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In-plane electronic anisotropy in underdoped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ revealed by partial detwinning in a magnetic field

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We present results of angle-dependent magnetoresistance measurements and direct optical images of underdoped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ which reveal partial detwinning by action of a 14 T magnetic field. The relative change in the twin domain population for the given field is modest, of order 5–15 %. The associated field-induced hysteretic changes in the resistivity are up to 5% for intermediate Co concentrations, implying a large in-plane resistivity anisotropy in the broken-symmetry state. Based on the generic anisotropic susceptibility of collinear antiferromagnets, we infer a smaller resistivity along the antiferromagnetic ordering direction. The observation of field-induced motion of twin boundaries indicates a substantial magnetoelastic coupling in this material.

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I. INTRODUCTION

High-temperature superconductivity emerges in the proximity of an antiferromagnetic (AF) ground state in several closely related families of iron pnictides.[1] Interestingly, the AF transition in most of these materials is either preceded by or coincidental with a structural transition, which lowers the system's symmetry from tetragonal to orthorhombic.[2] It is generally believed that insights into the physics of this antiferromagnetism will lead to a better understanding of the origin of high-temperature superconductivity in this family of compounds. To fully understand the magnetic ground state, it is crucial to measure the intrinsic in-plane anisotropy. However, the formation of structural twins in the orthorhombic crystals,[3,4] makes such a measurement very challenging, and one would ideally like to find a simple method to detwin these materials and hence reveal the underlying in-plane anisotropy.

In 2002, Lavrov and co-workers[5] performed a series of magnetotransport measurements accompanied by direct optical imaging on lightly doped La$_{2-x}$Sr$_x$CuO$_4$ which has an orthorhombic transition around 450 K, showing that it can be detwinned by a 14 T magnetic field. Encouraged by this result, we have explored the possibility of affecting the structural/magnetic domains of iron pnictides in a similar manner, choosing Co-doped BaFe$_2$As$_2$ as a starting point. By doping with cobalt the single structural/magnetic transition of the undoped parent compound splits into two: the system first undergoes a tetragonal to orthorhombic structural transition (T_s), then enters a collinear AF state at a lower temperature (T_N).[6–10] Homogeneous doping and large single crystals, with different transition temperatures T_s and T_N, makes this an ideal material for such a study.

In this paper, we show via a combination of angle-dependent magnetoresistance (MR) measurements and direct optical images how magnetic fields can be used to partially detwin underdoped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$. Our observations imply a surprisingly large in-plane electronic anisotropy below T_N. Based on the generic susceptibility anisotropy anticipated for the collinear AF structure, we infer a smaller resistivity along the AF ordering direction ($\rho_a < \rho_b$). These results have important consequences for theoretical treatments both of the electronic properties of underdoped iron pnictides and of the structural transition itself.

II. EXPERIMENTAL METHODS

Single crystals of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ were grown from a self-flux, as described previously.[6] Electrical contacts were made in a standard four-point configuration, with current contacts across the ends of the crystal, and voltage contacts on the exposed (001) surface. Angle-dependent magnetotransport measurements were made in fields up to 14 T. All the measurements were performed with both current and field parallel to the FeAs plane. Samples were cut into bar shapes of typical dimension 1 mm × 0.2 mm × 0.05 mm and the crystal axes determined by x-ray diffraction.[11] Except for the first set of experiments shown in Fig. 1, for which we deliberately varied the current orientation, all the measurements were taken with current along the orthorhombic a/b direction. Optical measurements were performed in a superconducting magnet cryostat, in which the field could be varied between 0 and 10 T. The sample was illuminated with linearly polarized light and viewed through an almost fully crossed polarizer in order to maximize the contrast of birefringence between neighboring domains. An aspheric objective lens with a numerical aperture (NA) = 0.68 was placed inside the cryostat to focus polarized light onto the sample. The sample was positioned inside the cryostat using piezoelectric slip-stick positioners, and mounted in an identical manner for both transport and optical measurements in such a way as to minimize external stress.

