Evidence for Pauli-limiting behaviour at high fields and enhanced upper critical fields near $T_c$ in several disordered FeAs based Superconductors

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Abstract

We report resistivity and upper critical field $B_{c2}(T)$ data for disordered (As deficient) LaO$_{9.0}$F$_{0.1}$FeAs$_{1-x}$ in a wide temperature and high field range up to 60 T. These samples exhibit a slightly enhanced superconducting transition at $T_c = 28.5$ K and a significantly enlarged slope $dB_{c2}/dT = -5.4$ T/K near $T_c$ which contrasts with a flattening of $B_{c2}(T)$ starting near 23 K above 30 T. The latter evidences Pauli limiting behaviour (PLB) with $B_{c2}(0) \approx 63$ T. We compare our results with $B_{c2}(T)$-data from the literature for clean and disordered samples. Whereas clean samples show almost no PLB for fields below 60 to 70 T, the hitherto unexplained pronounced flattening of $B_{c2}(T)$ for applied fields $H \parallel ab$ observed for several disordered closely related systems is interpreted also as a manifestation of PLB. Consequences are discussed in terms of disorder effects within the frames of (un)conventional superconductivity, respectively.

Key words: Ferro-pnictide superconductors, upper critical field, disorder, As-vacancies, Pauli-limiting, magnetic pair breaking

FeAs based superconductors show high transition temperatures $T_c \leq 57$ K and remarkably high upper critical fields $B_{c2}(0)$ exceeding often 70 T. Many of their basic properties and the underlying pairing mechanism are still not well understood. A study of $B_{c2}(T)$, in particular, studies on disordered FeAs superconductors are of large interest since for an unconventional pairing both $T_c$ and $dB_{c2}/dT$ at $T_c$ might be reduced by introducing disorder. Here, $B_{c2}(T)$ of As-deficient (AD) LaO$_{9.0}$F$_{0.1}$FeAs$_{1-x}$ samples are studied in fields up to 60 T.

Polycrystalline samples of LaO$_{9.0}$F$_{0.1}$FeAs were prepared by the standard solid state reaction [1]. Some samples have been wrapped in a Ta foil during the final annealing procedure. Ta acts as an As getter at high $T$ forming a solid solution of about 9.5 at.% As in Ta. This leads to an As loss in the samples resulting in an As/Fe ratio ~ 0.9. Due to disorder in the FeAs layer, a three times larger resistivity above $T_c$ at 31 K is found for the studied AD samples compared with a clean reference sample. Anyhow, the AD samples have, with $T_c = 28.5$ K, a higher $T_c$ than that of stoichiometric reference samples (27.7 K) [1].

For polycrystalline samples, only $B_{c2}^{ab}$ is accessible which is related to those grains oriented with their $ab$-planes along the applied field. $B_{c2}^{ab}$ was determined from resistance data by defining the onset of superconductivity (SC) at 90% of the resistance $R_N$ in the normal state. The $T$-dependence of the resistance of a typical AD samples is shown in Fig. 1 for applied magnetic fields up to 50 T. We confirmed that the pulsed field data were unaffected by sample heating [2]. $B_{c2}^{ab}(T)$ of our AD sample obtained from pulsed field measurements in the IFW and the FZD is shown in Fig. 2 together with $B_{c2}$ data reported for a clean reference sample [3]. The large slope $dB_{c2}^{ab}/dT = -5.4$ T/K at $T_c$ of our AD samples points to strong impurity scattering in accord with enhanced resistivity at 30 K. For the clean sample [3] the available data up to 45 T is well described by the WHH (Werthamer-Helfand-Hohenberg) model [4] for the orbital limited $B_{c2}(T)$. Whereas for the AD samples, the WHH model which predicts $B_{c2}^{ab}(0) = 0.697 T_c (dB_{c2}^{ab}/dT)|_{T_c} = 106$ T at $T = 0$, fits the experimental data up to 30 T, only. For applied fields $> 30$ T increasing deviations from the WHH curve are clearly visible for the $B_{c2}^{ab}(T)$ data. The flattening of $B_{c2}^{ab}(T)$ at high field points to its limitation by the Pauli spin paramagnetism. This effect is measured in the WHH model by the Maki parameter $\alpha = \sqrt{2}B_{c2}^{ab}(0)/B_p(0)$, where $B_p(0)$ is the Pauli limiting field.

