Superconductivity and phonon self-energy effects in \( \text{Fe}_{1+y}\text{Te}_{0.6}\text{Se}_{0.4} \)

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We study \( \text{Fe}_{1+y}\text{Te}_{0.6}\text{Se}_{0.4} \) multi-band superconductor with \( T_c = 14 \) K by polarization-resolved Raman spectroscopy. Deep in the superconducting state, we detect pair-breaking excitation at 45 cm\(^{-1}\) (2\( \Delta \approx 5.6 \) meV) in the \( XY(B_{2g}) \) scattering geometry, consistent with twice of the superconducting gap energy (3 meV) revealed by ARPES on the hole-like Fermi pocket with \( d_{xz}/d_{yz} \) character. We analyze the superconductivity induced phonon self-energy effects for the \( B_{1g}(\text{Fe}) \) phonon and estimate the electron-phonon coupling constant \( \lambda^T \approx 0.26 \), which is insufficient to explain superconductivity with \( T_c = 14 \) K.

**Introduction** – Since the discovery of the multi-band iron-based superconductors (FeSCs) in 2008 [1], a unified understanding of the pairing mechanism in FeSCs remains in a focus of attention [2–8]. One step towards understanding the pairing mechanism is to study the superconducting (SC) gaps on different pockets of the Fermi surface (FS) [5, 9].

The chalcogenide family \( \text{Fe}_{1+y}\text{Te}_{1-x}\text{Se}_x \) has simple stoichiometry and crystal structure which can be viewed as stacks of \( \text{FeTe}_{1-x}\text{Se}_x \) layers [Fig. 1(a)]. Superconductivity in this system was first found at 8 K in the non-magnetic \( \text{FeSe} \). With about 60\% isovalent substitution of Se for Te, \( T_c \) in \( \text{Fe}_{1+y}\text{Te}_{0.6}\text{Se}_{0.4} \) increases to 14 K [10]. Thus, the non-magnetic and tetragonal \( \text{Fe}_{1+y}\text{Te}_{0.6}\text{Se}_{0.4} \) is an ideal system to study the SC order parameter, without the effect of coexisting or interacting with other electronic orders.

Polarization-resolved Raman spectroscopy has been used to study the pair-breaking excitations in different symmetry channels [12–18] and for estimation of the electron-phonon coupling [19–21] in FeSCs. Previous Raman studies on \( \text{Fe}_{1+y}\text{Te}_{1-x}\text{Se}_x \) were focused on the lattice dynamics [22–25] and the magnetic excitations [25], while the superconducting features for \( \text{Fe}_{1+y}\text{Te}_{0.6}\text{Se}_{0.4} \) have not been well established.

In this work, we use polarization-resolved Raman spectroscopy to study the pair breaking excitations in \( \text{Fe}_{1+y}\text{Te}_{0.6}\text{Se}_{0.4} \). In the SC state, we identify the coherence peak at 45 cm\(^{-1}\) (2\( \Delta = 5.6 \) meV) in the \( XY(B_{2g}) \) scattering geometry with cross-polarized light along Fe-Te/Se directions. The peak energy is close to the twice of the gap energy (3 meV) on the hole-like FS pocket with \( d_{xz}/d_{yz} \) character around \( \Gamma \) point, as determined by ARPES. We investigate the superconductivity induced phonon self-energy effects for the \( B_{1g}(\text{Fe}) \) phonon mode and estimate the electron-phonon coupling constant \( \lambda^T \approx 0.26 \), which is very weak to explain \( T_c = 14 \) K in \( \text{Fe}_{1+y}\text{Te}_{0.6}\text{Se}_{0.4} \).

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**Experimental** – The single crystals of \( \text{Fe}_{1+y}\text{Te}_{0.6}\text{Se}_{0.4} \) were grown using modified Bridgman method [26]. The composition of \( \text{Fe}_{1+y}\text{Te}_{0.6}\text{Se}_{0.4} \) was determined on samples from the same growth batch by energy-dispersive x-ray (EDX) analysis. The nominal composition of excess Fe \((y) \) in \( \text{Fe}_{1+y}\text{Te}_{0.6}\text{Se}_{0.4} \) is about 2\%, while the real value of \( y \) is close to zero [26]. The magnetic susceptibility confirming a sharp SC transition at \( T_c = 14 \) K is shown in Fig. 1(d).

