J Supercond Nov Magn (2013) 26:2593–2596
DOI 10.1007/s10948-013-2141-4

ORIGINAL PAPER

Dynamics of Photoexcited Carriers in $Ba(Fe_{1-x}Co_x)_2As_2$ Single Crystals with Spin-Density-Wave Ordering

L. Stojchevska · T. Mertelj · Juin-Haw Chu · Ian R. Fisher · D. Mihailovic

Received: 2 December 2012 / Accepted: 14 February 2013 / Published online: 16 March 2013

Abstract We apply the femtosecond optical pump-probe spectroscopy to study the relaxation dynamics in photoexcited Co-doped $Ba(Fe_{1-x}Co_x)_2As_2$ single crystals in the underdoped spin-density wave (SDW) state region of the $x - T$ phase diagram. Underdoped SDW samples with Co-0 % and Co-2.5 % show a bottleneck in the relaxation as a consequence of the partial charge gap opening in the orthorhombic SDW phase, similar to previous results in other SDW iron-pnictides. Moreover, the charge gap magnitude decreases with increasing doping. The sample with Co-5.1 % displays both a SDW ordering and superconductivity at low T. We were able to observe a 2-fold anisotropy in our samples, existing up to ∼200 K, without any applied uniaxial stress. We associate the 2-fold symmetry breaking in nominally tetragonal phase with nematic ordering of the Fe d orbitals.

Keywords Femtosecond optical spectroscopy · Iron-based pnictides · Spin-density-wave systems · Carrier relaxation dynamics

L. Stojchevska (✉) · T. Mertelj · D. Mihailovic
Complex Matter Department, Jozef Stefan Institute, Jamova 39, 1000 Ljubljana, Slovenia
e-mail: ljupka.stojchevska@ijs.si

J.-H. Chu · I.R. Fisher
Geballe Laboratory for Advanced Materials and Department of Applied Physics, Stanford University, Stanford, CA 94305, USA

J.-H. Chu · I.R. Fisher
SLAC National Accelerator Laboratory, Stanford Institute for Materials and Energy Sciences, 2575 Sand Hill Road, Menlo Park, CA 94025, USA

1 Introduction

Discovery of the superconductivity in the iron-based pnictides [1–3] set up plenty of new challenges for the researchers worldwide. Even though there have been significant experimental and theoretical attempts to reveal the puzzles related to their properties, we still have not been able to answer all open questions. In addition, different experimental techniques [4–12] have revealed some unusual properties of the normal state, such as pseudogap and normal-state electronic nematic fluctuations. In the cuprates, the pseudogap is omnipresent [13–17], whereas in the iron-based pnictides its omnipresence is still questionable [18, 19]. Similarly as in cuprates, understanding the unusual normal state is important for explaining the mechanism leading to the superconductivity in iron-based pnictides.

It has been shown that the femtosecond optical spectroscopy is a powerful tool not only for real-time studies of the dynamics of photoinduced processes and determination of the life-time of the excited state, but also for separation of different processes since it can distinguish between different excitations by their different relaxation dynamics.

In this paper we present a femtosecond optical spectroscopy study of the quasi-particle relaxation dynamics in $Ba(Fe_{1-x}Co_x)_2As_2$ for the orthorhombic underdoped SDW ordering region of the phase diagram [20]. The aim of the experiments presented here [24] is systematic study of the temperature dependence of the optical reflectivity transients as a function of Co doping, for $x = 0$ %, 2.5 % and Co-5.1 %.

