

# Phase transition beneath the superconducting dome in $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$

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We present a theory for the large suppression of the superfluid-density,  $\rho_s$ , in  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  in the vicinity of a putative spin-density wave quantum critical point at a P-doping,  $x = x_c$ . We argue that the transition becomes weakly first-order in the vicinity of  $x_c$ , and disorder induces puddles of superconducting and antiferromagnetic regions at short length-scales; thus the system becomes an electronic micro-emulsion. We propose that frustrated Josephson couplings between the superconducting grains suppress  $\rho_s$ . In addition, the presence of ‘normal’ quasiparticles at the interface of the frustrated Josephson junctions resolves some seemingly contradictory observations between the metallic and the superconducting phases. We propose experiments to test our theory.

*Introduction.* - An important focus of the study of high temperature superconductivity (SC) has been on the role of antiferromagnetism (AFM) and its relation to SC [1]. There is clear evidence across many different families of compounds that SC appears in close proximity to an AFM phase [2]; these families include the iron-pnictides, the electron-doped cuprates and the heavy-fermion superconductors. Moreover, the optimal transition temperature ( $T_c$ ) of the SC is often situated where the normal state AFM quantum critical point (QCP) would have been located, in the absence of superconductivity. The experimental detection of the QCP is often challenging in the normal state, and more so in the superconducting state.

Recently, a number of measurements were reported in a member of the pnictide family,  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ , as a function of the isovalent P-doping,  $x$ . The experiments show a phase transition involving onset of spin-density wave (SDW) order in the normal state above  $T_c$ , which extrapolates to a  $T = 0$  SDW QCP (see [3] and references therein). These experiments include: (i) a sharp enhancement in the effective mass,  $m^*$ , upon approaching a critical doping from the overdoped side, as obtained from de Haas-van Alphen oscillations [4] and from the jump in the specific-heat at  $T_c$  [5], and, (ii) a vanishing Curie-Weiss temperature ( $\theta_{CW}$ ), extracted from the  $1/T_1T$  measurements using NMR.

As we will review below, a number of puzzling results have appeared from experiments investigating whether the SDW QCP actually survives “under the SC dome.” Here we propose a resolution of these puzzles by postulating a weakly first-order transition [6] for the onset of SDW order in the presence of SC order (see Fig. 1a). It is well known that ‘random bond’ disorder has a strong effect on symmetry-breaking first-order transitions [7], and ultimately replaces it with a disorder-induced second order transition in two dimensional systems. Our main claim is that the inhomogeneities associated with these highly relevant effects of disorder can resolve the experimental puzzles.

The fate of the normal state transition within the SC state was investigated by measurements [8] of the zero temperature London penetration depth,  $\lambda_L(0) \propto 1/\sqrt{\rho_s}$  ( $\rho_s \equiv$  superfluid-

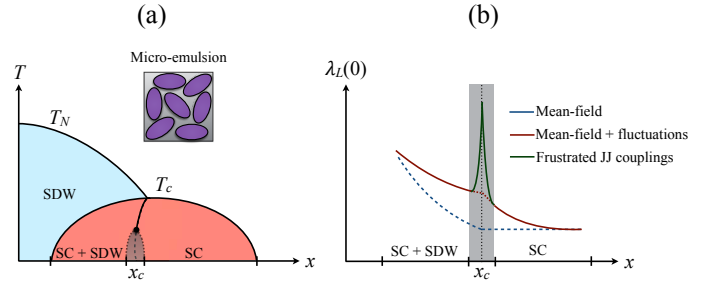


FIG. 1. (a) A cartoon phase-diagram showing the interplay between SDW and SC phases. The  $T_N$  line may survive as a continuous transition inside the SC phase up until the point marked ‘•’, below which it becomes weakly first-order (shown as black dashed line). The grey region depicts the regime where the system develops spatial inhomogeneity due to the presence of disorder. Inset: Emulsion with SC grains (purple) and SDW(+SC) regions (grey). We ignore the presence of other symmetry-broken phases in the diagram. (b) The behavior of  $\lambda_L(0)$  as a function of the tuning parameter,  $x$ , within different scenarios (see text for details).

