Ultrafast Dynamics Evidence of High Temperature Superconductivity in Single Unit Cell FeSe on $SrTiO_3$

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We report the time-resolved excited state ultrafast dynamics of single unit cell (1 UC) thick FeSe films on $SrTiO_3$ (STO), with FeTe capping layers. By measuring the photoexcited quasiparticles' density and lifetime, we unambiguously identify a superconducting (SC) phase transition, with a transition temperature T_c of 68 $(-5/+2)$ K and a SC gap of $\Delta(0) = 20.2 \pm 1.5$ meV. The obtained electron-phonon coupling strength λ is as large as 0.48, demonstrating the likely crucial role of electron-phonon coupling in the high temperature superconductivity of the 1 UC FeSe on STO systems. We further find a 0.05 THz coherent acoustic phonon branch in the capping layer, which provides an additional decay channel to the gluing bosons.

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Single unit cell (UC) thick FeSe films on STO with high T_c values [1] have recently attracted much attention. The T_c values have been reported to be as high as 55 ± 5 [2], 65 ± 5 [3], 60 ± 5 [4], and 58 ± 7 K [5,6] in angle-resolved photoemission spectroscopy experiments, 40 [1,7] and 109 K [8] in transport measurements, and 20 K (45 K onset) [9] in magnetization measurements, respectively. Electron-phonon ($e-$ph) coupling has been proposed [1] and analyzed to play a key role for the high T_c values [5,10,11]. While the inconsistency in the T_c values needs to be clarified, more importantly, the superconducting nature of the phase transition needs to be understood comprehensively by using different experimental methods [12].

As a powerful means to investigate ultrafast dynamics of excited states, gap-perceptive ultrafast optical spectroscopy provides information on comprehensive interactions among the quasiparticles and various excitations, such as $e-$ph coupling, and has often been used to probe and control novel quantum materials including superconductors [13–20]. In this Letter, ultrafast spectroscopy is used to investigate the 1 UC FeSe/STO covered by 2 UC FeTe capping layer sample (abbreviated as 2FeTe/1FeSe/STO, and the FeTe layers are used to protect the sample from air contamination). We have unambiguously identified the SC transition in this system. The T_c value, the SC energy gap $\Delta(T)$, the $e-$ph coupling strength λ, the density of thermal electrons n_{th}, as well as the electron temperature T_e have been obtained. We have also observed an acoustic phonon branch in the thin FeTe capping layer, which forms a side-decay channel to the gluing bosons, hence explains the inconsistency in the reported T_c values. Our results demonstrate the likely crucial role of $e-$ph coupling in the SC transition.

Ultrafast laser pulses of 800 nm central wavelength, 96 fs pulse width, and 250 kHz repetition rate were used to excite and probe the ultrafast dynamics of the photocarriers in the 2FeTe/1FeSe/STO sample from 5 K to room temperature. We measured the differential reflectivity, $\Delta R/R$, which was recorded as a function of the delay time of the probe beam [18,19]. The pump and probe beam fluences were kept at 268 and 30 μJ/cm^2, respectively. Compared with the frequently used 76–100 MHz laser systems, together owing to the sample transmission, the steady state laser heating (the so-called thermal effect) in our 250 kHz experiment is ~1000 times weaker. This allows us to use much higher

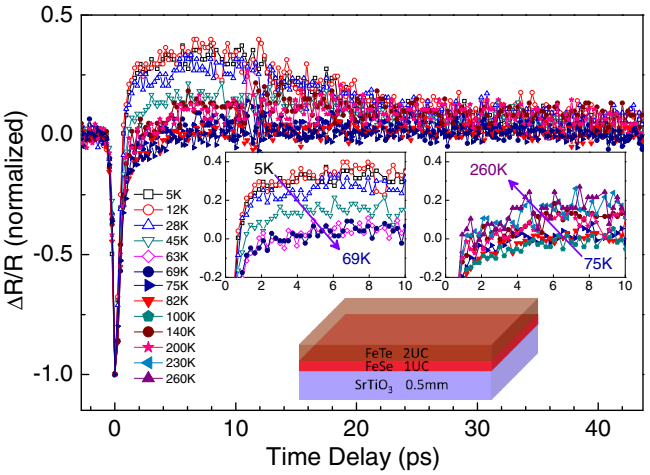

FIG. 1. Normalized photoinduced differential reflectivity. Scanning traces at different temperatures are illustrated, with zoom-in view shown in insets. The purple arrows mark the hump varying trends on temperature increase, respectively. Also shown is the schematic sample configuration.