2 Experimental Details

The experiments were performed using standard pump-probe technique as described in detail elsewhere [21]. The

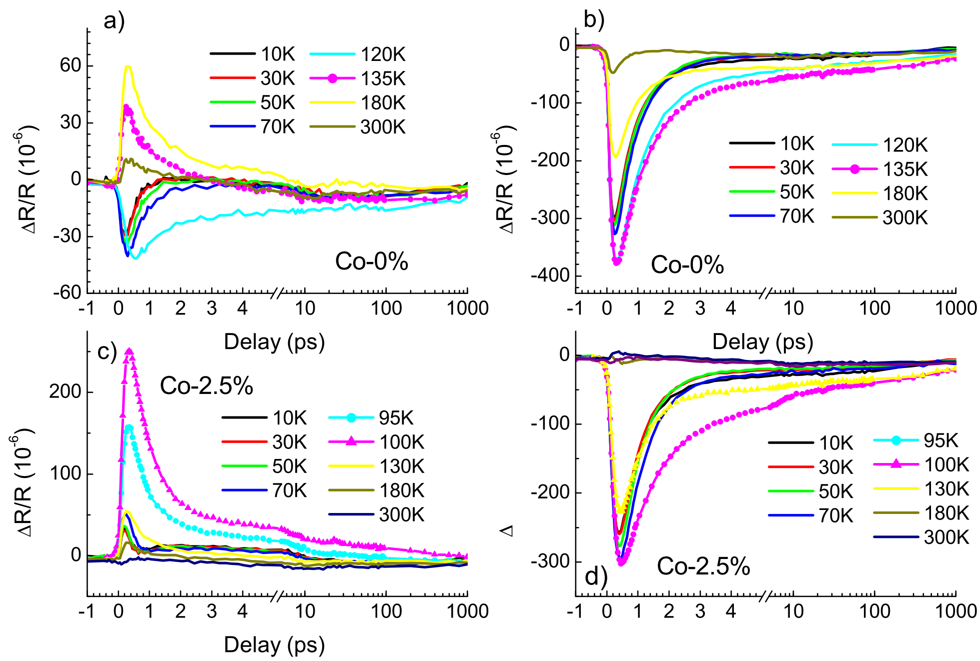

Fig. 1 The photoinduced reflectivity $\Delta R/R$ transients at 13 μJ/cm^2 pump fluence as a function of temperature at Co-0 % (**a**, **b**) and Co-2.5 % (**c**, **d**) doping. The *left column* corresponds to the $\mathcal{P}^+$ and the *right one* to the $\mathcal{P}^-$ polarization. *Full circled line* represents the T_{SDW}, where the *full triangled line* refers to T_s (obtained from the phase diagram [20])

samples were excited with 50 fs laser pulses generated from a 250-kHz Ti:sapphire regenerative amplifier seeded with Ti:sapphire oscillator. The pump photons were at 3.1 eV photon energy and for the probe we had 1.55 eV. Pump and probe beams were perpendicularly polarized to each other and oriented relatively to the crystals in a way to obtain maximum/minimum response amplitude at low temperatures.

Single crystals of $Ba(Fe_{1-x}Co_x)_2As_2$ with Co doping $x = 0$ %, 2.5 % and 5.1 % were grown from a self-flux and characterized as described elsewhere [9, 12, 20]. Samples with Co-0 % and Co-2.5 % belong to the underdoped region of the phase diagram with SDW ordering, where Co-5.1 % sample displays both a SDW and superconductivity at low T. Before mounting the crystals in a liquid-He flow cryostat, they were glued onto a copper holder and cleaved by a razor which resulted in a terrace like surface with a width of a few μm and step height of a few nm.

3 Results and Discussion

In Fig. 1 we plot the temperature dependence of the photoinduced reflectivity transients for underdoped Co-0 % and Co-2.5 % samples. One of the most astonishing feature of our experimental results is the observation of the 2-fold rotational anisotropy in nominally tetragonal phase at $T <$ 200 K. For easier following of the presented experimental results, we denote the polarization according to the low-T minimum and maximum $\Delta R/R$ peak as $\mathcal{P}^-$ and $\mathcal{P}^+$, respectively. Even though we were not able to determine the precise origin of the 2-fold rotational anisotropy, we believe that it might appear as a consequence of the anisotropic component of the surface strain induced by the laser heating in the presence of the unidirectional terraces on the surface of the cleaved crystals. Since the increase of the temperature in samples during the experiments is of the order of a few K and there is not any external magnetic field to order spins, we believe that the 2-fold symmetry breaking is related with nematic fluctuations or ordering of the Fe d orbitals.

