Mott behavior in $K_xFe_{2-y}Se_2$ superconductors studied by pump-probe spectroscopy

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We report on a signature indicating a transition to the orbital-selective Mott phase in superconducting $K_xFe_{2-y}Se_2$ single crystals by employing dual-color pump-probe spectroscopy. In addition to the multiexponential-decay recovery dynamics of photoinduced quasiparticles, a damped oscillatory component caused by coherent acoustic phonons emerges when the superconducting phase is suppressed at an increased temperature or excitation power. Upon raising the temperature to 150–170 K, the oscillatory component diminishes alongside a significant enhancement of the slow decay component in the recovery traces. These results can be understood as a gap opening in certain k directions, indicating that a vital role is played by electron correlation in iron-based superconductors.

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I. INTRODUCTION

Recently, the question of whether iron-based superconductors (FeSCs) are in close proximity to Mott behavior that is driven by electron correlation has been intensively studied in the field of high-temperature superconductivity [1–8]. So far, superconductivity in ferropnictides has been generally understood within the Fermi-surface nesting framework with weak and moderate correlations [9–12]. The recently discovered alkaline iron selenide superconductors [13–16], which have transition temperatures (T_C) above 30 K, have stimulated an interest in reevaluating the role played by electron correlations [1–6,17,18]. Their Fermi-surface structures, as revealed in the multiple angle-resolved photoemission spectroscopy (ARPES) experiments [19–22], are different from those predicted by weak-coupling theory. The ordered magnetic moment in $K_xFe_{2-y}Se_2$ (KFS) was found to be the largest among FeSCs [23]. The parent phase of such KFS superconductors has been found to be on the border between an insulating state and a magnetically ordered state despite the fact that an exact structure of the parent phase remains under debate [3,24–26]. These properties cannot be fully explained by the nesting mechanism. Instead, they behave similarly to doped Mott insulators such as cuprates [3–5,7,27].

Despite the fact that a metal-insulator crossover in the transport curves has been measured in the superconductor family of alkaline iron selenides [13–16], it remains quite challenging to directly identify the Mottness experimentally because of the coexistence of insulating and superconducting/metallic domains in the KFS superconducting samples [3,26,28–31]. The nature of multiple bands crossing the Fermi surface [19–22,32] makes this issue even more complicated. Mott-like physics has been observed in the insulating KFS samples; however, this is unlikely to give rise to the superconducting phase [3]. Very recently, Yi *et al.* suggested a scenario involving the orbital-selective Mott phase to explain the ARPES data on superconducting KFS samples [4] that is consistent with the prediction of a multiband theory assuming strong on-site Coulomb interactions [5]. Nevertheless, such Mott behavior is not a prerequisite for understanding the resistivity hump (T_H in the range of 50–250 K) observed in the transport curves, which can also be viewed as a result of parallel resistors [33,34]. Femtosecond pump-probe spectroscopy can provide new opportunities to investigate this issue by providing additional dynamical information about quasiparticles (QPs) [35–41], phase transitions [42], competing orders [43], and collective modes [44–46]. In particular, these time-resolved optical techniques can monitor the microstructures isolated from one another, adding vital information that is unattainable by common electrical characterizations.

In this paper, we report a femtosecond spectroscopic study on superconducting KFS single crystals that reveals ultrafast optical signatures of Mott-transition-like behavior at the normal state. When the superconducting phase is suppressed by increased temperature or excitation fluence, a damped oscillatory component emerges in the transient traces alongside the biexponential recovery dynamics of photoexcited QPs. Such an oscillatory signal is most likely induced by coherent acoustic phonons related to competing orders. Upon raising the temperature to 160 K, the oscillatory component diminishes concurrent with a dramatic increase of a slow exponential component in the recovery trace of the QP dynamics. The abnormal temperature-dependent behaviors are likely to be a signature of a gap opening at the 3d xy bands in KFS superconductors, as predicted by theory [4,5], which implies that a vital role is played by the electron correlations for superconductivity in KFS crystals.

