Evolution of the Fermi surface of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ on entering the superconducting dome

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Using the de Haas-van Alphen effect we have measured the evolution of the Fermi surface of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ as function of isoelectric substitution (As/P) for $0.41 < x < 1$ ($T_c$ up to 25 K). We find that the volume of electron and hole Fermi surfaces shrink linearly with decreasing $x$. This shrinking is accompanied by a strong increase in the quasiparticle effective mass as $x$ is tuned toward the maximum $T_c$. It is likely that these trends originate from the many-body interaction which gives rise to superconductivity, rather than the underlying one-electron bandstructure.

Superconductivity in the 122 iron-pnictide family XFe$_2$As$_2$ (where X=Ba,Sr or Ca), can be induced by a variety of means, including doping [1,2], pressure [3] or isoelectric substitution either on the Fe [4] or As sites [5]. The highest $T_c$ achievable by each of these routes is roughly the same. It has been suggested [6,7,8] that the interband coupling between the hole and electron sheets plays an important role in determining the magnetic or superconducting order formed at low temperature. Discovering how the Fermi surface evolves as a function of the various material parameters which drive the material from an antiferromagnetic spin density wave state, through the superconducting dome and eventually towards a paramagnetic non-superconducting metal, should therefore be an important step toward gaining a complete understanding of the mechanism that drives high temperature superconductivity in these materials.

In the antiferromagnetic state it is expected that the Fermi surface suffers a major reconstruction. This is supported experimentally by the observation [4,10,11] of very low frequency quantum oscillations in the undoped XFe$_2$As$_2$ compounds, corresponding to very small Fermi surface pockets which are 1-2% of the total Brillouin zone planar area. At the other extreme of the phase diagram, where the materials are paramagnetic and non-superconducting, quantum oscillation measurements of SrFe$_2$P$_2$ [12] and CaFe$_2$P$_2$ [13] show that the Fermi surface is in good agreement with conventional bandstructure calculations. Up to now, tracking the changes in the Fermi surface across the phase diagram using quantum oscillations has not been possible because of the additional disorder and high $H_{c2}$ introduced by doping.

Measurements on the low $T_c$ (6 K) superconducting iron-pnictide LaFePO [14,15] established that the Fermi surface is in broad agreement with bandstructure with moderate correlation enhancements of the effective mass. It is not clear whether the higher $T_c$ pnictide superconductors, which unlike LaFePO occur in close proximity to a magnetic phase, are also well described by bandstructure and whether the electronic correlations change significantly.

The substitution of P for As in the series BaFe$_2$(As$_{1-x}$P$_x$)$_2$ offers an elegant way to suppress magnetism and induce superconductivity without doping [10]. As P and As are isoelectric, there is no net change in the ratio of electrons to holes and the system remains a compensated metal for different $x$ (equal volumes for the electron and hole Fermi surfaces). This series has several remarkable properties which are similar to those observed in cuprate superconductors. Firstly, the temperature dependence of the resistivity changes from a quadratic ($\rho \sim T^2$) to linear behavior ($\rho \sim T$) as the system evolves from a conventional Fermi liquid ($x = 1$) towards the maximum $T_c$ ($x = 0.33$). Secondly, there is strong evidence from magnetic penetration depth [17], thermal conductivity [17] and NMR [18] measurements that for $x = 0.33$ the superconducting gap has line nodes.

In this Letter, we report the observation of quantum oscillation signals in samples of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ as $x$ is varied across the superconducting dome from $x = 1$ to $x = 0.41$ with $T_c \sim 25$ K ($\sim 0.8 T_{c\text{max}}$). Our data show that the Fermi surface shrinks and the quasiparticle masses become heavier as the material is tuned toward the magnetic order phase boundary, at which $T_c$ reaches its maximum. This implies that the significant change of electronic structure caused by many-body interactions plays a key role for the occurrence of unconventional high-$T_c$ superconductivity.

Single crystal samples of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ were grown as described in Ref. [10]. The $x$ values were determined by an energy dispersive X-ray analyzer. Measurements of
the quantum oscillations in magnetic torque [the de Haas-van Alphen (dHvA) effect] were made using a miniature piezo-resistive cantilever technique. Experiments were performed in: a dilution refrigerator system with DC fields up to 17 T (Osaka), a pumped $^3$He system with DC fields up to 30 T (Nijmegen) and 45 T (Tallahassee) and a pumped $^4$He system with pulsed fields up to 55 T (Toulouse). Our data are compared to band structure calculations which were performed using the wien2k package [19].

Fig. 1(a) shows raw magnetotorque $\tau(H)$ data for three values of $x$ measured up to 55 T at $T \approx 1.5$ K, with the magnetic field direction close to the c-axis. The torque response for the higher $T_c$ samples ($x = 0.33$, $T_c \approx 30$ K and $x = 0.41$, $T_c \approx 25$ K) is highly hysteretic. $\mu_0 H_{irr}$ increased substantially with $x$ reaching a maximum of 51.5 T for the highest $T_c$ sample ($x = 0.33$), which is close to the estimated $H_{c2}$ value [17]. By subtracting a smooth polynomial background the oscillatory dHvA signal is clearly seen above $H_{irr}$ (Fig. 1(b)) for all samples except for that with the highest $T_c$ ($x = 0.33$).