3.1 Temperature Dependence

The undoped $Ba(Fe_{1-x}Co_x)_2As_2$ samples exhibit similar response as in undoped $SrFe_2As_2$ [22] and SmFeAsO [23]. The initial relaxation dynamics in Co-0 % can be fitted well with a single exponential decay in both polarization, whereas in the Co-2.5 % and Co-5.1 % samples a multi-component relaxation is observed (see Fig. 2). To determine T-dependence of the $\Delta R/R$ transients, we apply three exponentially decaying component fit [25] and we denote components with A, B, and C. Component A with relaxation time $\tau_A \sim 0.2$ ps is visible below 150 K and it exists in all samples, except in Co-0 %. It is associated with the initial relaxation of the optically excited electrons towards the Fermi energy or the inter-band momentum scattering between the states near the Fermi energy at different parts if the Fermi surface. The second component B is present in all samples and its relaxation time τ_B shows a critical slowing down at T_s. At the end, by increasing the temperature above T_s component C becomes visible. Although it has the longest relaxation time, its behavior is similar to component B, both belonging to a single process with a non-exponential decay dynamics.

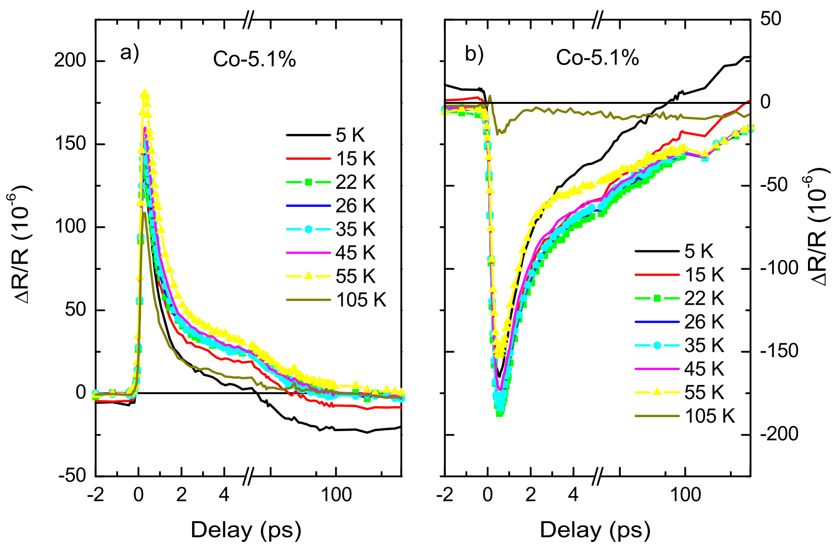

Fig. 2 Raw T-dependence of $\Delta R/R$ transients in Co-5.1 % sample, taken at 13 μJ/cm^2 pump fluence. (**a**) corresponds to the $\mathcal{P}^+$ and (**b**) to the $\mathcal{P}^-$ polarization. *Full circled line* represents the T_{SDW}, the *full triangled line* refers to T_s and *full squared line* applies to T_c (obtained from the phase diagram [20])

Table 1 Obtained charge gap magnitudes

x	$2\Delta(0)/k_B T_s$	$2\Delta_{\mathrm{PG}}$ (K)
0 %	9 ± 2	–
2.5 %	7 ± 2	–
5.1 %	4 ± 2	800 ± 100