II. EXPERIMENTAL MEASUREMENTS

The samples of superconducting KFS crystals, which have a superconducting transition temperature of $T_C \approx 32$ K and a resistivity hump at $T_H \approx 180$ K [Fig. 1(a)], were prepared by using a rapid quenching process, as previously reported [16,26]. To avoid the heating accumulation induced by the high pulse energy required in the study, we performed a series of dual-color pump-probe experiments with low-repetition (1 kHz) femtosecond pulses (~100 fs, Libra, Coherent) [47]. The sample was excited with a photon energy of 3.1 eV, and the QP dynamics was monitored by measuring the transient

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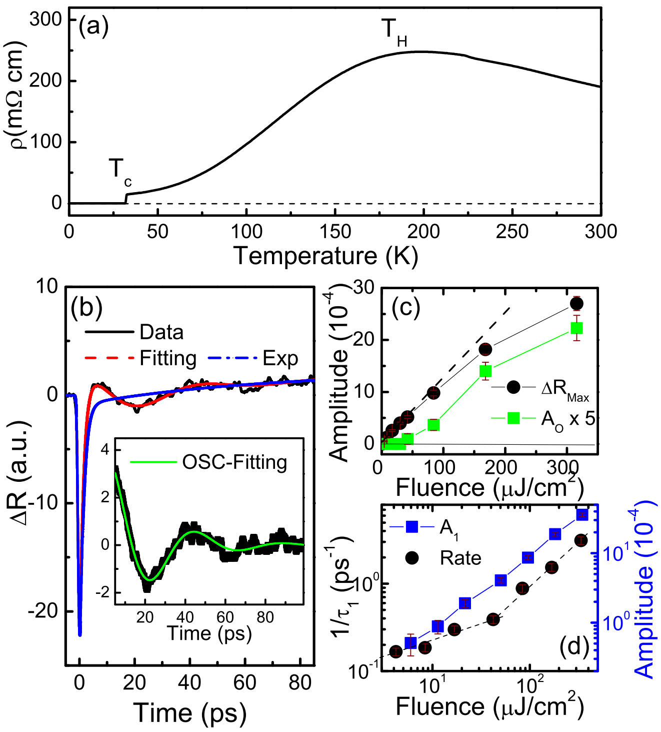

FIG. 1. (Color online) Fluence-dependent QP dynamics at the superconducting phase. (a) Temperature-dependent resistivity of a superconducting KFS single crystal. In addition to a superconducting transition at $T_C \sim 32$ K, a resistivity hump appears at $T_H \sim 180$ K. (b) Time-resolved $\Delta R/R$ recorded at 5 K with an excitation flux of $\sim$200 μJ/cm^2. The decay curve is analyzed [Eq. (1)]. The exponential-decay component and the oscillatory component (inset) are highlighted. (c) The fluence-dependent maximum signal ($\Delta R_{\max}$) and the amplitude of the oscillatory component (A_O). (d) The amplitude (A_1) and decay rate ($1/\tau_1$) of the fast component are plotted as functions of the incident fluence using a double-logarithm scale.

reflectivity change ($\Delta R/R$) using a probe photon energy of 1.55 eV. The pump beam ($\sim$1 mm in diameter) and the probe beam ($\sim$0.2 mm in diameter) coincide onto the sample. The fluence of the probe beam was set to be less than 5% of the pump beam fluence. The transient optical data were acquired by using a balanced detector and a lock-in amplifier. Bandpass filters were employed to exclude the scattered pump light. By using this configuration, the signal-to-noise ratio for detecting $\Delta R/R$ was better than 10^{-5}. To exclude the aging effect on the cleaved surfaces of the samples, we carefully loaded the samples into an optical cryostat (MicroCryostatHe, Oxford) to avoid exposing the samples to the ambient environment. We placed the optical cryostat into the argon tank where the samples were stored. The cleaved samples were glued onto the cold finger, and the cryostat was sealed before it was taken outside. After being taken outside, a vacuum was established within the cryostat for the temperature-dependent optical studies. We performed a temperature cycle test, and found reproducible experimental data in the first 48 h after the sample loading.

