TOPICAL REVIEW — Iron-based high temperature superconductors

Electron–phonon coupling in cuprate and iron-based superconductors revealed by Raman scattering*

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Electron–phonon coupling (EPC) in cuprate and iron-based superconducting systems, as revealed by Raman scattering, is briefly reviewed. We introduce how to extract the coupling information through phonon lineshape. Then we discuss the strength of EPC in different high-temperature superconductor (HTSC) systems and possible factors affecting the strength. A comparative study between Raman phonon theories and experiments allows us to gain insight into some crucial electronic properties, especially superconductivity. Finally, we summarize and compare EPC in the two existing HTSC systems, and discuss what role it may play in the HTSC.

Keywords: electron–phonon coupling, cuprate and iron-based superconducting systems, Raman scattering, high-temperature superconductor

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1. Introduction

Electron–phonon coupling (EPC) is one of the most common and fundamental interactions in solids. It not only dominates many basic dynamic processes like resistivity, thermal conductivity, etc., but also provides the pairing glue in conventional superconductors. However, in high-temperature superconductors (HTSCs), the question of whether EPC favors pairing remains controversial. Despite the controversies, many experiments have provided clear evidence for EPC in HTSC.

Superconductivity was discovered in mercury in 1911 by Onnes.[1] It was successively observed in many metals and alloys. However, the superconducting (SC) mechanism was not well understood for quite a long time. Gortor and Casimir proposed a phenomenological two-fluid model in 1934 to explain zero resistivity,[2] in which electrons are divided into two categories: normal electrons and SC ones carrying current without resistance. Afterwards, the London equations were proposed to describe electromagnetic properties of a superconductor,[3] providing a beautiful explanation for Meissner diamagnetic behavior. In 1950, the phenomenological Ginzburg–Landau equation was raised based on a general Landau's second-order phase transition theory, and it was considered to be a huge success in vortex dynamics and quantitative description of the SC transition. Bardeen, Cooper, and Schrieffer (BCS) proposed a microscopic pairing mechanism based on effective electron–phonon coupling,[4] which eventually established a solid foundation for understanding the commonly observed superconductivity phenomena.

When Müller and Bednorz reported superconductivity at 35 K in layered compound $La_{2-x}Ba_xCuO_4$ in 1986,[5] a new era of HTSC was launched. The temperature barrier of 77 K, the boiling point of liquid nitrogen, was soon broken.[6,7] The current record of the highest SC temperature was realized in the HgBaCaCuO system under high pressure.[8] In the early stage of HTSC study, many researchers attempted to understand the HTSC mechanism within the framework of BCS theories, but the small isotope effect revealed by experiments[9–12] failed to support the conventional EPC scenario. On the other hand, it was confirmed that cooper pairs are indeed the carriers of SC current in HTSC.[13–15] This means that a new concept of a pairing "glue" is needed for the HTSC systems, such as magnon,[16] exciton,[17,18] polaron,[19] bipolaron,[20,21] and antiferromagnetic (AF) fluctuations.[22,23] At the same time, a strong EPC in cuprate HTSC was reported by a lot of experimenters, which implies that EPC plays an important role in pairing, though not a dominant one. EPC is observed not only in the Raman channel, but also in other measurements like neutron scattering and angle-resolved photoemission spectroscopy (ARPES), as has been comprehensively reviewed in Refs. [24]–[29]. After over two decades, the discovery of high-T_c superconductivity in iron-based systems in 2008 was a great breakthrough in the HTSC study. It is generally expected that many unresolved problems in cuprates, including EPC, may be answered by comparing their behaviors with those of iron pnictides. Raman scattering, one of the most fundamental techniques in condensed matter physics, shows many unique advantages in EPC study. Through the changes of Raman phonons in lineshape, width, and frequency due to EPC, we can conveniently study the renormalization of electronic self-energy and precisely estimate the strength of EPC,

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and hence explore its relation to superconductivity.

The reviews of electronic, phononic, and magnonic properties, in which many precursor concepts and pictures are introduced, can be found in Refs. [30]–[32]. Raman scattering in Fe superconductors has been reviewed in Ref. [33]. In this paper, we summarize the existing experimental and theoretical results in both cuprate and iron-based superconductors from the point of view of Raman scattering, and discuss the connection between the EPC and the HTSC pairing mechanism. We will give a short introduction to the crystal structures and Raman-active modes of relevant compounds. Then, we will discuss EPC in both HTSC systems through Fano asymmetric lineshapes and the renormalization of self-energies upon crossing the SC and other electronic transitions.

