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Photoinduced femtosecond relaxation of antiferromagnetic orders in the iron pnictides revealed by ultrafast laser ellipsometry

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Photoinduced femtosecond relaxation of antiferromagnetic orders in the iron pnictides revealed by ultrafast laser ellipsometry

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Abstract. We report ultrafast softening of the antiferromagnetic order, ~150fs after the electron thermalization, which follows a two-step recovery pathway to reveal a distinct interplay of magnetism and the nematic order in iron pnictides.

1 INTRODUCTION

Ultrafast studies of collective magnetic phenomena can provide fundamental insights into nonequilibrium processes of correlated spins. The key to understand and control these phenomena is the capability to directly reveal the spin fluctuations associated with the dynamic processes with femtosecond resolution, beyond time-averaged mean field values obtained from those static measurements. Although ultrafast spin spectroscopy and measurements are progressing well in ferromagnetic materials, femtosecond spin dynamics and dynamic critical phenomena of antiferromagnetic (AFM) orders are rarely addressed. AFM orders are extremely relevant for many "unconventional" materials since they are the key normal state properties out of which many elusive collective phenomena emerge, e.g., high-temperature superconductivity and colossal magnetoresistance. The correlation mechanisms determining the former are likely to be responsible for the latter. Thus far, most prior work on the ultrafast relaxation of AFM orders have focused on conventional, band insulators, where hundreds of ps magnetization quenching dynamics is revealed.

One prime example of such materials is the recently discovered high-T_c iron-based superconductors (FeSCs) [1]. FeSCs' exhibit a layered FeAs crystal structure with a complex phase diagram including: superconductivity, antiferromagnetism, Ising-nematic phase, orbital order, tetragonal and orthorhombic crystal structures [2]. One of the most perplexing behaviors of FeSCs occurs in the undoped (parent) and weakly doped antiferromagnetic compounds, where a significant in-plane anisotropy (broken C4 rotational symmetry) emerges in the temperature region between the striped AFM transition and the structural tetragonal-to-orthorhombic transition ($T_N<T<T_S$). Exactly what correlation mechanism can substantially break the symmetry above T_N remains controversial. Thus far, experimental evidence for the anisotropic states in FeSCs has mostly been from stationary measurements, and has shown an onset of the anisotropy to appear well above T_S, which is inconsistent with the theoretical predictions of the Ising nematic order parameter [3]. Particularly, there is also a competing view for the roles of the spin vs. orbital orders as the origin of anisotropy.

2 EXPERIMENTAL METHODS

Here we explore dynamics of the AFM and anisotropic states that allow decoupling various correlations in the time domain to differentiate their contributions to the origin of the anisotropy. We have performed pump-probe experiments utilizing two-color ultrafast laser ellipsometry, ΔA, techniques to probe genuine anisotropy dynamics in parent and weakly Co-doped compounds of the Fe-pnictide superconductor family $BaFe_2As_2$. Our measurements directly reveal femtosecond spin relaxation time of photoexcited AFM orders, which exhibit distinct dependence on the transition temperatures, T_N and T_S.

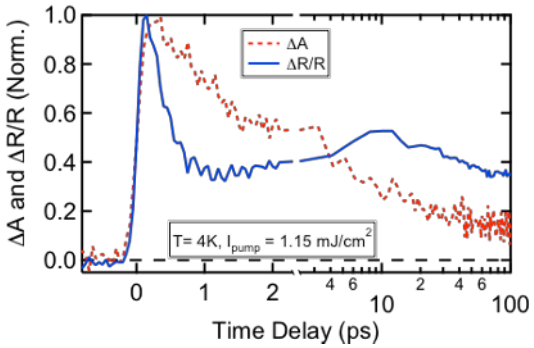

Fig. 1. Transient ellipticity ΔA and ΔR/R are shown for the first 2 ps and the extended time scales of 100 ps. The anisotropy and charge carriers show distinctly different relaxation dynamics in the time domain.

3 RESULTS AND DISCUSSIONS

A representative trace for the temporal profile of the ellipsometry and differential reflectivity, ΔR/R, is shown in Figure 1 for the parent compound $BaFe_2As_2$ at 4K. There are noticeably different anisotropy and charge relaxation dynamics both in the first few ps and also at extended time scales of 100 ps. The anisotropy has the slower initial decay with a time-constant ~2 ps after photoexcitation compared to the charge relaxation which is on the order of ~200 fs. The ΔR/R signal has some additional features not present in the ΔA signals, such as a periodical oscillation and a strongly damped, ps component.

Figure 2(a) further shows the peak amplitude of the ellipticity signal as a function of temperature. It is clearly visible that the signal quickly diminishes as the temperature approaches T_N = 136K, which indicates the anisotropy emerges at the onset of the antiferromagnetic order parameter. In accord, the anisotropy decay time-constant also shows a critical slowing down near T_N and indicates three-dimensionality. For the parent compound, photoinduced anisotropy signals show similar relaxation profiles below T_N, as demonstrated by the normalized traces at two temperatures, 4K and 130K.

We compared these results to that of an under-doped sample (4.7% doping) where the spin-density-wave/structural transition temperatures are far apart. Most intriguingly, the ultrafast ellipticity signals for the 4.7% doping show distinct transitions at both T_N~ 47K and T_S~ 66K. The fs relaxation component only exists below T_N and quickly diminishes above it, so we attribute this to the spin contributions. This provides a direct determination of the spin relaxation time of photo-melted AFM orders. In addition, the distinct fs component clearly shows the AFM orders strongly contribute to the anisotropic nematic phase, which brings the system to the verge of a magnetic transition in the undoped compound, or occurs before a magnetic transition to induce a magneto-structural phase transition in the underdoped case.

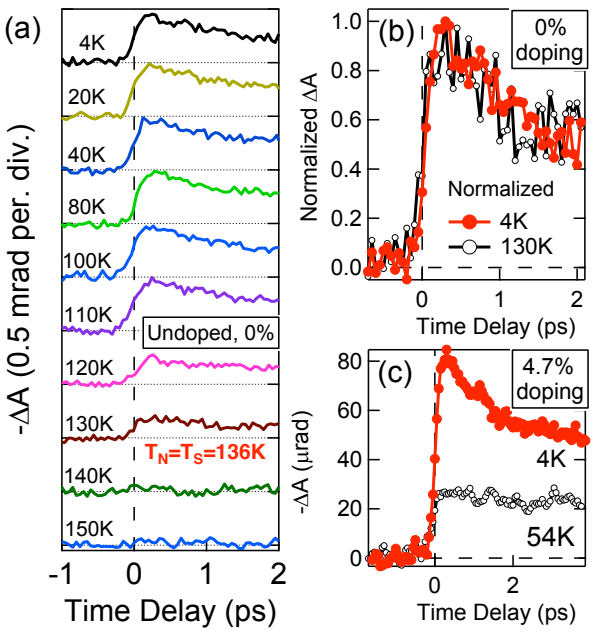

Fig. 2.(a) Temperature dependence of the transient ellipticity ΔA for the parent compound BaFe2As2 (0%) quickly diminishes with increasing the temperature and completely disappears at T_S/T_N. (b) Normalized ΔA for the paraent compound at two temperatures, 4K and 130K. (c) Transient ellipticity ΔA for an under-doped sample (4.7% doping) at two temperatures, 40K and 54K. The AFM transition temperature is ~47K.

4 CONCLUSIONS

In summary, we report fs spin relaxation in photoexcited undoped and weakly Cobalt doped $BaFe_2As_2$, which represents the first evidence to identify the dynamic interplay of antiferromagnetic correlation and the nematic phase in iron pnictides.

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