# Possibility of vortex lattice structural phase transition in the superconducting pnictide $Ba(Fe_{0.925}Co_{0.075})_2As_2$

R. Kopeliansky, A. Shaulov, B. Ya. Shapiro, and Y. Yeshurun Department of Physics, Institute for Superconductivity, Bar-Ilan University, Ramat-Gan, Israel

#### B. Rosenstein

Electrophysics Department, National Chiao Tung University, Hsinchu, Taiwan, Republic of China

#### J. J. Tu

Department of Physics, City College of New York, CUNY, New York, New York 10031, USA

#### L. J. Li, G. H. Cao, and Z. A. Xu

Department of Physics, Zhejiang University, Hangzhou 310027, China (Received 10 December 2009; revised manuscript received 25 January 2010; published 10 March 2010)

We present magnetic measurements in a single crystal of the newly discovered superconducting iron-pnictide Ba(Fe<sub>0.925</sub>Co<sub>0.075</sub>)<sub>2</sub>As<sub>2</sub>. The magnetization loops exhibit a second magnetization peak (SMP) similar to that observed in most high-temperature superconductors (HTSs). Magnetic relaxation measurements reveal a minimum in the normalized relaxation rate,  $S = d \ln M/d \ln t$ , located in between the SMP onset and the peak fields. The SMP in HTSs is commonly associated with the vortex order-disorder phase transition. However, in Ba(Fe<sub>0.925</sub>Co<sub>0.075</sub>)<sub>2</sub>As<sub>2</sub> the onset and peak fields, as well as the minimum point in S, exhibit strong temperature dependence down to low temperatures, excluding the possibility for such a transition. We suggest that the SMP in Ba(Fe<sub>0.925</sub>Co<sub>0.075</sub>)<sub>2</sub>As<sub>2</sub> is associated with a vortex structural phase transition from rhombic to square lattice taking place at field and temperatures corresponding to the minimum point of S. A theoretical fit to the transition line, based on a recent theoretical model for vortex structural phase transition, shows good agreement with the experimental results.

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# I. INTRODUCTION

The magnetization curves in the "122" Co-doped pnictides Ba(Fe<sub>1-x</sub>Co<sub>x</sub>)<sub>2</sub>As<sub>2</sub> exhibit an anomalous second magnetization peak<sup>1-5</sup> (SMP) similar to that observed in the superconducting cuprates.<sup>6–11</sup> In the latter, this anomaly has been commonly interpreted as indicating a vortex orderdisorder phase transition. Such a transition is theoretically characterized by a weak temperature dependence of the transition line in the low temperature region, far below the transition temperature,  $T_c$ ,  $^{12}$  as commonly observed in the cuprates, e.g.,  $YBa_2Cu_3O_{7-\delta}$ ,  $Bi_2Sr_2CaCu_2O_{8-\delta}$ , and  $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ . An exceptional behavior, however, is revealed in the high-T<sub>c</sub> superconductor La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> with x=0.126 and similar doping. <sup>13</sup> This material exhibits a broad SMP with characteristics that are strongly temperature dependent down to low temperatures. The SMP in La<sub>2-r</sub>Sr<sub>r</sub>CuO<sub>4</sub> was associated with a structural vortex phase transition, from a rhomb to square lattice, caused by softening of the "squash" vortex lattice elastic modulus  $c_{\text{sq}} = 2(c_{11} + c_{12}) - c_{66}.^{14}$ 

In this Brief Report we present magnetic measurements in Ba(Fe<sub>0.925</sub>Co<sub>0.075</sub>)<sub>2</sub>As<sub>2</sub> single crystal confirming the results of Prozorov *et al.*<sup>1</sup> and Shen *et al.*<sup>4</sup> who interpreted the SMP as signifying a crossover from elastic to plastic vortex creep.<sup>8</sup> We note, however, that such a crossover in the vortex dynamics may accompany a thermodynamic phase transition in the vortex lattice as is the case, for example, in YBaCu<sub>2</sub>O<sub>7-δ</sub>.<sup>8,9</sup> As the SMP has been so far associated with

vortex phase transition, it is natural to attempt such an interpretation also in  $Ba(Fe_{1-x}Co_x)_2As_2$ . We point to striking similarities between the magnetic behavior of this pnictide and the high- $T_c$  superconductor  $La_{2-x}Sr_xCuO_4$ . In particular, similar to  $La_{2-x}Sr_xCuO_4$ , the SMP in this pnictide exhibits strong temperature dependence down to low temperatures. We, therefore, propose a similar interpretation of the SMP in  $Ba(Fe_{0.925}Co_{0.075})_2As_2$ , i.e., a structural vortex phase transition from rhomb to square lattice, showing that the measured transition line can be well fitted to the theoretical predictions. <sup>14</sup>