2. Crystal structures and Raman phonon modes in cuprate and Fe-based superconductors

Generally, cuprate superconductors are classified and named according to the heavier elements, for example, rare-earth-, Y-, Bi-, Tl-, and Hg-systems. All the compounds contain one or more CuO planes and charge reservoirs. In the following, we will take $YBa_2Cu_3O_{7-x}$ (YBCO) as an example to illustrate Raman-active modes in cuprates. As shown in Fig. 1, two buckled CuO planes, charge reservoirs, and CuO chains reside on the opposite sides of yttrium symmetrically. The tetragonal phase (D_{4h}) at $x = 1$ and the orthorhombic phase (D_{2h}) at x=0 differ by about 1% in their a and b lattice parameters, and the latter can be seen as a pseudo-tetragonal structure. Thus, we adopt the D_{4h} point group for both phases in symmetry analysis. Raman-active modes are $4A_{1g}+B_{1g}+5E_g$ for D_{4h}. In D_{2h} case, A_{1g} and B_{1g} are combined into A_g modes and doubly degenerate E_g is split into B_{2g} and B_{3g} modes. The vibration patterns of several Raman modes in YBCO are illustrated in Fig. 2.

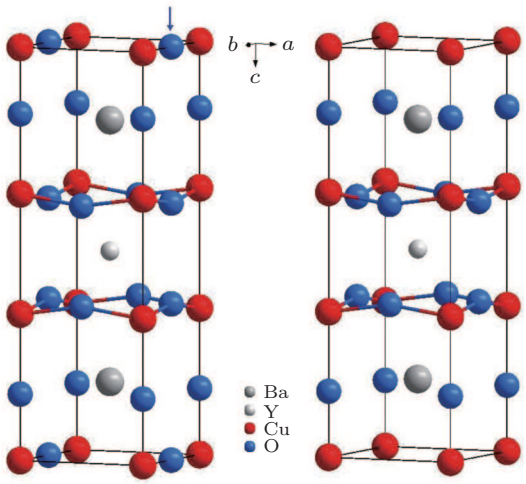


Fig. 1. Crystal structures of YBCO-123. The left is orthorhombic phase, and the arrow indicates the oxygen atom in the CuO chain. The right is tetragonal phase without a CuO chain.

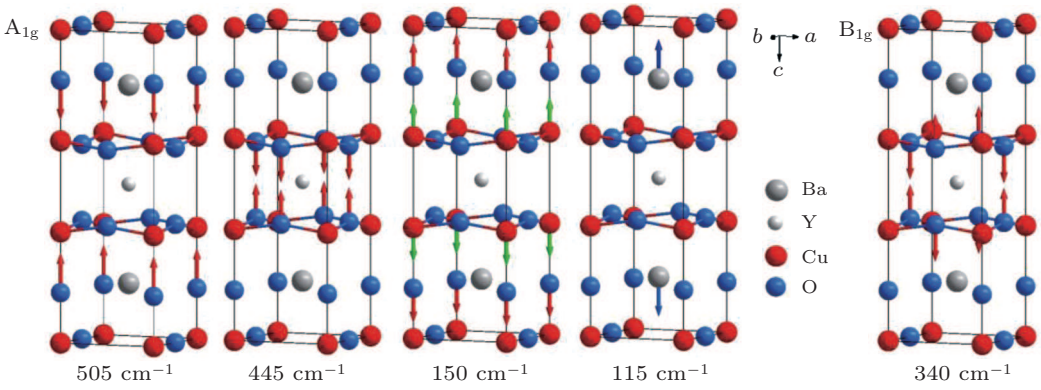


Fig. 2. Five Raman-active modes vibrating along c-axis.

Up to now, tens of Fe-based superconductors have been synthesized, all of which can basically be divided into six categories. 1) "1111", represented by F-doped LaFeAsO, was the first reported iron-based superconducting system. The highest T_c in the series can be raised to 56 K by rare-earth substitution.[34] 2) "122", represented by Co- or K-doped $BaFe_2As_2$, is the most commonly studied system due to the availability of high-quality crystals and the tunable doping levels.[35] 3) "111" refers to LiFeAs or NaFeAs with a maximum $T_c \sim 18$ K.[36] 4) "11" refers to Fe(Se,Te), the prototype of iron-based superconductor without poisonous element As.[37] 5) "21311" is isostructural to the 122 system but with a divalent group instead of Ba.[38] 6) Intercalated FeSe superconductor with a maximum T_c of 30 K, represented by $A_{0.8}Fe_{1.6}Fe_2$ (A = K, Tl, Cs,...), was discovered recently.[39] It is the Fe-deficient version of the 122 structure and exhibits a strong Fe–vacancy ordering. Similar to the structure of cuprates, the Fe-based compounds also have conducting FeAs(Se) layers and charge reservoirs. Unlike CuO planes, Fe ions are not located exactly in the same plane as As (Se) ions, but are rather sandwiched by As (Se) ions. Four Raman-active modes of $BaFe_2As_2$ are shown in Fig. 3. Barium, Fe, and As

