Ultrafast transient response and electron-phonon coupling in the iron-pnictide superconductor Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$

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The transient response of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$, $x=0.08$ was studied by pump-probe optical reflectivity. After ultrafast photoexcitation, hot electrons were found to relax with two different characteristic times, indicating the presence of two distinct decay channels: a faster one, of less than 1 ps in the considered pump fluence range and a slower one, corresponding to lattice thermalization and lasting $\cong 6$ ps. Our analysis indicates that the fast relaxation should be attributed to preferential scattering of the electrons with only a subset of the lattice-vibration modes with a second moment of the Eliashberg function $\lambda \langle \omega^2 \rangle \cong 64$ meV^2. The simultaneous excitation of a strong fully symmetric A_{1g} optical phonon corroborates this conclusion and makes it possible to deduce the value of $\lambda \cong 0.12$. This small value for the electron-phonon coupling confirms that a phonon-mediated process cannot be the only mechanism leading to the formation of superconducting pairs in this family of pnictides.

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The discovery of high-temperature superconductivity in iron-pnictide compounds in 2008 (Refs. [1] and [2]) has raised a lot of questions about the nature of this phenomenon. One of these questions concerns the role of electron-phonon (e-ph) coupling, which is at the heart of conventional Bardeen-Cooper-Shrieffer (BCS) superconductivity.[3]

For unconventional superconductors, electron-lattice interaction mechanisms have been extensively studied in cuprates; these materials present some similarities with pnictides such as the bidimensionality of the crystallographic structure with Cu-O planes instead of Fe-As ones and the presence of a magnetic phase in the underdoped part of the phase diagram.[4,5] While the electron pairing mechanism is still controversial in superconducting cuprates, some elements are today accepted: first, their high critical temperatures are not compatible with a BCS scheme; second, the e-ph interaction is anisotropic, as theoretically predicted[6] and verified by means of time-resolved experiments;[7–11] indeed, the e-ph coupling constant is strongly mode selective and ranges from $\lambda \cong 0.13$ to $\lambda \cong 0.55$. This selectivity is linked to the marked bidimensional layered structure of cuprates, leading to a preferential coupling between electrons coming from a specific k direction of the Fermi surface and one particular phonon mode.[6,8]

Much less information is available on e-ph coupling in pnictides. Theoretical works on the 1111 family (LaFeAsO$_{1-x}$F$_x$), employing density-functional perturbation theory, predicted an isotropic coupling, equally distributed for the whole phonon population and too small to be responsible for superconductivity through a BCS-type mechanism.[5,12] On the other hand, a very strong coupling ($\lambda \cong 1$) between electrons and the A_{1g} mode (consisting of a breathing movement of As atoms) was predicted by a model in which the electronic polarization of As atoms involve e-ph interaction.[13] For 122 compounds (doped AFe$_2$As$_2$, A=Ba, Sr, and Ca) spin fluctuations remaining in the paramagnetic

phase may enhance e-ph coupling.[14] As far as experimental results are concerned, the determination of e-ph coupling by pump-probe spectroscopies found a rather low value of $\lambda \cong 0.15$ in BaFe$_2$As$_2$,[15,16] raising up to $\lambda \cong 0.25$ in SrFe$_2$As$_2$ (Ref. [16]) and an intermediate value of $\lambda \cong 0.18$ in SmFeAsO.[17] All of these studies are in defavor of a BCS-type coupling as the origin of superconductivity but noteworthy none of them was performed on doped specimens presenting a superconducting transition.

In this work, we studied the iron-pnictide Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$, at almost optimal doping, $x=0.08$. This material is superconductor below $T_c=24$ K and does not present any magnetic transition.[18] Single-crystal samples were grown by the self-flux method[18] and fully characterized prior to our measurements. We carried out pump-probe reflectivity measurements using a mode-locked Ti:Sapphire system delivering laser pulses at 800 nm wavelength and 1 kHz repetition rate. The pulses duration was $\approx$40 fs. The experimental setup has been described in Ref. [19], allowing us to reach a signal-to-noise ratio up to 10^5. We kept the linearly polarized pump and probe beams orthogonal to each other; the pump beam was p polarized, incident at about 5° from the surface normal and was focused on a 100 μm diameter spot. The probe beam incidence angle was 15° with respect to the sample normal, and its size was 25 μm diameter, in order to probe a uniformly excited area. We used pump fluences between 1.3 and 3.2 mJ/cm^2. A continuous flow helium-gas cryostat allowed us to cool the samples down to $\approx$10 K. The samples were cleaved along the (001) crystallographic direction in order to obtain clean and optically flat surfaces.

