Distinct quasiparticle relaxation dynamics in an electron-doped superconductor, BaFe$_{1.9}$Ni$_{0.1}$As$_2$

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Abstract. We report an investigation of the distinct quasiparticle (QP) relaxation dynamics in an electron-doped superconductor, BaFe$_{1.9}$Ni$_{0.1}$As$_2$, employing femtosecond pump–probe measurements. Two distinct relaxation components and one sub-nanosecond long-lived component are clearly observed in our transient reflectivity spectra. The slow relaxation component, which is on a picosecond time scale, strongly correlates with the superconducting (SC) transition, and we attribute it to the recombination dynamics of Cooper pairs. We calculated the SC gap, $\Delta(0) \approx 5.7$ meV. The fast relaxation component, on a sub-picosecond scale, is ascribed to the QP relaxation from a range of large gap states to the state above the SC gap. We obtained the low end of these bandgaps to be $\Delta_G \approx 9$ meV. Furthermore, we estimated the electron–phonon (e–ph) coupling constant and the Coulomb pseudopotential from the fast relaxation lifetime. The small values of the e–ph constant and the negative Coulomb pseudopotential suggest that the e–ph interaction is not the dominant contribution of the SC transition.
1. Introduction

The recent discovery of high-temperature superconductivity in iron pnictides has attracted much attention because of their high superconducting (SC) transition temperature (as high as 56 K) and new possibilities to investigate unconventional SC mechanisms [1]–[9]. On the one hand, these new superconductors share similar features with cuprates, such as anti-ferromagnetic (AF) parent compounds, doping-induced superconductivity and their layered crystal structure. On the other hand, the materials differ from cuprates in their isotropic (s-wave) SC characteristics and the apparent multi-gap nature. (Ba, K)Fe$_2$As$_2$ was the first superconductor found in the 122 series (oxygen-free system); its SC transition temperature, $T_C$, can be as high as 38 K [10]. Its parent compound BaFe$_2$As$_2$ shows a first-order structural phase transition from tetragonal to orthorhombic with the simultaneous onset of long-range AF order around 140 K [10]. The presence of static AF ordering in the parent compounds and the remarkable similar doping-dependent phase diagram to that of the high-$T_C$ copper oxides suggest that local AF fluctuations may also play an important role in the superconductivity of these materials. Indeed, recent neutron scattering measurements on spin fluctuations of powder samples of SC Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ ($T_C = 38$ K) and crystalline samples of CeFeAsO$_{0.84}$F$_{0.16}$ ($T_C = 41$ K) show clear evidence for resonant-like magnetic peaks below $T_C$ at $\hbar \omega \sim 14$ meV and $\hbar \omega \sim 18.7$ meV, respectively [11]–[15].

Understanding the interplay between spin fluctuations and superconductivity in iron pnictides is important because spin fluctuations may indeed mediate electron pairing for superconductivity. The time-resolved optical spectroscopic techniques are ideal for the study of quasiparticle (QP) dynamics involving SC gaps, spin fluctuations and other ordering parameters [16]–[24], which are extremely important in establishing the microscopic picture of superconductivity in iron pnictides. For instance, femtosecond pump–probe spectroscopy has proved to be a superior tool for discriminating coexisting or competing phases such as the pseudogap phase [25], the suppression of superconductivity by the AF phase [26], and the competition between the spin density wave (SDW) and the precursor superconductivity phase [27]. In the SC states, SC gaps open in the QP density of states. In the time-resolved optical experiments, the coherent pump photons break Cooper pairs and excite the QPs into energy states far above the gaps. The subsequent relaxation processes (recombination of the photo-excited carriers back into the SC condensate) are detected by the probe pulses. Dynamic information on the low-lying electronic/spin states of superconductors can be obtained if the QP relaxation and recombination are studied on a picosecond time scale as a function of temperature, polarization and magnetic field.
In this work, we employ time-resolved optical reflection spectroscopy to study relaxation dynamics in an electron-doped SC single crystal, BaFe$_{1.9}$Ni$_{0.1}$As$_2$ (BFNA). We observe two distinct relaxations and one sub-nanosecond long-lived component in our transient reflectivity spectra below $T_C$. The slow relaxation component (picosecond scale) only exists below $T_C$ and it shows a direct connection with the SC gap, $\Delta(0) \sim 5.7$ meV. The fast component is ascribed to the QP relaxation from the higher excited state. Moreover, we estimate the electron–phonon (e–ph) coupling constant and the Coulomb pseudopotential. The small e–ph-coupling constant and negative Coulomb pseudopotential suggest that the e–ph interaction does not dominate the SC transition in BFNA. In addition, we attribute the sub-nanosecond long-lived component to the relaxation of heat via diffusion.