ions occupy 2a, 4d, and 4e positions, respectively. Barium ions allow a B_{1g} mode vibrating along the c axis and an in-plane E_g mode, and As ions allow an out-of-plane A_{1g} mode and an in-plane E_g mode. Similar Raman modes related to FeAs(Se) exist in the other Fe-based systems, but in 111 and 1111 systems, additional out-of-plane A_{1g}/B_{1g} and E_g modes may be observed because Li/Na, La, and O in charge reservoirs are off-center. More details can be found in Ref. [33].

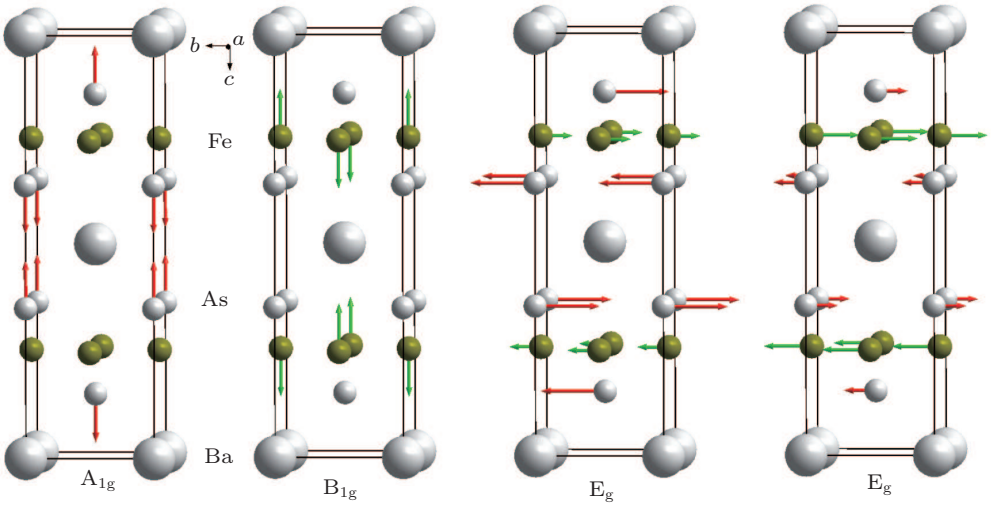


Fig. 3. Crystal structure of $BaFe_2As_2$ and its Raman modes.

3. Electron–phonon coupling in cuprate superconductors

3.1. Fano effect

The Fano effect was first proposed to explain the atomic absorption spectrum.[40] Then it was employed to understand the asymmetric phonon lineshape in heavily doped Si,[41,42] which is caused by the resonance between discrete phonon spectra and the electronic continuum. In Raman spectra, the Fano effect typically changes a Lorentzian phonon peak to an asymmetric one. If there is no interaction between phononic and electronic excitations, one can expect a simple superposition of both contributions. However, if resonance exists between them, lineshapes and widths will be modified. The scattering intensity modified by EPC is written as[43]

$$I(\omega)=I_C\frac{|q+\varepsilon|^2}{1+\varepsilon^2}+I_b(\omega), \tag{1}$$

where $\varepsilon=\omega-\omega_p/\Gamma_p$ and $\omega_p=\omega_p^0+\delta\omega_p$, ω_p^0 is the bare phonon frequency, q is a parameter quantitatively describing asymmetry, Γ_p is the phonon width (half width at half maximum), I_C is an intensity parameter, and $I_b(\omega)$ represents incoherent background. An asymmetric peak will get back to a Lorentzian lineshape at $q\to\infty$. By neglecting other renormalization mechanisms, one can further establish the relation between scattering intensity and microscopic parameters as