fluence of excitation and detection to maintain significant signal-to-noise ratio. Moreover, since each light pulse excites only about 0.03 electrons per UC of FeSe and the system relaxes to the SC ground state within ~100 ps (see below), nonthermal melting of the majority of the SC condensate will not occur. Cross-polarization detection was implemented to further enhance the signal-to-noise ratio.

Figure 1 summarizes the normalized photoinduced transient differential reflectivity of the sample, which is schematically illustrated in the inset (the details of the growth procedure are described in Refs. [1,7], and [21]). Here, $|\Delta R/R|$ is proportional to the density of photoexcited free carriers [14,15], since the absorption of the probe beam is linearly modified by the pump-induced variation in the quantum occupation number (consider Fermi's golden rule). It can be seen that the scanning traces after $t = 0$ ps are composed of both increasing and decreasing components, whose amplitude and lifetime vary with temperature. Prominently, a hump can be seen in the scan traces at $t > 0$ ps. The magnitude of the hump decreases and then increases as temperature rises up (see insets of Fig. 1), showing a change around 70 K. This is related to the SC transition as discussed below. We note that, in our control experiment [22], we found that 2 UC FeTe is of optimized number of capping layers, by which the ultrafast dynamics of the 1 UC FeSe layer can be clearly distinguished from that of the FeTe capping layers. And in the Supplemental Material we show that the positive component contributing to the SC response is from FeSe rather than FeTe [22].

To identify phase transitions through ultrafast carrier dynamics, we map out the transient reflectivity as a function of both time and temperature in Fig. 2. The colors are used as a guide to the eye to illustrate the phase transitions. A few temperature regions can be seen, corresponding to different phases. Particularly, both the initial (at 0–2 ps, shown in white and purple) and the proceeding (at >2 ps, shown in red and blue) relaxations show distinct changes around 68 K (Fig. 2). These color changes reflect the variations in the carrier density and lifetime in a phase transition, and qualitatively represent that of a SC transition. We further show a quantitative data analysis below, which unambiguously demonstrates that this is a SC transition, with a high T_c value of 68 $(+2/-5)$ K.

The time-resolved processes can be approximated by two exponential components: $\Delta R/R = A_{\text{fast}}\exp(-t/\tau_{\text{fast}}) + A_{\text{slow}}\exp(-t/\tau_{\text{slow}}) + A_0$, where A_{fast}, τ_{fast}, A_{slow}, and τ_{slow} are functions of temperature, which are obtained by fitting the experimental data, and A_0 is the background level due to the leakage of scattered pump noise. Note that we have included the convolution in our data fitting. The results are summarized in Fig. 3, where the fast component reflects the $e-\text{ph}$ interaction [Figs. 3(a) and 3(b)] and the slow component reflects the across-gap quasiparticle recombination, which is influenced by the phonon-induced pair breaking [Figs. 3(c) and 3(d)]. It can be seen that,

