Carrier relaxation dynamics in high temperature superconductors

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In this report we review recent experimental results on photoexcited carrier relaxation dynamics on high temperature superconductors (HTSC) probed by a femtosecond time-resolved optical spectroscopy, and compare the results with the data obtained on quasi two dimensional charge density waves. In these experiments, a femtosecond laser pump pulse excites electron-hole pairs via an inter-band transition in the material. These hot carriers rapidly release their energy via electron-electron and electron-phonon collisions reaching states near the Fermi energy within \( \sim 100 \) fs. If an energy gap is present in the low-energy density of states (DOS), it inhibits the final relaxation step and photoexcited carriers accumulate above the gap causing a transient change in reflectivity arising from excited state absorption. The relaxation and recombination processes of photoexcited quasiparticles, governed by the magnitude, anisotropy and the T-dependence of the low energy gap, are monitored by measuring the resulting photoinduced absorption as a function of time after the photoexcitation. This way, the studies of carrier relaxation dynamics give us direct information of the T-dependent changes in the low energy DOS. The technique is particularly useful to probe the systems with spatial inhomogeneities, where different local environments give rise to different relaxation rates. The data on series of HTSC-s show evidence for the coexistence of two distinct relaxation processes, whose T-dependences seem to be governed by two different energy scales: a T-independent pseudogap and a mean-field-like T-dependent gap that opens at \( T_c \). The data suggest the origin of the two-gap behavior is in the intrinsic microscopic spatial inhomogeneity of these materials.

I. INTRODUCTION

Femtosecond time-resolved optical spectroscopy has been shown in the last couple of years to present an excellent alternative to more conventional time-averaging frequency-domain spectroscopies for probing the changes in the low energy electronic structure in strongly correlated systems. In these experiments, a femtosecond laser pump pulse excites electron-hole pairs via an interband transition in the material. In a process which is similar in most materials including metals, semiconductors and superconductors, these hot carriers rapidly release their energy via electron-electron and electron-phonon collisions reaching states near the Fermi energy within 10-100 fs. Further relaxation and recombination dynamics, determined by measuring photoinduced changes in reflectivity or transmission as a function of time after photoexcitation, depends strongly on the nature of the low-lying electronic spectrum. In particular, the experimental technique was found to be sensitive to opening of the superconducting gap, \( \sim 100 \) fs. If an energy gap is present in the low-energy density of states (DOS), it inhibits the final relaxation step and photoexcited carriers accumulate above the gap causing a transient change in reflectivity arising from excited state absorption. The relaxation and recombination processes of photoexcited quasiparticles, governed by the magnitude, anisotropy and the T-dependence of the low energy gap, are monitored by measuring the resulting photoinduced absorption as a function of time after the photoexcitation. This way, the studies of carrier relaxation dynamics give us direct information of the T-dependent changes in the low energy DOS. The technique is particularly useful to probe the systems with spatial inhomogeneities, where different local environments give rise to different relaxation rates. The data on series of HTSC-s show evidence for the coexistence of two distinct relaxation processes, whose T-dependences seem to be governed by two different energy scales: a T-independent pseudogap and a mean-field-like T-dependent gap that opens at \( T_c \). The data suggest the origin of the two-gap behavior is in the intrinsic microscopic spatial inhomogeneity of these materials.

Since the optical penetration depth in these materials is on the order of 100 nm, the technique is essentially a bulk probe. Moreover, since the effective shutter speed is on the order of a picosecond, the technique is particularly useful to probe the systems with (dynamic) spatial inhomogeneities. In this case different local environments (that appear frozen on the timescale of picoseconds) give rise to different components in measured photoinduced reflectivity (transmissivity) traces. As the different components can have different time scales, temperature, photoexcitation intensity, and probe polarization or wavelength dependences, they can be easily extracted.