$$I(\omega)\sim\pi T_e^2\rho(\omega)\frac{\left(\omega-\omega_p^0+T_pV/T_e\right)^2}{\left[\omega-\omega_p^0-V^2R(\omega)\right]^2+\left[\pi V^2\rho(\omega)\right]^2}, \tag{2}$$

where T_e and T_p are electronic and phononic Raman vertexes, V is the electron–phonon coupling vertex, ω_p^0 is the bare phonon frequency, $\rho(\omega)$ and $R(\omega)$ are the imaginary and real parts of electronic polarizability, respectively. Comparison between Eqs. (1) and (2) gives

$$q=\frac{T_pV/T_e+V^2R(\omega)}{\pi V^2\rho(\omega)},$$
$$\varepsilon=\frac{\omega-\omega_p^0-V^2R(\omega)}{\pi V^2\rho(\omega)}.$$

Then, the EPC strength and other parameters can be obtained by fitting phonon peaks with the above expressions.

3.2. Fano effect in cuprates

The asymmetry in YBCO has been studied in depth, especially with regard to the buckling B_{1g} mode of oxygen. Thomsen *et al.* and Cooper *et al.* first reported the asymmetry of the B_{1g} mode at 340 cm^{-1},[44,45] which was confirmed by further Raman measurements.[46–56] As shown in Fig. 4, the B_{1g} mode exhibits a clear asymmetry, which can be perfectly fitted by a Fano lineshape. The fitting reveals quite a large EPC strength ($q\sim-6$), which implies that electronic properties may be substantially affected by EPC in YBCO. It seems that EPC in YBCO strongly depends on oxygen content. Actually, EPC is much larger in the superconducting YBCO at $x\sim7$ and the B_{1g} mode even goes back to a symmetric Lorentzian lineshape in the insulating phase at $x\sim6$.[32] The evolution of asymmetric factor $1/q$ with oxygen content is shown in Fig. 5.[56] The reduction of asymmetry at small x may be a reflection of low carrier concentration. Consistent with YBCO, no asymmetry was observed in $PrBa_2Cu_3O_7$ which has a very low carrier concentration.[57] However, in $RBa_2Cu_3O_7$ with

very close T_c's, different rare-earth R ions produce different q values.[48] This means that asymmetry is also related to other factors, not only carrier concentration. It has been reported that q even depends on polarization geometry.[58] Fano resonance was also reported for the mode at 112 cm^{-1} in YBCO, which was assigned as the vibration of Ba along the c axis (see Fig. 2). Strangely, its asymmetry remains unchanged with different rare-earth ions and carrier concentrations.[59] Hence, the mode has recently been considered irrelevant to superconductivity.

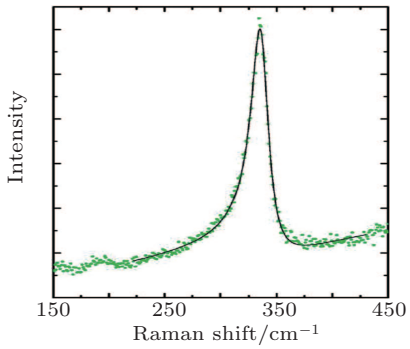


Fig. 4. Fano asymmetric fitting to B_{1g} mode at 340 cm^{-1} in YBCO. The temperature is 16.6 K. The fitting gives $\omega = 336.3 \pm 0.1$ cm^{-1}, $q = -6.0 \pm 0.2$, HWHF $\gamma = 9.6 \pm 0.1$ cm^{-1}.[51]

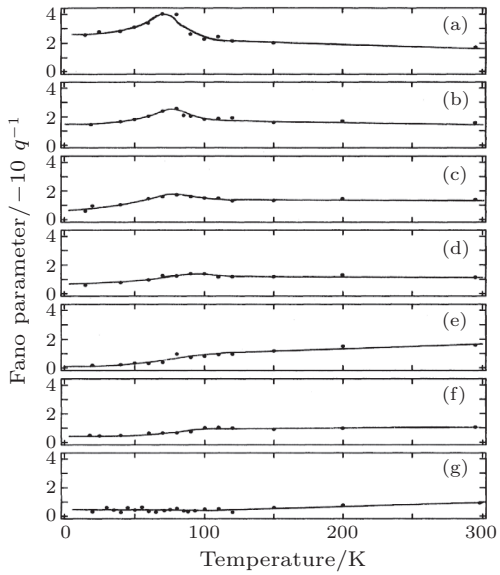


Fig. 5. Temperature dependence of asymmetric factor extracted from B_{1g} phonons. The oxygen contents are 7, 6.99, 6.93, 6 .9, 6.86, 6.81, and 6.68 from (a) to (g).[56]