density), as a function of  $x$ . A sharp peak in  $\lambda_L(0)$ , corresponding to a nearly ten-fold suppression in  $\rho_s$ , was observed at  $x = x_c$ , strongly indicating a QCP within the SC state [9]. However, it has been subsequently argued [10] that this peak cannot be due to a second-order quantum phase transition associated with the onset of SDW order in the presence of a superconductor with gapped quasiparticle excitations [11, 12]. Indeed, general arguments [10] establish that a large class of such second-order transitions actually have a *monotonic* variation in  $\lambda_L(0)$  across the QCP (see dashed-blue/solid-red curves in Fig. 1b). In contrast, the most striking feature observed is the sharp decrease in  $\lambda_L(0)$  on the ordered side of the transition, where large portions of the Fermi-surface are lost due to reconstruction from long-range SDW.

A related, and equally surprising, experimental observation in this material is the apparent violation of optical sum-rules. A remarkable, but straightforward, consequence of these sum-rules is that  $\lambda_L(0)$  is related to the normal state

d.c. conductivity,  $\sigma_{dc}$ , just above  $T_c$ . When a conventional metal with a Drude-like conductivity becomes superconducting without any sub-gap excitations (*i.e.* the density of states,  $N(E) \sim E/\sqrt{E^2 - \Delta^2}$  for  $E > \Delta$  and 0 otherwise), all of the low energy spectral weight goes into the superfluid density at  $\omega = 0$ , which manifests itself as a delta-function peak. From conservation of spectral weight,

$$\rho_s \sim \sigma_{dc}\Delta \sim \sigma_{dc}T_c, \quad (1)$$

where we have assumed that  $\Delta$  scales with  $T_c$ . The last relation, often referred to as Homes' law, has been verified to be true across many different superconducting materials, including the cuprates [13]. The above relation also implies that if  $\rho_s$  behaves non-monotonically as a function of  $x$ , then  $\sigma_{dc}$  should have a similar dependence on  $x$ , as long as the variation in  $T_c$  is small. In the case of  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ , where the variation in  $T_c$  is negligible around optimal doping, while  $\rho_s$  behaves highly non-monotonically across  $x_c$ , transport experiments have not seen any evidence of a non-monotonic variation in  $\sigma_{dc}$  [14]. Moreover, recent measurements of optical conductivity [15] in near optimal  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  suggest that the normal state spectral weight below the gap frequency is consistent with the small penetration depth measured for samples with  $x$  far from the critical value, rather than the much larger  $\lambda_L(0)$  observed near  $x = x_c$ .

In order for the above observations to be consistent with optical sum-rules, the uncondensed part of the spectral weight is transferred either (*i*) to very high frequencies (as is known to happen for  $c$ -axis optical conductivity in the underdoped cuprates [16]), or, (*ii*) to very low frequencies (but not to  $\rho_s$ ), which goes undetected in optical conductivity experiments. The latter possibility is plausible within the scenario that we develop here.

We analyze the above experiments by assuming a weakly first-order transition [6], and argue that the presence of quenched disorder leads to formation of a *micro-emulsion* at small scales [7]. The system consists of SC puddles, where some of the puddles additionally have SDW order (see Fig. 1a inset). The SDW(+SC) regions, which have a locally well-developed antiferromagnetic moment but no long-range orientational order, act as barriers between the different SC grains. Upon moving deeper into the ordered side of the transition, the SDW(+SC) regions start to percolate and crossover to a state with long-range SDW order; this is the regime with a microscopically coexistent SC+SDW. As a function of decreasing  $x$ , the micro-emulsion is therefore a transitional state (shown as grey region in Fig. 1a) between a pure SC and a coexistent SC+SDW. We note that the granular nature of superconductivity should have no effect on the bulk  $T_c$  in the presence of percolating SC channels.

*Model.* - When the system is well described in the vicinity of  $x_c$  by a micro-emulsion as explained above, the phase fluctuations associated with the SC grains (shown as purple regions in Fig. 1a inset), can be modeled by the following effective

theory,

$$H_\theta = - \sum_{a,b} J_{ab} \cos(\theta_a - \theta_b), \quad (2)$$

where  $J_{ab}$  represent the Josephson junction (JJ) couplings between grains 'a' and 'b'. We have ignored the capacitive contributions.

