Measurement and analysis of the Hall effect of A-Fe$_2$As$_2$ single crystals with A = Ba, Ca or Sr.

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We report measurements of the Hall coefficient $R_H$ for single crystals of AFe$_2$As$_2$ with A = Ba, Ca or Sr which are the anti-ferromagnetic parent compounds of some high temperature pnictide superconductors. We show that $R_H$ of Sr-122 is consistent with high field quantum oscillation data. Our $R_H(T)$ data can also be used to estimate values of the spin density wave gap, giving $\Delta_{SDW}(0) = 710 \pm 70$ K for Sr-122 and $435 \pm 20$ K for Ba-122.

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The discovery of high temperature superconductivity in the iron pnictides has generated considerable scientific interest. They are correlated electron systems with quasi-2D structure, and show both similarities with, and notable differences from, the cuprates. Here we report measurements of the Hall coefficient $R_H$ for three isostructural ‘122’ iron arsenides that all have a structural phase transition at a temperature $T_S$. It is believed that anti-ferromagnetic (AF) order also sets in at $T_S$, and that superconductivity occurs when this is suppressed by doping with K [4], Na or Co or by applying pressure [5].

For for Ba-122 this flattens out as $T \to 0$, and $R_H(T)$ increases approximately linearly by a factor of 3 between 20 and 60-80 K followed by notable flattening well below $T_S$. This flattening could be associated with the proposed spin density wave (SDW) state. Below 120 K $R_H$ is negative for all the samples but for Ca-122 and the higher purity Sr(I) crystal there is a region between $T_S$ - 40 and $T_S$ where $R_H > 0$, suggesting a different balance between electron and hole contributions there. Above $T_S$, $|R_H|$ for Ca-122 rises slightly and then becomes constant. The other two compounds show a linear increase in $|R_H|$ between 300 K and $T_S$.

Three QO studies of in AFe$_2$As$_2$ in magnetic fields as high as 55 T have been reported [11, 13, 15] and the results analyzed in terms of small ellipsoidal pockets. We find that our Hall data are consistent with the QO results, especially for Sr-122. A priori they could be very different because there is an even number of electrons per unit cell in both the high- T tetragonal phase and the low- T phase where there is a commensurate anti-ferromagnetic
(AF) wave vector $\mathbf{Q}^{(1)}$ [11] that halves the in-plane Brillouin zone area. In both cases the number densities of electrons ($n_e$) and holes ($n_h$) will be the same and their contributions to $R_H$ should be weighted by the corresponding electrical conductivities $\sigma$, e.g. for two bands:

$$R_H = \frac{|R_{H1}| \sigma_h^2 - |R_{H2}| \sigma_e^2}{(\sigma_e + \sigma_h)^2} \tag{1}$$

So if $n_h$ and $n_e$ are the same, and $\sigma_h \simeq \sigma_e$, $R_H$ can be very small indeed, and will not give a valid measure of the carrier concentration. It might well vary strongly from sample to sample depending on the precise balance between $e$ and $h$ contributions. We see from Table I that there is actually good agreement between our values of $R_H$ (10 K) and those in the literature despite the differences in the $RR$ values mentioned earlier. Another possible source of discrepancies between high field QO and lower field Hall effect work is magnetic breakdown (MB) [16]. In a weak-coupling SDW system, the energy gap ($\Delta_{SDW}$) is small [17]; $\Delta_{SDW} = 1.76k_BT_S$ since the gap equation for an SDW has the same form as in BCS superconductivity. In practice, and perhaps surprisingly, any MB effects seem to be small.

For a single ellipsoidal pocket, the standard, nearly free electron formulae $R_H = 1/ ne$ and $\sigma_i = ne^2 \tau/m_i$ are still valid [18]. Here $m_i$ is the effective mass along a principal axis ($i$) of the ellipsoid, and $n = 2V_k/(2\pi)^3$, where $V_k$ is its volume in $k$-space. For Sr-122 three pockets labelled $\alpha$, $\beta$ and $\gamma$ were detected [11] with frequencies of $370 \pm 20$, $140 \pm 20$ and $70 \pm 20$ T and in-plane $k$ space areas of $1.38$, $0.52$ and $0.26$ % of the paramagnetic Brillouin zone area, $(2\pi/a)^2$, where the in-plane lattice parameter $a = 3.93$ Å. Angular studies showed that the ellipsoids were elongated along the crystallographic $c$-axis with axis ratios ($r$) of 1.4, 6.1 and 3.3 for $\alpha$, $\beta$ and $\gamma$ respectively. Later three similar frequencies and smaller $m^*$ values were found for Ba-122 crystals [14]. Band-structure calculations [11] [14] predict 4 inequivalent pockets in the AF Brillouin zone arising from imperfect nesting of the AF vector $Q = (\pi/2, \pi/2, 0)$. As discussed below, analysis of the Hall data suggests that the largest electron pockets are absent for Sr-122.