In this report we summarize some of the recent results on various high-temperature superconductors (HTSC), where the data show evidence for the coexistence of two distinct relaxation processes whose T-dependences seem to be governed by different energy gaps: a T-independent pseudogap and a mean-field-like T-dependent gap that opens at \( T_c \). The data suggest the origin of the two-gap behavior is in the intrinsic microscopic spatial inhomogeneity of these materials. In additions we present experimental data on two prototype quasi two dimensional (2D) charge density wave (CDW) dichalcogenides \( 1T-TaS_2 \) and \( 2H-TaSe_2 \), and compare these results with the data on HTSC.
II. EXPERIMENTAL DETAILS

In these experiments, a Ti:sapphire mode-locked laser operating at a 78 MHz repetition rate and pulse length of 50 fs was used as a source of both pump and probe pulse trains. The wavelength of the pulses was centered at approximately \( \lambda \approx 800\text{nm} \) (1.58eV) and the intensity ratio of pump and probe pulses was approximately 100:1. The pump and probe beams were crossed on the sample’s surface, where the angle of incidence of both beams was less than 10\(^\circ\). The diameter of both beams on the surface was \( \sim 100\mu\text{m} \). The typical energy density of pump pulses was \( \sim 1\ \mu\text{J/cm}^2 \), which produces a weak perturbation of the electronic system with approximately \( 3\times10^{10} \) thermalized photoexcited carriers per pulse (the approximation is based on the assumption that each photon creates \( h\omega/\Delta \) thermalized photoexcited carriers, where \( \Delta \approx 40\text{ meV} \) is of the order of the superconducting or CDW gap). The train of the pump pulses was modulated at 200kHz with an acousto-optic modulator and the small photoinduced changes in reflectivity or transmission were resolved out of noise with the aid of phase-sensitive detection. The pump and probe beams were cross-polarized to reduce scattering of pump beam into the detector (avalanche photodiode).

A detailed description of the experimental technique can be found in Ref. [2].

III. CARRIER DYNAMICS IN CUPRATES AND 2-D CHARGE DENSITY WAVES

In this section we discuss some recent experimental results on carrier relaxation dynamics on HTSC and low dimensional CDW’s with particular focus on comparison of the dynamics in the two systems and its implications. The comparison of carrier relaxation dynamics between the two systems is particularly interesting, since HTSC and quasi-2D CDW’s are thought to exhibit some important similarities. Both are layered, highly anisotropic materials which are often described in terms of a quasi-2D Fermi surface (FS) in their normal state. In HTSCs, it is commonly believed that the superconducting gap has nodes along certain directions on the FS due to the \( d \)-wave component of the order parameter, whereas in 2D-CDW systems a CDW gap is also expected only along certain wavevectors, remaining gapless (and metallic) on other regions of the FS. The low-energy single particle excitations in the two classes of compounds might therefore be expected to show some important common features related to reduced dimensionality. However, the validity of the Fermi-liquid (FL) concept when applied to low-temperature properties in HTSCs has repeatedly been brought into question, suggesting that new insight into the physics of quasi-2D systems may be gained by investigating the low-energy electronic gap structure and carrier recombination dynamics of quasi-2D CDW dichalcogenides with femtosecond spectroscopy.

Figure 1 shows the raw photoinduced reflectivity data on high temperature superconductor \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) (YBCO), compared to two quasi two dimensional charge density wave systems 1T-TaS\(_2\) and 2H-TaSe\(_2\). The data are plotted with solid symbols, together with solid lines representing the best fit obtained using the function \( \Delta R/R = f(t)[A \exp(-t/\tau_1) + B \exp(-t/\tau_2) + .. + D] \). Here \( f(t) = H(t)[1 - \exp(-t/\tau_{ee})] \) represents the finite risetime, with \( H(t) \) being the convoluted Heaviside step function accounting for the finite laser pulse width, and \( \tau_{ee} \) is the electron-electron thermalization time. The decay dynamics is described by one or more exponential decays with corresponding amplitudes \( A, B, .. \) and relaxation times \( \tau_1, \tau_2, .. \) and a possible long lived component with amplitude \( D \) (lifetime of the long lived component is shown to be longer then the separation between two successive excitation pulses of 12 ns).

In the case of charge density waves, the oscillatory component with frequency \( \omega \) and damping constant \( \Gamma \) has been observed, superimposed on the decaying transient. The oscillatory transient was found to be due to coherently excited collective mode - amplitudon, in very good agreement with Raman spectroscopy data [3, 4], and will not be discussed in detail here. In this report we focus on the picosecond decay dynamics, described by the temperature, photoexcitation intensity and the probe polarization dependence of amplitudes \( (A,B,..) \) and relaxation times \( (\tau_1, \tau_2,..) \), whose temperature dependences are shown in Fig. 2.