In general, the magnitude of the $\Delta R/R$ signal is proportional to the density of the photoinduced QPs [48]. In superconductors, the QPs are excited through avalanche multiplication caused by electron-electron collisions [48], where one incident photon creates a large number of QPs. The number of QPs created per absorbed photon is proportional to the ratio between the excitation photon energy and the gap value near the Fermi level [48]. Because the insulator $K_2Fe_4Se_5$ phase has a large gap (500 meV) [3,25], the contribution from the insulating domains is negligible.

III. RESULTS AND DISCUSSION

The pump photon energy is sufficiently large to excite the electronic transitions, which causes a swift decrease of $\Delta R/R$. The time-dependent $\Delta R/R$ exhibits the dynamics of QPs in superconductors. We recorded the transient traces of $\Delta R/R$ as functions of time, temperature, and laser fluence. In the superconducting phase, the QP relaxation is dominated by an exponential-decay component. An oscillatory component emerges when the laser fluence reaches a threshold (Figs. 1 and 2). Upon increasing the temperature, a slow decay component gradually becomes important at the normal state (Fig. 3). The normal-state QP dynamics exhibits a transition within the temperature range of 150–170 K, characterized by a significant decrease of the amplitude of the oscillatory component and a corresponding increase of the amplitude of the slow decay component.

To compare the data recorded at every temperature, we analyzed the recovery curves with a widely used phenomenological model consisting of one oscillatory and two exponential-decay components in the form of [45,46]

$$\Delta R(t)/R = A_1 e^{-t/\tau_1} + A_2 e^{-t/\tau_2} + A_3 + A_O e^{-t/\tau_O}\cos[2\pi f(t+t_0)], \quad (1)$$

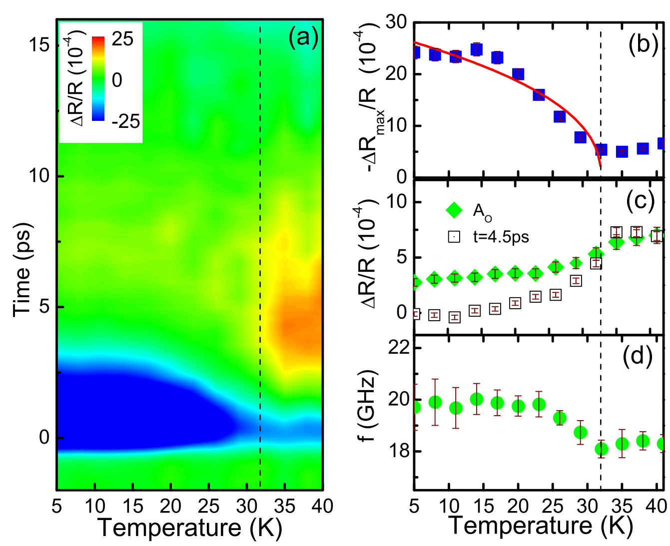

FIG. 2. (Color online) Temperature-dependent QP dynamics in KFS superconductors. (a) The differential reflectivity variation is plotted as functions of temperature and delay time. The maximum value of the initial reflectivity variation (b), the signal magnitude at $t \sim 4.5$ ps and the fitted amplitude of the oscillatory component (c), and the oscillatory frequency (d) are plotted vs temperature. The red line in (b) is the curve fitting to the Mattis-Bardeen formula. The dashed lines indicate the superconducting transition temperature.

