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Quasiparticle Scattering Induced by Charge Doping of Iron-Pnictide Superconductors Probed by Collective 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 $BaFe_2(As_{1-x}P_x)_2$ is consistent with the observation of a linear temperature dependence of the low-temperature penetration depth in this material.

With the advent of the superconducting iron pnictides [1–6] and chalcogenides, there are currently two classes of high temperature superconducting materials, the other being the cuprates. In both classes, superconductivity appears upon partial substitution of one or more elements of a magnetic parent material. Further substitution has the critical temperature T_c go through a maximum, and back to zero on the overdoped side of the temperature-composition phase diagram. Pnictides are specific in that this phenomenology may be induced either by charge doping or by isovalent substitutions. Known examples of the latter are the partial replacement of As by P [7,8], or Fe by Ru [9] in the $BaFe_2As_2$ "122" type materials, while charge doping is achieved by replacing O by F in the RBaFeO "1111" type materials (R is a rare earth element) [1–5], and Ba by K, or Fe by a transition metal ion in the 122's [6]. Introduction of either type of substitution causes important changes in band structure [8,10,11]; charge doping cannot be reduced to a rigid shift of the Fermi level in these multiband superconductors. Finally, dopant atoms act as scattering impurities, which in the weak scattering (Born) limit would couple quasiparticle excitations on different Fermi surface sheets, with possible repercussions [12–15] for the type of superconducting order parameter that may be realized [16–18], as well as for the diminishing T_c in the overdoped region of the phase diagram due to pair breaking [19].

In this Letter, we focus on the latter aspect of the problem, and argue that charged dopant atoms act as scattering impurities for quasiparticles, while isovalent substitutions do not. The approach used is that of pinning of the vortex lattice by the impurities. The dimension of the vortex cores, of the order of the coherence length $\xi \sim 2$ nm, implies a high sensitivity not only to extrinsic but also to intrinsic disorder in superconductors. Thus, in electron-doped $PrFeAsO_{1-y}$, $NdFeAsO_{1-x}F_x$, and $Ba(Fe_{1-x}Co_x)_2As_2$, as in hole-doped $Ba_{1-x}K_xFe_2As_2$, the critical current density j_c is consistently described in terms of collective pinning mediated by spatial fluctuations of the quasiparticle mean free path [20–22]. The impurity density accounting for pinning closely corresponds to the dopant atom concentration. Analysis of j_c allows one to estimate the scattering cross section and scattering phase angle δ_0 of the defects, which turns out to be best described by the Born limit. On the other hand, isovalently doped $BaFe_2(As_{1-x}P_x)_2$ is characterized by a monotonic power-law decrease of j_c as function of magnetic flux density B, indicative of pinning solely by nm-scale disorder [23].

Critical current densities of single crystalline $PrFeAsO_{0.9}$ (with $T_c \sim 35$ K) [23–26], $NdFeAsO_{0.9}F_{0.1}$ ($T_c \sim 36$ K) [23,27–29], $Ba_{0.45}K_{0.55}Fe_2As_2$ ($T_c \sim 34$ K) [30], and $BaFe_2(As_xP_{1-x})_2$ [31,32] were obtained from local measurements of the magnetic flux density perpendicular to the crystal surface $B_\perp$ and the flux density gradient $dB_\perp/dx \propto j_c$. Previous work has shown j_c of superconducting iron-pnictide crystals to be spatially inhomogeneous [23]. 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 of the flux density [23,33]. Measurements in fields up to 2 T were performed using micron-sized Hall probe arrays, tailored in a pseudomorphic GaAlAs/GaAs heterostructure [26]. The 10 Hall sensors of the array, spaced by 20 μm, had an active area of 3×3 μm^2, while an 11th sensor was used for the measurement of the applied field.