Several scenarios have been proposed to understand the origin of the Fano lineshape in YBCO.[60–62] Opel *et al.* suggested that EPC for the 340 cm^{-1} B_{1g} mode is associated with the buckling of CuO planes.[63,64] As shown in Fig. 6, Y^{3+} and Ba^{2+} ions are located on the two sides of CuO planes, which gives rise to an effective electric field vertical to CuO planes. The field causes the buckling of CuO planes and a charge transfer between neighboring oxygen in an out-of-phase vibration of oxygen-like 340 cm^{-1} B_{1g} mode. In this picture, B_{1g} phonons couple with the transferred electrons and the Fano lineshape is manifested. EPC strength obtained by fitting B_{1g} mode coincides with that estimated by vertical electric field, which is calculated using the experimental buckling parameters.[65,66] Generally, carrier doping determines the distance between Fermi level and van Hove singularity, and hence affects EPC. This may explain why the asymmetry changes with oxygen content as demonstrated in Fig. 5. There is no such electric field and buckling of CuO planes due to local charge imbalance in Bi-2212 system. By doping Y into Bi-2212 to induce a charge imbalance, Opel *et al.* found that EPC strength is increased by almost one order of magnitude.[63] At the same time, T_c goes down from 91.5 K to 57.0 K. This seems to imply that EPC does not favor superconductivity, which appears to be supported by some theoretical scenarios.[67]

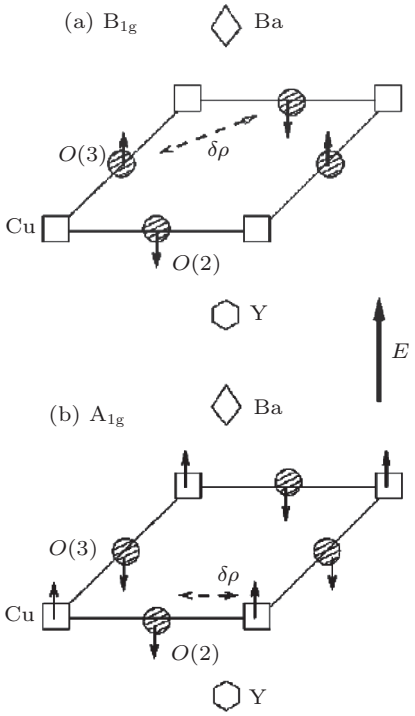


Fig. 6. B_{1g} and A_{1g} modes and the electric field induced by Ba^{2+} and Y^{3+} in YBCO. The eletric field causes a charge transfer between neighboring oxygen.[63]

3.3. Renormalization of self-energy induced by superconducting transition

3.3.1. Phonon frequencies and widths

As discussed above, the imaginary and real parts of electronic self-energy and polarizability can be extracted from phonon lineshapes, through which we can also obtain electronic properties across an electronic transition, for example, an SC transition.

In fact, phononic self-energy effect has been extensively investigated in YBCO. Phonon softening at the SC

transition was first reported in polycrystals[68] and then observed in single crystals.[43] It was further confirmed by other techniques.[45,69,70] Phonon hardening was also found at the SC transition in A_{1g} mode,[71] an in-phase vibration of oxygen in CuO planes along the c axis. The softening and hardening were believed to be intimately connected with superconductivity.[32] The reasons consist of three aspects. 1) No structural change that could bring about a phonon frequency shift of 2% was actually observed.[72,73] 2) The phonon anomaly does not appear in the non-SC $YBa_2Cu_3O_6$ and $PrBa_2Cu_3O_7$.[57,74] 3) The temperature T_s at which the phonon anomaly occurs, is found to be associated with T_c through the relation $dT_s/dH = dT_c/dH_{c2}$. This means that the phonon anomaly will disappear if superconductivity is killed by magnetic field.[75] Klein and Dierker presented a qualitative understanding based on the BCS theory.[76] Zeyher and Zwicknagl explained the phonon anomaly in the framework of the Eliashberg theory.[77,78] The basic idea is that EPC will induce a renormalization of phononic self-energy across an SC transition. The imaginary and real parts reflect the changes in width and frequency, respectively, which can be written as[71]

$$\Delta\Sigma_\nu/\omega_\nu = \lambda_\nu f(\omega_\nu/2\Delta),$$

where $\Delta\Sigma$ is the change of phonon self-energy, ν denotes the phonon modes and λ_ν is the corresponding EPC constant, f is the function of reduced frequency, $\tilde{\omega} \equiv \omega_\nu/2\Delta$, and Δ is the SC gap. In the BCS weak coupling limit, one obtains

$$f(\tilde{\omega}) \equiv \begin{cases} -2u/\sin 2u, & \sin u \equiv \tilde{\omega} < 1, \\ (2\nu - i\pi)/\sinh 2\nu, & \cosh \nu \equiv \tilde{\omega} < 1. \end{cases}$$