To perform the analysis of the temperature dependence of the transients and therefore estimate the magnitude of the charge gap we apply a bottleneck model (see Fig. 3). Since the reflectivity transients display a characteristic shape related with the appearance of the bottleneck in the relaxation dynamics below T_s [13]:

$$n_{\mathrm{pe}} \propto 1/\left[\left(1+\frac{k_{\mathrm{B}}T}{2\Delta(T)}\right)\left(1+g_{\mathrm{ph}}\sqrt{\frac{k_{\mathrm{B}}T}{\Delta(T)}}\times \exp\left(-\frac{k_{\mathrm{B}}T}{\Delta(T)}\right)\right)\right] \tag{1}$$

$$\Delta R = \left(\gamma_0 + \eta\Delta(T^2)\right)n_{\mathrm{pe}} \tag{2}$$

The magnitudes of the charge gap (its values are summarized in Table 1) are consistent with previous results [17, 23] and decrease with increasing x as a consequence of a decrease of the stability of the SDW state.

In the superconducting sample Co-5.1 % (Figs. 2 and 4) we observe an additional component in the relaxation dynamics, visible below critical temperature T_c. Similarly to the previous studies [21], by changing the laser pulse intensity, the superconducting state is destroyed and the characteristic superconducting signal saturates when a certain laser fluence threshold $\mathcal{F}_T$ is reached. The SC component can be obtained by subtraction of the normal state $\Delta R/R$ signal above T_c from the transient measured at 5 K (Fig. 4).

Above T_s, tails in the T-dependent $\Delta R/R$ transients are visible which cannot be fitted with (2). They are related with the presence of a bottleneck in the relaxation and therefore

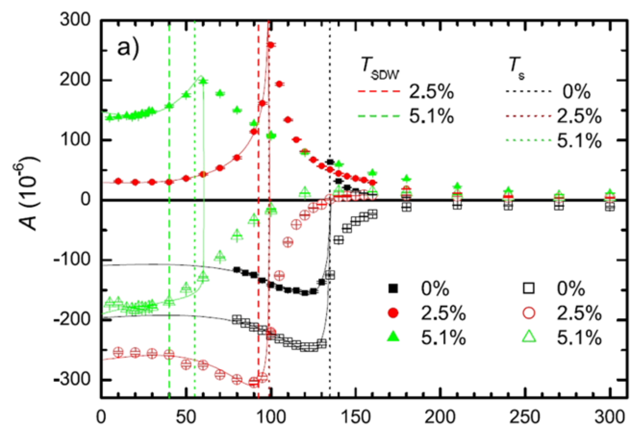

Fig. 3 The photoinduced reflectivity $\Delta R/R$ transients amplitudes as a function of temperature at different dopings. The *full and open symbols* correspond to the $\mathcal{P}^+$ and $\mathcal{P}^-$ polarization, respectively. *Thin lines* are fits of Eq. (2) discussed in the text. *Vertical lines* indicate T_s, T_{SDW} and T_c, obtained from Ref. [20]

with a pseudogap at the Fermi energy. In order to estimate the magnitude of the pseudogap, we fit the amplitudes of the T-dependence of $\Delta R/R$ signal with

$$\Delta R \propto n_{\mathrm{pe}} \propto \left[1 + g_{\mathrm{ph}}\exp\left(-\frac{k_{\mathrm{B}}T}{\Delta_{\mathrm{PG}}(T)}\right)\right]^{-1} \tag{3}$$

It is important to mention that estimated value of the pseudogap magnitude is in good agreement with previous studies [23] and has similar value as the spin pseudogap magnitude obtained from the Knight shift [6].