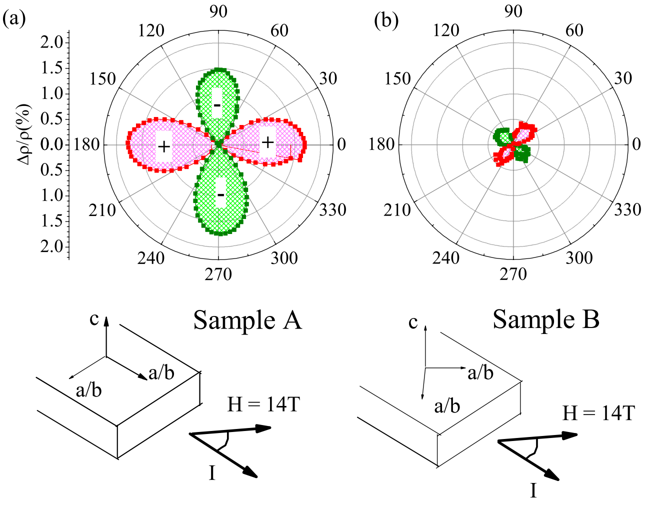

FIG. 1. (Color online) The in-plane magnetoresistance $\Delta\rho_{ab}/\rho_{ab}$ (%) of Ba$(\mathrm{Fe}_{1-x}\mathrm{Co}_x)_2\mathrm{As}_2$ for x=2.5% for two specific current orientations (samples A and B). The angle independent part of the magnetoresistance for sample B has been subtracted for clarity. The data were taken at 84 K, below both T_s and T_N for this cobalt concentration. The geometry of the measurement is indicated below each panel. Both current and field are in the *ab* plane and the angle of the magnetic field is measured relative to the current direction. For sample A current is applied parallel to the orthorhombic a/b axis whereas for sample B current is applied at 45° to the orthorhombic a/b axis.

III. RESULTS

For the first set of experiments, samples of Ba$(\mathrm{Fe}_{1-x}\mathrm{Co}_x)_2\mathrm{As}_2$ were cut such that the direction of the current varied with respect to the crystal axes. Representative data are shown in Fig. 1 for x=2.5% with current running along the orthorhombic [100] and [010] directions (sample A) and along the orthorhombic [110] direction (sample B). The configuration is illustrated schematically below each panel. At 84 K, which is below both the structural and magnetic transitions for this cobalt concentration (T_s =99 ± 0.5 K and T_N=93 ± 0.5 K, respectively[8]), we applied 14 T and rotated the field within the FeAs plane to measure the resistivity as a function of angle. The angle-dependent MR of sample A is shown in the polar plot Fig. 1(a). It has a twofold symmetry (as reported previously by Wang *et al.*[12]) with a positive MR for fields aligned parallel to the current and negative when the field is perpendicular to the current. The angle-dependent MR of sample B is shown in the polar plot Fig. 1(b). It also has a twofold symmetry but the magnitude is much smaller and the angle is shifted by 45°. Clearly the twofold MR is tied to the crystal axes and not to the current orientation.

To investigate the origin of this angle-dependent MR, we performed a detailed temperature-dependent map close to the magnetic and structural transition temperatures. Representative data for x=2.5% and 3.6%, in a 14 T magnetic field and using the "sample A" configuration (i.e., current along orthorhombic a/b axis) are shown in Fig. 2. These data are shown together with the temperature derivative of resistivity, which can be used to determine the two transition temperatures.[6,8,9]