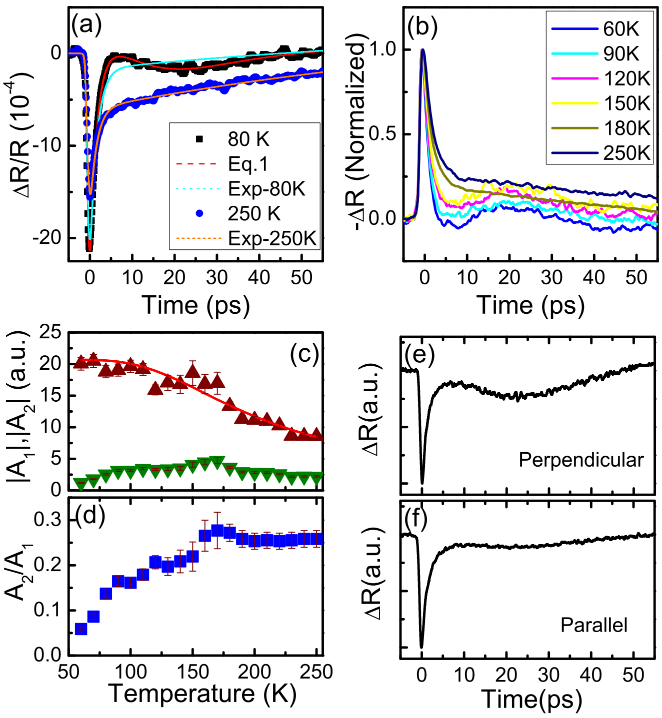

FIG. 3. (Color online) QP dynamics at the normal state. (a) The time-resolved differential reflectivity recorded at 80 and 250 K, respectively. (b) The normalized, time-resolved differential reflectivity variation recorded at different temperatures. (c) The temperature-dependent amplitudes of the fast and slow components. The solid line is a fitting curve with a temperature-independent pseudogap. (d) The temperature-dependent amplitude ratio (A_2/A_1). (e) and (f) are the traces recorded at 150 K with the pump and probe beams configured with perpendicular and parallel polarizations, respectively.

where A_1 and A_2 are the amplitudes of the two exponential-decay components with lifetimes τ_1 (<10 ps) and τ_2 (<100 ps) caused by the excited electrons and phonons, respectively [45,46], and the constant A_3 represents a very slow decay component with a lifetime exceeding the time window. The last term shows a damped oscillatory component with an amplitude A_O, damping lifetime τ_O, oscillatory frequency f, and an initial phase of $2\pi f t_0$.

A. Superconducting phase

The recovery dynamics at the superconducting phase are strongly dependent on the pump fluence. Figures 1(c) and 1(d) plot the fluence-dependent results obtained by fitting the data using Eq. (1). The maximum value of the photoinduced amplitude $\Delta R/R$ is linearly dependent on the fluence under a weak excitation. Above a threshold value $I_{th} \approx 30 - 50\ \mu\text{J/cm}^2$, the signal amplitude deviates from linearity [Fig. 3(c)]. Such an onset of saturation has been recognized as a signature of the vaporization of the superconducting phase in high-T_C superconductors [39,40,42,49]. The decay dynamics of $\Delta R/R$ is dominated by the fast exponential component (A_1). The decay rate ($1/\tau_1$) is also dependent on the incident fluence [Fig. 1(d)], indicating that an important role is played by electron-electron interactions for the recombination of photoexcited QPs [37]. The fluence dependence of the decay rate ($1/\tau_1$) exhibits a slope change at a fluence of approximately I_{th}. It is also worth noting that the oscillatory amplitude becomes detectable when the excitation fluence reaches a similar threshold. The significant changes of the QP dynamics at I_{th}, including the saturation of the maximum amplitude, the slope variation of the decay rate, and the appearance of the oscillation component, should be attributed to the vaporization of the superconducting phase [38,42]. The threshold I_{th} observed here is comparable to (or even higher than) the fluence required to vaporize the superconducting phase [39,40,42,49].

From the above results, it is likely that the oscillatory component emerges when the superconducting phase is depleted. For further confirmation, we studied the oscillatory component by increasing the temperature across T_C to suppress the superconductivity. The magnitude of $\Delta R/R$ at a fluence of $\sim 200\ \mu\text{J/cm}^2$ is plotted in Fig. 2(a) as functions of delay time and temperature. The signal has an initial negative value at approximately zero time delay ($t \sim 0.2$ ps) and a positive value at a delay time of $t \sim 4.5$ ps induced by the oscillatory component. These two features indicate different temperature dependences [Figs. 2(b) and 2(c)]. The initial negative value becomes less dominant when the temperature increases [Fig. 2(b)]. Because the fluence is large enough to destroy the superconducting phase, the temperature-dependent magnitude can be analyzed using the Mattis-Bardeen formula in the form of [39,40]

$$\frac{\Delta R(T)}{R} \propto \left(\frac{\Delta(T)}{\hbar\omega}\right)^2 \log\left(\frac{3.3\hbar\omega}{\Delta(T)}\right), \qquad (2)$$

where $\hbar\omega$ is the probe-photon energy and $\Delta(T)$ is the temperature-dependent superconducting gap. By assuming $\Delta(T) = \Delta_0(1 + T/T_C)^{1/2}$, Eq. (2) can reproduce the experimental data with a superconducting gap value of 12 meV, which is approximately the value obtained by the ARPES measurement [Fig. 2(b)] [21].

