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ORIGINAL PAPER

Quasiparticle Dynamics in FeSe Superconductors Studied by Femtosecond Spectroscopy

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Abstract The ultrafast quasiparticle dynamics in FeSe single crystals were measured by using dual-color transient reflectivity measurements ($\Delta R/R$) from 4.4 to 290 K. In general, the typical $\Delta R/R$ of FeSe includes two significant components. One is the relaxation of photoinduced quasiparticles, which has been used to estimate the electron–phonon coupling strength ($\lambda = 0.16$). The other is the oscillation component due to the acoustic phonon. Moreover, the acoustic phonon's energy estimated from the period of oscillation in $\Delta R/R$ markedly shrinks around 90 K, which is the so-called phonon softening.

Keywords Fe-based superconductors · Electron–phonon coupling strength · Ultrafast quasiparticle dynamics

1 Introduction

The discovery of $\mathrm{LaFeAsO}_{1-x}\mathrm{F}_x$ with $T_\mathrm{c} \sim 26$ K [1] initiated the investigations of the diverse family of Fe-based superconductors (FeSC), e.g., $\mathrm{Ba}_{1-x}\mathrm{K}_x\mathrm{As}_2\mathrm{Fe}_2$ (122-type) with $T_\mathrm{c} \leq 38$ K [2], LiFeAs (111-type) with $T_\mathrm{c} \leq 18$ K [3], and FeSe (11-type) with $T_\mathrm{c} \leq 10$ K [4]. Among various FeSCs, the iron chalcogenide FeSe [4] stands out due to its structure simplicity, which consists of iron-chalcogenide layers stacking one by another with the same Fe^{+2} charge state as the iron pnictides. This so-called "11" system is so simple that it could be the key structure to understanding the origin of high-T_c superconductivity [5]. There has been considerable concern over the interplay between electronic structure, phonons, magnetism and superconductivity in 11-type FeSe. Therefore, further studies of their quasiparticle dynamics are indispensable to understanding the high-T_c mechanism in FeSCs. Here we report the time-resolved femtosecond spectroscopy of FeSe single crystals to elucidate the electronic structure and the quasiparticle (QP) dynamics.

2 Experiments

In this study, the FeSe single crystals were grown in evacuated quartz ampoules using a $\mathrm{KCl/AlCl}_3$ flux [6]. The crystalline structure of the samples was examined by X-ray diffraction. The low temperature feature related to superconducting transition is at $T_\mathrm{c} = 8.8$ K.

The femtosecond spectroscopy measurement was performed by using a dual-color pump–probe system (for the laser light source, the repetition rate is 5.2 MHz, the wavelength is 800 nm, and the pulse duration is 100 fs) and an avalanche photodetector with the standard lock-in technique. The fluences of the pump beam and the probe beam are 9.92 and 1.40 $\mu\mathrm{J/cm}^2$, respectively. The pump pulses have corresponding photon energy (3.1 eV) where the higher absorption occurred in the absorption spectrum of FeSe [7]

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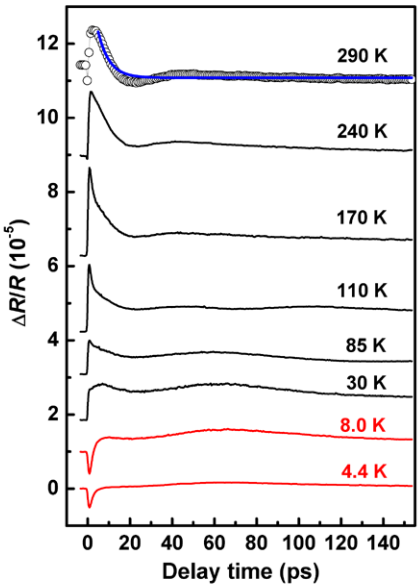

Fig. 1 Temperature-dependent $\Delta R/R$ curves in a FeSe single crystal. The *solid line* at 290 K is the fitting curve using an exponential decay function (Color figure online)

and hence can generate electronic excitations. The photoinduced QP dynamics is studied by measuring the photoinduced transient reflectivity changes ($\Delta R/R$) of a probe beam with photon energy of 1.55 eV.