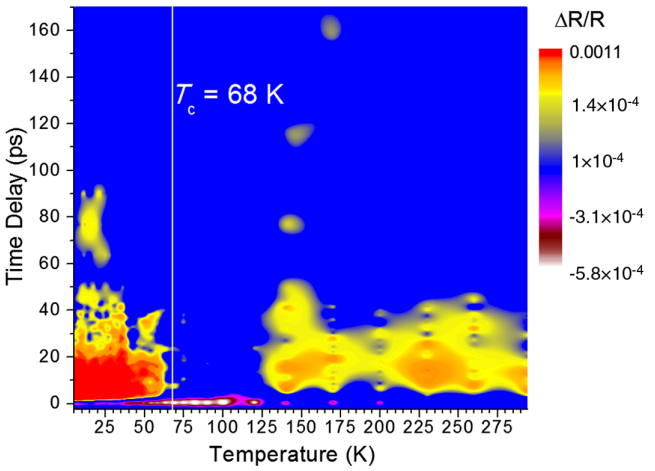

FIG. 2. Qualitative evidence of the SC transition. The two dimensional mapping of the differential reflectivity as a function of both temperature and time illustrates qualitatively that a SC phase transition exists at ~68 K. The white line marks the SC T_c. The temporal step sizes are 0.23 ps for the $-2.8-43.5$ ps range and 3.33 ps for the 43.5–173 ps range; the temperature step size varies, as explicitly shown in Fig. 3. Appropriate interpolation is used to make smoother coloring.

with increasing temperature, A_{slow} decreases steadily at $T < 68$ K, drops abruptly at approaching 68 K, and then levels off at $T > 68$ K to nearly a constant [Fig. 3(c)]. Moreover, a distinct divergence in τ_{slow} is clearly seen at a slightly lower temperature than 68 K [Fig. 3(d)]. The simultaneous observation of the two prominent changes strongly suggests a SC phase transition [14,16,17,23,24]. Quasiequilibrium between quasiparticles and bosons are formed in the SC state and their shared lifetime is determined by the so-called phonon bottleneck effect (see below discussion). At across the T_c, the closing of the SC gap certainly causes prominent changes in both the density and lifetime of the quasiparticles. Note that usually T_c is slightly higher than the temperature corresponding to the lifetime singularity [14,23,24]. We ascribe the 103 K peak in Fig. 3(d) to the antiferromagnetic to paramagnetic phase transition of the FeTe layer. It does not correspond to a SC transition, because no simultaneous change in A_{slow} was found. In addition, the 160 ps lifetime [Fig. 3(d)] is nearly identical to that for a separate 10 UC FeTe/STO sample (see details in the Supplemental Material [22]). The alternative possibility that $T_c = 103$ K is also straightforwardly ruled out by time-domain data fitting results [22]. Despite the strong background of FeTe, we have clearly observed the SC transition of FeSe. This demonstrates that our method is single-layer sensitive and might apply to other interface investigations [25].

Microscopically, after the coherent photoexcitation, the quasiparticles at high excited state far above the Fermi surface relax to right above the SC gap to form a quasiequilibrium quantum mixture with the high frequency phonons (abbreviated as HFP, with $E_{\text{ph}} > 2\Delta$, where Δ is the SC gap). The balance between the formation and

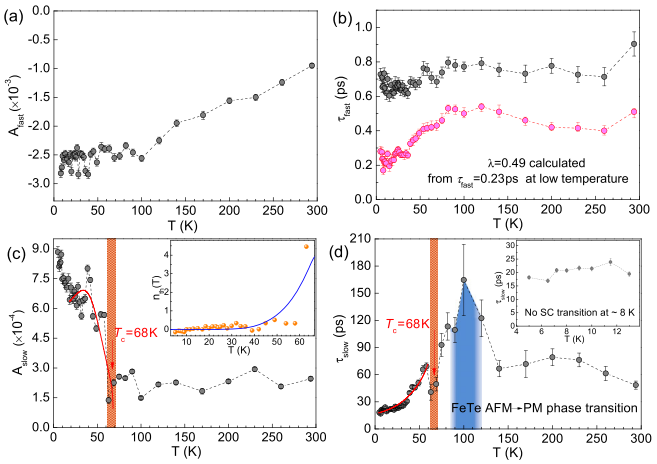

FIG. 3. Quantitative evidence for SC transition at $68(-5/+2)$ K. Temperature dependences of the amplitude (a) and lifetime (b) of the fast component, where the red dots in (b) are the deconvoluted data. Temperature dependences of the amplitude (c) and lifetime (d) of the slow component, where the red lines are theoretical simulation curves. The orange stripes mark the T_c value of 63–70 K. The light-blue stripe marks the non-SC phase transition in the FeTe capping layer. Inset: Density of thermally excited quasiparticles (c) and zoom-in view of the data around 8 K (d).

deformation of the Cooper pairs evolves in a way dictated by the phonon population—the phonon bottleneck effect. This effect is most directly and effectively observed using time-resolved ultrafast spectroscopy and can be understood using the Rothwarf-Taylor description of the $e-$ph balancing [26]. In the model, HFPs break Cooper pairs during the electron condensation process, thus together with the quasiparticles constitute a dynamic equilibrium that can only be mitigated by the decay and escape of the HFPs. This is known as the phonon bottleneck effect, which can be identified from Fig. 3(d) (see the larger $\tau_{\rm slow}$ value around 60 K).