2. Experimental

Single crystals with nominal formula BFNA of typical sizes $\sim 3 \times 2 \times 0.3$ mm$^3$ were grown by the self-flux method [28]. The actual Ni concentration was confirmed to be 0.1 by energy dispersive x-ray microanalysis. The $ab$-plane resistivity was measured using a standard four-probe method and the result is shown in figure 1. The SC phase transition, which takes place at 20 K in our sample, is very sharp with a transition region of $<0.7$ K, indicating the excellent quality of the single crystal. For the time-resolved reflectivity measurements, a Ti:sapphire laser system delivering 100 fs short pulses at an 80 MHz repetition rate at 800 nm was used as the source of both pump and probe beams. The average pump power was $<5$ mW, giving a pump fluence of $\sim 0.2 \mu$J cm$^{-2}$. The probe intensity was 10 times lower. We have already calibrated the temperature increase due to the heating effect in our time-resolved reflectivity data from 4 to 180 K. The pump beam was modulated at 2 KHz with an optical chopper and a lock-in amplifier was used to measure the transient reflectivity change, $\Delta R/R$, of the probe beam.
Figure 2. Time-resolved reflectivity of BFNA at temperatures above (b) and below $T_c$ (a). The solid lines represent the fitting of the $\Delta R/R$ signal. The $\Delta R/R$ signal can be better fitted using two-exponential decay. The intensity difference between the initial value before the incoming pulse and the value around 100 fs indicates a long-lived component with a sub-nanosecond time scale lying in the $\Delta R/R$ signal.

3. Results and discussion

Figure 2 shows typical temporal traces of the transient reflectivity change at various temperatures near $T_c$ in BFNA. The data can be fitted by two exponentials: $\Delta R/R = A_0 + A_{fast} \exp (-t/\tau_{fast}) + A_{slow} \exp (-t/\tau_{slow})$. In figure 2(a), the three components in the reflectivity signal are demonstrated. Below $T_c$, the fast component, $\tau_{fast}$, is of the order of 0.2 ps, while the slow component, $\tau_{slow}$, is of the order of 10 ps. In addition, a very long-lived relaxation component is clearly observable and it has negative amplitude and a sub-nanosecond time constant. First, we discuss the slow relaxation component. The slow relaxation process is evidently related to SC phase transition since its lifetime changes sharply and its amplitude remarkably decreases when the temperature is approaching $T_c$ from below. We attribute the component to the Cooper-pair recombination after the photo-excitation. We extract the relaxation time, $\tau_{slow}$, and photo-induced reflectivity amplitude, $A_{slow}$, as shown in figures 3(a) and (b). According to the Kabanov model [25], the temperature dependence of the relaxation time, $\tau_{slow}$, can be described as

$$\tau_{slow} \propto \ln \left\{ 1 / \left( (E_i/2N(0)[\Delta(0)]^2) + e^{-\Delta(T)/k_B T} \right) \right\} \left[ \Delta(T) \right]^2,$$

where $E_i$ is the incident energy density, $N(0)$ is the density of the state at $E_f$ and $\Delta(T)$ is the SC gap obeying the Bardeen–Cooper–Schrieffer (BCS) temperature dependence. Figure 3(a) shows
Figure 3. The relaxation time, $\tau_{\text{slow}}$, (a) and amplitude, $A_{\text{slow}}$, (b) extracted from the fitting of the $\Delta R/R$ signal for the slow component versus $T/T_c$. The $\tau_{\text{slow}}$ shows a sharp increase when the temperature is approaching $T_c$ and a sudden jump to the low value above $T_c$, which can be described by the model related to the BCS SC gap (red solid line). The amplitude of the slow component, $A_{\text{slow}}$, matches the theoretical curve (red solid line in (b)) with the SC gap $\Delta(0) \sim 3.3k_B T_c$ obtained from the fitting of $\tau_{\text{slow}}$ (red solid line in (a)).