The Josephson current across the junction will be given by  $I_s = J_{ab} \sin(\theta_a - \theta_b)$ , and  $J_{ab}$  may therefore be interpreted as the lattice version of the local superfluid density,  $\rho_s(\mathbf{r})$ , *i.e.*  $\mathbf{J}_s(\mathbf{r}) = \rho_s(\mathbf{r}) \mathbf{v}_s(\mathbf{r})$ , with  $\mathbf{J}_s(\mathbf{r})$ ,  $\mathbf{v}_s(\mathbf{r})$  representing the superfluid-current and velocity respectively. Having a frustrated JJ (also known as a  $\pi$ -junction) with a negative value of  $J_{ab}$  leads to a local suppression in  $\rho_s$ . Similar ideas have been discussed in the past in a variety of contexts (see Refs. [17] for a specific example), though the mechanism considered here will be different. We shall now propose an explicit scenario under which a suppression in  $\rho_s$  arises in the vicinity of putative magnetic QCPs, utilizing the SC gap structure in the material under question.

The basic idea is as follows: suppose that the tunneling of electrons between the two grains is mediated by the SDW moment in the intervening region [18], and is accompanied by a transfer of finite momentum that scatters them from a hole-like to an electron-like pocket. Because the SC gaps on the two pockets have a relative phase-difference of  $\pi$ , the JJ coupling will be frustrated [19].

Let us first focus on a single grain. In order to capture the multi-band nature of the SCs, we introduce two superconducting order parameters,  $\Delta_i$  with  $i = \pm$  to model the  $s^\pm$  state on the two pockets. Microscopically, these belong to regions in the grain having different momenta,  $\mathbf{k}_\parallel$ , parallel to the junction. The gaps are related to the microscopic degrees of freedom [20] via the following relation,

$$\Delta_i(z) = \frac{1}{A} \sum_{\mathbf{k}_\parallel \in \mathcal{R}_i} V_{\mathbf{k}_\parallel, \mathbf{k}'_\parallel} \langle \psi_{\mathbf{k}'_\parallel \uparrow} \psi_{-\mathbf{k}_\parallel \downarrow} \rangle, \quad (3)$$

where  $\psi_{\mathbf{k}_\parallel \sigma}^\dagger$  creates an electron at position  $z$  with momentum  $\mathbf{k}_\parallel$  parallel to the junction and spin  $\sigma$ .  $V_{\mathbf{k}_\parallel, \mathbf{k}'_\parallel}$  is the pairing interaction in the Cooper channel and  $z$  is the coordinate perpendicular to the junction with area  $A$ . The regions  $\mathcal{R}_i$  are defined as,  $\mathcal{R}_+ = \{\mathbf{k}_\parallel | k_0 > |\mathbf{k}_\parallel|\}$  and  $\mathcal{R}_- = \{\mathbf{k}_\parallel | k_0 \leq |\mathbf{k}_\parallel|\}$ , where  $k_0$  is an arbitrary momentum scale chosen such that  $\Delta_+ > 0$ ,  $\Delta_- < 0$  (see Fig. 2 for an illustration). We'll assume that such a prescription is valid for each grain, with possibly different values of  $k_0$ .

Let us then write down a model for the two coupled SC grains with an intervening proximity coupled SDW that has a well developed moment,  $\mathbf{n}$ . Our notation is as follows: we use  $\alpha = a, b$  to denote the grain index and  $i = \pm$  to denote the band index within each grain. From now on, we relabel  $\mathbf{k}_\parallel$  as  $\mathbf{k}$ . We introduce the Nambu spinor,  $\Psi_{i, \mathbf{k}, \sigma}^{\alpha \dagger} = (\psi_{i, \mathbf{k}, \sigma}^{\dagger \alpha} \epsilon_{\sigma \sigma'} \psi_{i, -\mathbf{k}, \sigma'}^\alpha)$ , where now  $\psi_{i, \mathbf{k}, \sigma}^{\dagger \alpha}$  creates an electron with momentum  $\mathbf{k}$  parallel to the junction and at a position  $z$  (label suppressed), which

