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Structural transition dynamics in $FeSe_{0.8}Te_{0.2}$ thin film

Alexander Bartenev[1], Roman S. Kolodka[1], Camilo Verbel[1], Adrian Rua[1], Larry Theran[2], Felix Fernandez[1], Armando Rua[1], Sergiy Lysenko[1]

[1]*Department of Physics, University of Puerto Rico, Mayaguez, Puerto Rico 00681, USA*
[2]*Department of Electrical Engineering, University of Puerto Rico, Mayaguez, Puerto Rico 00681, USA*
e-mail: alexander.bartenev@upr.edu

Abstract: Time-resolved ultrafast reflectivity measurements along with the two-temperature model analysis reveal a complex interplay between optical nonlinearities, structural phase transition, electronic correlations, electron-phonon energy transfer, and the second moment of Eliashberg function in $FeSe_{0.8}Te_{0.2}$.

$FeSe_{0.8}Te_{0.2}$ is an iron-based superconductor with complex pressure- and doping-dependent electronic and structural properties, and with a certain correlation between suppressed nematicity and enhanced superconductivity [1]. This system has attracted increasing attention in recent years due to the remarkable superconducting temperature T_c, exceeding 100 K for single-layer FeSe films, attributed to interface-enhanced electron-phonon (*e-ph*) coupling [2]. The estimation of this coupling strength can be derived from the second moment of the Eliashberg function $\lambda\langle\omega^2\rangle$, as demonstrated previously [3, 4]. However, the relationship between *e-ph* coupling and sample temperature is not yet comprehensively understood.

In this work, we present ultrafast temperature-dependent optical dynamics of $FeSe_{0.8}Te_{0.2}$ upon femtosecond light excitation. Transient optical response simulated in terms of a two-temperature model (TTM) [5] reveals interplay between laser excitation and the second moment of Eliashberg function near the structural transition temperature T_S.

The $FeSe_{0.8}Te_{0.2}$ epitaxial film of 288 nm thickness was grown on CaF_2 single-crystal substrate by pulsed laser deposition. The pump-probe ultrafast optical spectroscopy measurements were performed in reflection geometry with a Ti:Sapphire femtosecond laser system operated at 1 kHz repetition rate, producing 35 fs laser pulses with a central wavelength of λ=800 nm, and 6 mJ/cm^2 fluence. The sample was mounted inside an optical cryostat with computer-controlled temperature adjustment.

Transient reflectivity $\Delta R(t)/R(0)$ of $FeSe_{0.8}Te_{0.2}$ measured within a temperature range from 8 K to 140 K was modeled by the TTM [Figs. 1(a-c)], as defined by the system of equations:

$$C_e(T_e)\frac{\partial T_e}{\partial t} = -G(T_e - T_L) + S(t) \quad (1)$$

$$C_L(T_L)\frac{\partial T_L}{\partial t} = G(T_e - T_L) \quad (2)$$

The TTM was solved numerically for electronic T_e and lattice T_L temperatures [Fig. 1(b)] versus time t, using the laser source term S, and the electronic C_e and lattice C_L heat capacities estimated from [6, 7]. An additional fitting algorithm for *e-ph* coupling constant G was applied. The transient reflectivity signal was modeled using the thermoreflectance equation:

$$\frac{\Delta R(t)}{R(0)} = a\Delta T_e(t) + b\Delta T_L(t) \quad (3)$$

Figure 1(a) shows a reflectivity signal at 8 K plotted along with the TTM fit. Constants a and b fitted from Eq. (3) indicate a sensitivity of transient reflectivity to changes in the electronic ΔT_e and lattice ΔT_L temperatures. The temperature dependent transient reflectivity data obtained from 8 K to 140 K [Fig. 1(c)] was also fitted to obtain these coefficients.

Figure 1(d) shows the temperature dependence of the fitting constants a and b in Eq. (3). This dependence illustrates the relative contribution of electronic and lattice temperatures to the transient reflectivity signal, revealing significant changes near the temperature of the orthorhombic-tetragonal structural transition, T_s=60 K for this compound [1]. The coefficient a corresponds to the electronic contribution and has lower values in the tetragonal crystal phase above T_s, as compared to Low-T orthorhombic phase. This is attributed to the change in the density of states (DOS) of $FeSe_{0.8}Te_{0.2}$. The coefficient b corresponds to the lattice contribution and shows the opposite trend: higher values in High-T tetragonal phase and lower values in the orthorhombic phase. The temperature dependence of a and b indicates a significant change in electronic coupling with the crystal lattice across the structural phase transition. Using Allen's theory [8] we obtain the second moment of the Eliashberg function $\lambda\langle\omega^2\rangle$ using TTM