The amplitude of the oscillatory component (A_O) increases dramatically when the temperature surpasses T_C [Fig. 2(c)]. Because of the lack of either a static magnetic order or excitonic resonance in the superconducting phase, such low-frequency oscillations observed in superconductors have generally been recognized as a result of coherent acoustic phonon vibrations caused by stimulated Brillouin scattering [38,45,46]. In principle, this phonon mode can come from either the superconducting domains or the insulating domains in the inhomogeneous KFS superconductors. If caused by the insulating domains, a gradual increase from zero should be expected with increasing excitation fluence, which is in contrast to the threshold behavior observed here [Fig. 1(c)]. The fluence and temperature dependences of the oscillatory amplitude indicate that the contribution from the insulating phase can be ruled out here. The abrupt change of the signal at T_C [Fig. 2(c)] is a distinct signature of the correlated interplay between the superconducting phase and the oscillatory component. Thus, we can safely conclude that the oscillatory component is caused by the contribution from the superconducting domains, which is strongly associated with the competing orders because it is activated when the superconductivity is suppressed. Under stimulated Brillouin scattering [38], the activation of the oscillatory phonon component in competing orders can be plausibly understood using energy-conservation arguments. The amplitude of stimulated Brillouin scattering is linearly proportional to the degree of thermal expansion. For the superconducting phase, the photoexcited carriers lose most of their energy through the breaking of Cooper pairs. When superconductivity is suppressed, a large fraction of the energy quickly dissipates to the lattice, inducing through-plane thermal gradients that result in the oscillation component in the pump-probe traces.

Moreover, we observed that the oscillation frequency, i.e., the phonon energy [Fig. 2(d)], gradually decreases with increasing temperature at T_C. Such lattice hardening in the superconducting phase is consistent with ultrasound experiments on superconducting iron arsenides [50], confirming the origin of the superconducting domains.

B. Normal-state phase

Next, we shift our attention to the normal state at higher temperatures. The experimental data obtained at 80 and 240 K are compared in Fig. 3(a). We carefully analyze the experimental results using Eq. (1) and highlight the exponential-decay components. There are three major differences between the results at the two temperatures: At higher temperatures, (1) the fast decay component becomes less dominant, (2) a slow decay component with a lifetime of ~80 ps becomes more pronounced, and (3) the oscillatory component becomes negligible. To clarify the temperature-dependent behaviors, the time-resolved traces of $\Delta R/R$ recorded at different temperatures are compared with a normalized amplitude scale [Fig. 3(b)].

Upon raising the temperature, the amplitude (A_1) of the fast component gradually decreases [Fig. 3(c)]. Such temperature-dependent behavior of the electronic component has been regarded as a pseudogaplike feature. The temperature dependence of the signal's amplitude can be reproduced by the bottleneck model from Kabanov *et al.* [39,48] with a temperature-independent gap Δ_{PG} [red solid line, Fig. 3(c)]. The fitted value of $\Delta_{PG}/k_B \sim 730$ K is comparable with the value derived in the NMR study [51].

The unexpected rise of the slow component with increased temperature was not observed in earlier ultrafast studies on other FeSCs. Because this component gradually becomes important with increasing temperature, it cannot be explained by a pseudogap. The ratio A_2/A_1 increases and becomes constant at temperatures slightly below T_H [Figs. 3(c) and 3(d)], implying that the recombination probability of the photoexcited QPs through this slow channel increases with increasing temperature. Such temperature-dependent QP dynamics might be associated with the modification of the band structure by a gap opening [52].