4 Conclusions

We present detailed study of the relaxation dynamics in $Ba(Fe_{1-x}Co_x)_2As_2$ single crystals by using femtosecond optical spectroscopy. In the SDW state we observe a bot-

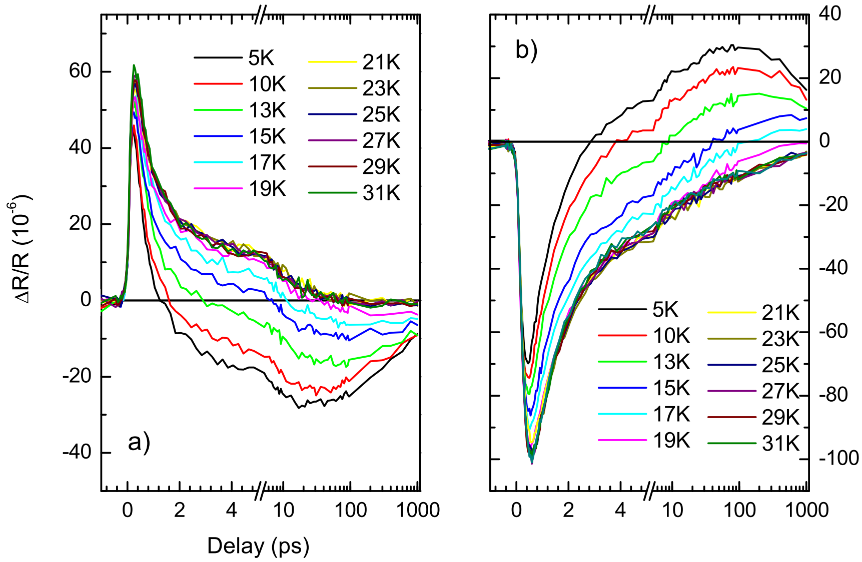

Fig. 4 Low temperature photoinduced reflectivity $\Delta R/R$ transients at 3.9 μJ/cm^2 pump fluence in Co-5.1 % where (**a**) corresponds to the $\mathcal{P}^+$ and (**b**) to the $\mathcal{P}^-$ polarization

tleneck in the relaxation dynamics due to the opening of a partial charge gap, similarly as in other iron-based pnictides. Its magnitude $2\Delta(0)/k_B T_s$ decreases with increasing of Co-doping, in agreement with a decrease of the orthorhombic/SDW state stability. We associate observed 2-fold symmetry breaking with nematic fluctuations or ordering of the Fe d orbitals. Breaking of the 4-fold symmetry appears due to the anisotropic boundary which introduce an anisotropic surface strain. Moreover, strain appears as a consequence of the local crystal expansion due to the local thermal load. We relate the anisotropy in nominally tetragonal phase of the strain to the unidirectional terraces on the cleaved surface, characterized with the atomic force microscope (AFM). Regarding the superconductivity existing in Co-5.1 % sample, an additional SC component in the relaxation dynamics is appearing, consistent with the previous related work. Moreover, the saturation of the amplitude of the SC signal for pump fluences above $\mathcal{F}_T$ is linked with non-thermal destruction of the SC state.