The \( \text{Fe}_{1+y}\text{Te}_{0.6}\text{Se}_{0.4} \) single crystals used for Raman measurements were cleaved in nitrogen gas atmosphere in a glove bag, then immediately loaded into connected continuous helium flow optical cryostat and quickly cooled down to 250 K within 5 minutes to avoid surface contamination. All the Raman scattering measurements...
were performed using the Kr⁺ laser line at 647.1 nm (1.92 eV) in a quasi-back scattering geometry along the crystallographic c-axis with an instrumental resolution about 1.5 cm⁻¹. The excitation laser beam was focused into a 50 × 100 μm² spot on the ab-surface, with the incident power around 10 mW and 2 mW for normal state and superconducting state measurements, respectively. The scattered light was collected and analyzed by a triple-stage aberration corrected Raman spectrometer and recorded using a liquid nitrogen-cooled charge-coupled detector. The Raman spectra were corrected for the spectrometer response and the detector. The temperature shown in this paper has been corrected for laser heating. An empirical heating coefficient 1K/nW was applied according to FeSe [14, 27, 28].

In this manuscript, we define the X and Y directions along the two-Fe unit cell basis vectors (at 45° degrees from the Fe-Fe directions) in the tetragonal phase, whereas X' and Y' are along the Fe-Fe directions [Fig. 1(b)]. Raman spectra were recorded for (τ) scattering geometry [18, 30], the Raman response in (µν) continuum [18, 30], the Raman response in (X'X') and (X'Y') polarization geometries in the ab-plane, where ̂e₁ and ̂e₂ represent the incident and scattered light polarization, respectively.

For crystals with point group symmetry D₄h, the XX, X'X' and XY geometries probe A₁g+B₁g, A₂g+B₁g and A₂g+B₂g channels, respectively [29]. Assuming the A₂g response is negligible [12] and using the background estimated from the X'Y'-symmetry electronic continuum [18, 30], the Raman response in (µν) scattering geometry χ”(ω,T) can be obtained: χ”(ω,T) = I(ω,T)/[1+n(ω,T)], where I(ω,T) is the Raman intensity after background subtraction and n(ω,T) is the Bose-Einstein factor.

The normal state – The Fe₁₊yTe₀.₆Se₀.₄ crystal structure belongs to the space group P4/nmm (point group D₄h). The Γ point Raman active modes are \( \Gamma_{\text{Raman}} = A_{1g} + B_{1g} + 2E_{g} \). The A₁g and B₁g modes are related with Te/Se and Fe atoms c-axis lattice vibrations, respectively.

In Fig. 2, we show the normal state Raman spectra for four in-plane scattering geometries at 25 K. The observed mode at around 162 cm⁻¹ and the mode at around 207 cm⁻¹ in XX scattering geometry are assigned to A₁g(Te/Se) and B₁g(Fe) modes, respectively [24, 25]. The comparison of the phonon frequencies and line-widths obtained in this work with previous studies [24, 25] are summarized in Table I. The phonon frequency for both modes and the line-width for the B₁g(Fe) mode are consistent with Ref. [24, 25]. In contrast, the line-width for the A₁g(Te/Se) mode in this work is about 3-4 times sharper than in Ref. [24, 25]. We note that the spectra shown in Fig. 2 lack the two features reported in Ref. [25]: the low frequency bump/tail appearing in all in-plane scattering geometries and a substantial ‘leakage’ intensity of A₁g phonon into cross scattering geometries. The absence of these two features indicates the quality of cleaved surface in this study.

The superconducting state – Before looking into the Raman features observed in the SC state for Fe₁₊yTe₀.₆Se₀.₄, we recall the SC gap values obtained by complementary spectroscopic probes. Angle-resolved photoemission spectroscopy (ARPES) revealed nodeless SC gaps with small or negligible in-plane anisotropy on both hole-like and electron-like FS pockets [26, 32, 33]. In addition to the SC gap of 1.8 meV for the topological surface state around the Γ point [33], two close to isotropic gap values for bulk bands with \( d_{x^2-y^2} \) character were reported: a 3 meV gap for the hole-like FS pocket centered around the Γ point [26] and a larger, about 4.2 meV gap for the electron pocket around the M point [32]. The scanning tunneling spectroscopy (STS) showed SC coherence peaks at 1.5 meV and a shoulder at 2.5 meV [34]. The infrared spectroscopy (IR) revealed two SC gaps at 2.47 meV and 5.08 meV [35]. The measurements of specific heat confirmed the nodeless nature of the SC order parameter with an averaged gap value 2.3 meV [36]. At the energy scale of the sum of SC gaps on the hole-like and electron pockets with \( d_{x^2-y^2} \) character, a 6.5 meV neutron-spin-resonance mode was reported below \( T_c \) at \( Q = (\pi, \pi) \) [2-Fe BZ] by inelastic neutron scattering (INS) measurements [37]. The values of the SC gaps and bosonic modes deduced from different spectroscopies are summarized in Table II.
TABLE II. The summary of the values for SC gaps and bosonic modes deduced from ARPES, STS, IR, Specific heat, Raman scattering, and INS measurements for Fe\textsubscript{1+y}Te\textsubscript{0.6}Se\textsubscript{0.4} with similar compositions. $\Delta_1$, $\Delta_2$ and $\Delta_3$ represent three energy scales of gap values. $E_{CM}$ represents the energy of the bosonic mode. All energies are given in units of meV.