Figure 1 shows hysteresis cycles of the local "self-field" $B_s \equiv B_\perp - \mu_0 H_a$ (where $\mu_0 \equiv 4\pi \times 10^{-7}$ H m^{-1}) versus H_a for a variety of iron-pnictide superconductors, at the same reduced temperature $T/T_c = 0.2$. A salient
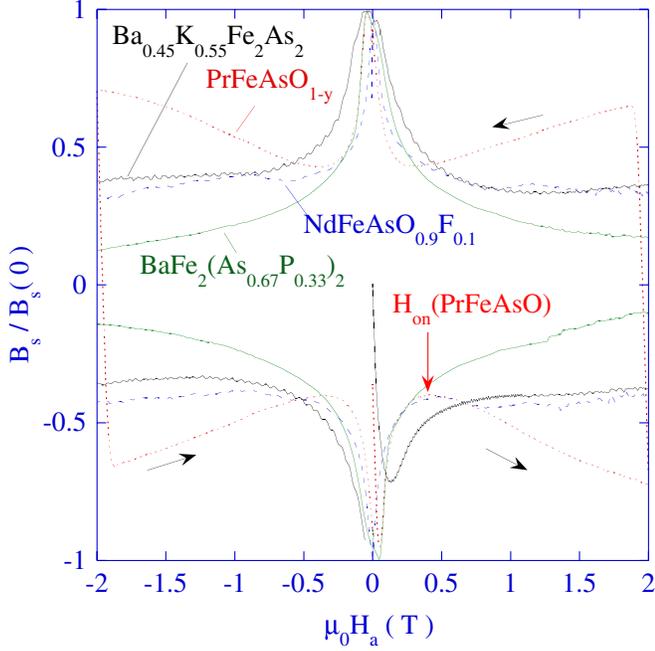
FIG. 1 (color online).   Normalized hysteresis loops of the local "self-field," measured on the center of the top surfaces of $PrFeAsO_{1-y}$, $NdFeAsO_{0.9}F_{0.1}$, $Ba_{0.45}K_{0.55}Fe_2As_2$, and $BaFe_2(As_{0.67}P_{0.33})_2$ single crystals, at reduced temperature $T/T_c = 0.3$. Arrows indicate the direction in which the cycles are traversed.

feature of the hysteresis loops is the presence of a pronounced peak at small field. In the "1111" family of iron-pnictide superconductors [23], as in the $Ba(Fe_{1-x}Co_x)_2As_2$ [34–36] and $Ba_{1-x}K_xFe_2As_2$ [37] "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 [23]. On the contrary, in isovalently doped $BaFe_2(As_{0.67}P_{0.33})_2$, the hysteresis loop width shows a monotonic decrease.

Figure 2 shows the field dependence of j_c 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 [38], while data for $Ba(Fe_{0.9}Co_{0.1})_2As_2$ and $Ba_{0.6}K_{0.4}Fe_2As_2$ are taken from Refs. [35,37], respectively. In what follows, we describe j_c as the superposition of two contributions, j_c^{coll} and $j_c^s(B)$. The former accounts for the constant hysteresis width at higher fields, and the latter for the low-field peak. In all materials, the peak has the shape of a plateau,

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

followed by a power-law decrease, such that

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

with $0.5 < \beta < 0.63$. The behavior of $j_c^s(B)$ is that expected for vortex pinning by sparse pointlike defects of radius larger than ξ, which are inevitably present in any

real, imperfect crystal [39–41]. An analysis of data on the $RFeAsO$ 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 [23]. Oppositely, the field-independent j_c^{coll} is attributed to atomic scale fluctuations of the dopant atom positions (collective pinning) [20]. The different field dependence of the critical current contributions (Fig. 2) allows one to extract both as a function of temperature. Figure 3(a) shows the T dependence of the field-independent contribution j_c^{coll}, nonzero in the charge-doped compounds, but absent for all investigated isovalent substitutions x in $BaFe_2(As_{1-x}P_x)_2$.

We quantitatively describe j_c^{coll} by treating the dopant atoms as point defects responsible for quasiparticle scattering. The elementary pinning force of such defects can be written

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

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 wave vector, D_v is the effective range of the potential, and $g(\rho_D)$ is the Gor'kov function [20–22]. 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 quasiparticle mean-free path, n_d the defect density, and $\xi_0 \approx 1.35\xi(0)$ the (temperature-independent) Bardeen-Cooper-Schrieffer coherence length [20–22]. 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 [20,22], it reads [23]
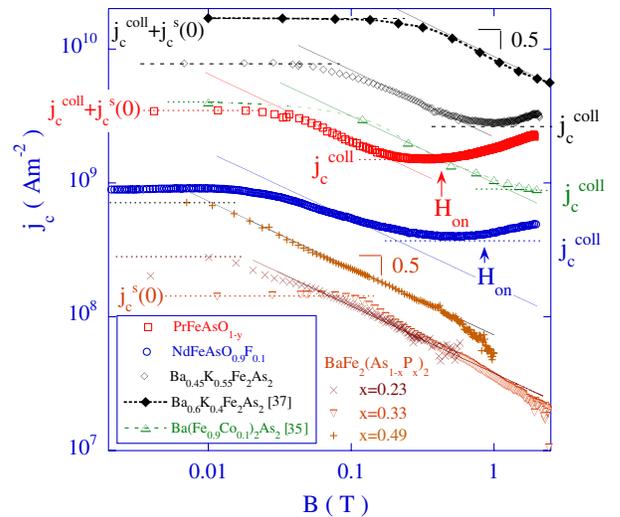