Figure 2 a) and d) presents the generic temperature dependence of relaxation times and amplitudes of the photoinduced transient obtained on HTSC over the last decade or so [5, 6]. At temperatures below \( T_c \), the relaxation dynamics is showing a two-component behavior, with the two components having the same or opposite signs depending on the probe photon wavelength and/or polarization of the probe light with respect to the crystal axis. The component that is present only below \( T_c \) shows vanishing amplitude (dashed line in Fig. 2 d)) and diverging relaxation time (Fig. 2 a)) as the temperature approaches \( T_c \). This behavior is attributed to opening of the superconducting gap and corresponding quasiparticle (QP) relaxation bottleneck that occurs due to the fact that there are no ingap states available in the process of QP recombination to the condensate. The model [7] is based on the fact that near \( T_c \) the gap is small and the number of thermally excited QPs is large compared to the number of photoexcited ones. In this case the process of recombination of two QPs to the condensate is fast. However, since recombination of two QPs creates high energy photon, which can further excite QP pairs, the relaxation dynamics near \( T_c \) is governed by anharmonic phonon decay. As the gap \( \Delta(T) \) is closing, the specific heat of phonons with energy less than the gap energy is becoming smaller, and the relaxation time increases as \( \tau \propto 1/\Delta(T) \). The relaxation time of the second
component has been found to show only weak temperature dependence. The studies of T-dependence of the amplitude of the transient as a function of doping have revealed, that it is associated with the pseudogap energy scale. The coexistence of the two relaxation component below \( T_c \) implies some kind of microscopic phase separation, with coexisting areas of high carrier and low carrier densities.

In order to test the predictions of the theoretical model and the implications of the experimental results on the physics of high temperature superconductors, we have applied the same technique to study 1D-CDW and quasi 2D-CDW compound. In the following we discuss the observed T-dependences of the relaxation dynamics in 1T-TaS\(_2\) and 2H-TaSe\(_2\) in view with the spectroscopic data on associated changes in the low energy density of states.

At room temperature 1T-TaS\(_2\) is in a nearly-commensurate \((hc-)\) CDW phase. Around \( T_{nc-c} = 200 \) K it undergoes a strongly first-order ”lock-in” transition to a \( c\)-CDW state. In spite of the expected appearance of a gap in parts of the Fermi surface due to nesting at \( T_{nc-c} \), the whole FS was found to exhibit a ”pseudogap” feature already at room temperature with a finite density of states (DOS) at \( E_F \). Upon lowering the temperature through \( T_{nc-c} \), a further abrupt decrease in the DOS is observed near \( E_F \), accompanied by an order of magnitude increase in resistivity. Yet in spite of the presence of a CDW gap at low temperatures 1T-TaS\(_2\) is reported to have a small but finite DOS at \( E_F \) in the low-temperature \( c\)-phase. The picosecond relaxation dynamics in 1T-TaS\(_2\) requires a \( \text{stretch exponential decay} \) fit, \( A \exp(-(t/\tau)^p) \), with \( p \approx 0.5 \) to fit the data adequately. (Although the SP transient in 1T-TaSe\(_2\) can also be fit by the sum of two exponentials, the two components have the same \( T \)-dependence, which suggests that we are dealing with a single relaxation process with non-exponential dynamics rather than two distinctly independent processes).

The observation of a stretch exponential decay - which typically describes systems with a spread of relaxation times - is consistent with the observed finite DOS at \( E_F \) below \( T_{\tau-n-c} \). Since \( \tau \approx 1/\Delta \) the observed stretch exponential decay actually implies a near-Gaussian spread of \( 1/\Delta \). The \( T \)-dependences of the amplitude \( \tau \) (using \( s = 0.5 \) for \( 1T\)-TaS\(_2\)) are plotted in Fig. 2 e) and b), respectively. The relaxation dynamics are clearly strongly affected by the lock-in transition around 200K. We observe an abrupt hysteretic change of both amplitude and \( \tau \) around \( T_{nc-c} \), consistent with the hysteresis observed in transport measurements. Above 230 K the photoinduced transient is fast and very weak.