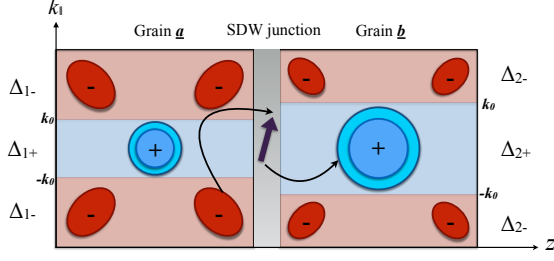


FIG. 2. A cartoon of a frustrated  $\pi$ -junction between two superconducting grains with a SDW(+SC) barrier. The SDW moment imparts a finite momentum transfer along the direction of the interface while scattering electrons from the electron (hole) pocket on one grain to the hole (electron) pocket on the other grain.

belongs to a region of band “ $r$ ” within grain “ $\alpha$ ”. The effective Hamiltonian is given by,

$$H_{\text{eff}} = H_{\Delta} + H_T, \quad (4)$$

$$H_{\Delta} = \sum_{\alpha, i, \mathbf{k}} \Psi_{i, \mathbf{k}, \sigma}^{\alpha \dagger} \left[ \varepsilon_{i\alpha, \mathbf{k}} \hat{\tau}^z + \Delta_{i\alpha, \mathbf{k}} \hat{\tau}^x \right] \Psi_{i, \mathbf{k}, \sigma}^{\alpha}, \quad (5)$$

$$H_T = g \sum_{\mathbf{k}} \mathbf{n} \cdot \left( \Psi_{+, \mathbf{k}, \sigma}^{\alpha \dagger} [\sigma_{\sigma\sigma'} \otimes \hat{\tau}^0] \Psi_{-, \mathbf{k}, \sigma'}^{\beta} + \Psi_{-, \mathbf{k}, \sigma}^{\alpha \dagger} [\sigma_{\sigma\sigma'} \otimes \hat{\tau}^0] \Psi_{+, \mathbf{k}, \sigma'}^{\beta} \right) + \text{H.c.}, \quad (6)$$

where  $g$  is the tunneling matrix element,  $\hat{\tau}^i$  ( $i = 0, x, y, z$ ) act in Nambu space and  $\hat{\sigma}^i$  ( $i = 0, x, y, z$ ) act in spin space.

In the above,  $H_{\Delta}$  corresponds to the bare pairing Hamiltonian written for the  $\pm$  bands within each of the two grains.  $H_T$  represents the SDW moment mediated hopping of electrons from one grain to the other (represented by the  $a, b$  superscripts) and simultaneously scattering from one band to the other (represented by the  $\pm$  subscripts). Therefore,  $\mathbf{n}$  imparts a finite momentum (along the interface) to the electrons when it scatters them from the electron (hole) pocket on one grain to the hole (electron) pocket on the other grain (shown as the black arrows in Fig. 2).

*Results.*- Using the Ambegaokar-Baratoff relation [19], we can write the Josephson coupling (at  $T = 0$ ) between the two grains as,

$$J_{ab} = \frac{g^2 \langle \mathbf{n}^2 \rangle}{\pi^2} \left[ \sum_{\ell \in a, \ell' \in b} \Delta_{\ell} \Delta_{\ell'} \int_0^{\infty} \frac{d\varepsilon_{\ell}}{E_{\ell}} \int_0^{\infty} \frac{d\varepsilon_{\ell'}}{E_{\ell'}} \frac{1}{E_{\ell} + E_{\ell'}} \right] \quad (7)$$

where  $E_{\ell}^2 = \varepsilon_{\ell}^2 + \Delta_{\ell}^2$  and  $\ell, \ell'$  represent the band indices on the different grains. Since  $\Delta_{\ell} \Delta_{\ell'} < 0$ , the coupling  $J_{ab} < 0$ . Note that the specific nature of the frustrated tunneling arises from the same spin-fluctuation mediated mechanism that is predominantly responsible for the  $s^{\pm}$ -pairing symmetry [2]. However, there will also be a direct tunneling term (not included in Eqn. 4) in the Hamiltonian, which does not scatter the electrons from one pocket to the other, as they hop across the junction. The contribution to the JJ coupling from this term will be unfrustrated (i.e.  $J_{ab} > 0$ ).

