y}$ ,  $\text{NdFeAsO}_{1-x}\text{F}_x$ ,  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ , and  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$ , but is conspicuously absent in isovalently doped  $\text{BaFe}_2(\text{As}_{0.67}\text{P}_{0.33})_2$ , see Fig. 3c. This means that charged dopant atoms (or vacancies, in the case of  $\text{PrFeAsO}_{1-y}$ ) lead to collective pinning of vortices, but uncharged dopant atoms do not.

In order to quantitatively describe  $j_c^{coll}$ , we treat the dopant atoms as point defects responsible for quasi-particle scattering. The elementary pinning force of such

defects can be written

$$f_p \sim 0.3g(\rho_D)\varepsilon_0 (\sigma_{tr}/\pi\xi^2) (\xi_0/\xi), \quad (3)$$

where  $\sigma_{tr} = (2\pi/k_F^2) \sin^2 \delta_0 = \pi D_v^2$  is the transport scattering cross-section,  $k_F$  is the Fermi wavevector,  $D_v$  is the effective range of the potential, and  $g(\rho_D)$  is the Gor'kov function [21, 22, 40]. The disorder parameter  $\rho_D = \hbar v_F/2\pi T_c l \sim \xi_0/l$ , with  $v_F$  the Fermi velocity,  $l = (n_d \sigma_{tr})^{-1}$  the quasi-particle mean free path, and  $\xi_0 \approx 1.35\xi(0)$  the (temperature-independent) Bardeen-Cooper-Schrieffer coherence length [21, 22, 40]. The critical current arises from the local density fluctuations of the defects, and is therefore determined by the second moment of the elementary pinning force,  $\langle f_p^2 \rangle$ . Applying the theory of collective pinning [21, 40], it reads [26]

$$j_c^{coll} \approx j_0 \left[ \frac{0.1n_d D_v^4}{\varepsilon_\lambda \xi} \left( \frac{\xi_0}{\xi} \right)^2 \right]^{2/3} \propto \left[ \frac{\lambda(0)}{\lambda(T)} \right]^2 \left( 1 - \frac{T}{T_c} \right)^\alpha, \quad (4)$$

where  $j_0 \equiv \Phi_0/\sqrt{3}\pi\mu_0\lambda_{ab}^2\xi$  is the depairing current density, and  $\varepsilon_\lambda \equiv \lambda_{ab}/\lambda_c$  the penetration depth anisotropy. Eq. 4 does not depend on the symmetry of the superconducting ground state. However, the exponent  $\alpha \sim 2$  does depend on the different weight that distinct Fermi surface sheets have in contributing to superconductivity in different compounds. Here it is treated as a phenomenological parameter, obtained from the ratio of  $ab$ -plane and  $c$ -axis penetration depths in the different compounds [25, 28], while  $\lambda(0)/\lambda(T)$  was published in Refs. [25, 28] and [24].

Figure 3b shows that the temperature dependence of  $j_c^{coll}$  is very well described by Eq. (4). In  $\text{PrFeAsO}_{1-y}$ , its magnitude is accurately reproduced by inserting the oxygen ion radius  $D_v = 1.46 \times 10^{-10}$  m and the defect density  $n_d \approx 1.5 \times 10^{27}$  m $^{-3}$ . This corresponds to the oxygen vacancy concentration at our doping level,  $y \sim 0.1$ . Thus, the collective pinning contribution to the critical current density of the  $\text{PrFeAsO}_{1-y}$  compound is well described by the quasi-particle mean-free path fluctuation mechanism of Refs. [21, 22]. The same holds true for  $\text{NdFeAsO}_{1-x}\text{F}_x$  and  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$ . If one takes defect densities corresponding to the dopant atom concentration,  $n_d \sim 1.5 \times 10^{27}$ ,  $\sim 1 \times 10^{27}$ , and  $4 \times 10^{27}$  m $^{-3}$  respectively, very satisfactory fits to  $j_c^{coll}(T)$  can be