Quantitatively, we simulate the results in Fig. 3 by the model used by Kabanov *et al.* for a Cooper-pairing superconductor. The gap-sensitive nature of the ultrafast dynamics is reflected, for an $e-$boson interacting system, through the temperature-dependent differential reflectivity as [14]

$$|\Delta R/R| \propto \frac{\varepsilon_I/[\Delta(T)+k_BT/2]}{1+\frac{2\nu}{N(0)\hbar\Omega_c}\sqrt{\frac{2k_BT}{\pi\Delta(T)}}e^{-\Delta(T)/k_BT}}, \qquad (1)$$

where ε_I is the absorbed laser energy density per unit cell, Ω_c is the cutoff frequency of the phonons, ν is the effective number of interacting phonon branches per unit cell, $N(0)$ is the density of states at the Fermi surface, and we have assumed a symmetric [1,2,22] gap $\Delta(T)=\Delta(0)[1\text{-}(T/T_c)]^{1/2}$ nearby T_c. With Eq. (1) we fit the data in Fig. 3(c) and find that the fitting curve for $A_{\rm slow}$ has a sharp decrease ending at 68 K. As for other conventional, cuprate, and iron-based superconductors, this ending temperature is the T_c of a SC phase transition [14,17,23,24]. In parallel, using the same model, the gap-dependent lifetime of the quasiparticles (and phonons in equilibrium) can be described as [14]

$$\tau_{\rm slow}=\tau_{\rm ph}=\frac{\hbar^2\omega^2\ln\{1/[\varepsilon_I/2N(0)\Delta(0)^2+e^{-\Delta(T)/k_BT}]\}}{12\Gamma_\omega\Delta(T)^2}, \qquad (2)$$

where ω is the frequency of the HFP and Γ_ω is the phonon linewidth. With Eq. (2) we fit the $\tau_{\rm slow}$ data in Fig. 3(d), which exhibits a distinct singularity around 60 K. Multiple tries of the fitting parameters give a 63 K $< T_c <$ 70 K bound, with the best optimized value of $T_c=68$ K. Note that by considering the systematic error in the temperature measurements, the T_c value of the sample can only be slightly higher than, but no lower than, 68 K.

Simultaneously, the above analysis yields $\Delta(0)=3.2$–$3.7\ k_BT_c$ (k_B is the Boltzmann constant), i.e., $\Delta(0)=20.2\pm1.5$ meV, which is in excellent agreement with the reported 20.1 meV [1], 19 meV [3] and 3.0–3.5 k_BT_c [3] values. In the angle-resolved photoemission spectroscopy measurements of Refs. [2–5], removal of the capping layer, or annealing, or storing for 4–6 days prior to measurements occurs, all causes a change in the gap. In transport measurements, the electrical contact frequently causes defects to the very thin active layer. Generally speaking, the variation in the gap and T_c results from the sample-to-sample quality fluctuation in the films where the measurements were conducted. Compared to these, ultrafast spectroscopy provides a noninvasive means of investigating high T_c superconductors. Hence, it is not surprising that we have observed higher gap and T_c values than in the *ex situ* transport measurements, given the same sample quality. Without the capping layer, we expect that our ultrafast spectroscopy will give higher T_c values for *in situ* measurements. Also obtained is the density of thermal quasiparticles $n_{\rm th}$ [see inset of Fig. 3(c)], which increases prominently as the temperature approaches T_c.