3 Results and Discussion

Figure 1 shows the typical transient reflectivity changes ($\Delta R/R$) taken at various temperatures on a FeSe single crystal. Above 230 K, there is a fast negative response with a relaxation time of about 1.5 ps together with a long period oscillation. When the temperature decreases below 230 K, a positive and slow response appears and $\Delta R/R$ gradually becomes smaller until $T = 90$ K. Below 90 K, the slow positive response disappears and is replaced by a complicated mixture of the positive and negative components. For $T < T_c$ (8.8 K), a negative response appears as that one in the region above 230 K. In order to figure out the QP relaxation processes after excitation, we try to fit the $\Delta R/R$ curves as shown in Fig. 1 and obtain the relaxation time of QPs. In the case of Co-doped $BaFe_2As_2$, the symmetric A_{1g} mode is coherently excited by photoexcitation and efficiently coupled [8]. Consequently, we take the A_{1g} mode into account in the present case of FeSe, which is the strongest phonon mode in electron-phonon spectral function, $\alpha^2F(\omega)$ [9]. Then, we can obtain the electron-phonon coupling strength, $\lambda = 0.16$, in FeSe from Allen's model [10]. This value is consistent with the theoretical results of $\lambda = 0.17$ [9] obtained by using linear response within the generalized gradient approximation (GGA).

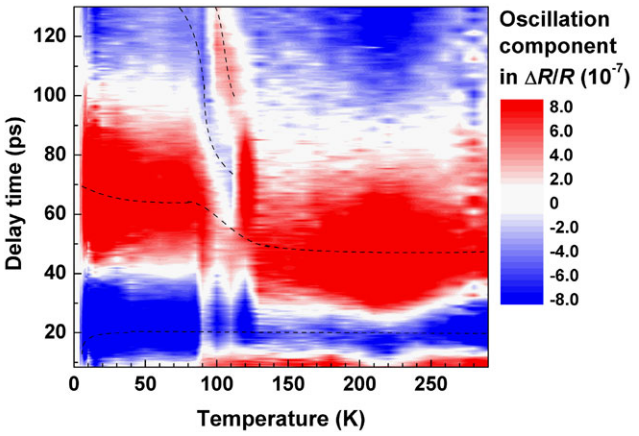

Fig. 2 Temperature-dependent oscillation component of $\Delta R/R$ in a FeSe single crystal, which was obtained by subtracted the decay background (solid line in Fig. 1) from $\Delta R/R$ in Fig. 1. *Dashed lines* are a guide to the eyes (Color figure online)

However, the temperature-dependent $\Delta R/R$ in FeSe cannot be solely fitted by an exponential decay function as shown in the case of 290 K in Fig. 1. By subtracting the decay background (e.g., the solid line in Fig. 1), a significant oscillation component is clearly observed as shown in Fig. 2. This oscillation is caused by the propagation of strain pulses inside a FeSe single crystal, namely the interference between the probe beams reflected from the crystal surface and the wave front of the propagating strain pulse [11]. At high temperatures, the damping time is very short and the oscillation sustains only for one period. However, the number of oscillation period markedly increase around 100 K in FeSe; hence the damping time becomes much longer. Besides, the oscillation period significantly increases below 90 K. This means that the longitudinal-acoustic (LA) phonons can propagate further into the interior of FeSe crystals with the orthorhombic structure. According to the difference between the first trough (at 23.52 ps) and the second trough (at 72.78 ps) at $T = 110$ K in Fig. 2, the phonon frequency is found to be 20.3 GHz. The phonon energy is estimated to be ∼0.087 meV. It is worth to note that the phonon energy drops by 60 % around 90 K where a structural phase transition occurs and by 6 % at superconducting transition temperature, which is consistent with the larger distance between the first trough and first peak in Fig. 2.

Very recently, the phonon softening near the structure transition in $BaFe_2As_2$ and Co-doped $BaFe_2As_2$ was observed by inelastic X-ray scattering [12] and resonant ultrasound spectroscopy [13, 14], respectively. Fernandes et al. [13] found the 16 % softening of shear modulus in $BaFe_{1.84}Co_{0.16}As_2$ at $T_c = 22$ K. For the non-superconducting case of $BaFe_2As_2$, however, the rather large softening of 90 % was observed around 130 K where is the structural and AFM phase transition temperature. Here, the larger phonon softening due to structural phase transition and rather small phonon softening due to the superconducting phase transition are also observed in 11-type FeSe. These results suggest that the reduction of phonon energy at both the structural and the superconducting phase transitions is a general feature in FeSCs and may participate in the superconductive pairing, albeit not the mechanism responsible for high T_c in FeSCs.

4 Summary

We have studied the ultrafast quasiparticle dynamics and phonon softening in FeSe single crystals by dual-color femtosecond spectroscopy. The relaxation time of quasiparticles reveals an electron–phonon coupling strength $\lambda = 0.16$. Moreover, the energy of LA phonons at 110 K was estimated to be 0.087 meV from the oscillation component of $\Delta R/R$, which markedly softens around both the structural phase transition and superconducting transition. Our results provide the vital understanding of the role of phonons in Fe-based superconductors.

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