FIG. 2 (color online).   Critical current density as function of magnetic flux density for $PrFeAsO_{1-y}$ ($\square$), $NdFeAsO_{0.9}F_{0.1}$ ($\bigcirc$), $Ba(Fe_{0.9}Co_{0.1})_2As_2$ ($\triangle$) [35], $Ba_{1-x}K_xFe_2As_2$ ($\blacklozenge$, $\lozenge$), and $BaFe_2(As_{1-x}P_x)_2$ single crystals of different x, at $T/T_c = 0.3$. Drawn lines indicate the power-law dependence $AB_\perp^{-\beta}$ of the pinning contribution from sparse pointlike defects.
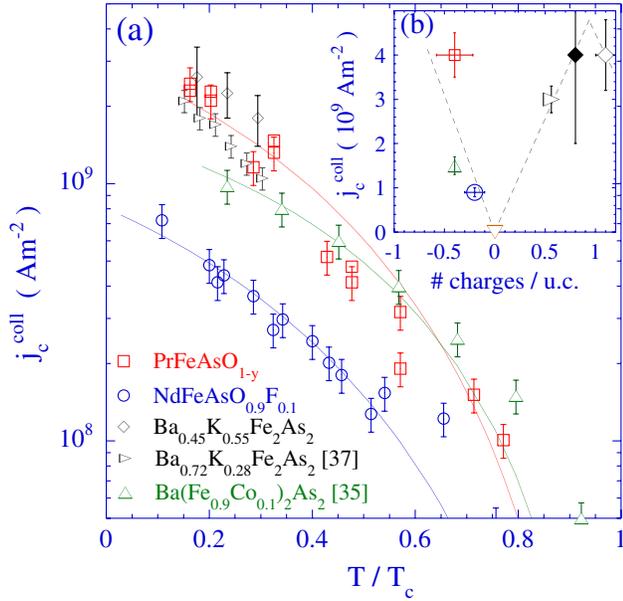
FIG. 3 (color online). (a) Collective pinning contribution j_c^{coll} in the charge-doped compounds PrFeAsO_{1-y}, $\text{NdFeAsO}_{0.9}\text{F}_{0.1}$, $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$ [35], and $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ single crystals. Drawn lines are fits to Eq. (4). (b) The value of j_c^{coll}, extrapolated to $T/T_c = 0.1$, as function of the number of dopant charges per unit cell. $j_c^{\text{coll}} = 0$ for $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ ($\triangledown$). A point for $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ [37] has been added [$\blacklozenge$, not shown in Fig. 3(a)].

$$j_c^{\text{coll}} \approx j_0 \left[\frac{0.01 n_d \sigma_{\text{tr}}^2}{\varepsilon_\lambda \xi} \left(\frac{\xi_0}{\xi}\right)^2 \right]^{2/3} \propto \left[\frac{\lambda(0)}{\lambda(T)}\right]^2 \left(1 - \frac{T}{T_c}\right)^\alpha, \quad (4)$$

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. Equation (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 [26,28], while $\lambda(0)/\lambda(T)$ was published in Refs. [25,26,28].

Figure 3(a) shows that the temperature dependence of j_c^{coll} is very well described by Eq. (4). In PrFeAsO_{1-y},

its magnitude is accurately reproduced by inserting $\sigma_{\text{tr}} = \pi D_v^2$, with the oxygen ion radius $D_v = 1.46$ Å and $n_d \approx 1.5 \times 10^{27}$ m^{-3}. This corresponds to the oxygen vacancy concentration at the doping level, $y \sim 0.1$. Thus, the collective pinning contribution to the critical current density of the PrFeAsO_{1-y} compound is well described by the quasiparticle mean-free path fluctuation mechanism of Refs. [20–22]. 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 4×10^{27} m^{-3}, respectively, very satisfactory fits to $j_c^{\text{coll}}(T)$ can be 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 consider only 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 the collective pinning approach [20], and $n_d D_v^3 \ll 1$, which means that background scattering is irrelevant—each defect can be considered independent [21].