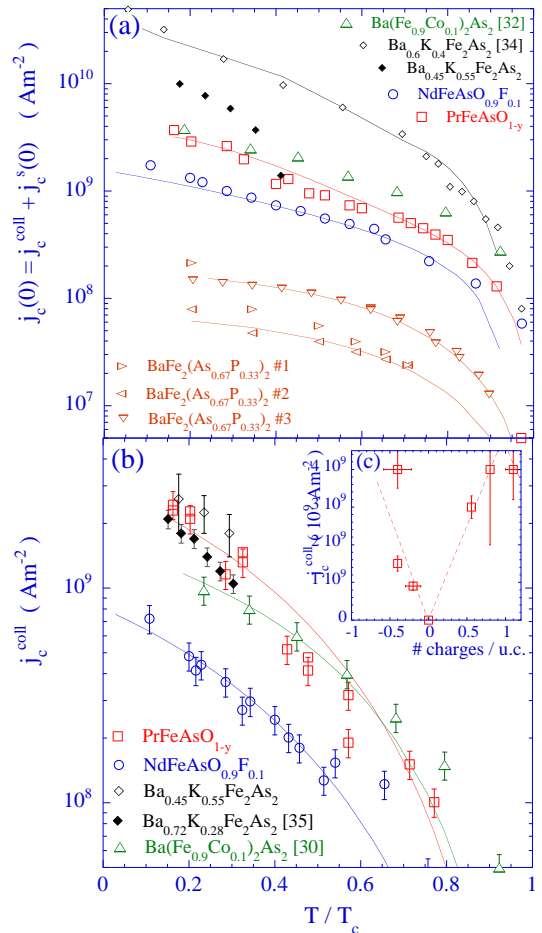


FIG. 3: (Color online) (a) Zero field  $j_c$  of  $\text{PrFeAsO}_{1-y}$ ,  $\text{NdFeAsO}_{0.9}\text{F}_{0.1}$ ,  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ ,  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  [32], and three different  $\text{BaFe}_2(\text{As}_{0.67}\text{P}_{0.33})_2$  single crystals. Drawn lines indicate fits to Eq. (1), with  $j_c^s(0)$  taken from Ref. [26] [ $j_c^{coll} = 0$  for  $\text{BaFe}_2(\text{As}_{0.67}\text{P}_{0.33})_2$ ]. (b) Collective pinning contribution  $j_c^{coll}$  in the charge-doped compounds. Drawn lines are fits to Eq. (4). (c) The low- $T$  value of  $j_c^{coll}$  as function of the number of dopant charges per unit cell.

obtained using the scattering cross-sections of Table I. As far as  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  is concerned,  $j_c^s$  exceeds  $j_c^{coll}$  by more than an order of magnitude, which prohibits a reliable determination of the latter at high temperature. Therefore, we only consider the low- $T$  magnitude of  $j_c^{coll}$  for this compound. The dopant atom densities lead to values  $n_d\xi_0^3$  that are largely in excess of unity, justifying

Compound	$k_F$ ( $\text{\AA}^{-1}$ )	$\xi_0$ (nm)	$n_d$ ( $\text{nm}^{-3}$ )	$\sigma_{tr}$ ( $\text{m}^2$ )	$D_v$ ( $\text{\AA}$ )	$n_d D_v^3$	$n_d \xi_0^3$	$\sin \delta_0$	$\Gamma$ (meV)	$l$ (nm)
$\text{PrFeAsO}_{1-y}$	0.33	2.4	1.5	$6.7 \times 10^{-20}$	1.46	$5 \times 10^{-3}$	21	0.3(2)	10	10
$\text{NdFeAsO}_{0.9}\text{F}_{0.1}$	0.33	3.3	1.5	$2.5 \times 10^{-20}$	0.9	$1 \times 10^{-3}$	54	0.2	4	25
$\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$	0.25	1.6	2	$2.5 \times 10^{-20}$	0.9	$1.5 \times 10^{-3}$	8	0.17	5	20
$\text{Ba}_{0.72}\text{K}_{0.28}\text{Fe}_2\text{As}_2$ [35]	0.4	2.4	2.8	$1.5 \times 10^{-20}$	0.7	$1 \times 10^{-3}$	38	0.1(4)	3	23
$\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ [34]	0.5	2.2	4	$2.5 \pm 1.3 \times 10^{-20}$	$0.8 \pm 0.2$	$2 \times 10^{-3}$	43	0.2	8	10
$\text{Ba}_{0.45}\text{K}_{0.55}\text{Fe}_2\text{As}_2$	0.5	2.2	5.5	$1.5 \times 10^{-20}$	0.7	$2 \times 10^{-3}$	59	0.2	10	12
$\text{BaFe}_2(\text{As}_{0.67}\text{P}_{0.33})_2$	0.3 [20]	1.6	3.3	$< 1.5 \times 10^{-22}$	$< 0.1$	$< 1 \times 10^{-6}$	14	—	—	—

