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Ultrafast Nematic Dynamics of FeSe Film

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Abstract: Femtosecond spectroscopy of FeSe film shows distinct transient nematic behavior below and above superconducting critical temperature. Results reveal correlations between photoinduced nematicity, quasiparticle formation, superconducting and pseudogap openings, emphasizing electronic correlations and preformed electron pairing.

The discovery of high-temperature superconductivity in single-layer FeSe attracted considerable attention in recent years [1]. It was found that the strain and interfacial charge transfer in FeSe film plays a major role in elevating superconducting critical temperature T_c up to 83 K [2], while the bulk FeSe has T_c ~8 K [3]. The FeSe film deposited on the CaF_2 substrate showed growth of T_c up to 11.4 K with increasing film thickness [4]. At T_s=90 K, the FeSe undergoes tetragonal to the orthorhombic structural transition. This transition contributes to the nematic properties of FeSe. The ultrafast spectroscopy of FeSe shows significant transient nematicity of this material [5, 6]. However, the information about the photoinduced nematicity of FeSe in superconducting state and its relation to *e-ph* coupling is not yet comprehensively understood.

In this work, we present ultrafast temperature-dependent dynamics of photoinduced nematicity of thin FeSe film with a relatively high-temperature superconductivity transition point of ~40K, significantly higher than reported previously for FeSe/CaF_2. Transient dynamics show a distinct nematic response below and above T_c and T_s upon femtosecond laser excitation and is associated with the formation of quasiparticles, the opening of pseudogap, and the superconducting (SC) gap in FeSe.

The deposition process was carried out by ablating a premixed FeSe target composition at a 10^{-6} Torr chamber pressure with an excimer laser with a fluence of 1.5 J/cm^2. FeSe was grown in its tetragonal phase [Fig.1(a)] on the CaF_2 substrate at temperature 573 K. A set of thin films was deposited with thicknesses *d* up to 40 nm. It was found that the films with *d*<40 nm showed an absence of metallic conductivity across all temperatures. This was associated with the island-like structure of the film. However, at *d*=40 nm the film has shown substantially elevated T_c=40K.

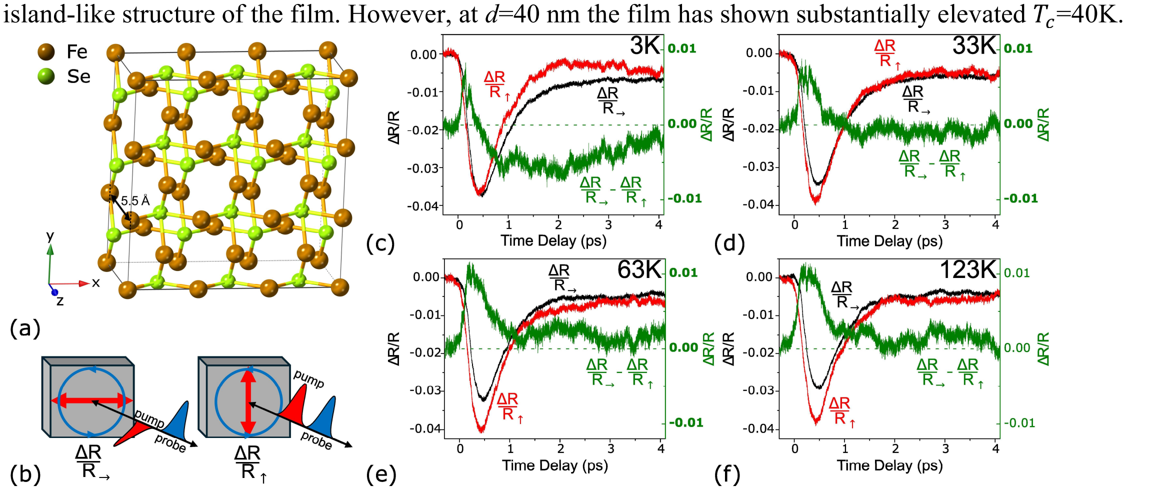

Fig. 1. (a) The crystal structure of FeSe. (b) Schematic diagrams of pump-probe experiment. Probe pulse has circular polarization, pump is linearly-polarized, oriented either horizontally $\left(\frac{\Delta R}{R_{\rightarrow}}\right)$ or vertically $\left(\frac{\Delta R}{R_{\uparrow}}\right)$. (c) Transient reflectivity signals of FeSe within 4 ps scale at 3K, (d) at 33K, (e) at 63K, and (f) at 123K. Green line shows difference between transient reflectivity signals $\left(\frac{\Delta R}{R_{\rightarrow}} - \frac{\Delta R}{R_{\uparrow}}\right)$.