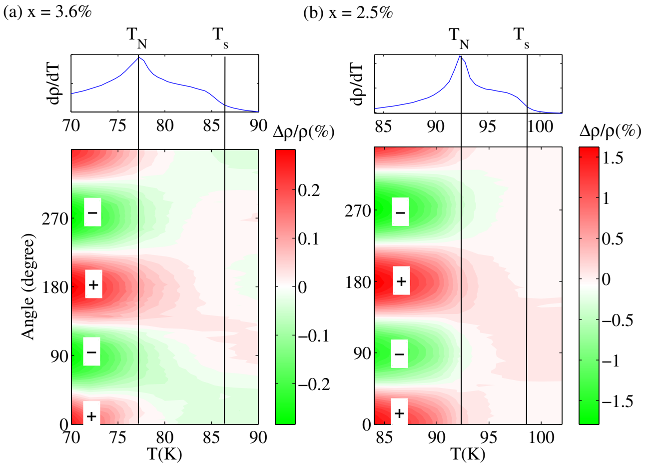

FIG. 2. (Color online) The angle-dependent magnetoresistance $(\rho_{ab}(\theta,14T)-\overline{\rho_{ab}})/\overline{\rho_{ab}}$ [where $\overline{\rho_{ab}}$ is the average of $\rho_{ab}(\theta,14T)$ over θ] as a function of angle and temperature of Ba$(\mathrm{Fe}_{1-x}\mathrm{Co}_x)_2\mathrm{As}_2$ for (a) x=3.6% and (b) x=2.5%. The magnitude of MR is indicated by the color scale. Vertical lines indicate T_s and T_N, as determined for these samples by the derivative of the resistivity (shown above each panel).

The magnitude of the twofold MR clearly rises sharply as the samples are cooled below T_N, although a very weak signal is visible up to almost T_s for the higher cobalt concentrations.[13]

To further investigate this effect, MR measurements were performed as a function of angle and field down to even lower temperatures. Representative data for x=1.6% are shown in Fig. 3. For temperatures close to T_N, the MR follows an almost perfect sinusoidal angle dependence with minimal hysteresis as the angle between the field and the current is swept from 0° to 360° and back to 0 again [Fig. 3(b)]. The MR follows a B^2 field dependence for the entire field range with a slight indication of some small hysteresis as the field is cycled back to zero [Fig. 3(a)]. Upon cooling to lower temperatures [for instance, to 20 K, as shown in panels (c) and (d) of Fig. 3], the field dependence of the MR develops an apparent threshold behavior, evidenced by a distinct upward (downward) kink in the MR for fields parallel (perpendicular) to the current. The MR also develops a substantial hysteresis. Both effects are also evident in the angle dependence of the MR [Fig. 3(d)], which develops distinct shoulders, presumably related to the threshold behavior observed in the field dependence. The absolute value of the MR in 14 T is comparable at the two temperatures shown. It is therefore unlikely that the predominant effect arises from orbital motion of the conduction electrons, which is ordinarily suppressed at higher temperatures according to Kohler's rule.[14] Rather, the behavior shown in Fig. 3 is indicative of field-driven changes in the sample. The angle dependence implies that the projection of the applied field on to specific crystal orientations must exceed a specific threshold value to induce these changes at low temperatures.

The changes described above appear to be thermally assisted, such that the associated relaxation after the field is cycled to zero is much slower (essentially frozen on laboratory time scales) at lower temperatures, resulting in a larger hysteresis. Evidence supporting this view can be obtained