The EPC constant λ_ν contributed by the ν-th mode is defined as

$$\lambda_\nu \equiv 2N(0)\langle |g_{\nu kk}|^2 \rangle_{FS}/\omega_\nu,$$

where $N(0)$ is the electronic density of states, and g are the EPC matrix elements. For a phonon with $\omega_\nu > 2\Delta$, its frequency will be shifted up and the width broadened, while a phonon with $\omega_\nu < 2\Delta$ will behave exactly in the opposite way. For a d-wave symmetry, the calculations in the clean limit were different from the above results,[79] i.e., a phonon with $\omega_\nu < 2\Delta$ will exhibit a softening rather than a hardening and its width will be broadened. Devereaux calculated the temperature dependence of optical phonon modes at low temperatures.[80] It was found that the EPC strength strongly depends on the symmetry of the phonon mode. For B_{1g} mode, the real and imaginary parts of the self-energy reach their maximums at $\omega/2\Delta \approx 1$, while the peak shifts to $\omega/2\Delta \approx 0.5$ for A_{1g} mode. He further pointed out that for a d-wave symmetry below T_c, the widths of B_{1g} and A_{1g} modes show a T^3 and linear temperature dependence, respectively. In an isotropic s-wave superconductor, both decay exponentially with temperature. This may be used to estimate the SC gap.[71] The early studies on phonon anomaly are summarized in Ref. [32]. We introduce some later research in the following.

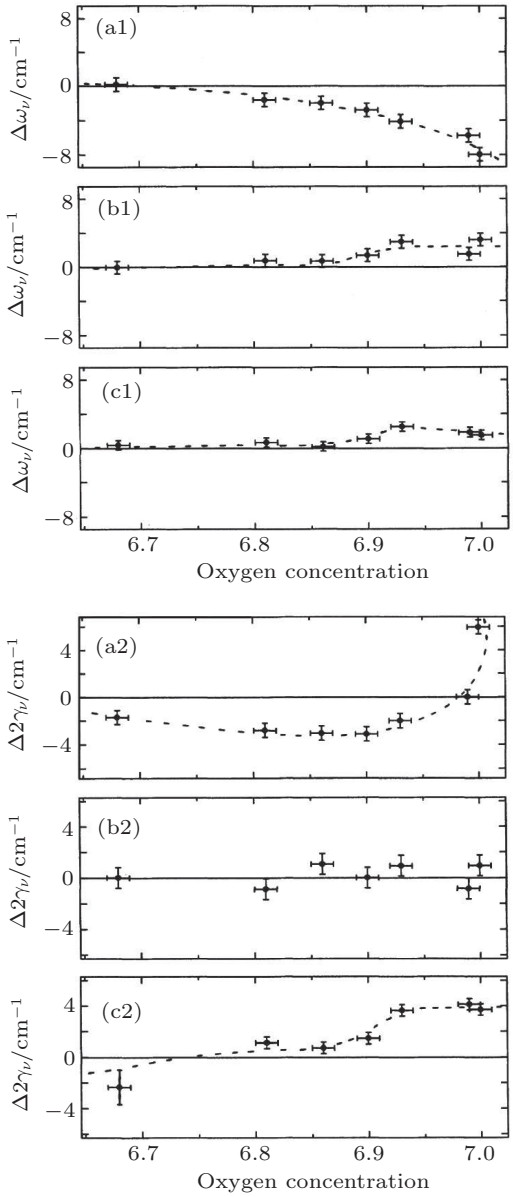


Fig. 7. The evolution of frequencies and widths of Raman phonons at (a1, a2) 340, (b1, b2) 400, and (c1, c2) 500 cm^{-1} with oxygen content.[56]