From the values of $r$ and the $k$ space areas we obtain $V_k$ for each type of ellipsoidal pocket. Each pocket in the reduced AF Brillouin zone gives $n_\alpha = 0.0053(5)$, $n_\beta = -0.0054$ and $n_\gamma = -0.0010(1)$ carriers per formula unit ($f.u.$). Here the + and − signs denote $h$ and $e$ contributions to $R_H$ respectively. Within the error bars these satisfy $\Sigma n_\alpha = \Sigma n_h$ and imposing this as a precise constraint does not affect our conclusions. Both papers [11] [14] identify $\alpha$ as a “tear-shaped” hole pocket, and $\beta$ as an ellipsoidal electron pocket. The small $\gamma$ pocket is identified as electron-like for Sr-122 in Ref. 11 and as hole-like for Ba-122 in Ref. 13. Using the analysis described below we get good agreement with our Hall data if the $\gamma$ pocket is electron-like for Sr-122.

In order to calculate $R_H$ we need to find appropriate values of $\sigma_\alpha$, $\sigma_\beta$ and $\sigma_\gamma$ for use in Eqn. 1. Experimentally [19] it was found that the isotropic mean free path approximation worked well for Sr$_2$RuO$_4$. Here we are dealing with small values of $k_F$ but any impurity potentials will still be screened over short distances $(d)$ because of the underlying metallic state that is weakly perturbed by a SDW. In this limit, $k_Fd \ll 1$ we expect a $k$-independent scattering cross-section, $4\pi d^2$ [20], which will indeed give a mean free path that is independent of $k$. The relative contributions to $\sigma$ in Eqn. 1 are then given by $n_\alpha/k_F^2$, etc. where $k_F^2$ is the in-plane Fermi wavevector of the $\alpha$ pocket and give a negative value for $R_H$. Band-structure calculations [11] [14] seem to give two pockets of each type in the AF Brillouin zone. Taking this into account leads to an effective electron concentration $n_{eff} = 0.039$ per $f.u.$ This is in excellent agreement with the value given in Table I for Sr-122 for a field of 5 T at low $T$. Although $\Sigma n_\alpha = \Sigma n_h$ within error bars, there is little $e - h$ cancellation in $R_H$ because the $\alpha$ and $\beta$ pockets have very different eccentricities $r$, which means that $\sigma_\alpha \neq \sigma_\beta$. If instead we included the fourth and largest electron pocket in this analysis, using the eccentricity implied by the band-structure calculations, then the calculated value of $n_{eff} = 0.11$ per $f.u.$, which is much larger than the experimentally determined values.

The above analysis has all been based on $R_H$ data taken at 5 T. As shown in Fig. 3 $R_H$ is in fact field-dependent. We do not understand this, but although the three materials show completely different $H$-dependence, it could still be a general property of the SDW state because of the $e - h$ compensation described above. For the Sr-122 crystal $R_H$ increases by nearly 50% be-
TABLE I: Parameters obtained in the present work or used in the analysis. $T_S$ values were obtained on cooling; for $A = Ca$ the warming transition is 2K higher. $RR$ is the resistivity ratio $\rho(300)/\rho(10)$. $R_H$ is the Hall coefficient measured at 5T, $n_{eff}$ is the corresponding carrier concentration in a single band analysis. $\mu_H$ is the Hall mobility, $R_H \sigma$, and $\beta$ is the $T^2$ coefficient of the inverse mobility obtained from fits to $1/\mu_H = \alpha + \beta T^2$ between 10 and 60K. Sr(I) is from growth batch 170 and Sr(II) batch 105.