From the fast Fourier transform (FFT) spectra of the oscillatory data (Fig. 1(c)) we can extract the dHvA frequencies $F$. These are related to the extremal cross-sectional areas, $A_k$, of the Fermi surface orbits giving rise to the oscillations via the Onsager relation, $F = (\hbar/2\pi e) A_k$. From the evolution of these frequencies (see Fig. 2) as the magnetic field is rotated from being along the $c$-axis ($\theta = 0^\circ$) towards being perpendicular the $c$ axis ($\theta = 90^\circ$), we can deduce the shape of the Fermi surface.

Band structure calculations of the Fermi surface of the end members of the BaFe$_2$(As$_{1-x}$P$_x$)$_2$ series are shown in Fig. 2. For both BaFe$_2$As$_2$ and BaFe$_2$P$_2$ the calculations were done with the experimental lattice parameters and pnictide $z$ position in the non-magnetic state. The two electron sheets at the zone corner are quite similar in size and shape in both compounds but there are significant differences between the hole sheets. In BaFe$_2$As$_2$ three concentric quasi-two-dimensional hole tubes are located at the zone center, whereas in BaFe$_2$P$_2$ the inner one of these tubes is absent whereas the outer tube has become extremely warped. The Fermi surface of BaFe$_2$P$_2$ is quite similar to that for SrFe$_2$P$_2$ [12].

For SrFe$_2$P$_2$ [12], CaFe$_2$P$_2$ [13] and LaFePO [14] it was observed that the strongest dHvA signals came from the electron sheets and so this is likely also to be the case for BaFe$_2$P$_2$. Thus, we assign the two strongest peaks ($\alpha$ and $\beta$) to the inner and outer electron sheet, respectively (see Fig. 1(c)). The observed frequencies for $x = 1$ are somewhat smaller than those predicted by the calculations and a rigid band shift of 45 – 75 meV is needed to bring the orbits into agreement with experiment. This is quite comparable to the shifts 50 – 60 meV needed for the electron sheets in SrFe$_2$P$_2$ [12]. Although there are other weak peaks in the FFT which could come from the hole sheets, further measurements with better signal to noise ratio are needed to confirm these.

As the sample composition is varied towards optimal doping, the signal from the electron sheets is reduced but
which are fitted (solid line) to the Lifshitz-Kosevich formu- 
la for the sufficiently high fields (see Fig. 1(c)); the mean free path 
two peaks are clearly visible over the full doping range in 
linearly with decreasing that the frequency of both these electron orbits decreases 
from 0.92 to 0.81 (see Fig. 2(a)) by 170 meV. The bare mass then decreases 
to determine the effective masses, \( m^*/m_b \) therefore increases from \( \sim 2 \) to \( \sim 4 \) as \( x \) decreases 
from 1 to 0.41. Note that conventional electron-phonon coupling is weak in these materials, and is estimated to 
only account for \( \sim 25\% \) of the observed mass enhancement \[22\].

The shrinking of the electron and hole sheets as \( x \) decreases could come from either the underlying one-
electron bandstructure or alternatively it could be driven 
by many-body correlation effects. We have estimated the 
change in the electron sheet area by calculating the band-
structure for both BaFe\(_2\)As\(_2\) and BaFe\(_2\)P\(_2\) with values of the lattice constants and \( z \) fixed to the experimental values 
appropriate for the various values of \( x \). Experimentally it is found that these parameters follow Vegard’s law and scale linearly with \( x \) \[16\]. We find that for both the 
As and P material the \( \alpha \) and \( \beta \) frequencies vary relatively 
little with \( x \). In Fig. 3(a) we show a weighted average of these calculated frequencies, \( F_{\text{av}} = xF_{\text{P}} + (1 - x)F_{\text{As}} \) where \( F_{\text{P}}/F_{\text{As}} \) refers to the (average maximal and minimal extremal) frequencies calculated with the atom set 
to P or As respectively. The calculated changes are much 
smaller than we observe experimentally, so it seems un-
likely that the shrinking electron sheets can be explained 
by conventional bandstructure theory.

An alternative is that the shrinking is driven by many-
body effects. Recently, Ortenzi et al. \[23\] suggested that 
in LaFePO the shrinking of the electron and hole Fermi-
surfaces and enhancement of effective masses can be ex-
plained by the interaction between the electron and hole 
bands. The observed correlation of the Fermi surface ar-
as with the increase in effective mass and increase in \( T_c \) 
would be consistent with this model; however, it remains
FIG. 3: color online. (a) Experimental (solid symbols) average electron sheet frequencies (\( \alpha \) and \( \beta \)) versus P content, \( x \). The data for \( x = 0 \) are taken from Ref. [9]. The dashed lines show bandstructure predictions. b) The variation with \( x \) of the quasiparticle effective masses, \( m^* \) and (c) \( T_c \) after Ref. [16]. The vertical dashed line marks the location of the onset of the appearance of magnetism at \( T = 0 \).

In summary, we have presented dHvA data which shows the evolution of the Fermi surface of \( \text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2 \) as \( x \) is varied towards \( T_c^{\text{max}} \). We find that the volume of the electron sheets (and via charge neutrality also the hole sheets) shrink linearly and the effective masses become strongly enhanced with decreasing \( x \). It seems unlikely that these changes are a simple consequence of the one-electron bandstructure but instead they likely originate from many-body interactions. These changes may be intimately related to the high \( T_c \) unconventional superconductivity in this system.

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