In the early Raman measurements, the changes in phonon frequencies and widths have already been noticed,[71,81] but the magnitude of renormalization seems to be very sensitive to oxygen content.[82] The evolution of phonon frequency and width with temperature in YBCO has been systematically explored,[46,53,56,82–84] as shown in Fig. 7. A key question raised from the measurements is whether the renormalization is determined by oxygen content or by carrier concentration. In Ca-doped YBCO-123 and 124 samples, frequency changes induced by SC transitions follow oxygen content but not carrier concentration,[52] as shown in Fig. 8. However, Raman measurements in the slightly Ca-doped samples shown in Fig. 9 rather suggests that the frequency change at T_c is ex-

actly proportional to carrier concentration.[85–87] Generally, it is recognized that both phonon self-energy normalization and SC gap are tuned by carrier concentration. The dramatic increase of EPC strength in the overdoped range is considered to be an indication of SC gap size, which is also supported by theories.[79] The reduction of normalization in the underdoping range may be related to the pseudogap.[88] In optimally doped YBCO, Ba A_{1g} mode at 120 cm^{-1} and oxygen B_{1g} mode at 340 cm^{-1} demonstrate renormalization across T_c. In underdoped YBCO, phonon anomalies of the B_{1g} mode are removed while A_{1g} phonon hardening occurs at 150 K and its width rapidly decreases, which is regarded as a sign of the opening of the pseudogap.[89]

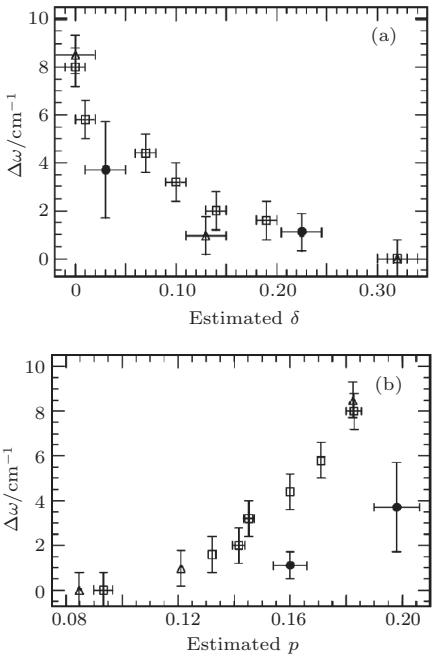


Fig. 8. The frequency softening of B_{1g} mode in Ca doped YBCO in the SC state with (a) oxygen defects and (b) carrier concentration.[52]

In oxygen-sufficient YBCO, different lineshapes and peak positions of B_{1g} mode were measured in XX and XY configurations.[58] Assuming constant electronic Raman vertex T_e and phonon Raman vertex T_p, Fano fitting gives different frequencies, widths, and q. These parameters indicate anomalies at T_c. This could not be explained by pure d-wave pairing. However, it was argued that the assumption about electronic and phononic vertexes is not reasonable.[90] After taking into account both contributions simultaneously, it was concluded that about 10%–15% s-waves were mixed into the electron pairing.[50] The renormalization of B_{1g} mode was also reported in YBCO films,[91] but not observed in NdBaCuO crystals.[92]

Besides the B_{1g} mode at 340 cm^{-1}, much attention has also been paid to the Ba A_{1g}mode at 120 cm^{-1}. In the YBCO-124 system, the mode shows a clear softening and broadening, which has been associated with the opening of a small gap at 113 cm^{-1}.[93] The anomalies of A_{1g} mode at T_c are strongly suppressed in Ni- and Zn-doped YBCO-124. The relation between gap size and the normalized frequencies and widths follows the predictions by Zeyher and Zwicknagl based on s-waves.[78] McCarty *et al.* reported anomalies of the modes at 440 and 500 cm^{-1} at T_c, and further estimated the SC gap.[94] Two Raman-inactive modes in underdoped YBCO-123 exhibit a significant resonance and an obvious softening in the SC state.[54] The origin of the strange EPC remains unknown.

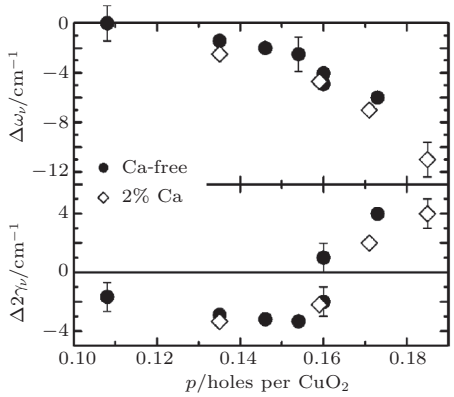


Fig. 9. Carrier concentration dependence of renormalization of B_{1g} phonon in YBCO, where $\Delta\omega \equiv \omega(30\ \mathrm{K}) - \omega(100\ \mathrm{K})$, and $2\Delta\gamma \equiv 2\gamma(T_0) - \Gamma_{\mathrm{AH}}(T_0)$.[51]