Within the same temperature range, the magnitude of the oscillatory component exhibits a critical point behavior. We subtract the exponential decay from the total signal and only plot the oscillatory component in Fig. 4(a) to better exhibit the temperature-dependent behaviors. The amplitude and frequency, as analyzed using the Fourier transformation, are shown in Figs. 4(b) and 4(c), respectively. The oscillation amplitude becomes less pronounced and eventually undetectable at higher temperatures [Fig. 4(b)]. Aside from the slight difference of the damping lifetime and the initial phase

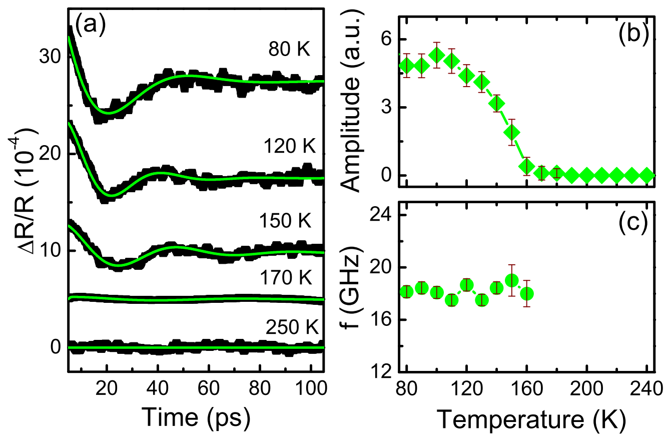

FIG. 4. (Color online) Temperature dependence of the oscillatory component. (a) The oscillatory component after subtracting the exponential-decay components from the total signal recorded at different temperatures. The traces are vertically shifted for clarity. Temperature-dependent amplitude (b) and frequency (c) of the oscillatory component.

during the procedure, the frequency variation is insignificant in this temperature range [Fig. 4(c)]. In the past few years, the oscillatory behavior, including coherent optical/acoustic phonons at relatively high/low frequencies, has been observed in the pump-probe traces of a number of ferropnicitides [38,40,44] and ferroselenides [36,45,46]. In a small number of temperature-dependent studies, the frequency variation of the coherent acoustic oscillations has been observed at the onset of the superconducting transition [45,46], which is consistent with the study of resonant ultrasound spectroscopy [50]. In the KFS superconductors studied here, we observe the variations of both the amplitude and frequency at approximately T_C, which are similar to the previous study [Figs. 2(b) and 2(c)]. At the normal state, the temperature-dependent magnitude of the coherent acoustic oscillation as well as significant variations of both the amplitude and frequency have been argued to result from the structure phase transition or the electronic nematicity in FeSCs [36,45,46]. However, in this study, the temperature dependence of the oscillation frequency is not distinct [Fig. 4(c)] in spite of a significant depletion of the oscillation amplitude from 120 to 170 K. In principle, the reduction of the phonon response may be induced by a structure change, however, this is unlikely in the present study. In this temperature range, no structure transition or nematicity fluctuation have been identified in KFS superconductors [25,53]. Within this temperature range, the conductivity transition has been explained as being a result of parallel resistors consisting of insulating and superconducting/metallic domains [33,34]. However, such a framework cannot fully explain the abrupt change in the optical response observed here. The pump-probe measurements simultaneously monitor the separated domains, and the ultrafast dynamical behaviors in either the insulator or the metal should gradually change with temperature. These facts imply that the change of the electronic band structure may be a significant influence for the temperature dependences of the slow decay component and the oscillatory component. In the following, we attempt to explain the abnormal temperature dependence of the coherent acoustic oscillation within the framework of band structure modification by a gap opening [52].

In strongly correlated electronic systems, the signal of the coherent phonons displayed in the pump-probe traces has been employed to monitor the Mott transition as reported by Kim *et al.* [54]. Moreover, the metal-to-insulator transition is always accompanied by a slow decay process in the QP dynamics [52]. Recently, an orbital-selective Mott transition was identified in the normal-state phase of the FeSCs [4,5,55], where a number of the d orbitals undergo metal-to-insulator-like transitions, i.e., a gap opening at certain k directions. Such a scenario can provide comprehensive explanations for the observed phenomena. The photoinduced reflectivity changes revealed in the time-resolved traces are related to the opening of a gap (or pseudogap) in the density of states near the Fermi energy [48]. The QP dynamics in FeSCs are band dependent and have been previously investigated in iron arsenides [37]. Multiple orbitals contribute spectral weights near the Fermi surface in FKS samples [19–22]. When raising the temperature, the spectral weight near the Fermi surface for the $3d$ xy orbitals diminishes while the other orbitals remain metallic [4,5]. For KFS superconductors, all the $3d$ bands are metallic at low temperatures slightly above T_C. The photoexcited QPs in metals reach an equilibrium with energy dissipation to the lattice caused by electron-phonon interactions, which induce a coherent lattice vibration that manifests itself as an oscillation in the time-resolved trace of $\Delta R/R$.