2H-TaSe\(_2\) on the other hand is expected to bear close resemblance to cuprates. It exhibits metallic properties above room temperature. Upon cooling it undergoes a second order phase transition to an incommensurate \((s-)\) CDW state at \( T_{n-s} = 122 \) K. This phase transition is reportedly accompanied by the appearance of a gap in the Fermi surface (FS) centered at the K point, but apparently - according to photoemission studies - remains gapless on the part of the FS centered at the L point. The transition is accompanied by a decrease in the scattering rate and a corresponding \text{drop} in resistivity. The onset of a \( c\)-CDW phase at \( T_{\tau-c} = 88K \) leaves the excitation spectrum as well as the transport and thermodynamic properties almost unaffected. Since below \( T_{\tau-c} \), the system is expected to have a highly anisotropic gap with gapless regions over parts of the FS, the relaxation dynamics in 2H-TaSe\(_2\) is expected to be non-exponential as in 1T-TaS\(_2\). However, the relaxation dynamics in 2H-TaSe\(_2\) over the entire \( T \)-range can be described well by the single-exponential decay. Moreover, below \( T_{\tau-c} \) both the \( T \)-dependence of amplitude and relaxation time \( \tau \) obtained from the single exponential fit to the data on 2H-TaSe\(_2\), plotted in Fig. 2 f) and c), show extremely good agreement with the prediction of the model for carrier relaxation across a well-defined temperature-dependent gap. The theoretical fit to the data using the model by Kabanov et al. with a BCS \( T \)-dependence of the gap is superimposed (lines), giving the value of the gap of \( 2\Delta(0) = 70 \) ± 10 meV. The value is somewhat smaller than the maximum gap obtained from tunneling, but in good agreement with the maximum gap value of 65 meV from ARPES. Above \( \sim 140 \) K the relaxation time and \( S(T) \) show only a weak \( T \)-dependence with \( \tau \approx 0.1 \) – 0.17 ps. This time is close to the expected electron-phonon thermalization time (in case there is no gap in the DOS, like in metals, the photoinduced reflectivity/transmissivity dynamics is governed by energy transfer from thermalized electron subsystem to the lattice). On the other hand, the behavior of amplitude just above \( T_{\tau-n-c} = 120 \) K is rather unusual, showing a rapid increase with increasing temperature between 120 and \( \sim 140 \) K. This could be attributed to the presence of short-range segments of ordered CDW, as observed in quasi-1D CDW K\(_{0.3}\)MoO\(_4\).

The emerging picture based on the time-domain measurements on HTSC and 2H-TaSe\(_2\) presented here is one in which the low-temperature state shows a \text{clear large gap} in the excitation spectrum on the femtosecond timescales (not just a depression in the DOS such as is observed in time-averaged experiments). There is also clear evidence for very slow relaxation of excitations whose energy is less than the maximum gap - in both HTSC and 2H-TaSe\(_2\). The observed behavior is in clear contradiction with a FL interpretation, where the QP relaxation would be expected to occur primarily in the gapless regions of the FS (in the nodes for the case of superconductors). The observation of only a large gap on the femtosecond timescale implies that there are certain momenta associated with the gapless regions which are either inaccessible to quasiparticles, or - implying a breakdown of the FL picture altogether - simply that \text{extended} states with these wavevectors do not exist at all. The latter behavior is consistent with the notion of fluctuating \text{locally ordered} regions in space, in which case it becomes clear why one cannot speak of FL-like
quasiparticle excitations with well defined momenta. The precursor "pseudogap state" appears to be associated with the fluctuating presence of fully gapped short-range-ordered CDW patches or segments, similar to the locally gapped regions in real space arising from a statistically fluctuating population of pre-formed pairs in HTSCs.

**IV. LOW TEMPERATURE RELAXATION TIME IN CUPRATES**

In this section we analyze the relaxation dynamics in high temperature superconductors at low temperatures ($T \ll T_C$) and low excitation fluences, that has gained considerable interest in the last couple of years. It has been shown that at extremely low fluences the low temperature relaxation time depends on photoexcitation intensity ($P$), and in the limit of $P \rightarrow 0$ the relaxation time diverges as $T \rightarrow 0$ K. This has been consistently observed in the extremely low excitation regime on Bi2212, Tl2201, Hg1223, underdoped YBCO, Bi2201, and LASCO.