The correlation between the collective pinning contribution j_c^{coll} to the critical current density and the nominal number of dopant charges per unit cell is shown in Fig. 3(b). Since there are two formula units per unit cell, the number of dopant charges is defined as twice the product of the dopant valency and the doping fraction x. In $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$, j_c^{coll} is unmeasurably small, implying a qualitative difference between charge-doped and isovalently substituted iron-pnictide superconductors.

The collective pinning effect implies that the core charge on the doping impurities is incompletely screened, consistent with a Thomas-Fermi screening length, ~ 1 nm. From Table I, one sees that scattering by charged dopants in the iron-pnictide superconductors is rather in the Born limit ($\sin\delta_0 \ll 1$). Therefore, if so-called $s_\pm$ superconductivity [16–19], with a sign change of the order parameter on different Fermi surface sheets, is realized in these materials, the scattering would be detrimental [12,13,15]. A crude assessment of the pair-breaking effect can be made using the quasiparticle 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] estimated from experiment (see Table I).

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}.

Compound	k_F (Å^{-1})	ξ_0 (nm)	n_d (nm^{-3})	σ_{tr} (Å^2)	D_v (Å)	$n_d D_v^3$	$n_d \xi_0^3$	$\sin\delta_0$	Γ (meV)	l (nm)
PrFeAsO_{1-y}	0.33	2.4	1.5	6.7	1.46	5×10^{-3}	21	0.3(2)	10	10
$\text{NdFeAsO}_{0.9}\text{F}_{0.1}$	0.33	3.3	1.5	2.5	0.9	1×10^{-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	0.9	1.5×10^{-3}	8	0.17	5	20
$\text{Ba}_{0.72}\text{K}_{0.28}\text{Fe}_2\text{As}_2$ [42]	0.4	2.4	2.8	1.5	0.7	1×10^{-3}	38	0.1(4)	3	23
$\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ [37]	0.5	2.2	4	2.5 ± 1.3	0.8 ± 0.2	2×10^{-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	0.7	2×10^{-3}	59	0.2	10	12
$\text{BaFe}_2(\text{As}_{0.67}\text{P}_{0.33})_2$	0.3 [8]	1.6	3.3	$<1.5 \times 10^{-2}$	<0.1	$<1 \times 10^{-6}$	14	$\cdots$	$\cdots$	$\cdots$

These turn out to be of the order $\Gamma \sim 1.7T_c$ for NdFeAsO$_{0.9}$F$_{0.1}$ and Ba$_{0.78}$K$_{0.22}$Fe$_2$As$_2$, 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 Ψ the digamma function) [43], this implies that the T_c's of the charge-doped pnictides would be reduced by a factor 2–5 from a hypothetical T_{c0} in the absence of disorder. Moreover, superconductivity should become gapless at impurity densities much less than the actual dopant concentration [15]. Within the hypothesis of nodal extended s-wave superconductivity [13], the obtained scattering rates imply a $T_c/T_{c0} \sim 0.5$–0.7. Finally, for fully gapped, nonsign changing multiband s-wave superconductivity (''s_{++}''), the dopant atoms or vacancies are not pair breaking, and their effect is the averaging of the gap components on different Fermi surface sheets. A different situation occurs in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ material, which is characterized by the *absence* of quasiparticle scattering. Isoelectronic dopant disorder is benign to superconductivity with order parameter nodes, as this was observed by penetration depth measurements [32]. 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 three-dimensional nature of superconductivity in the low-field limit due to the contribution of the more dispersive holelike sheets [26], centered on the Γ point [44].

In conclusion, it is shown that the analysis of collective vortex pinning provides clues as to microscopic scattering mechanisms in superconductors. In that, the analysis of the critical current density adds another transport property to the spectrum of techniques available for the quantification of disorder effects in superconductors. Applied to iron-pnictide superconductors, we find strong indications that charged atomic sized defects, including dopant atoms, are responsible for quasiparticle scattering in the Born limit. The presence of such defects in charge-doped pnictides should have consequences for suggested $s_{\pm}$ superconductivity in these materials. On the other hand, isovalently doped BaFe$_2$(As$_{1-x}$P$_x$)$_2$, which has a superconducting ground state with gap nodes [32], is characterized by the absence of such quasiparticle scattering.

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