parameters. Figure 1(e) shows that the $\lambda\langle\omega^2\rangle$ undergoes noticeable change during the transition in $FeSe_{0.8}Te_{0.2}$. The electron thermalization time $\tau_{e\text{-}e}$ [Fig.1(f)] shows a significant drop with temperature rise in the Low-T phase up to 23 K, slightly above T_c, indicating suppression of electronic correlations. At the same time, it doesn't change significantly near T_s, and after the phase transition in the High-T phase. It is nearly constant in the tetragonal phase. Both $\lambda\langle\omega^2\rangle$ and $\tau_{e\text{-}e}$ are consistent with our previous results, obtained using other methods [4].

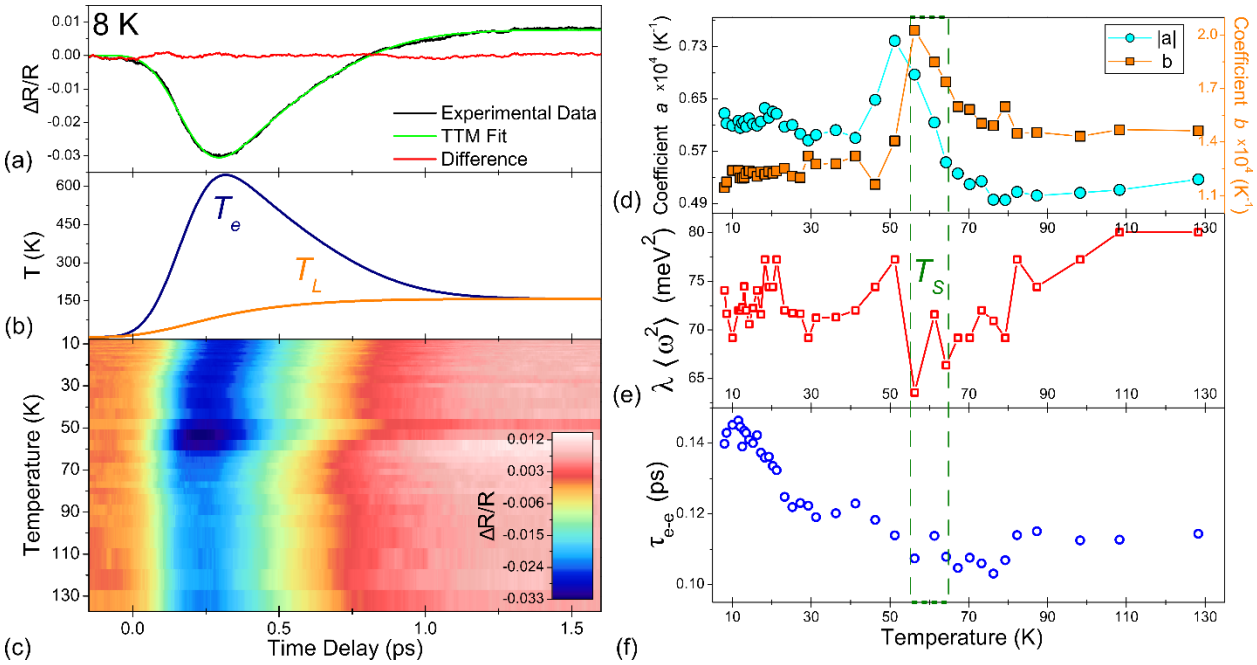

Fig. 1. (a) Transient reflectivity of $FeSe_{0.8}Te_{0.2}$ at 8 K along with the TTM fit and their difference. (b) The evolution of temperatures for electronic and the lattice subsystems at 8 K; (c) Transient reflectivity signal for $FeSe_{0.8}Te_{0.2}$ measured in the range from 8 K to 140 K. (d) Thermoreflectance coefficients a and b; (e) Second moment of Eliashberg function; (f) Electron thermalization time $\tau_{e\text{-}e}$.

In summary, the nonequilibrium dynamics of $FeSe_{0.8}Te_{0.2}$ was studied using ultrafast reflectivity measurements and TTM analysis. The temperature dependence of a and b coefficients suggests a significant change in electronic coupling with lattice across the structural phase transition, associated with the change in DOS. The electronic anisotropy of the Low-T nematic phase could result in lower electron-phonon coupling. The observed evolution of $\lambda\langle\omega^2\rangle$, a and b during the structural transformation of $FeSe_{0.8}Te_{0.2}$ reveal a complex interplay between electronic correlations, *e-ph* coupling, DOS, and optical nonlinearity.

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