Observation of a coherent optical phonon in the iron pnictide superconductor $Ba(Fe_{1-x}Co_x)_2As_2$ (x=0.06 and 0.08)

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We report the observation of a coherent lattice oscillation in a pnictide superconductor. A coherent fully symmetric optical phonon was detected in $Ba(Fe_{1-x}Co_x)_2As_2$ (x=0.06 and x=0.08) using time-resolved pump-probe reflectivity with 40 fs time resolution. The analysis of the phonon parameters for various excitation fluences reveals no evident difference below and above the critical temperature, suggesting that the A_{1g} mode is not involved in the superconducting phase transition.

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Understanding the mechanism of high-temperature superconductivity is one of the most important open issues in condensed matter physics. A huge effort has been and is being devoted to the exploration of all the physical aspects of the materials presenting this property such as cuprates and pnictides. The study of lattice vibrations and of their role in the physics of these materials is an important part of this effort. While conventional superconductivity found a comprehensive description and explanation in the framework of the Bardeen-Cooper-Schrieffer (BCS) theory,[1] where the attractive interaction between electrons forming Cooper pairs is mediated by lattice oscillations, the role of phonons in unconventional superconductivity is still controversial.

The study of the behavior of specific phonon modes could shine some light on this open issue: in particular, coherent phonons can be excited and analyzed using pump-probe experimental schemes which became possible thanks to the advent of femtosecond lasers. By laser-pumping the system, a highly excited electronic state is photoinduced: afterwards, excited electrons and lattice interact through electron-phonon coupling, which may result in coherent atomic displacements[2–4] corresponding to one or several specific phonon modes. This approach has been employed on insulators, metals, semiconductors, and strongly correlated systems,[5,6] elucidating the role of phonons in phenomena such as metal-insulator[7,8] and ferroelectric[9] transitions.

High T_C superconductors have also been the subject of many pump-probe studies,[10,11] but it must be noted that the excitation of coherent optical phonons by means of time-resolved experiments has been only rarely reported. It was observed only in $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films[12–15] and single crystals[16] and in $NdBa_2Cu_3O_{7-\delta}$ single crystals.[17] No coherent phonons were detected so far in the few time-resolved reflectivity studies performed on iron-pnictide superconducting materials.[11,18,19]

In this Brief Report, we report the observation of a coherent lattice oscillation in an iron-pnictide high temperature superconductor, namely a fully symmetric optical phonon in $Ba(Fe_{1-x}Co_x)_2As_2$, for two dopings x=0.06 and x=0.08, observed with time-resolved reflectivity. Doped $BaFe_2As_2$ are with LaFeAsO the most studied compounds in this family of superconducting materials, which have triggered a worldwide intensive research effort since their recent discovery.[20,21] These compounds display a very interesting phase diagram, where structural, spin, and charge degrees of freedom compete.[22,23] An obvious similarity between cuprates and pnictides is their marked bidimensional structure, characterized by Cu-O and Fe-As layers, respectively. Nevertheless, distinct differences have already been identified: iron based pnictides exhibit metallic behavior and a spin-density wave state at low doping level,[24,25] instead of the antiferromagnetic Mott-Hubbard insulator state found in cuprate compounds. The compound studied here is Co-doped $BaFe_2As_2$, which presents a superconducting state for a range of doping between ≈3% and ≈15%. The undoped system exhibits a spin density wave state below 135 K and this phase progressively disappears during the onset of the superconducting phase, vanishing at a doping level of 7%. Single crystal samples were grown by the self-flux method,[26] and fully characterized prior to our measurements. We analyzed the coherent optical phonon in $Ba(Fe_{1-x}Co_x)_2As_2$ by means of time-resolved reflectivity, studying two doping levels x=0.06 and x=0.08, which become superconducting at T_C=21 and T_C=24 K,[26] respectively.