We interpret the low temperature (low photoexcitation fluence) increase of relaxation time in terms of QP recombination bottleneck. Namely, the assumption that the two-particle recombination is fast compared to the anharmonic phonon decay (that determines the decay of photoexcited population density ($n_{pe}$) at temperatures close to $T_c$), can be violated at low temperatures, when the gap is large and the density of thermally excited QPs ($n_{th}$) is small. It can lead to a situation when the recombination time becomes longer than the anharmonic phonon decay time, and in this case the relaxation time of photoexcited QP density is governed by bi-particle recombination process. To show this, we have to bare in mind that in this low excitation fluence experiments the number of thermally excited QPs is, except at very low temperatures, much larger than the number of photoexcited ones. In this limit, the rate equation describing the dynamics of QP density $n$ can be written as

$$\frac{\partial n}{\partial t} = -\beta \left( n^2 - n_{th}^2 \right) ; \quad n = n_{th} + n_{pe},$$

(1)

where $\beta$ is a constant. Due to the fact, that at low excitation densities $n_{th} \gg n_{pe}$ in most of the temperature range, we can linearize the equation to obtain

$$\frac{\partial n_{pe}}{\partial t} = -\frac{n_{pe}}{\tau_R} ; \quad \tau_R = (2\beta n_{th})^{-1}.$$  

(2)

Since $n_{th} \propto \exp(-\Delta/T)$, where $\Delta$ is the superconducting gap, the relaxation time is expected to increase exponentially at low temperatures. In the limit where $n_{th} \gg n_{pe}$ the relaxation dynamics is expected to be exponential. On the other hand, at very low temperatures, when $n_{pe} \gg n_{th}$, the Eq.(1) cannot be linearized and the dynamics is non-exponential. More rigorously, the relaxation time in the limit $n_{th} \gg n_{pe}$ and $T \ll T_c$ can be obtained explicitly by considering the kinetic equation for QPs:

$$\tau_R = \frac{\hbar \Omega_c^2}{32\pi \lambda \Delta^2 \sqrt{\pi \Delta k_B T / 2}} \exp \left( \frac{\Delta}{k_B T} \right).$$

(3)

Here $\lambda$ is the dimensionless electron-phonon coupling constant, for HTSC typically on the order of 1, and $\Omega_c$ is a typical phonon cutoff frequency. There is a subtle point to Eq.(3) that needs to be mentioned: temperature $T$ in Eq.(3) is actually the temperature of phonons with energy less than $2\Delta$, and can be slightly higher than the equilibrium sample temperature. These corrections can be important at very low temperatures (see Ref. [9] for details).

Fig. 3 a) shows the raw photoinduced reflectivity data (symbols) on Hg1223 at different temperatures, together with fits (solid lines) using a single exponential decay. We should note that even though the lowest sample holder temperature in the experiments was 4 K, the actual lowest temperature in the illuminated spot is substantially higher (~ 40 K) due to heating induced by excitation pulse trains. The temperature increase is determined by thermal conductivity of the sample, and can be easily accounted for, giving the uncertainty in temperature of ±2 K. The extracted relaxation time as a function of temperature is plotted in Fig. 3 b) using solid symbols, compared to the theoretical predictions for $\tau$ governed by recombination bottleneck [Eq.(8)] - solid line, and $\tau$ determined by phonon bottleneck (Eq.(28) of Ref.[1]) - dashed line. As we can see, the recombination bottleneck describes well the low temperature behavior. At higher temperatures a crossover to phonon bottleneck determined relaxation is expected. A crossover from high temperature relaxation to low temperature recombination picture is expected to highly depend on the magnitude of the superconducting gap $\Delta$. Since the gap value in YBCO, determined from tunneling data is lower than that of Bi2212 and Hg1223 the crossover in YBCO is expected to be lower in temperature (or photoexcitation intensity). At lower temperatures, when $n_{pe} \gtrsim n_{th}$, the relaxation should be non-exponential. Indeed the crossover to non-exponential relaxation was reported at very low temperatures in Bi2212 and Tl2201.
We should mention an alternative model for carrier relaxation dynamics in cuprates recently suggested by Segre et al. Analyzing the dependence of $\tau$ on temperature and photoexcitation intensity $P$, the authors argue that the decay in $\Delta R/R$ is determined by QP thermalization, rather than recombination. This is clearly inconsistent with published data on ultrafast conductivity dynamics on YBCO in the THz spectral region, where the recovery of the condensate due to recombination of QPs has been measured directly, and was found to be on a picosecond time-scale. In fact, the $T$-dependence of the condensate recovery time on optimally doped YBCO was found to be identical to the recovery time extracted from all-optical pump-probe experiments at 1.5 eV. Moreover, similar one-to-one relation between the relaxation dynamics measured with the two experimental techniques has recently been found also on MgB$_2$. Furthermore, in Fig. 3 of Ref. [16] it is shown that the relaxation rate ($1/\tau$) is proportional to the amplitude of the signal. Such behavior is well known in superconductors and arises from QP recombination to the condensate. Originally it was discussed on a phenomenological level by Rothwarf and Taylor and later derived from kinetic equations for superconductors. We believe that the linear intensity dependence of the relaxation rate is consistent with a QP recombination mechanism presented above, rather than QP thermalization, as proposed by Segre et al.