|         | $\Delta_1$ | $\Delta_2$ | $\Delta_3$ | $E_{CM}$ |
|---------|-------------|-------------|-------------|----------|
| ARPES   | 1.8 [33]    | 3 [26]      | 4.2 [32]    |          |
| STS     | 1.5 [34, 38]| 2.5 [34]    |             |          |
| IR      | 2.47 [35]   | 5.08 [35]   |             |          |
| Specific heat | 2.3 [36]   |             |             |          |
| Raman   | 2.8 (this work) |             |             | 6.5 [37] |
| INS     |             |             |             |          |

In Fig. 3, we present the Raman response above and below $T_c$ for three scattering geometries. For the XX and X‘Y‘ scattering geometries, the electronic continuum barely changes upon cooling below $T_c$. In contrast, for XY scattering geometry a clear peak at around 45 cm$^{-1}$, which we relate to the pair-breaking excitation, emerges below $T_c$. Since the peak’s energy is about 5.6 meV, based on the Table II, we assign it to the gap $2\Delta_2$ on the hole-like FS pocket with the $d_{xz}/d_{yz}$ character [39] around $\Gamma$ point with a typical gap value $\Delta_2 = 3$ meV from the ARPES measurement [26]. The pair-breaking peak associated with the larger gap $2\Delta_3 = 8.4$ meV on the electron pockets around $M$ point is not detected in the $B_{2g}$ channel. In contrast, in optimally-doped Ba(Fe\textsubscript{0.93}Co\textsubscript{0.06})\textsubscript{2}As\textsubscript{2}, a similar single peak observed in $B_{2g}$ channel was interpreted as a pair-breaking peak originated from the electron pockets around $M$ point [12]. While Fe\textsubscript{1+y}Te\textsubscript{0.6}Se\textsubscript{0.4} and Ba(Fe\textsubscript{0.93}Co\textsubscript{0.06})\textsubscript{2}As\textsubscript{2} share similar FS topology, these differences suggest that the Raman vertex in the $B_{2g}$ channel for multi-band FeSCs is rather complex.

Phonon self-energy effects – The frequency and line-width for the $B_{1g}(Fe)$ phonon are presented in Fig. 4(b-c). Above $T_c$, the $B_{1g}(Fe)$ phonon shows a conventional temperature dependence: hardening and sharpening upon cooling following the anharmonic phonon decay model [40–42]. Below $T_c$, in contrast to the $A_{1g}(Te/Se)$ phonon which barely changes upon cooling across $T_c$ [inset of Fig. 4(a)], the $B_{1g}(Fe)$ mode shows abnormal behavior in the SC state. The mode’s energy and line-width show additional hardening and sharpening in the SC state [Fig. 4(a)].

The $B_{1g}(Fe)$ phonon energy is around 26 meV. It is much larger than the twice of the maximum gap $2\Delta_3 = 8.4$ meV in Fe\textsubscript{1+y}Te\textsubscript{0.6}Se\textsubscript{0.4} [32]. For a phonon with $\omega > 2\Delta$, such hardening upon entering into SC state was also reported for MgB\textsubscript{2} [43], for cuprate superconductors [44–49], and for FeSCs [19–21]. The effects were explained within the Zeyher-Zwicknagl’s model [50] as a consequence of electron-phonon coupling. When the SC gap opens below $T_c$, the electronic density-of-states around the Fermi level is reorganized and pushed above the $2\Delta$ energies to the proximity of the phonon energy, then the real part of the phonon self-energy shifts to higher energies due to the renormalization by electron-phonon coupling.

However, the Zeyher-Zwicknagl’s model predicts that the line-width for a phonon with $\omega > 2\Delta$ should be broadened upon entering the SC state as the phonon has additional decay channels. To the contrary, the line-width for the $B_{1g}(Fe)$ phonon for Fe\textsubscript{1+y}Te\textsubscript{0.6}Se\textsubscript{0.4} further decreases in the SC state. Similar sharpening of the $B_{1g}(Fe)$ phonon in the SC state was also observed for NaFe\textsubscript{0.97}Co\textsubscript{0.03}As [20], Ba\textsubscript{0.72}K\textsubscript{0.28}Fe\textsubscript{2}As\textsubscript{2} [19] and Sr\textsubscript{0.85}K\textsubscript{0.15}Fe\textsubscript{2}As\textsubscript{2} [19].