The experiments were carried out using a mode-locked Ti:sapphire system delivering laser pulses at 800 nm wavelength and 1 kHz repetition rate. The pulses duration was ≈40–50 fs. The experimental setup is the same as described in Ref. 4 allowing us to reach a signal-to-noise ratio up to 10^5. The pump beam was linearly p polarized, incident at about 5° from the surface normal, and was focused on a 100 μm diameter spot. On the other hand, the probe beam was s polarized and arrived at an incidence angle of approximately 15° with respect to the sample normal. The probe beam was focused on a smaller spot of around 25 μm diameter, in order to probe a uniformly excited area. We used pump fluences between 1.9 mJ/cm^2 and 5.9 mJ/cm^2, while the probe energy density was fixed at 0.95 mJ/cm^2. At lower probe energy density, the signal-to-noise ratio was not high enough to allow signal changes detection. We used a charge-coupled device camera to rigorously verify the pump and probe spatial overlap. A continuous flow helium-gas cryostat allowed us to cool the samples down to ≈10 K. The samples were cleaved along the (001) crystallographic direc-

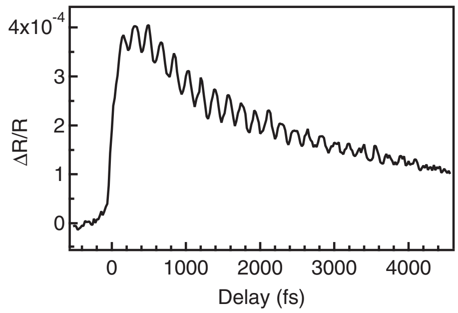

FIG. 1. Time-resolved reflectivity curve on $Ba(Fe_{1-x}Co_x)_2As_2$, $x=0.08$ at 10 K.

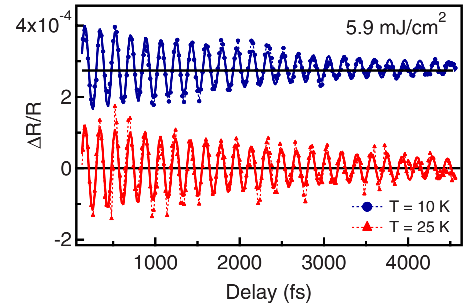

FIG. 2. (Color online) Coherent optical lattice oscillation on $Ba(Fe_{1-x}Co_x)_2As_2$, $x=0.08$ in the superconducting and metallic phases at 10 and 25 K, respectively. The curve at 10 K is vertically offset for clarity. The solid line is the oscillatory component of the fitting function.

tion in order to obtain clean and optically flat surfaces.

Figure 1 shows a typical reflectivity behavior versus the pump-probe time delay in the initial superconducting phase (the crystal temperature was 10 K), at a pump fluence of $3.2\ \mathrm{mJ/cm^2}$. A similar behavior is obtained at 25 K. The signal is characterized by a first increase just after the pump pulse arrival, due to fast electronic excitation. The signal reaches its maximum variation in around 100 fs. For longer time delays, the signal has mainly two contributions, i.e., an exponential relaxation, probably due to electron-phonon thermalization, superimposed to an oscillatory component with a period of 180 fs. We will focus hereafter only on this oscillating part, which is the signature of the coherent phonon.

The oscillation frequency is 5.56 THz, which corresponds to an energy of about 23 meV. By comparison with Raman data,[27] this coherent phonon can be identified as the fully symmetric A_{1g} optical mode. Its frequency does not change with the fluence in the measured range. This mode corresponds to a pure breathing displacement of As atoms,[28] and it is the only full symmetric mode in this structure.