V. CONCLUSIONS

The time-resolved experiments on high temperature superconductors and quasi 2D charge density waves show that irrespective of the fundamental underlying cause for the instability, these quasi-2D materials, show a transition from a high-temperature uniform metallic state to a low-temperature correlated state via the formation of a dynamically inhomogeneous intermediate state. Since the timescale of the measurement is on the order of a picosecond, and the inhomogeneities appear to be frozen on this timescale, different local environments give rise to different relaxation dynamics. In this way these measurements on high temperature superconductors reveal the coexistence of high and low carrier density regions, with the recovery dynamics being governed by superconducting gap and pseudo-gap respectively. Since the lateral dimension of the inhomogeneities, as determined for example by EXAFS, is on the order of several unit cells, the interpretation of the data in terms of quasi-2D Fermi liquid is questionable. In fact, the observation of a clear large gap in these experiments implies that there are certain momenta associated with the gapless regions which are either inaccessible to quasiparticles, or simply that extended states with these wavevectors do not exist at all. The time-averaged response (such as is observed in ARPES or infrared spectra) may then be thought of as the superposition of the different components in the inhomogeneous state, while the observed anisotropy reveals the directionality of the interaction which leads to the formation of long range order.

Carrier dynamics studies on quasi-2D charge density waves have been performed, to elucidate the carrier relaxation dynamics in cuprates. We confirm, that the dynamics is sensitive to the opening of the gap in DOS. In 1T-TaS$_2$ the relaxation dynamics is non-exponential, consistent with the spectroscopic data showing finite DOS at $E_F$ at all temperatures. On the other hand, single exponential decay dynamics, and the $T$-dependence of amplitude and relaxation time in 2H-TaSe$_2$ suggests similar behavior as in cuprates, where low temperature correlated state is formed via local precursor CDW segments.

Finally, we analyze the low-temperature, low excitation fluence dynamics in cuprates, where the relaxation time was found to increase exponentially as $T \rightarrow 0$. We show, that in this limit the recovery dynamics of photoexcited quasiparticle density is governed by bi-particle recombination process.

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VI. FIGURE CAPTIONS

Figure 1: Photoinduced reflectivity traces taken at different temperatures for a) optimally doped YBCO, b) 1T-TaS$_2$, and 2H-TaSe$_2$. In b) and c) the traces are offset for clarity. Inset to panel a): the two-exponential decay dynamics revealed on a semi-log scale. Insets to panels b) and c): The phase diagrams of bulk 1T-TaS$_2$ and 2H-TaSe$_2$. 
Figure 2: Panels a) – c): the temperature dependences of picosecond relaxation times of optimally doped YBCO, 1T-TaS$_2$, and 2H-TaSe$_2$. Panels d) – f): the corresponding T-dependences of amplitudes of photoinduced changes. Lines are fits to the data using the model - see text. Note the hysteresis observed in T-dependence of photoinduced transient on 1T-TaS$_2$.

Figure 3: a) The photoinduced reflectivity on Hg1223 taken at various temperatures below and above $T_c = 120$ K. Solid lines are fits to the data using single exponential decay. b) The temperature dependence of the relaxation time $\tau$ taken on Hg1223, compared to the theoretical curves for bi-particle recombination bottleneck model (solid curve) and phonon bottleneck model - dashed curve.

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YBCO

1T-TaS$_2$

2H-TaSe$_2$

Figure 2

Amp [norm.]

$\tau$ [ps]

Amp [norm.]

$\tau$ [ps]

$T$ [K]

$T$ [K]
Figure 3

(a) 

- \( \Delta R/R \times 10^{-4} \) vs. Time [ps]
- Symbols for different temperatures:
  - ■ 42 K
  - ○ 70 K
  - ▼ 115 K
  - △ 150 K

(b) 

- \( \tau \) [ps] vs. Temperature [K]
- \( \tau_{ph} \)
- \( \tau_R \)