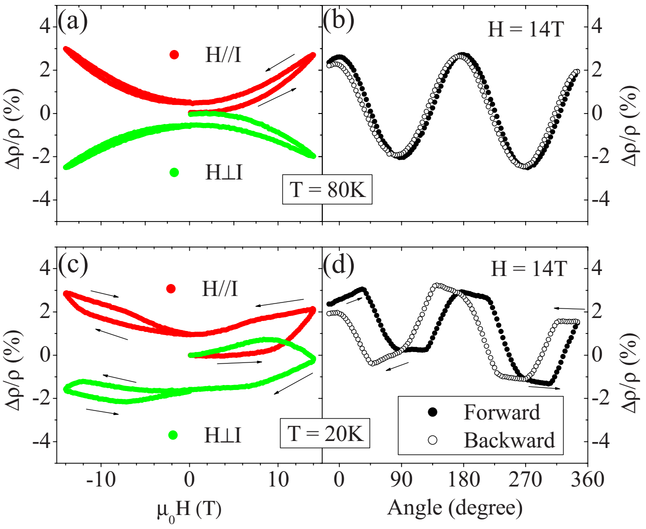

FIG. 3. (Color online) Representative magnetoresistance data for $Ba(Fe_{1-x}Co_x)_2As_2$ as a function of field and angle for $x=1.6\%$ at $T=80$ K [panels (a) and (b)] and $T=20$ K [panels (c) and (d)]. For all cases the current is aligned parallel to orthorhombic a/b axes and field is applied parallel to the *ab* plane (i.e., sample A configuration). For field sweeps, the magnetic field was swept from 0 to 14 T, then 14 to −14 T, then back to 0T following an initial zero-field cool. Data were taken for fields aligned parallel (red) and perpendicular (green) to the current. Angle sweeps were performed in a field of 14 T following an initial zero-field cool. Data were taken continuously as the angle was increased from 0° to 360° and then back to zero. The nonzero offset in (c) and (d) results from a slight mixing of out of plane field component due to a small sample misalignment.

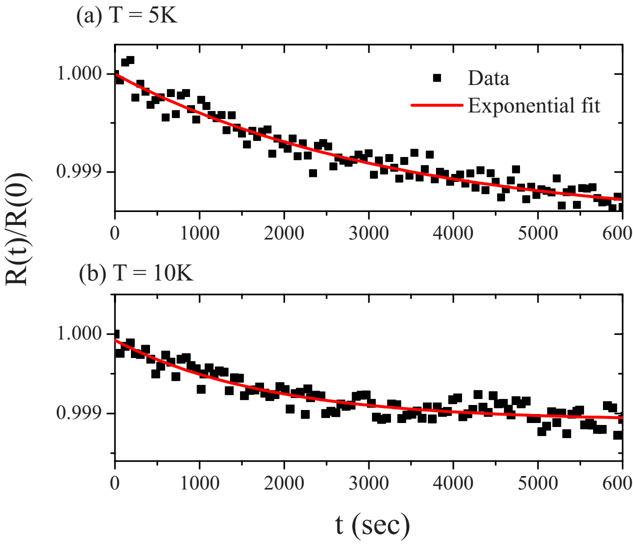

FIG. 4. (Color online) Time dependence of the resistivity for a representative sample with $x=1.6\%$, revealing thermal relaxation of the field-induced changes in the resistivity. The sample was initially cooled to 2 K in 14 T, before the field was cycled to zero. The sample was then warmed to (a) 5 K and (b) 10 K, and the resistivity measured as a function of time after the temperature had stabilized. Data are fit by an exponential relaxation with resulting time constants of $3.3\pm1.8\times10^3$ and $1.8\pm0.4\times10^3$ s, respectively.

from time-dependent measurements. Figure 4 shows resistivity data as a function of time for a sample with $x=1.6\%$. The sample was cooled from above T_s to 2 K in a field of 14 T (with the field parallel to the current, following the sample A configuration), at which temperature the field was cycled to zero. The sample was then warmed to various temperatures (data are shown for 5 and 10 K, in Fig. 4) and the resistivity measured as a function of time. Clear evidence for relaxation is observed with a relaxation time that grows with decreasing temperature.

In order to maximize the field-induced changes in the resistivity, samples can be cooled through the structural/magnetic transitions in an applied magnetic field. Representative data are shown in Fig. 5 for $x=1.6\%$ and 2.5% for fields applied both parallel and perpendicular to the current (still employing the sample A configuration). Measurements were taken while cooling in an applied field of 14 T (field cool FC), and then while warming in zero field (zero field warm, ZFW) after cycling the field to zero at base temperature. The resistivity difference induced by field cooling along one orientation can be as large as 5% at low temperature, even after the field is cycled to zero. The FC MR and ZFW resistivity difference for the two field configurations are also plotted in (b) and (d) for the two cobalt concentrations. The sign of MR for the two field orientations is opposite, and the absolute value appears to follow the magnetic order parameter, developing rapidly below T_N. A positive background in the FC MR data can be observed for both orientations, the magnitude of which increases as temperature is decreased, which is likely due to the ordinary metallic MR. In contrast, the resistivity difference in ZFW cycles appears to be rather symmetric. Its magnitude converges rapidly as the temperature is increased, consistent with the thermally assisted relaxation described above, and with the hysteresis effect observed in field sweeps as a function of temperature.