TABLE I: Fundamental parameters and contribution of dopant disorder to the elastic scattering parameters of various iron pnictide superconductors, as deduced from the collective pinning part of the critical current density,  $j_c^{coll}$ .



the collective pinning approach [21], and  $n_d D_v^3 \ll 1$ , which means that background scattering is irrelevant – each defect can be considered independent [22]. Moreover, an estimate of the Thomas-Fermi screening length,  $\sim 1$  nm, shows that charged defects are badly screened, so that Coulomb scattering is indeed important.

The obtained parameters mean that quasi-particle scattering by charged impurities in the iron-pnictide superconductors is to be situated rather in the Born limit ( $\sin \delta_0 \ll 1$ ). Such weakly scattering defects are thought to be detrimental to  $s_{\pm}$  superconductivity [6, 7, 13]. A crude estimate of the pair-breaking effect the dopant atoms would have can be made using the quasi-particle scattering rates  $\Gamma \sim n_d [\pi N_n(0)]^{-1} \sin^2 \delta_0$  (with  $N_n(0) = mk_F/\pi^2 \hbar^2$  the density of states and  $m$  the electronic mass) that can be extracted from experiment (see Table I). These turn out to be of the order  $\Gamma \sim 1.7T_c$  for NdFeAsO<sub>0.9</sub>F<sub>0.1</sub> and Ba<sub>0.78</sub>K<sub>0.22</sub>Fe<sub>2</sub>As<sub>2</sub>, and  $\Gamma \sim 4T_c$  for the other charge-doped compounds. When inserted in the Abrikosov-Gor'kov relation,  $\ln(T_c/T_{c0}) = \Psi(\frac{1}{2}) - \Psi(\frac{1}{2} + \Gamma/2\pi k_B T_c)$  (with  $\Psi$  the digamma function) [6, 7, 13, 41], this implies that, for  $s_{\pm}$ -superconductivity, the  $T_c$ 's of the charge-doped pnictides under investigation would be reduced by a factor 2-5 from a hypothetical  $T_{c0}$  in the absence of disorder. Moreover, superconductivity would be expected to become gapless at impurity densities much less than the actual dopant concentration [13]. Within the hypothesis of nodal extended  $s$ -wave superconductivity [7], the obtained scattering rates imply a  $T_c/T_{c0} \sim 0.5 - 0.7$ . Finally, for fully gapped, non-sign changing multiband  $s$ -wave superconductivity ( $s_{++}$ ), the weakly scattering dopant atoms or vacancies are not pair-breaking, and their effect would be the averaging of the gap components on different Fermi surface sheets.

A clearly different situation occurs in the isovalently doped BaFe<sub>2</sub>(As<sub>0.67</sub>P<sub>0.33</sub>)<sub>2</sub> material, which is apparently characterized by the *absence* of quasi-particle scattering. Isoelectronic dopant disorder is therefore benign to superconductivity with order parameter nodes, as this was observed by penetration depth measurements [19]. Furthermore, our analysis shows no clear distinction between scattering centers in the FeAs planes (such as Co), and out-of-plane defects, which attests to the clear three-dimensional nature of superconductivity in the low-field limit due to the contribution of the more dispersive hole-like sheets [25], centered on the  $\Gamma$ -point [42]. Table I shows that, if anything, the effect of out-of-plane defects [7] is stronger, because they act in the charge reservoir layer where superconductivity is weaker.

In conclusion, an analysis of vortex pinning in iron pnictide superconductors strongly suggests that charged atomic sized defects, including dopant atoms, are responsible for quasi-particle scattering in the Born limit, as well as for collective pinning. The presence of such defects in charge-doped pnictides casts doubt on the premise of  $s_{\pm}$  superconductivity in these materials. On

the other hand, isovalently doped BaFe<sub>2</sub>(As<sub>0.67</sub>P<sub>0.33</sub>)<sub>2</sub>, which has a superconducting ground state with gap nodes [19], is characterized by the absence of such quasi-particle scattering.

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