#### II. EXPERIMENTAL

A parallelepiped shaped single crystal of  ${\rm Ba(Fe_{0.925}Co_{0.075})_2As_2}$ , with dimensions  $2.4\times1\times0.19~{\rm mm^3}$  and  $T_c{\sim}25~{\rm K}$ , was grown by the self-flux method. Shapenetization measurements, as a function of field, temperature, and time, were performed using a commercial superconducting quantum interference device (SQUID) (Quantum Design MPMS-5S). All measurements were done after zero-field cooling the sample to the measured temperature and then applying external magnetic field parallel to the crystallographic c axis.

#### III. RESULTS

Figure 1 exhibits magnetization loops measured in Ba(Fe<sub>0.925</sub>Co<sub>0.075</sub>)<sub>2</sub>As<sub>2</sub> at various temperatures between 19.5

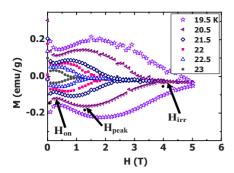


FIG. 1. (Color online) Magnetization loops at the indicated temperatures. Arrows point to three characteristic features of the loop.

and 23 K. A broad SMP is apparent at all temperatures. The arrows in the figure point to three characteristic fields:  $H_{\rm on}$ , the onset of the SMP,  $H_{\rm peak}$ , the peak field, and  $H_{\rm irr}$ , the irreversibility field. At lower temperatures (not shown in the figure), only the onset of the SMP is observed as the peak is shifted beyond the measured field range.

Relaxation measurements at constant temperature and field reveal logarithmic increase in the magnetization with time. The field dependence of the normalized relaxation rate,  $S=d \ln(M)/d \ln(t)$ , is shown in Fig. 2(a) for several temperatures. One notices a decrease in S to a minimum point in each curve which is interpreted in the following as indicating softening of the vortex lattice giving rise to enhanced pinning and slower relaxation.

A typical temperature dependence of the normalized relaxation rate is shown in Fig. 2(b) for several fields. In the cuprates, *S* usually *increases* monotonically with

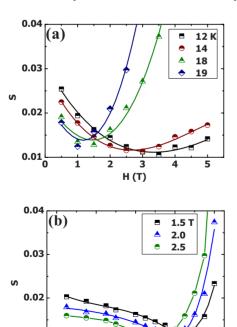


FIG. 2. (Color online) (a) Normalized relaxation rate S vs H at the indicated temperatures. (b) S vs T at the indicated fields. Solid lines are guides for the eyes.

T (K)

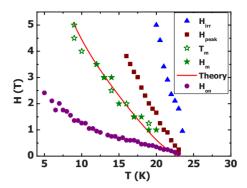


FIG. 3. (Color online) H-T plot of the characteristic fields for Ba(Fe<sub>0.925</sub>Co<sub>0.075</sub>)<sub>2</sub>As<sub>2</sub>: H<sub>on</sub> (circles), H<sub>m</sub> (full stars), H<sub>peak</sub> (squares), H<sub>irr</sub> (triangles), and the characteristic temperature T<sub>m</sub> (hollow stars). H<sub>m</sub> vs T and T<sub>m</sub> vs H form a line that is well fitted to Eq. (1) (solid line) that describes rhomb-to-square structural phase transition.

temperature.  $^{16,17}$  In Ba(Fe<sub>0.925</sub>Co<sub>0.075</sub>)<sub>2</sub>As<sub>2</sub>, however, *S decreases* as temperature increases, reaching a *minimum* point at  $T_{\rm m}$  and then *increases* sharply. This figure indicates that softening of the vortex matter, and the consequent enhanced pinning, is also achieved on warming the crystal. As we show below, the minima in  $T_{\rm m}$  vs H and  $H_{\rm m}$  vs T coincide in the field-temperature diagram suggesting that this behavior is associated with a thermodynamic transition.