Direct optical measurements were also performed to observe the twinning domains on the surface of $Ba(Fe_{1-x}Co_x)_2As_2$. Representative images of a 2.5% sample at $T=40$ K are shown in Fig. 6. Data were taken in zero field and also after sweeping to 10 T. The presence of vertical stripes in both images indicates that the sample was twinned in zero and 10 T field. However, the relative density of the two domains is changed by application of the magnetic field. To track the difference in domain distributions, we plot the intensity profile across the boundaries, since the intensity depends on the crystal axes' orientation with respect to the light polarization. The twinning boundaries between two adjacent domains can then be determined from the maximum of the derivative of the intensity, from which we calculate the percentage of the volume of the domains of high intensity $[f=V_b/(V_a+V_b)]$. For the two images shown in Fig. 6, f increases from $54\pm1\%$ to $61\pm1\%$ a difference $\Delta f=7\%$ upon application of 10 T. Several regions in the optical images were analyzed, yielding percentage differences Δf ranging from 5% to 15%. Despite the large range in Δf, which is due to the spatial variation in domain distribution, it is always positive, providing convincing evidence for partial field-induced detwinning.

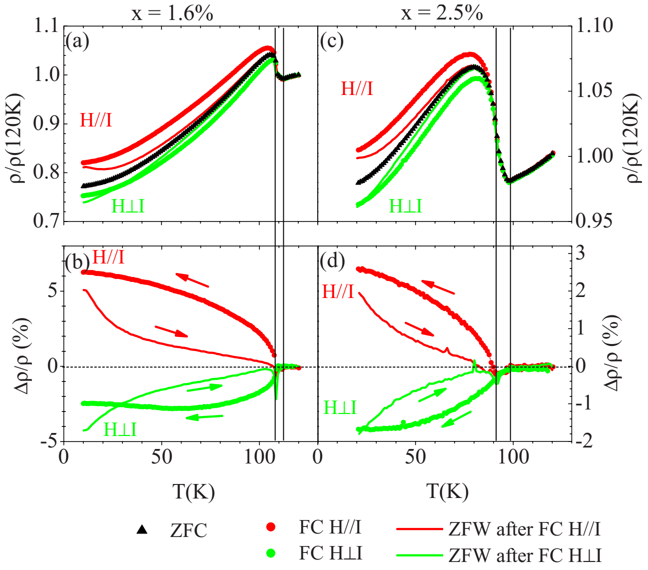

FIG. 5. (Color online) Temperature dependence of the resistivity for representative samples with $x=1.6\%$ [panels (a) and (b)] and $x=2.5\%$ [panels (c) and (d)]. Data were taken during an initial field cool (FC, solid symbols) in 14 T, after which the field was cycled to zero and the resistivity measured in zero field while warming (ZFW, thin lines). The current is aligned parallel to the a/b orthorhombic axes and the field applied either parallel (red) or perpendicular (green) to the current. For comparison, data are also shown for the same samples while cooled in zero field (ZFC, black symbols). The FC MR $[(\rho_{FC}-\rho_{ZFC})/\rho_{ZFC}]$ and ZFW resistivity difference induced by field cooling $[(\rho_{ZFW}-\rho_{ZFC})/\rho_{ZFC}]$ for the two field configurations are also plotted in (b) for $x=1.6\%$ and (d) for $x=2.5\%$. Vertical lines indicate T_s and T_N.