![Figure 1](image-url)  
Figure 1: (Color online) $T$-dependence of the resistance from both DC and pulsed field measurements.
The paramagnetically limited upper critical field, $B_{c2}^p$, is given by $B_{c2}^p(0) = B_{c2}^p(0)(1 + \alpha^2)^{0.5}$. For our AD samples, a reasonable fit of the experimental data to this model has been obtained for $\alpha = 1.31$ (see Fig. 2) and yields $B_{c2}^p(0) = 63$ T. In contrast, $B_{c2}(T)$ data for clean LaO$_{0.93}$F$_{0.07}$FeAs samples [3] (see Fig. 3) show almost no PLB below 70 T. But for several possibly disordered closely related systems, a similar flattening of $B_{c2}(T)$ as we found for our AD samples has been reported for applied fields $H \parallel ab$. This is shown in Fig. 3 using normalized dimensionless units $b^* (T) = B_{c2}(t)/[T_c(dB_{c2}/dT)]_T$ and $t = T/T_c$. The data in Fig. 3 are well described by the WHH model with the obtained Maki parameters $\alpha$. The deviation of $b^* (T)$ at low $T$ from $b^* (0)$ for $\alpha = 0$ increases with $\alpha$ due to rising paramagnetic pair-breaking. In this context the partial substitution of Fe by 4d and 5d transition metals (e.g. Ru, Rh, Ir, and Pd) [12]-[15] is of interest. Due to their larger atomic sizes compared with that of Fe$^{2+}$, also a stronger disorder effect compared with the 3d substitutions by Co and Ni should be expected. For all these cases we predict PLB, too, at variance with large WHH estimates on the basis of the enhanced initial slopes [13]-[14].

For AD samples we found hints for an enhanced Pauli paramagnetism from $\mu$SR experiments [2]. Their improved SC at high $T$ and low fields could be understood within conventional $s_{\pm}$-wave SC by enhanced disorder. In contrast, for clean FeAs superconductors an unconventional $s_\pm$-wave scenario has been proposed [16]. Based on our results for $B_{c2}(T)$, two alternative scenarios of opposite disorder effects might be suggested: (i) an impurity-driven change of the pairing state from $s_\pm$ to conventional $s_{\pm}$-wave SC and (ii) a special impurity-driven stabilization of the $s_\pm$ state where the As-vacancies are assumed to scatter mainly within the bands, only, but not in between them. Concerning the impurity scattering and SC gap symmetry a recent $^{75}$As-NMR study on a disordered AD sample [17] revealed $T_1^{-1} \propto T^5$ for the spin-lattice relaxation rate below $T_c$ compared with a $T^3$-law for a 'clean' sample is of considerable interest.

The PLB found here suggests to continue measurements at least up to 70 T to elucidate, whether there is still much room for increasing $B_{c2}$ beyond 80 T. Improving the low-field properties of FeAs superconductors by introducing As vacancies opens new preparation routes for optimising their properties.

Note added. Finishing the present paper we have learned about a very interesting $B_{c2}$-study by Lee et al. [18] on O deficient F-free and F doped Sm-1111 single crystals with high $T_c$-values of 50.5 and 42 K, respectively. Using the same approach and probing the same field range as we, the authors detected for $B_{c2}$ PLB, too ($\alpha = 2.3$ and 2.7). Thus, at present in total at least seven different ferro-pnictide superconductors exhibit PLB which now seems to become a rather general feature.

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