|      | Ba | Ca | Sr(I) | Sr(II) |
|------|----|----|-------|--------|
| $V_{T,u}$ ($\AA^3$) | 102 | 89 | 95 | 95 |
| $T_S$ (K) | 135.4 | 168.6 | 194.6 | 190.8 |
| $RR$ | 2.65 | 6.2 | 1.85 | 1.5 |
| $RR$ (other work) | (4.3)$^a$ | (11)$^b$ | (4.7)$^c$ | - |
| $R_H$($295$)($10^{-10}$m$^3$/C) | -7.6 | -3.0 | -3.0 | -2.3 |
| $R_H$(TS+$5$)($10^{-10}$m$^3$/C) | -21.0 | -2.5 | -5.5 | -4.1 |
| $R_H$(10)($10^{-10}$m$^3$/C) | -292 | -99 | -162 | -120 |
| $R_H$(10)($10^{-10}$m$^3$/C) | (-220)$^a$ | (-95)$^b$ | (-135)$^c$ | - |
| $n_{eff}$($10^5$)/f.u. | 0.022 | 0.056 | 0.037 | 0.049 |
| $\mu_H(295)$(cm$^2$/Vs) | 2.8 | 0.98 | 0.95 | - |
| $\mu_H(T_S+5)$(cm$^2$/Vs) | 9.3 | 1.3 | 1.9 | - |
| $\mu_H(0)$(cm$^2$/Vs at 1T) | 450 | 200 | 200 | 50 |
| $\beta(T)(10^{-6}$Vs/cm$^2$K$^2$) | 0.8 | 2.9 | - | 5.3 |
| $\beta(5T)(10^{-6}$Vs/cm$^2$K$^2$) | 0.9 | 3.3 | 4.1 | 5.1 |

$^a$ Ref.$^5$, $^b$ Ref.$^9$, $^c$ Ref.$^7$, $^8$

FIG. 3: Color online: left-hand panels, normalized magnetic field dependence of the measured Hall coefficient $|R_H|$ for the three compounds studied here, at several fixed $T$. Right hand panels, inverse Hall mobility vs. $T^2$ for the 3 compounds studied.

between 1 and 5T. In principle this could be caused by one or more of the pockets approaching the usual high field condition, $\mu_H H$, i.e. $\omega_c \tau \sim 1$ at 5T. But for the same crystals we did not observe any QOs in fields from 12 to 15T down to 1.4K. Therefore any explanation along these lines seems to require a very small pocket of frequency $\approx 50$T or less whose period was too long to be observed in our measurements. For Ca-122 the $H$ dependence is much smaller and of opposite sign, while that for Ba-122 is larger and also has the opposite sign to that in Sr-122. High field orbital effects are somewhat more likely here because Ba-122 has a higher average mobility and our Ca-122 crystal has comparable mobility to the Sr crystal but shows a much smaller $H$-dependence.

Applying the same analysis to available data for Ba-122 [14] does not give such straightforward results. If the largest $e$-pocket is again absent [14], we can satisfy $\Sigma n_e = \Sigma n_h$ and account for the low field value of $R_H$ by assuming that the $\beta$ pocket has $r = 3$, rather than 5 $\pm$ 1 [14], that $\gamma$ is in fact an $e$-pocket and by assuming that the mean free path on the $\beta$ and $\gamma$ electron pockets is 20% larger than that on the $\alpha$ pockets. For Ca-122 [15], the two pockets observed have very different volumes so we have not attempted to calculate $R_H$.

FIG. 4: Color online: (a) plots of effective carrier concentration $n_{eff}$ vs. $T^2$ for Sr-122 and Ba-122. The lines show the fits to $C + DT^2$. (b) $n_{eff}$ vs. $T^3$ for Ca-122 and the corresponding fits for three values of applied magnetic field (T). (c) - (e) semi-log plots $n_{eff}(T)$ vs. $1/T$ before and after subtraction of the limiting low $T$ dependence. For Sr and Ba, black triangles show data at 5T while crosses show data at 1T. For Ba black triangles, inverted triangles and crosses show 5T, 1T and 11T data respectively. The thicker, slightly curved, (red) lines show calculated weak-coupling BCS behavior for the $T_S$ values shown. The slopes of the black lines are used to find the activation energy, i.e. $\Delta_{SDW}(0)$. 

FIG. 4: Color online: (a) plots of effective carrier concentration $n_{eff}$ vs. $T^2$ for Sr-122 and Ba-122. The lines show the fits to $C + DT^2$. (b) $n_{eff}$ vs. $T^3$ for Ca-122 and the corresponding fits for three values of applied magnetic field (T). (c) - (e) semi-log plots $n_{eff}(T)$ vs. $1/T$ before and after subtraction of the limiting low $T$ dependence. For Sr and Ba, black triangles show data at 5T while crosses show data at 1T. For Ba black triangles, inverted triangles and crosses show 5T, 1T and 11T data respectively. The thicker, slightly curved, (red) lines show calculated weak-coupling BCS behavior for the $T_S$ values shown. The slopes of the black lines are used to find the activation energy, i.e. $\Delta_{SDW}(0)$. 