The fit of $\tau_{\text{slow}}$. We obtain the SC gap, $\Delta(0)$, to be $\sim 3.3k_B T_c$. The result is further confirmed by the temperature dependence of $A_{\text{slow}}$. Based on $N(0) \cong 2 \text{eV}^{-1} \text{cell}^{-1} \text{spin}^{-1}$, [29], we estimated $E_1 \cong 4.8 \times 10^{-5} \text{eV}^{-1} \text{cell}^{-1} \text{spin}^{-1}$. The photo-induced signal amplitude is proportional to the photo-induced QP density by considering the bottleneck condition and the energy conservation law. The density of the thermally excited QPs is defined by $n(T) \propto \sqrt{\Delta(T)/T} \cdot \exp\left(-\frac{\Delta(T)}{k_B T}\right)$. We ascribe the slow relaxation below $T_c$ to the QP relaxation across the superconductor gap. As a result, the significant drop of $A_{\text{slow}}$ near $T_c$ can be understood as the annihilation of the QPs as the temperature increases. To describe the $T$-dependence amplitude, $A_{\text{slow}}$, we take advantage of the formula [25]

$$A_{\text{slow}} \propto \frac{E_1 / (\Delta(T) + k_B T/2)}{1 + (2\nu / N(0)\hbar \Omega_c) \sqrt{2k_B T / \pi \Delta(T)} \cdot \exp\left[-\Delta(T)/k_B T\right]} \cdot \exp\left[-\frac{\Delta(T)}{k_B T}\right],$$

where $\Omega_c$ is the photon frequency cutoff and $\nu$ is the effective number of phonon modes per unit cell participating in the relaxation. By using $\Delta(0) \sim 3.3k_B T_c$ obtained from the fitting of $\tau_{\text{slow}}$ and $\Omega_c \cong 60 \text{meV}$, [29], we obtain a theoretical fit (red solid line) of $A_{\text{slow}}$ in figure 3(b), which gives $\nu = 2.6 \pm 0.5$.

Figure 4(a) shows the photo-induced reflectivity amplitude, $A_{\text{fast}}$, and relaxation time, $\tau_{\text{fast}}$, of the fast relaxation component in BFNA. The amplitude of the fast relaxation component, $A_{\text{fast}}$, presents a clear $T$-dependent behavior but the relaxation time $\tau_{\text{fast}}$ has no obvious dependence on temperature. We ascribe the fast relaxation component to the QP relaxation from a range of
Figure 4. Temperature dependence of amplitude, $A_{\text{fast}}$, (a) and the intensity of the sub-nanosecond long-lived component (b). The inset in (a) shows the relaxation time, $\tau_{\text{fast}}$, of the fast component. The amplitude of the fast component, $A_{\text{fast}}$, can be described by the Kabanov bottleneck model (red solid line in (a)), while $\tau_{\text{fast}}$ shows no obvious temperature dependence. The inset in (b) shows a picture of the two-channel relaxation process of the QP.

Large gap states to the state above the SC gap. The photo-induced reflectivity amplitude, which is related to the QP density in the excited state of a gapped system under bottleneck conditions, takes the form

$$A_{\text{fast}} \propto \left[1 + \frac{2\nu}{N(0)\hbar \bar{\alpha}_c} \cdot \exp\left(-\frac{\Delta G}{k_B T}\right)\right]^{-1}$$

(3)

where $\Delta G$ is the effective energy of the bandgap (the fit is shown in figure 4(a)). By taking the parameters we estimated in the slow relaxation, we obtain $\Delta G = 9 \pm 1$ meV. Due to the fact that the relaxation time, $\tau_{\text{fast}}$, related to $\Delta G$ is much faster than that of the SC gap, $\Delta G$ corresponds to a higher lying excited state. One possible explanation is that the relaxation process is governed by a relaxation cascade from the upper bound of $\Delta G$ to that of the SC gap shown in the inset of figure 4(b) and then relaxes to the ground state via the SC gap. However, it is hard to rule out the possibility of the direct relaxation from $\Delta G$ to the ground state. Therefore, it is possible that the two relaxation channels coexisted in parallel in the relaxation process.