We point out that our time resolution allows the detection of all four Raman active modes A_{1g}, B_{1g}, and two E_{1g}. Since only the A_{1g} mode is excited, this suggests that the phonon excitation mechanism is not impulsive stimulated Raman scattering,[29] which is commonly accepted as the excitation process in insulators and some semiconductors. Following this theory, all optical phonon modes may be excited and their relative excitations depend on the crystal orientation with respect to the laser pump polarization. Here, a possible scenario for coherent phonon excitation may be either displacive excitation of coherent phonons (DECP) (Ref. 30) or electronic temperature gradient,[4] which both successfully describe the selective excitation of the completely symmetric mode if it exists, as experimentally observed in several metallic systems. The excitation of coherent A_{1g} phonons in superconducting YBCO has been also described successfully by a generalized DECP mechanism, proposed by Mazin *et al.*,[31] which takes explicitly into account the ions displacement induced by the superconductivity. It should also be noted that when the crystal space group does not allow the existence of an A_{1g} phonon, other modes may be coherently excited, as for example in Zn.[32] In order to better compare the phonon behavior below and above the superconducting transition, in Fig. 2 we show the coherent oscillations for the two cases. The two curves (dotted lines with markers) were obtained by removing from the experimental data the exponential component of the fit. The solid lines are the oscillatory components of the fitting functions. We applied a vertical offset to the data taken at 10 K for clarity. This figure allows a direct comparison between the two cases, below and above the critical temperature: we notice that the phonon initial phase, amplitude, frequency, and damping time are very similar.

We fit the reflectivity changes behavior $\Delta R(t)$ shown in Fig. 2 by a damped harmonic oscillator,

$$\frac{\Delta R(t)}{R} = A_{ph} \sin(\omega t + \phi) e^{-t/\tau}. \qquad (1)$$

Here, R is the unperturbed reflectivity, t is the pump-probe time delay, whereas A_{ph}, ω, ϕ, and γ are the phonon amplitude, frequency, initial phase, and damping time, respectively.

Figures 3(a) and 3(b) show phonon damping time and amplitude for various excitation fluences for the same crystal temperatures, 10 and 25 K, respectively. We can immediately notice that the behavior of these phonon parameters shows a clear continuity as a function of the pump fluence, confirming the reliability of the optical response in this fluence range, and allowing us to explore this response in more detail.

In particular, the phonon amplitude increases with pump fluence at either temperature, as expected since the initial atomic displacement depends on the temperature gradient of the photo excited carriers, which increases as a function of the excitation level. The behavior at both temperatures is linear, but with different slopes: in particular, it is interesting to note that at the higher temperature the phonon amplitude is larger than at the lower temperature. This cannot be interpreted as a simple thermal effect, since usually a decrease of crystal temperature produces an increase of phonon amplitude.[33] This is confirmed in Fig. 3(c), which shows the phonon amplitude as a function of the temperature for a pump fluence of $4.5\ \mathrm{mJ/cm^2}$ for $x=0.06$; this amplitude slightly increases from 300 to 50 K; conversely, the points below T_C present a smaller oscillation amplitude than in the normal phase. Overall, we consistently found a reduction in the phonon oscillation amplitude while going from above to