FIG. 3: Color online: left-hand panels, normalized magnetic field dependence of the measured Hall coefficient $|R_H|$ for the three compounds studied here, at several fixed $T$. Right hand panels, inverse Hall mobility vs. $T^2$ for the 3 compounds studied.
403 and 115 K for Ba-122. As shown in Table I the $\beta T^2$ term in $1/\mu_H(T)$ is a factor 4 larger for Sr-122, reflecting the smaller $E_F$ values listed above.

We now address the $T$ dependence of $R_H$ at low $T$ and propose a method of estimating the SDW gap from our data. An important clue here is that for Ba-122 the electronic heat capacity above $T_S$ is $\sim 8$ times larger than the low $T$ value [23]. This suggests that, although the small pockets do give a finite density of states (DOS) at $E_F$, any square-root singularities in the DOS from the SDW will be much larger. As shown in Fig. 4, for Sr-122 and Ba-122 we can fit $n_{e\perp}(T)$ below 40-50 K to a $C + DT^2$ law. This $DT^2$ term could arise partly from the low values of $E_F$ combined with the constraint $\Sigma n_e = \Sigma n_h$, leading to changes in $n_e$ or $n_h$ as $T$ is increased. A second possible reason is that in Eqn. 1, $\sigma_{n,\beta}$ could have different $T^2$ terms from $e-e$ scattering; a third is that the pockets expand as the SDW gap decreases. While the latter must be true at higher $T$, in Fig. 4 we also show $n_{e\perp}$ vs. $1/T$ plots calculated [25] for the fully nested case where there are no pockets and the energy gap $2\Delta_{SDW}(T)$ has the usual BCS s-wave $T$-dependence. These plots are linear below $T_{SDW}/2$ implying that $\Delta_{SDW}$ is effectively constant and any expansion of the pockets is not important below $T_{SDW}/2$. As shown in Fig. 4, subtraction of the $C + DT^2$ terms leads to linear regions in plots of $\log n_{e\perp}$ vs. $1/T$. From the slopes of these lines we find $\Delta_{SDW}(0) = 710 \pm 70$ K for Sr-122, a factor of 2.1 larger than the BCS value, and $435 \pm 20$ K for Ba-122, a factor of 1.8 larger than BCS. For Ca-122 the value of $\Delta_{SDW}(0)$ is less certain. As shown in Fig. 4b, $n_{e\perp} = C + DT^3$ gives a good fit below 50 K. Subtracting this gives $\Delta_{SDW}(0) = 1150 \pm 150$ K, a factor 3.9 larger than BCS. On the other hand if $n_{e\perp}$ is forced to fit $C + DT^2$ at low $T$, then a much smaller value of $\Delta_{SDW}(0) \sim 1.2$ times the BCS value, is obtained.

Single crystal optical reflectivity data taken at 10 K have been analyzed in terms of two broad Lorentz-Drude peaks centered at 360 and 890 cm$^{-1}$ for Ba-122 [23, 24] and at 500 and 1360 cm$^{-1}$ for Sr-122 [23]. The lower peaks have similar energies to our values, $\Delta_{SDW}(0) = 300 \pm 15$ cm$^{-1}$ for Ba-122 and 490 $\pm 50$ cm$^{-1}$ for Sr-122. Moreover, there are clear changes in slope in the raw reflectivity data [23] at ca. 270 and 600 cm$^{-1}$ for Ba-122 and at 410 and 900 cm$^{-1}$ for Sr-122 that are absent above $T_S$. We therefore suggest that the lower reflectivity anomaly could correspond to the onset of electron excitations between the $e-h$ pockets at $E_F$ and the square root singularities from the SDW at $E_F \pm \Delta_{SDW}$ which would have an energy of $\Delta_{SDW}(0)$, while the upper root anomalies could be a measure of $2\Delta_{SDW}(0)$.

In summary, our Hall measurements on Sr-122 are surprisingly consistent with high-field quantum oscillation data, and subject to some minor modifications, with those observed for Ba-122. We have been able to make approximate estimates of the SDW gaps from the Hall data. For Sr and Ba we propose that these are consistent with optical reflectivity although detailed calculations of the latter are still needed.

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[25] Here $n_{eff}$ is the number of electrons excited above the gap. There will be an equal number of holes below the gap, but usually enough $e - h$ asymmetry to give $n_{eff} \sim \exp(-\Delta/T)$.

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