The ratio of the tunneling amplitudes in the two different channels is non-universal and depends on various microscopic details. In particular, the emulsion is associated with a distribution of Josephson-couplings,  $\mathcal{P}(J)$ , with a mean coupling strength,  $\langle J \rangle = \bar{J}$ . If a substantial fraction of the JJ couplings become negative due to the mechanism proposed above,  $\bar{J}$  will be small, and the superfluid density will be suppressed (see green curve in Fig. 1b).

We now turn to a resolution of the apparent sum rule violation in the optical conductivity. Frustrated  $\pi$ -junctions host gapless states at the interface between the two grains [21, 22], giving rise to a finite density of states around zero energy (see Fig 3 inset). As a result of the gapless ‘normal’-fluid component at the interface, a fraction  $f$  of the spectral weight will be displaced from the superfluid-density to non-zero frequencies (shaded region in Fig. 3). For  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ , it appears that  $f \sim 0.9$  in the vicinity of the putative QCP, in order for there to be a ten-fold suppression in  $\rho_s$ . The weight of the condensate,  $\rho_s$ , is then proportional to  $\bar{J}(1 - f)$ .

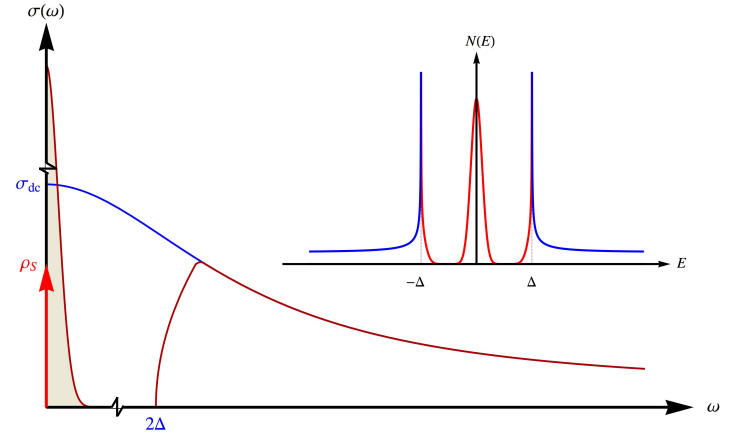


FIG. 3. Plot of the optical conductivity,  $\sigma(\omega)$ , for a Drude metal (blue curve) and a superconductor with a large number of sub-gap excitations (red curve). The shaded region corresponds to the displaced spectral weight from the superfluid-density,  $\rho_s$  (red arrow). Inset: Density of states,  $N(E)$  vs.  $E$ , for a conventional gapped superconductor (blue curve). A superconductor with a large number of sub-gap states has a finite  $N(E)$  below  $\Delta$  (red curve).

It is clear from our proposed form of the optical conductivity,  $\sigma(\omega)$ , in Fig 3 that Homes’ relation in Eq. 1 will not be satisfied. In particular,  $\sigma_{\text{dc}}$  (which is a property of the normal state), could continue to behave monotonically as a function of the isovalent-doping across  $x_c$ , while the abundance of low-energy excitations in the immediate vicinity of  $x_c$  would give rise to a non-monotonic variation in the superfluid density. This allows for an unusual way of rearranging spectral weight in the *superconducting* state below the gap, without violating optical sum-rules. Moreover, since approximately 90% of the sub-gap spectral weight in the vicinity of  $x_c$  needs to remain uncondensed,  $\sigma_0 = \lim_{\omega \rightarrow 0} \sigma(\omega)$  will be huge. Reflectivity measurements in the sub-gap frequency range cannot

distinguish this narrow conductivity peak centered on  $\omega = 0$  from a true delta-function.