When the temperature increases, the orbital-selective Mott transition opens a gap (larger than 50 meV) at the $3d$ xy bands [4], which is the likely origin of the slow decay component. With such a gap, the QP relaxation slows down because of the QP relaxation bottleneck [39,40,43]. The onset of the opening at the xy bands leads to the emergence of the slow decay component [Fig. 3(c)]. In addition to the bottleneck effect, the location effect of the photoexcited carrier by in-gap states may also slow down the recombination of QPs. However, the effect of the carrier location appears to be less important here because the in-gap states also exist at low temperatures. A number of additional processes, such as interband transitions and intervalley scatterings, are involved in the recombination of QPs trapped at the gap states [56]. Consequently, less energy will drive the lattice vibrations, leading to the depletion of the oscillatory amplitude. The fast decay component, mainly caused by the metallic bands and hot QPs, becomes less important at higher temperatures.

From the above discussion, the temperature-dependent behavior of the QP dynamics [52] and the coherent phonon dynamics [54] can be plausibly attributed to the orbital-selective Mott transition in KFS superconductors. While the gap only opens at certain k directions, the optical response should be anisotropic, which, in principle, can be identified with polarization-dependent optical measurements. However, different from the terahertz or far-infrared optical study that provided a resonant optical response [57], the probe photon energy (1.55 eV) used here is much higher than the gap values, which are on the order of $\sim$10 meV or less. Because the buildup of the $\Delta R/R$ signal involves a cascade of scattering processes as photoexcited QPs relax to the gap states [58], it is very challenging to establish a direct relationship between the orbital selectiveness and the polarization dependence of $\Delta R/R$. Figures 3(e) and 3(f) compare our experimental data recorded under two different polarization configurations. The signal, particularly the oscillatory component, exhibits a strong polarization dependence, which implies possible anisotropic QP dynamics associated with the electronic band structure. Nevertheless, a concise assignment of the polarization to specific orbitals must be determined in future studies. However, the experimental data are consistent with the scenario of a k-dependent gap opening.

Similar results obtained from different sample batches reveal the generality of the above optical features of Mott behavior in KFS superconductors. Physically, the Mott phase, induced by strong correlation effects, can dramatically modify the transport properties. The coexistence of Mott-localized electrons at the $3d$ xy orbitals and the itinerant electrons at other $3d$ orbitals involved in an orbital-selective Mott transition should be reflected in the transport properties. Concerning the insulating behavior above T_H in the transport curve, a number of mechanisms have been proposed to explain the normal-state transport properties including the models of coherence-incoherence crossover [59,60] and parallel resistors. Our data are not in conflict with these models. By considering the temperature dependence of the resistivity in the metallic domains, the Mott behavior may be incorporated in these models to achieve a better understanding of the transport properties.

IV. SUMMARY AND CONCLUSIONS

We have observed signatures of a transition to the orbital-selective Mott phase in superconducting KFS single crystals by employing dual-color pump-probe spectroscopy. When the superconducting phase is suppressed by increasing the temperature or increasing the excitation fluence, the coherent acoustic phonon signal, characterized by a damped oscillatory component, appears in the time-resolved traces of $\Delta R/R$. When the temperature is increased to 150–170 K, the oscillatory signal diminishes while a slow decay component becomes pronounced. These experimental results can be qualitatively explained by the orbital-selective Mott physics in KFS superconductors using a band normalization at the $3d$ xy bands where the opening of a gap slows down the QP recombination. Our results provide valuable information about the competing orders and confirm the proximity to Mott behavior induced by the electron correlation for superconductivity in KFS superconductors.

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