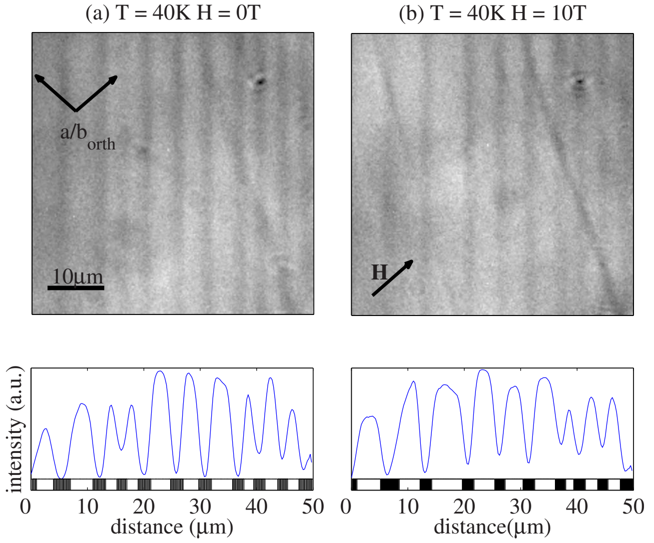

FIG. 6. (Color online) Representative optical images of a $\mathrm{Ba(Fe_{1-x}Co_x)_2As_2}$ sample for $x=2.5\%$, revealing the partial detwinning effect of an in-plane magnetic field. The images (obtained as described in the main text) were taken at $T=40$ K below both T_s and T_N. The initial image (a) was taken in zero field, following a zero field cool from above T_N. The field was then swept to 10 T, at which field the second image (b) was taken. Horizontal intensity profiles, shown below each image, were calculated by integrating vertically over the image area after background subtraction and noise filtering. Boundaries between domains were estimated as described in the main text, resulting in an estimate of the relative fraction of the two domains indicated by black and white stripes below the figures. The field, which was oriented along the orthorhombic a/b axes, has clearly moved the twin boundaries, favoring one set of twin domains (light) over the other (dark).

IV. DISCUSSION

The origin of the detwinning effect described above is presumably related to the anisotropic in-plane susceptibility ($\chi_a \neq \chi_b$) that must develop below T_N, and indicates a substantial magnetoelastic coupling. Since the magnetic structure is collinear, with moments oriented along the long a axis (referred to the orthorhombic unit cell),[2] we can anticipate that $\chi_b > \chi_a$. In this case, fields oriented along the a/b axis of a twinned crystal will favor domains with the b axis oriented along the field direction. Therefore for currents applied along the a/b axis and fields parallel to the current, the resistivity comprises a larger component of ρ_b than ρ_a. For fields aligned perpendicular to the current the opposite is true. Assuming $\mathrm{Ba(Fe_{1-x}Co_x)_2As_2}$ possesses the generic anisotropic susceptibility of collinear antiferromagnets, the observation of a positive MR for fields aligned parallel to the current, in concert with the relatively small degree of partial detwinning evidenced from optical images, implies a substantial in-plane resistivity anisotropy below T_N with $\rho_a < \rho_b$.[15] Subsequent measurements of mechanically detwinned samples of Co-doped $\mathrm{BaFe_2As_2}$ confirm this deduction.[16]

This result has some important consequences. Intuitively one might expect to find that $\rho_a > \rho_b$, both because the a-axis lattice constant is larger than the b-axis lattice constant (which all else being equal would lead to a larger orbital overlap), and also because the spin-density-wave wave vector is directed along the a axis (whereas the magnetic order is ferromagnetic along the b direction, which would ordinarily lead to a lower resistivity). Local-density approximation (LDA) calculations of the reconstructed Fermi surface for $\mathrm{BaFe_2As_2}$ (incorporating a negative U to reduce the moment to the observed value)[17] indicate a vanishingly small anisotropy in the plasma frequency.[18] The observed strong anisotropy in the resistivity for Co-doped $\mathrm{BaFe_2As_2}$ points toward either an unanticipated k dependence of the scattering rate, or a stronger variation in the orbital character around the Fermi surface than is predicted by LDA.

V. CONCLUSIONS

In summary, we have shown how a magnetic field can be used to partially detwin single crystals of underdoped iron pnictides, opening an avenue for research in to their anisotropic electronic properties. Our initial experiments have focused on the electron-doped system $\mathrm{Ba(Fe_{1-x}Co_x)_2As_2}$, but given the generic collinear AF structure found in this family of compounds, it is likely that the effect is quite general, limited only by details of the twin-boundary pinning and the strength of field available. Given the modest degree of detwinning achieved by the 14 T laboratory field, our experiments reveal a surprisingly large in-plane resistivity anisotropy. Based on the anticipated in-plane susceptibility anisotropy associated with the collinear AF structure, we infer a smaller resistivity along the AF ordering direction. Understanding the source of this anisotropy will be a key step toward establishing the origin of superconductivity in these materials.