We can directly obtain λ from the quasiparticle lifetime. The relaxation dynamics is determined by the $e-$ph coupling with $\lambda\langle\omega^2\rangle=(\pi/3)(k_BT_e/\hbar\tau_{e-\rm ph})$, where $\tau_{e-\rm ph}$ can be directly measured by our pump-probe spectroscopy [i.e., $\tau_{\rm fast}=0.23$ ps, see the red dots in Fig. 3(b), which are the deconvolution results of the raw $\tau_{\rm fast}$ values] and $\lambda\langle\omega^2\rangle$ is the second moment of the Eliashberg function [27]. From the laser fluence we estimate that $T_e=902$ K. We speculate that the $e-$ph coupling is mainly between the FeSe electrons and the STO (or the interface) phonons, owing to the vast atoms at the STO (and the interface) contributing to the phonon bath. For example, single layer FeSe grown on graphene is not superconducting [21]. Taking the experimentally measured low optical phonon frequency near Γ for bulk STO [28], $\hbar^2\langle\omega^2\rangle=22^2(\text{meV})^2$, we obtain that $\lambda=0.48$. As a comparison, it was measured that $\lambda=0.16$ for bulk FeSe by also using ultrafast spectroscopy [16]. Even if taking the FeSe A_{1g} phonon as in Ref. [16] to calculate, the value

of λ is even larger and still much larger than the bulk value. We thus identify that $e-$ph coupling plays a more crucial role in the SC mechanism for the 1 UC FeSe/STO system.

To date, in the bulk FeSe [29] and other iron-based superconductors, electron-electron interactions are widely believed to mediate superconductivity (e.g., Refs. [30–34]). However, the electronic structure of the 1 UC FeSe/STO systems is qualitatively different [2]. Moreover, owing to the strong interaction with the substrate, the electron-electron correlations are found to be significantly different [35]. Alternatively, interface interaction was theoretically illustrated to enhance $e-$ph coupling in a variety of ways [10,11]. Here our experiment demonstrates a much larger λ ($=0.48$) in the 1 UC FeSe/STO systems, leading us to believe that the $e-$ph interaction plays the likely critical role in the high T_c superconductivity, even if the role of electron-electron correlations cannot be excluded.

To further probe the pairing glue and why a capping layer leads to the reduction of T_c [12], we investigated the effect of the capping layer. We prepared a separate sample of 2 UC FeTe on STO (abbreviated as 2 UC FeTe/STO), where we unambiguously generated and detected a coherent acoustic phonon branch [18,19,36,37] in the FeTe layer. Figure 4(a) reveals a clear periodic phonon oscillation that superimposes on a single-exponential electronic decay. After removal of the electronic relaxations, the coherent acoustic phonon waves are explicitly displayed in the lower panel of Fig. 4(a). A phonon frequency of 0.05 THz (0.2 meV) is obtained by Fourier transformation in the inset of Fig. 4(a). The corresponding period of the acoustic phonon wave is 20 ps, of which the lifetimes are 274 ps at 4.5 K, 262 ps at 24 K, and 200 ps at 110 K, respectively. An identical acoustic phonon mode is also observed in a 10 UC FeTe/STO sample [22], where the oscillation is more prominent due to the thicker FeTe layer. Note that observing a coherent acoustic phonon mode in a single layer of atoms has rarely been reported [37] due to the weak signal and strong background.

We contemplate that this observed acoustic phonon branch in the FeTe capping layer forms a decay channel for the gluing bosons in the SC condensate, and thus reduces the T_c value [12] of the 2FeTe/1FeSe/STO sample. This scenario is schematically illustrated in Figs. 4(b) and 4(c). Although many kinds of bosons can be the glue, we take phonons as an example to illustrate the physics picture. The optical phonons in the STO and interface tunnel through the single FeSe layer to decay into the acoustic phonon branch in the FeTe capping layer. In the momentum space, the relaxation of the high energy optical phonons (possibly the gluing boson) is boosted by the additional decay channel due to the FeTe acoustic phonons. Here all features are drawn to energy scale and the phonon band structure is based on the calculation in Ref. [28]. Although other origins such as defect, impurity, interference, and magnetic scattering may also play a role in reducing T_c, anharmonic decay of the optical phonons into acoustic phonons is both intrinsic and efficient [38]. Moreover, additional channels of anharmonic decay will