Moreover, we are able to estimate the e–ph coupling constant, $\lambda$, from the fast relaxation dynamics of photo-excited QPs at room temperature if we assume that the e–ph interaction dominates the fast relaxation process in BFNA. In our time-resolved experiment, the electrons are driven to a state of inequilibrium when the pump pulse is absorbed by the sample and subsequently the electron temperature rises far above that of the lattice. As a result, the electrons will lose energy to the lattice through phonon emission. The electron temperature equilibrates with a rate ruled by the strength of e–ph coupling. According to the relationship between the
electron temperature $T_e$ and the lattice temperature $T_l$ [30], a nonlinear differential equation for $T_e$ can be written as

$$T_e(t) \frac{d^2 T_e(t)}{dt^2} + \left[ \frac{d T_e(t)}{dt} \right]^2 + \frac{3h}{\pi k_B} \lambda \langle \omega^2 \rangle \left[ 1 + \frac{\gamma}{C_l} T_e(t) \right] \frac{d T_e(t)}{dt} = 0,$$

(4)

where $C_l$ is the lattice specific heat and $\gamma$ is a constant that can be obtained from the electron specific heat $C_e = \gamma T$. The differential equation can be solved analytically by $T_e(t) = A e^{-(3h\gamma/2\pi k_B C_l)\lambda \langle \omega^2 \rangle t} + B$, where $A$ and $B$ are determined by $T_e(0)$ and $T_e(\infty)$. $\Delta R$ varies in response to both $\Delta T_e$ and $\Delta T_l$ as $\Delta R = a \Delta T_e(t) + b \Delta T_l(t)$. However, the specific heat of the lattice is much larger than that of electrons and thereby we only consider the electron heating effect in $\Delta R$ (the contribution of $T_l$ variation in $\Delta R$ should be very weak and decays much longer ($\gg 10$ ps)). The relaxation rate is $\sim 1$ ps at room temperature. By substituting the parameters $C_l = 25.5$ J mol$^{-1}$ K, $\gamma = 23.8$ mJ mol$^{-1}$ K$^{-2}$ [31], we estimated $\lambda \langle \omega^2 \rangle \approx 84.8$ meV$^2$. Based on the mean phonon frequency $\langle \hbar \omega \rangle = 23$ meV [32], we extract the e–ph coupling constant $\lambda$ to be $\sim 0.16$. Here, we have used the approximation $\langle \omega^2 \rangle \approx \langle \omega \rangle^2$.

Within the strong-coupling theory, the McMillan formula for the SC transition temperature in an s-wave superconductor is [33]

$$T_c = \frac{\theta_D}{\ln \left( \frac{1.04(1+\lambda)}{\lambda - \mu^*(1+0.62)} \right)} \exp \left[ \frac{-1.04(1+\lambda)}{\lambda - \mu^*(1+0.62)} \right].$$

(5)

where the renormalization factor, $(1+\lambda)$, arises from the s-wave channel of the e–ph coupling. The value $\theta_D \sim 140$ K from a recent thermal conductivity measurement [34] was used in the calculation. Thus, we estimated the Coulomb pseudopotential $\mu^*$ to be $\sim -0.4$. The negative $\mu^*$ implies that the e–ph interaction alone is not strong enough to induce an SC transition in BFNA.

Finally, we discuss the long-lived component in our photo-induced reflectivity spectra as shown in figure 4(b). The amplitude of the long-lived component $A_{\text{Long}}$ is plotted as a function of temperature. $A_{\text{Long}}$ reaches the maximum at low temperatures and drops quickly to the minimum at high temperature, which is roughly inverse to the behavior of overall specific heat. Therefore, we attribute the slow decay process to the relaxation of heat via diffusion.

### 4. Conclusion

In summary, we show that there are two distinct relaxations and one sub-nanosecond long-lived component in our transient reflectivity spectra in the BFNA superconductor. The slow relaxation gives us an SC gap of $\sim 5.7$ meV. The fast decay component implies a cascade relaxation from this higher excited state to the SC gap subsequently to the ground state. The calculated negative Coulomb pseudopotential suggests that the e–ph interaction does not dominate the SC transition in BFNA. It is necessary to include both the e–ph interactions and non-phonon dominating interactions to describe the SC phase transition in BFNA. The sub-nanosecond long-lived component is attributed to the relaxation of heat via diffusion.

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