The above scenario will give rise to a number of interesting low temperature thermodynamic and transport properties, as we now discuss. First of all, there should be a striking enhancement in the low-temperature thermal conductivity and specific-heat, as a function of  $x$  in the narrow vicinity of  $x_c$ , due to the ‘normal’-component. It is important to recall that this material has loop-like nodes on the electron-pockets [12]. However, the geometry of the electron-pockets and the magnitude of the gap do not change substantially in the vicinity of  $x_c$ , and therefore it is unlikely that the contribution to the above quantities from the nodal-quasiparticles will have a drastic modification. It should therefore be relatively straightforward to disentangle the contribution arising from the nodal versus the ‘normal’ quasiparticles. Studying the NMR-spectra as a function of decreasing temperature (across  $T_c$ ) and down to sufficiently low temperatures in the vicinity of  $x_c$  should also reveal the spatial inhomogeneity associated with the SDW regions. Finally, we note that a promising direction for future studies would be to measure the magnetic-field distribution due to the propagating currents in the emulsion using NV-based magnetometers [23].

*Discussion.*- The theoretical study in this paper is motivated by a number of remarkable experiments carried out in  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ , as a function of  $x$  in the normal and superconducting phases. Our primary objectives were two-fold — to provide an explanation for the striking enhancement of the London penetration depth in the vicinity of a putative SDW QCP in the SC state, and, to resolve a number of apparent discrepancies between properties of the metallic normal state, just above  $T_c$ , and the low-temperature properties of the SC. We stress that neither of these two experimental observations can be explained by invoking the critical fluctuations associated with a symmetry-broken order-parameter in a superconductor.

It is quite likely that the true SDW criticality is masked by a weak first-order phase transition in the superconducting state at  $T = 0$ . Quenched disorder would then naturally give rise to an *emulsion* at small length scales with puddles of SC and SDW(+SC). It is then, in principle, possible for the SDW moments at the interface of the SC grains to generate frustrated Josephson couplings, which would in turn deplete the local superfluid-density. The ten-fold suppression in the superfluid density requires a sufficiently large fraction of the Josephson couplings in the emulsion to become negative. Furthermore, the interface of these frustrated junctions host gapless states, which would contribute to a finite density of states around zero energy and help explain the dichotomy between the normal-state d.c. conductivity and the low temperature penetration depth.

Our proposed scenario naturally calls for a number of experimental tests that should be carried out in the near future, which should directly look for both the spatial inhomogeneities associated with the emulsion, and probe the gapless excitations using thermodynamic probes, as explained above.

In addition it will be interesting to verify that the optical sum rule is satisfied in a conventional way leading to Homes’ law away from the narrow vicinity of  $x_c$ .

In the superconducting state of the electron-doped material,  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ ,  $\lambda_L(0)$  behaves monotonically as a function of  $x$  across the putative QCP [24]. Electron-doping leads to significantly higher amounts of disorder compared to the isovalently-doped case, and would therefore lead to puddles with typically much smaller size. Our proposed mechanism for the strong suppression of the superfluid-density in the isovalently-doped material relies on the existence of an emulsion with puddles of appreciable size, in the presence of an optimal amount of disorder. A comparison of the NMR spectra in the narrow vicinity of the putative QCP in the electron and isovalently doped materials would shed light on these microscopic differences between the two families.

Finally, though we have hypothesized that the SDW onset transition *inside* the SC is, in the absence of disorder, a weak first order transition, we emphasize that the normal state properties are consistent with the presence of a “hidden” QCP around optimal doping [4, 5, 25]. It is plausible that in the normal state, different experimental techniques are probing the critical fluctuations associated with not one, but distinct QCPs as a function of  $x$ . For instance,  $m^*$  extracted from high-field quantum oscillations is dominated by the vicinity of ‘hot-spots’, where quasiparticles are strongly damped due to coupling to the SDW fluctuations [26]. On the other hand, strong critical fluctuations associated with the nematic order-parameter [27], that couple to the entire Fermi-surface, would dominate  $m^*$  extracted at zero-field from the jump in the specific heat at  $T_c$ .

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