The transient photoinduced nematicity was studied by using a femtosecond laser system with a central wavelength of 800 nm, pulse width of 35 fs, and a repetition rate of 1 kHz. The pump-probe transient reflectivity was measured with a linearly polarized pump with a fluence of 6.5 mJ/cm², as shown in Fig. 1(b). The nematicity of FeSe was monitored by illumination of the sample by a circularly-polarized probe beam with a subsequent linear analyzer installed to control the transient ellipticity of the reflected light associated with nematic order. The sample was mounted inside an optical cryostat with computer-controlled temperature adjustment.

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The transient reflectivity of FeSe [Figs. 1(c)-1(f)] shows similar dynamics for the material doped by Te [7, 8]. A significant difference in transient reflectivity of FeSe at orthogonal polarizations of laser pump reveals photoinduced nematicity in FeSe at an ultrafast time scale. The difference in reflectivity signals $\left(\frac{\Delta R}{R_{\rightarrow}} - \frac{\Delta R}{R_{\uparrow}}\right)$ shows a buildup at t=140 fs followed by rapid decay associated with transient nematic behavior. The dynamics of FeSe in the SC state at 3K show a significant difference to the dynamics at temperatures near T_c and higher. Thus, the formation of the SC state reveals substantially different nematic dynamics below and above T_c.

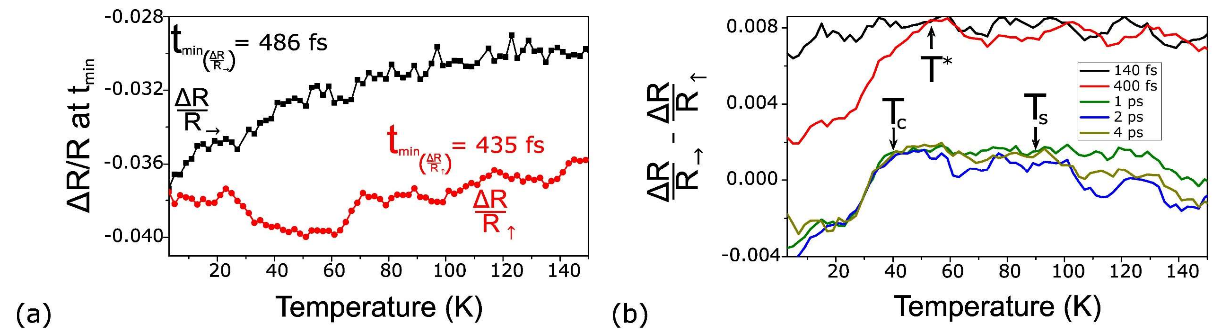

Fig. 2. (a) Temperature dependence of the minimal value of transient reflectivity signal for orthogonal pump polarizations. (b) Evolution of the difference $\frac{\Delta R}{R_{\rightarrow}} - \frac{\Delta R}{R_{\uparrow}}$ at various time delays as a function of sample temperature.

To clarify the main characteristics of the transient nematicity across a broad range of FeSe temperatures, we map the amplitude of the transient reflectivity signals $\frac{\Delta R}{R_{\rightarrow}}$ and $\frac{\Delta R}{R_{\uparrow}}$ in their minima separated by 51 fs [Fig. 2(a)]. While the $\frac{\Delta R}{R_{\rightarrow}}$ signal shows monotonic growth with temperature, the $\frac{\Delta R}{R_{\uparrow}}$ demonstrates a more sophisticated trend that correlates with the formation of SC gap. However, the most pronounced clear correlation between FeSe nematicity and formation of SC gap at T_c, structural transition at T_s, and formation of pseudogap at T^* was obtained from evolution of the signal difference $\frac{\Delta R}{R_{\rightarrow}} - \frac{\Delta R}{R_{\uparrow}}$ at various time delays [Fig. 2(b)]. Here we note that the transient nematicity associated with the formation of SC and pseudogap appears at the time-delay of 1 ps and above, while the transient signal associated with the preformed Cooper pairs scenario appears at the time scale of 400 fs, and is associated rather with fully electronic correlation origin in FeSe.

In summary, the ultrafast nonequilibrium nematic dynamics of FeSe reveals the relationship between nematicity, the formation of quasiparticles, and the opening of the SC and pseudogap in FeSe. The study emphasizes a noticeable difference in nematic dynamics below and above T_c, with the transient signals indicating the presence of electronic correlations and preformed pairing scenario.

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