The time-resolved reflectivity curves are presented in Fig. 1, where all the curves have been normalized to have the same value after the pump excitation. The transient reflectivity response consists of a fast increase at zero delay, corresponding to the excitation of electrons by the pump pulse.

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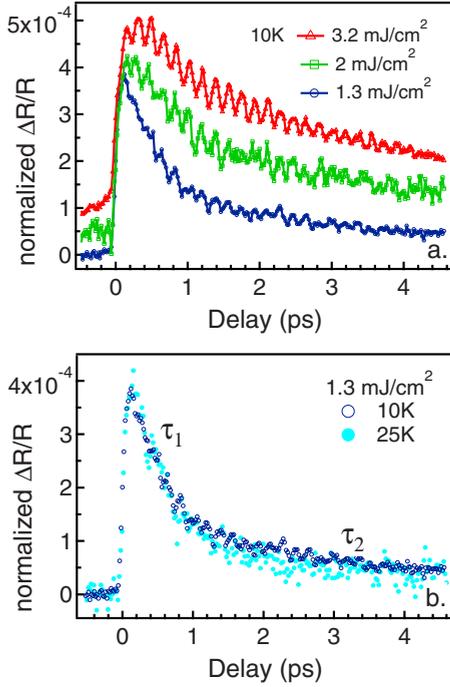
FIG. 1. (Color online) Time-resolved reflectivity curves on Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$: (a) at T_i=10 K (SC phase) for different pump fluences (an offset has been applied for clarity); (b) for F =1.3 mJ/cm^2 as a function of T_i, across T_c. All the curves have been normalized to show the same peak value after the excitation pulse.

FIG. 2. (Color online) (a) Fitting by a biexponential decay function and two components of the fit, for T_i=10 K and F =1.3 mJ/cm^2; (b) Relaxation times τ_1 and τ_2 as a function of the maximum electronic temperatures.

Then the relaxation dynamics occurs and the recovery of unperturbed value takes place in several tens of picoseconds. After the reflectivity increase due to the electron heating, we observe an oscillation, attributed to a coherent A_{1g} mode; its complete study has already been presented in Ref. 20. Here we focus on the transient reflectivity-relaxation dynamics occurring on the picosecond time scale.

This relaxation is composed of two different decay channels; indeed, the curve cannot be described by a single exponential decay but two of them are necessary. The faster one is hereafter called τ_1 and the slower one τ_2. They are both represented as a function of the maximum electronic temperature T_e (derivated as explained below) in Fig. 2, for the two considered initial temperatures T_i. τ_1 values range from $\cong$0.5 to $\cong$1.1 ps; τ_2 lies between $\cong$4.5 and $\cong$6.7 ps.

Several pump-probe reflectivity measurements performed on iron-pnictide family compounds report on the existence of different relaxation times linked to the opening of a gap either the superconducting (SC) gap or the spin-density wave one.[15–17,21,22] This kind of analysis is based on the Rothwarf-Taylor model,[23] describing the relaxation of electrons excited from the SC ground state across the gap and their coupling to high-energy phonon modes. It should be noted that the pump fluence used in this work is much higher (by several orders of magnitude) than all the previous studies. In this excitation-energy density range (mJ/cm^2) the measurements are sensitive to the relaxation due to electron-phonon coupling while for lower fluences (μJ/cm^2) one has direct access to Cooper-pair recombination.

The strong excitation-level effect can be seen by calculating the electronic temperature reached just after excitation. This can be done as a function of the pump fluence F by (see, for example, Ref. 19, and references therein)

$$T_e = \left\langle \sqrt{T_i^2 + \frac{2(1-R)F}{l_s\gamma}e^{-z/l_s}} \right\rangle, \qquad (1)$$

where $R \cong 0.32$ at 1.55 eV (Ref. 24) is the unperturbed reflectivity and γ the linear coefficient of heat capacity due to the electronic subsystem. The mean value is taken for the depth z going from the sample surface down to the skin depth l_s=26 nm [we used the value measured in Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ (Ref. 25)]. Prior to our time-resolved reflectivity measurements, we performed high-resolution, low-temperature heat-capacity measurements in samples coming from the same batches. The results are in excellent agreement with previous studies.[26,27] For $T_c < T \ll \Theta_D$ [$\Theta_D \cong 300$ K (Ref. 27) is the Debye temperature], the heat capacity behaves as $C(T) = \gamma T + \beta T^3$. We then obtained the accurate electronic heat capacity $C_e = \gamma T_e$ with γ =21 mJ/mol/K^2 and the lattice one $C_V = \beta T_l^3$ with β =0.40 mJ/mol/K^4.