In Fig. 3 we plot the characteristic fields  $H_{\rm irr}$ ,  $H_{\rm peak}$ ,  $H_{\rm m}$ ,  $H_{\rm on}$ , and  $T_{\rm m}$  in the field-temperature plane. We note that  $H_{\rm m}$  and  $T_{\rm m}$  are located approximately halfway between the onset and the peak of the broad SMP. The most significant observation depicted in Fig. 3 is the strong concave-shaped decrease with temperature of the characteristic fields associated with the SMP.

#### IV. DISCUSSION

The lines in Fig. 3 describing the temperature dependence of the characteristic fields associated with the SMP resemble the behavior of the melting line<sup>18</sup> or the depinning line.<sup>19</sup> However, neither of these is a valid interpretation, since the magnetization is strongly irreversible well above these lines. The common interpretation of the SMP as signifying an order-disorder vortex phase transition 10-12 is also excluded, as in this case the transition line must show weak temperature dependence at low temperatures. In the following we suggest that, similar to La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub>, <sup>14</sup> the SMP in Ba(Fe<sub>0.925</sub>Co<sub>0.075</sub>)<sub>2</sub>As<sub>2</sub> signifies a structural vortex phase transition from a rhomb lattice at low fields to a square lattice above a transition field,  $H_{\rm spt}$ . The scenario of a structural vortex phase transition in  ${\rm La_{2-x}Sr_xCuO_4}$  has been supported by the small angle neutron-scattering experiments of Gilardi et al.<sup>20</sup> showing a crossover from triangular to square coordination of the vortex structure with increasing magnetic field. The square structure originates from the fourfold symmetry of the intervortex interaction, which in the case of La<sub>2-r</sub>Sr<sub>r</sub>CuO<sub>4</sub> is provided by a *d*-wave order parameter.<sup>20</sup> Vortex-vortex interaction with fourfold symmetry may also be found in s-wave superconductors with fourfold symmetry in the Fermi velocity, as is the case in the pnictides.<sup>21</sup> The anisotropic interaction with fourfold symmetry may induce a rhombic rather than hexagonal Abrikosov lattice, which eventually may transform into a square vortex lattice when the distance between vortices is sufficiently small. We note that rhomb to square structural vortex phase transition was identified also in the borocarbides<sup>22,23</sup> exhibiting *s*-wave symmetry in the gap<sup>24</sup> but fourfold symmetry in the Fermi velocity.<sup>22</sup>

Thermal fluctuations on a mesoscopic scale assist in breaking the rhomb symmetry reducing  $H_{\rm spt}$  as temperature increases. On approaching the structural phase transition, the elastic squash modulus,  $c_{\rm sq} = 2(c_{11} + c_{12}) - c_{66}$ , is vanishing, <sup>14</sup> enabling vortices to be located at pinning sites. This vortex state with enhanced pinning is characterized by increase in the critical current,  $J_c$ , and slower magnetic relaxation, S, around  $H_{\rm spt}$ . When the increase in  $J_c$  is large enough compared to its natural decrease with field, it is borne out in the experiment as a second magnetization peak and a minimum in the normalized relaxation rate.

The square to rhomb structural phase transition line was calculated by minimizing the free energy for the square lattice with respect to the variational parameters, the elastic moduli  $c_{11}$ ,  $c_{66}$ , and  $c_{\rm sq}$ , yielding  $^{14}$ 

$$H_{\text{spt}} = A(\eta) \frac{T_0(\eta) - T}{C^{\nu-1} T^{\nu}}, \quad C = \frac{4\pi^3 \lambda^2}{L_z \phi_0^2},$$
 (1)

where A and  $T_0 \! < \! T_c$  are constants depending on the Ginzburg-Landau parameter  $\kappa$  and the anisotropy parameter  $\eta$  characterizing the deviations of the vortex-vortex interactions potential from rotational symmetry.  $\lambda$  is the London penetration depth,  $\phi_0$  is the flux quanta, and  $L_z$  is a numerical parameter that defines the effective superconducting layer width in which thermal fluctuations are considered.