To quantitatively estimate the SC induced self-energy effects and the electron-phonon coupling constant, we use the following model to fit the $B_{1g}(Fe)$ phonon: $\chi''(\omega) \propto 4\omega_0^2\Sigma''(\omega - \omega_0^2 - 2\omega\Sigma')^2 + 4(\omega_0\Sigma')^2$\textsuperscript{-1}, where $\omega_0$ is the bare phonon frequency and $\Sigma = \Sigma' + i\Sigma''$ is complex phonon self-energy [50]. The phonon appears at $\omega_{ph} = \sqrt{\omega_0^2 + 2\omega\Sigma'}$ if $\Sigma''$ is small. The fitting results are presented in Fig. 4(b-c) [42].

We compute the electron-phonon coupling constant $\lambda'_{B_{1g}}$ for the $B_{1g}(Fe)$ phonon at $\Gamma$ point [51]: $\lambda = -\kappa \sin u/u$, where $\kappa = [(\Sigma'(5K) - \Sigma'(15K)) - i(\Sigma''(5K) - \Sigma''(15K))]/\omega_{ph}(15K)$ and $u \equiv \pi + 2i\cosh^{-1}[\omega_{ph}(15K)/2\Delta]$. With $2\Delta = 45$ cm$^{-1}$-obtained
(b) and (c) represent by anharmonic decay model [42]. The dashed vertical lines in (b) and (c) represent the fitting of the normal state phonon corrected for the instrumental resolution. The solid lines in (b)-(c) T-dependence of the phonon scattering geometry. The top inset of (a) shows the pair-breaking peak energy in the bottom inset of (a) illustrates the atomic displacement of and 5 K in X from the pair-breaking peak energy in the X物料.

FIG. 4. (a) $B_{1g}(Fe)$ phonon for Fe$_{1+y}$Te$_{0.6}$Se$_{0.4}$ at 15 K and 5 K in X’Y’ scattering geometry. The top inset of (a) illustrates the atomic displacement of $B_{1g}(Fe)$ phonon. The bottom inset of (a) shows the $A_{1g}(Te/Se)$ phonon at 25 K and 5 K in XX scattering geometry. (b)-(c) T-dependence of the $B_{1g}(Fe)$ phonon energy $\omega_{ph}$ and line-width $2\Sigma''$. $\Sigma''$ has been corrected for the instrumental resolution. The solid lines in (b) and (c) represent the fitting of the normal state phonon by anharmonic decay model [42]. The dashed vertical lines in (b) and (c) represent $T_c$.

from the pair-breaking peak energy in the $B_{2g}$ channel, we derive a weak electron-phonon coupling constant $\lambda^{B_{1g}} \approx 0.026$. Since the $A_{1g}(Te/Se)$ phonon shows negligible SC-induced self-energy effect [inset of Fig. 4(a)], $B_{1g}(Fe)$ phonon is the only phonon that shows such an effect. Therefore, we can use the $\lambda^{B_{1g}}$ as an approximation for the averaged electron-phonon coupling constant $\lambda^X$ at BZ zone center [21].

By comparison, a much larger electron-phonon coupling constant $\lambda^X \approx 0.3$ was reported for a conventional phonon-mediated superconductor MgB$_2$ with $T_c = 39$ K [43]. Furthermore, for V$_3$Ga, an s-wave superconductor with a similar $T_c = 14.2$ K, the electron-phonon coupling constant was estimated to be $\lambda \approx 0.9$ by optical measurements [52]. Therefore, the electron-phonon coupling constant $\lambda^X \approx 0.026$ is clearly insufficient to cause $T_c = 14$ K superconductivity in Fe$_{1+y}$Te$_{0.6}$Se$_{0.4}$.

Conclusions – In summary, we present polarization-resolved Raman spectroscopic study of single crystal Fe$_{1+y}$Te$_{0.6}$Se$_{0.4}$ superconductor with $T_c = 14$ K.

In the SC state, we observe a distinct pair-breaking peak at 45 cm$^{-1}$ ($2\Delta = 5.6$ meV) in the $B_{2g}$ channel corresponding to the twice of the gap energy (3 meV) on the hole-like FS pocket with $d_{xz}/d_{yz}$ character around $\Gamma$ point reported by ARPES measurements.

We analyze the superconductivity induced phonon self-energy effects for the $B_{1g}(Fe)$ phonon and compute the electron-phonon coupling constant $\lambda^X \approx 0.026$, which is small for Fe$_{1+y}$Te$_{0.6}$Se$_{0.4}$ with $T_c = 14$ K.

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