The maximum T_e ranges from 1100 to 1740 K, far above the initial temperature. In Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ and the pump-fluence range considered here, the second term under the square root of Eq. (1) is dominant with respect to the first one, T_i; therefore, T_e is much more strongly affected by variations in F than by these of T_i.

The fact that τ_1 is unchanged across the superconducting phase transition [Fig. 1(b)] is a direct proof that it is not linked to a electronic relaxation across a gap, as expected

considering the high T_e. Moreover, its rather linear dependence as a function of T_e (Fig. 2) reflects the behavior of an e-ph relaxation time.[28] On the other hand, the slow relaxation time τ_2 slightly increases as a function of T_e.

The presence of two decay times suggests a preferential coupling between electrons and some phonon modes. As in cuprates, the marked layered crystallographic structure could induce such effects, and it is actually theoretically predicted to occur with the A_{1g} mode coherently excited here,[13] where As atoms move perpendicularly to the Fe-As plane. Excited electrons first thermalize over the time scale τ_1 with the more coupled phonon modes, which in turn relax their excess energy over the time scale τ_2. The most likely mechanism for this second decay time is thermalization with the remaining phonon modes; spin-lattice interactions appear unlikely, since the $x=0.08$ doped sample under study does not show any magnetic transition, but can not be completely ruled out due to the present antiferromagnetic fluctuations.

In order to extract the e-ph coupling constant, we used a three temperature model to simulate the evolution of electrons, "preferential phonons" and "remaining phonons" as a function of the delay time.

The three temperature model equations are[28,7]

$$2C_e \frac{\partial T_e}{\partial t} = \frac{2(1-R)}{l_s} I(t) - g(T_e - T_1),$$

$$C_1 \frac{\partial T_1}{\partial t} = g(T_e - T_1) - g_l(T_1 - T_2),$$

$$C_2 \frac{\partial T_2}{\partial t} = g_l(T_1 - T_2), \qquad (2)$$

where indexes e stand for electrons, 1 for "preferentially coupled phonons" and 2 for the rest of phonons. $I(t)$ is the laser intensity, exciting electrons only at time zero; g is the e-ph coupling constant which governs the fast decaying exponential and g_l the phonon-phonon diffusion governing the slow one. Phonon heat capacities C_1 and C_2 are taken to be partial of the total lattice one, i.e., $C_1 = \alpha C_V$ and $C_2 = (1 - \alpha)C_V$, α representing the fraction of preferentially coupled phonons. The whole temperature range dependence of C_V has been taken into account by performing interpolation between the measured low-temperature behavior and the high-temperature values following the experimental curve in Ref. 27. We neglected electron diffusion because of the quasibidimensionality of the system, which reduces thermal diffu-
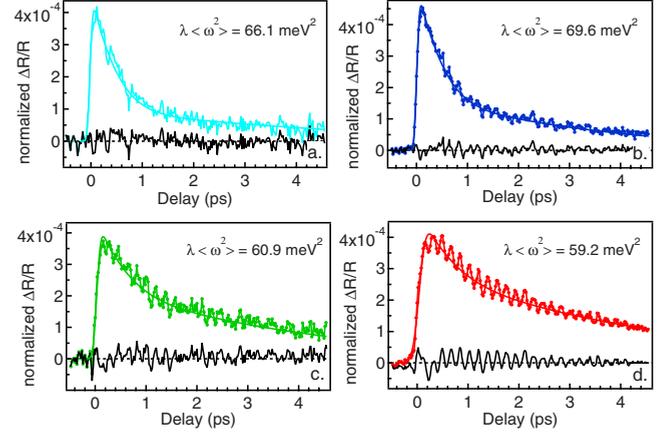
FIG. 3. (Color online) Three temperatures model simulations of the transient reflectivity curves. (a) $T_i=25$ K, $F=1.3$ mJ/cm^2; (b) $T_i=10$ K, $F=1.3$ mJ/cm^2; (c) $T_i=10$ K, $F=2$ mJ/cm^2; and (d) $T_i=10$ K, $F=3.2$ mJ/cm^2. The difference between experimental curve and simulation are shown in black.

sion along the (001) direction. Once the evolutions of T_e, T_1, and T_2 are known, we estimate the change of reflectivity to be a linear combination of electronic and average lattice temperature $\langle T_{latt} \rangle = \alpha T_1 + (1-\alpha)T_2$, following the theoretical derivation in Ref. 19,

$$\frac{\Delta R(t)}{R} = A_e T_e(t) + A_{latt} \langle T_{latt}(t) \rangle. \qquad (3)$$