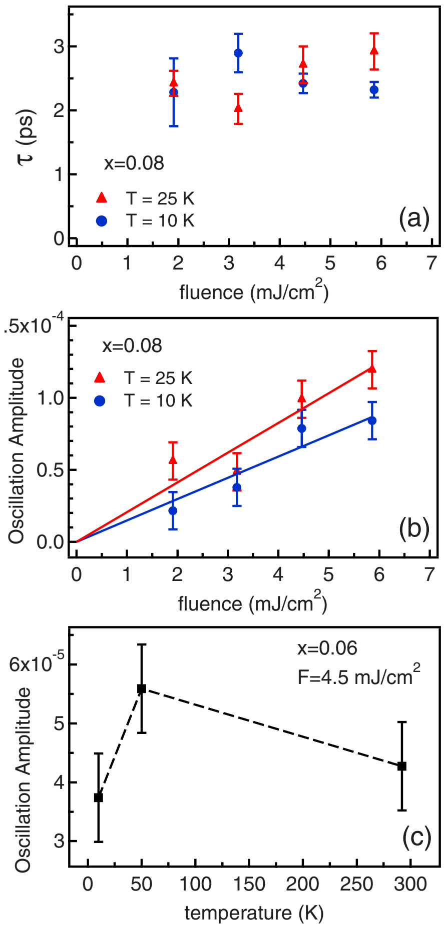

FIG. 3. (Color online) Phonon (a) damping time and (b) amplitude as a function of the pump fluence in $Ba(Fe_{1-x}Co_x)_2As_2$ $x=0.08$ for the superconducting and the metallic phases. The fitting line going through zero is shown on the amplitude graph. (c) Phonon amplitude versus temperature in $Ba(Fe_{1-x}Co_x)_2As_2$, $x=0.06$, for a pump fluence of 4.5 mJ/cm^2.

below T_C for all samples and fluences presented in Figs. 3(b) and 3(c). This lowering in coherent ions displacement below T_C could be related to the breaking of superconducting pairs induced by the pump pulse.

The phonon damping could be mainly due to two reasons. The first damping mechanism consists of the annihilation of one phonon resulting on the creation of two phonons of lower energy and higher wave vector, in order to fulfill the required energy and momentum conservation. This phonon-phonon transition is due to anharmonic effects. Increasing the excitation fluence results in a larger phonon amplitude, which brings the oscillations into an anharmonic regime, increasing the probability per unit time of phonon-phonon transitions. The second way for phonon damping is the phonon scattering by crystal impurities. As shown in Fig. 3(a), the phonon damping time at both temperatures remains approximately constant with fluence, ranging between 2 and 3 ps, whereas in most pump-probe reflectivity measurements the phonon damping time is reduced when the excitation fluence increases. This result indicates that the main phonon damping process in our $Ba(Fe_{1-x}Co_x)_2As_2$ single crystals is scattering by impurities. It also suggests that the pump fluence range used here is far below the anaharmonic regime excitation level.

The behavior of another phonon parameter, the frequency, corroborates the fact that the excited system is in the harmonic regime. Actually, the frequency of the coherent lattice oscillation is similar to the one measured with Raman spectroscopy, which means that we do not detect any redshift of the A_{1g} mode. Usually, when the pump fluence increases so that it brings the system into an anharmonic regime, atomic bonds become softer. Since we do not observe any softening of the phonon, it means that we stay in the low-excited state.

The only phonon parameter which is affected by the superconducting phase transition is the phonon amplitude. This dependence is due to the different electronic environment in the metallic and superconducting phases. However, the fact that neither the damping time nor the phonon frequency change between the two phases seems to indicate that the fully symmetric mode does not participate in the superconducting phase transition. On the other hand, this conclusion cannot be generalized, as we only deal with a coherent phonon with wave vector close to the center of Brillouin zone. We remark also that other phonon modes could instead participate to the phase transition.

In conclusion, we observed the coherent fully symmetric A_{1g} optical phonon by using time-resolved reflectivity in the superconducting and metallic phases of $Ba(Fe_{1-x}Co_x)_2As_2$, $x=0.06$ and $x=0.08$. The measured phonon frequency is 5.56 THz, which is in good agreement with standard Raman measurements and does not depend on the thermodynamic phase. The results show that at temperatures corresponding to the superconducting phase the phonon amplitude has a linear behavior versus the excitation fluence, but with a smaller slope with respect to the metallic phase. Instead, the phonon damping time does not show any dependence on the thermodynamic phase. Moreover, the phonon damping time is constant with fluence, indicating that these measurements were performed in a low-excited state, without reaching any anharmonic regime. The independence of the phonon frequency and damping time on the thermodynamic phase suggests that the A_{1g} optical phonon does not contribute to the superconducting phase transition. The observation of a coherent optical phonon in $Ba(Fe_{1-x}Co_x)_2As_2$ discloses an interesting opportunity for more detailed investigations, which may clarify the role of specific lattice oscillation in the physical behavior of iron-pnictide superconductors.

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