References

1. Kamihara, Y., Hiramatsu, H., Hirano, M., Kawamura, R., Yanagi, H., Kamiya, T., Hosono, H.: J. Am. Chem. Soc. **128**, 10012 (2006)
2. Kamihara, Y., Watanabe, T., Hirano, M., Hosono, H., et al.: J. Am. Chem. Soc. **130**, 3296 (2008)
3. Ren, Z., Che, G., Dong, X., Yang, J., Lu, W., Yi, W., Shen, X., Li, Z., Sin, L., Zhou, F., et al.: Europhys. Lett. **83**, 17002 (2008)
4. Liu, R.H., Wu, G., Wu, T., Fang, D.F., Chen, H., Li, S.Y., Liu, K., Xie, Y.L., Wang, X.F., Yang, R.L., Ding, L., He, C., Feng, D.L., Chen, X.H.: Phys. Rev. Lett. **101**, 087001 (2008)
5. Ahilan, K., Ning, F.L., Imai, T., Sefat, A.S., Jin, R., McGuire, M.A., Sales, B.C., Mandrus, D.: Phys. Rev. B **78**, 100501 (2008)
6. Ning, F., Ahilan, K., Imai, T., Sefat, A.S., Jin, R., McGuire, M.A., Sales, B.C., Mandrus, D.: J. Phys. Soc. Jpn. **78**, 013711 (2009)
7. Hess, C., Kondrat, A., Narduzzo, A., Hamann-Borrero, J.E., Klingeler, R., Werner, J., Behr, G., Buchner, B.: Europhys. Lett. **87**, 17005 (2009)
8. Mertelj, T., Kabanov, V., Gadermaier, C., Zhigadlo, N., Katrych, S., Karpinski, J., Mihailovic, D.: Phys. Rev. Lett. **102**, 117002 (2009)
9. Chu, J.-H., Analytis, J.G., De Greve, K., McMahon, P.L., Islam, Z., Yamamoto, Y., Fisher, I.R.: Science **329**, 824 (2010)
10. Chuang, T.-M., Allan, M.P., Lee, J., Xie, Y., Ni, N., Bud'ko, S.L., Boebinger, G.S., Canfield, P.C., Davis, J.C.: Science **327**, 181 (2010)
11. Tanatar, M.A., Ni, N., Thaler, A., Bud'ko, S.L., Canfield, P.C., Prozorov, R.: Phys. Rev. B **82**, 134528 (2010)
12. Dusza, A., Lucarelli, A., Pfuner, F., Chu, J.-H., Fisher, I.R., Degiorgi, L.: Europhys. Lett. **93**, 37002 (2011)
13. Kabanov, V.V., Demsar, J., Podobnik, B., Mihailovic, D.: Phys. Rev. B **59**, 1497 (1999)
14. Demsar, J., Podbik, B., Kabanov, V.V., Wolf, T., Mihailovic, D.: Phys. Rev. Lett. **82**, 4918 (1999)
15. Kusar, P., Demsar, J., Mihailovic, D., Sugai, S.: Phys. Rev. B **72**, 014544 (2005)
16. Kabanov, V.V., Demsar, J., Mihailovic, D.: Phys. Rev. Lett. **95**, 147002 (2005)
17. Stojchevska, L., Kusar, P., Mertelj, T., Kabanov, V.V., Toda, Y., Yao, X., Mihailovic, D.: Phys. Rev. B **84**, 180507 (2011)
18. Rullier-Albenque, F., Colson, D., Forget, A., Alloul, H.: Phys. Rev. Lett. **103**, 057001 (2009)
19. Lee, H.-S., Bartkowiak, M., Park, J.-H., Lee, J.-Y., Kim, J.-Y., Sung, N.-H., Cho, B.K., Jung, C.-U., Lee, H.-J.: Phys. Rev. B **80**, 144512 (2009)
20. Chu, J.-H., Analytis, J.G., Kucharczyk, C., Fisher, I.R.: Phys. Rev. B **79**, 014506 (2009)
21. Kusar, P., Kabanov, V.V., Demsar, J., Mertelj, T., Sugai, S., Mihailovic, D.: Phys. Rev. Lett. **101**, 227001 (2008)
22. Stojchevska, L., Kusar, P., Mertelj, T., Kabanov, V.V., Lin, X., Cao, G.H., Xu, Z.A., Mihailovic, D.: Phys. Rev. B **82**, 012505 (2010)
23. Mertelj, T., Kusar, P., Kabanov, V.V., Stojchevska, L., Zhigadlo, N.D., Katrych, S., Bukowski, Z., Karpinski, J., Weyeneth, S., Mihailovic, D.: Phys. Rev. B **81**, 224504 (2010)
24. Stojchevska, L., Mertelj, T., Chu, J.-H., Fisher, I.R., Mihailovic, D.: Phys. Rev. B **86**, 024519 (2012)
25. Mihailovic, D., Demsar, J.: In: Time-Resolved Optical Studies of Quasiparticle Dynamics in High-Temperature Superconductors, Spectroscopy of Superconducting Materials, pp. 230–244. American Chemical Society, Washington (1999)