Finally, the simulations are convolved with a Gaussian to take into account the finite pulse duration. Results of simulation and comparison with experimental data are given in Fig. 3, where we can notice a good agreement between experiments and theory. The relevant parameters used to simulate the transient reflectivity are given in Table I. The slight disagreement at low delay time between simulation and experimental curve at the highest pump fluence [Fig. 3(d)] comes probably from the electronic heat diffusion. This parameter has not been taken into account in our equations, and would increase with the electronic temperature, enlarging the curve just after excitation.

The obtained fraction of preferentially coupled phonons lies between 0.35 and 0.4, which is higher than the value 0.2 reported in cuprates.[7] An explanation for this may be related to the bidimensional structure of these materials, which may be an important factor in the selection of some phonon modes to be more efficiently coupled with electrons. The

TABLE I. Maximum electronic temperatures, fraction of strongly coupled phonons, and electron-phonon coupling constants obtained by three temperature model simulations.

T_i (K)	F (mJ/cm^2)	T_e^{max} (K)	α	$\lambda\langle\omega^2\rangle$ (meV^2)	λ
25	1.27	1100	0.4	66.1	0.125
10	1.27	1100	0.4	69.6	0.132
	1.91	1350	0.35	60.9	0.115
	3.18	1740	0.35	59.2	0.112

crystallographic structure of iron pnictides is much less bidimensional than $Bi_2Sr_2CaCu_2O_{8+\delta}$, especially for Co-doped $BaFe_2As_2$ (see, for example, Refs. 29 and 30): therefore the selectivity of phonons is less marked, and a more homogeneous coupling is observed. We notice here that the presence of two distinct decay times in $SrFe_2As_2$ (Ref. 16) also lead to the conclusion of selective e-ph coupling.

As a result of our analysis, we obtain an e-ph coupling $6.8 < g < 8.0$ mJ/K/s/m^3. By using the relation $g = \frac{6\hbar\gamma}{\pi k_B}\lambda\langle\omega^2\rangle$,[28] we obtain for the second moment of the Eliashberg function $\lambda\langle\omega^2\rangle \cong 64$ meV^2, in good agreement with Refs. 15 and 16.

In the estimation of $\langle\omega^2\rangle$, we also have to take into account the fact that some vibrational modes are more efficiently coupled than others. The most natural choice is to take the frequency of the fully symmetric A_{1g} mode, which is coherently excited by our photoexcitation and consequently efficiently coupled. Moreover, it is the exact counterpart of the preferentially coupled mode of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (the so-called buckling mode, where O atoms oscillate orthogonally to the Cu-O layer[6,8]). Then we used the A_{1g} energy, 23 meV,[20] as $\langle\hbar\omega\rangle$, which gives $\lambda \cong 0.12$. We notice that, by taking the average of the whole phonon spectrum as measured by neutron diffusion,[31] we obtain an even smaller but very close value of $\lambda \cong 0.10$; the coupling given here is therefore a higher limit estimation of λ.

With this value for λ, we can evaluate the critical temperature T_c using the McMillan formula,[32] derived for isotropic systems and moderate e-ph coupling,

$$T_c = \frac{\langle\omega\rangle}{1.20}\exp\left[-\frac{1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right]. \quad (4)$$

By taking $\langle\hbar\omega\rangle = 23$ meV and $\mu^* = 0$,[33] we obtain $T_c \cong 0.03$ K, which is far below the actual T_c of about 24 K: therefore, the electron pair formation cannot be explained only by electron-lattice interactions. Our results confirm that the scenario of an e-ph mediated superconductivity mechanism can be ruled out for these compounds.[12]

In conclusion, we reported an ultrafast transient reflectivity study on optimally doped $Ba(Fe_{1-x}Co_x)_2As_2$. The fact that two different time scales need to be taken into account to describe the relaxation dynamics, as well as the simultaneous observation of a strong coherently excited optical phonon, indicate that a preferential coupling exists with some vibrational modes. On the basis of our analysis, we can estimate a value of $\lambda\langle\omega^2\rangle \cong 64$ meV^2: if we consider a more efficient coupling with low-energy modes and especially with the coherently excited A_{1g} one, a value for the e-ph coupling constant of $\cong 0.12$ is obtained. This low value cannot explain a T_c of 24 K within a standard BCS framework, and confirms that other kind of mechanisms are responsible for superconductivity in these materials.

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