We suggest that the line in Fig. 3 defined by  $H_{\rm m}$  and  $T_{\rm m}$ , which is located in between  $H_{\rm on}$  and  $H_{\rm peak}$  of the broad SMP, signifies a structural vortex phase transition line in Ba(Fe<sub>0.925</sub>Co<sub>0.075</sub>)<sub>2</sub>As<sub>2</sub>. The solid line in Fig. 3 is a theoretical fit to these points based on Eq. (1), using A,  $T_0$ , and v as fitting parameters. This fit yields  $T_0$ =23 K $\sim$ 0.92 $T_c$  and v=0.95. As shown in Ref. 14, the anisotropy parameter,  $\eta$ , can be calculated from the fitting parameters A and  $T_0$ . This calculation yields  $\eta$ =0.04. It is interesting to note that the values of  $T_0/T_c$ ,  $\eta$ , and v found for Ba(Fe<sub>0.925</sub>Co<sub>0.075</sub>)<sub>2</sub>As<sub>2</sub> are similar to that reported for La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> (Ref. 14) (0.95, 0.03, and 0.9, respectively). By passing we note that La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> and Ba(Fe<sub>0.925</sub>Co<sub>0.075</sub>)<sub>2</sub>As<sub>2</sub> also have similar transition temperatures,  $T_c$  (30 and 25 K, respectively) and Ginzburg-Landau parameter  $\kappa$  (75 and 65, respectively).

We also note that unlike the behavior in the borocarbides, the slope of the transition line in La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> and Ba(Fe<sub>0.925</sub>Co<sub>0.075</sub>)<sub>2</sub>As<sub>2</sub> is negative consistent with the thermodynamic expectation that the more symmetric phase occurs at higher temperature. The positive slope of the transition line in the borocarbides has been attributed to strong

disorder. <sup>14</sup> In addition, the behavior of the transition line in  $La_{2-x}Sr_xCuO_4$  and  $Ba(Fe_{0.925}Co_{0.075})_2As_2$  is probably more affected by thermal fluctuations on a mesoscopic scale that are much stronger in high- $T_c$  superconductors.

neutron-scattering,<sup>25</sup> Recent small angle Bitter-decoration,<sup>25</sup> and scanning tunneling microscopy<sup>26</sup> studies of the vortex matter were performed on similar Ba(Fe<sub>1-x</sub>Co<sub>x</sub>)<sub>2</sub>As<sub>2</sub> pnictides. These studies seem to imply that the structure of the vortex lattice depends on the external magnetic field; while Ref. 25 reported a hexagonal vortex lattice for relatively low fields ( $\sim$ 4 mT), the data of Ref. 26, measured at relatively high fields ( $\sim 9$  T), show a disordered lattice. The latter observation does not contradict our scenario of rhomb to square vortex structural phase transition which is expected only in perfect, clean crystals. In crystals with defects, softening of the vortex lattice associated with the structural phase transition allows better pinning and thus leads to a disordered state rather than an ordered square lattice. Our pnictide crystal as well as the one measured in Ref. 26 are probably less clean than the La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> crystal in which the high field square lattice was observed.<sup>20</sup> This is reflected in the relatively low critical current measured in  $La_{2-r}Sr_rCuO_4$ —less than  $10^5$  A/cm<sup>2</sup> at 4.2 K at all fields<sup>27</sup> while in our prictide crystal  $J_c$  exceeds  $10^6$  A/cm<sup>2</sup> at low fields. The high field vortex phase in our sample is most probably also disordered, as reflected by the increase in the relaxation rate above the transition. However, as argued above, the underlying mechanism creating this disorder state is not an order-disorder transition but softening of the vortex lattice associated with a structural phase transition. More systematic studies of the vortex lattice structure in cleaner crystals are necessary to conclusively decide whether a rhomb to square structural phase transition takes place in  $Ba(Fe_{1-x}Co_x)_2As_2$ .

## V. SUMMARY AND CONCLUSION

We interpret the anomalous slowing down of the vortex dynamics in Ba(Fe<sub>0.925</sub>Co<sub>0.075</sub>)<sub>2</sub>As<sub>2</sub> as signifying softening of the vortex lattice on approaching a structural vortex phase transition. We support this interpretation by (i) eliminating possibilities of order-disorder, melting, and depinning lines; (ii) pointing to similarities between the pnictides and La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> crystals in which such a transition was predicted theoretically and confirmed experimentally; (iii) showing a theoretical fit to the experimental transition line for the pnictides crystal based on a theory for structural phase transition. A direct experimental evidence for this interpretation is a challenge for future neutron-scattering experiments in clean crystals.

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