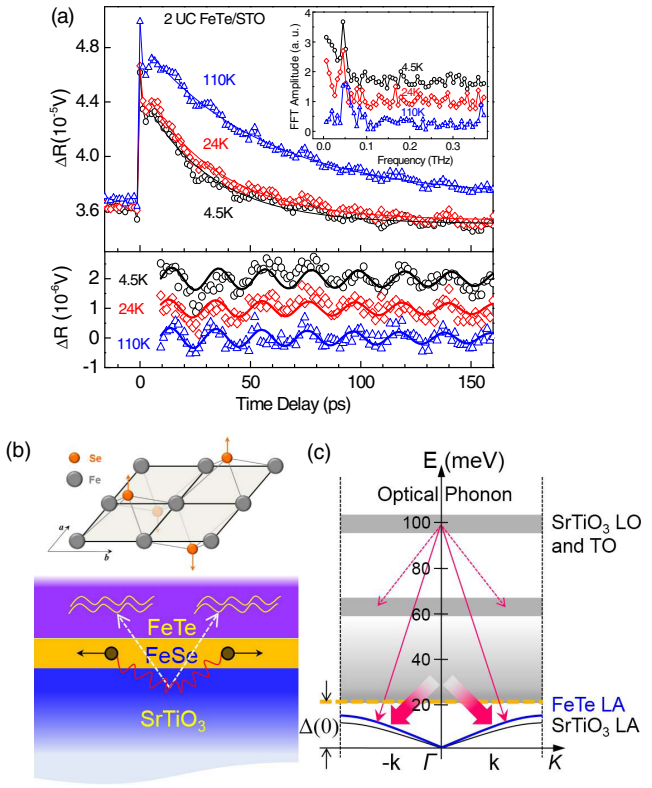

FIG. 4. Coherent acoustic phonon in the capping layer and the decay of the gluing bosons. (a) Ultrafast dynamics of the 2 UC FeTe/STO sample. Solid curves: single exponential fittings. The superimposed coherent acoustic phonons are further displayed in the lower panel. Inset: Fourier transformation. (b) Schematic phonon decay tunneling through the FeSe layer. Red wave: optical phonons in STO and at the interface. Yellow waves: acoustic phonons in the FeTe capping layer. (c) Corresponding K-space schematic diagram. Thick red arrows: new channels of relaxation. LO, TO, and LA: longitudinal optical, transverse optical, and longitudinal acoustic phonons.

result in additional broadening of the optical phonon branch [38], thus leading to less sharp SC transition. This is possibly reflected in the transport measurements [1,7,8].

In Fig. 4(a), it can be seen the quasiparticle dynamics of 2 UC FeTe/STO contains only a single exponential decay. We show its temperature dependence in Fig. S7 [22], whereby the amplitude and lifetime do not show characteristics of a superconductor. Thus, the SC response we observed is not due to the FeTe layer even though it is reduced in thickness. Furthermore, our additional experiments on reflections and control experiments on 2 UC FeTe/STO show that FeSe dominates the reflection signal and the SC response clearly comes from FeSe rather than FeTe [22].

In concluding remark, we have observed a high temperature SC transition with $T_c = 68\ (-5/+2)$ K in a 2FeTe/1FeSe/STO sample through the investigation of the quasiparticle ultrafast dynamics. This is the highest T_c value reported in capped single-layer FeSe samples. Gap energy of $\Delta(0) = 20.2 \pm 1.5$ meV, i.e., $\Delta(0) = 3.2$–3.7 $k_B T_c$, is observed. Stronger than bulk material $e-$ph

coupling is identified with $\lambda = 0.48$, which reveals the likely crucial role of $e-\mathrm{ph}$ coupling in the SC mechanism. Moreover, we have observed a 0.05 THz coherent acoustic phonon branch in the FeTe capping layer, which enhances the decay of the pairing bosons and well explains why a capping layer restrains T_c. Our experimental results, condensed from the excited state evolution, constitute an independent evidence of the high T_c SC transition of the 1 UC FeSe/STO systems. Our investigation demonstrates that ultrafast optical spectroscopy can be single-layer sensitive to phase transition, which paves way for noninvasive investigation of single-layer or interface quantum materials.

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