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[1] See review by K. Ishida, Y. Nakai, and H. Hosono, J. Phys. Soc. Jpn. **78**, 062001 (2009), and references therein.

[2] See recent review by J. W. Lynn and P. Dai, Physica C **469**, 469 (2009), and references therein.

[3] C. Ma, H. X. Yang, H. F. Tian, H. L Shi, J. B. Lu, Z. W. Wang, L. J. Zeng, G. F. Chen, N. L. Wang, and J. Q. Li, Phys. Rev. B **79**, 060506(R) (2009).

[4] M. A. Tanatar, A. Kreyssig, S. Nandi, N. Ni, S. L. Budko, P. C. Canfield, A. I. Goldman, and R. Prozorov, Phys. Rev. B **79**, 180508(R) (2009).

[5] A. N. Lavrov, S. Komiya, and Y. Ando, Nature (London) **418**, 385 (2002).

[6] J.-H. Chu, J. G. Analytis, C. Kucharczyk, and I. R. Fisher, Phys. Rev. B **79**, 014506 (2009).

[7] N. Ni, M. E. Tillman, J.-Q. Yan, A. Kracher, S. T. Hannahs, S. L. Budko, and P. C. Canfield, Phys. Rev. B **78**, 214515 (2008).

[8] C. Lester, Jiun-Haw Chu, J. G. Analytis, S. C. Capelli, A. S. Erickson, C. L. Condron, M. F. Toney, I. R. Fisher, and S. M. Hayden, Phys. Rev. B **79**, 144523 (2009).

[9] D. K. Pratt, W. Tian, A. Kreyssig, J. L. Zarestky, S. Nandi, N. Ni, S. L. Budko, P. C. Canfield, A. I. Goldman, and R. J. McQueeney, Phys. Rev. Lett. **103**, 087001 (2009).

[10] A. D. Christianson, M. D. Lumsden, S. E. Nagler, G. J. MacDougall, M. A. McGuire, A. S. Sefat, R. Jin, B. C. Sales, and D. Mandrus, Phys. Rev. Lett. **103**, 087002 (2009).

[11] The susceptibility of $\mathrm{Ba(Fe_{1-x}Co_x)_2As_2}$ is typically very small ($\sim 10^{-3}$ emu/mol) (Ref. 6), therefore the difference in demagnetization along different directions is negligible.

[12] X. F. Wang, T. Wu, G. Wu, R. H. Liu, H. Chen, Y. L. Xie, and X. H. Chen, New J. Phys. **11**, 045003 (2009).

[13] Variation in the absolute value of the MR for different samples with the same Co concentration indicates that the extent of the partial detwinning effect that we observe depends sensitively on extrinsic parameters, including strain. Consequently, we do not associate any significance to the difference in the MR between samples with different Co concentrations.

[14] A. B. Pippard, *Magnetoresistance in Metals* (Cambridge University Press, London, 1989).

[15] Partial detwinning reduces the twin boundary density and therefore presumably decreases the contribution to the overall scattering rate from twin boundary scattering. However this effect cannot explain the permanent *increase* in resistivity for fields parallel to the current at low temperature. Moreover the twin boundaries extend along orthorhombic [110] direction (Refs. 3 and 4) hence any scattering process would contribute equally to the resistivity along *a* and *b* axes, inconsistent with the observed anisotropy.

[16] J. Chu, J. Analytis, K. De Greve, P. McMahon, Z. Islam, Y. Yamamoto, and I. Fisher, arXiv:1002.3364 (unpublished).

[17] J. G. Analytis, R. D. McDonald, J.-H. Chu, S. C. Riggs, A. F. Bangura, C. Kucharczyk, M. Johannes, and I. R. Fisher, Phys. Rev. B **80**, 064507 (2009).

[18] L. Fang, H. Luo, P. Cheng, Z. Wang, Y. Jia, G. Mu, B. Shen, I. Mazin, L. Shan, C. Ren, and H. Wen, Phys. Rev. B **80**, 140508(R) (2009).