The studies of phonon renormalization in Bi-2212 system are relatively dispersive. Burns *et al.* and Boekholt *et al.* reported that a softening occurs in the A_{1g} mode of O(3) in SrO at 465 cm^{-1}.[95,96] This was also seen by Martin *et al.*[97,98] Below T_c, Leach *et al.* observed an anomalous drop of width of B_{1g} mode at 285 cm^{-1},[99] which is exactly the same as B_{1g} mode at 340 cm^{-1} in YBCO. However, no frequency change is found for the B_{1g} mode. These results are still waiting for a consistent understanding.

In Hg-1201, the softening of A_{1g} mode at 568 cm^{-1} below T_c was observed and accompanied by a broadening.[100] Actually, phonon anomalies exist at T_c in the Hg-1223 system.[101] Unlike YBCO, neither renormalization nor the Fano lineshape was seen in the B_{1g} mode of oxygen in CuO planes at 245 cm^{-1}. This may be due to the flat,[102] rather buckled CuO planes.[63,64] Interestingly, a large asymmetry was found in the A_{1g} mode of in-plane oxygen. The EPC observed in Hg-1234 may be the strongest among cuprates, as shown in Fig. 10.[103] A sharp drop in frequency was probed at T_c in the A_{1g} modes of Ca at 390 cm^{-1} and in-plane oxygen. At the same time, their widths are doubled across SC transition. Using Allen formula,[104] the EPC strength is about 0.08. Suppose that the EPC strengths of all the 60 modes are roughly at the same level, we will obtain a total EPC strength of $\lambda \sim 6$, which is high enough to explain the high T_c.

All the results mentioned above lead to the conclusion that unambiguous phonon renormalization induced by SC

transition can be seen in most cuprate systems. The magnitude of renormalization depends on many factors, such as cuprate systems, phonon frequency and symmetry, and doping. The most prominent manifestations are the in-plane oxygen B_{1g} mode in YBCO-123, A_{1g} modes in YBCO-124, and the Hg-system. Phonon renormalization caused by EPC provides an insight into pairing mechanism.

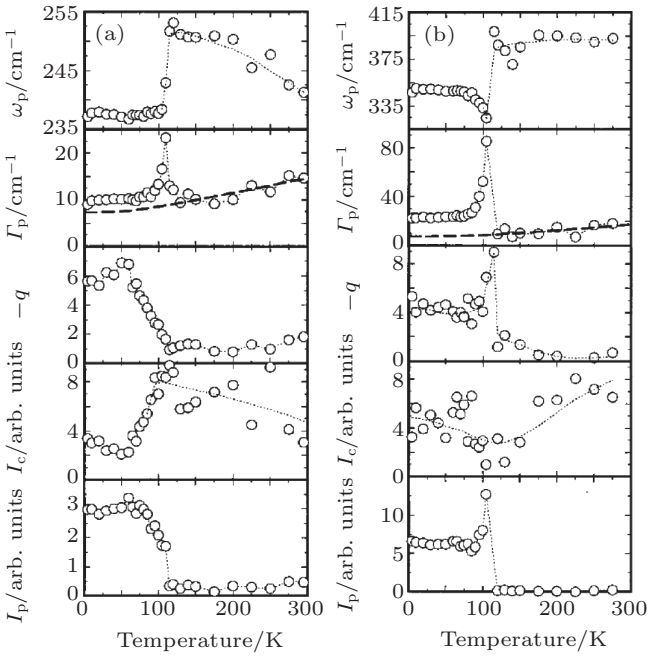


Fig. 10. Fitting results of A_{1g} modes at (a) 240 cm^{-1} and (b) 390 cm^{-1}. The dashed lines are the fitting to the data in the normal state.[103]

3.3.2. Phonon intensities

Besides phonon frequency and width, large changes in phonon intensity were also found in many cuprate superconductors when crossing T_c.[29,101,103,105–110] A representative example is Hg-1234,[103] as shown in Fig. 11. In the normal state, the modes at 240 cm^{-1} and 390 cm^{-1} are quite weak and the modes at 487 cm^{-1} and 575 cm^{-1} are almost invisible. All the four modes have an A_{1g} symmetry. The modes are dramatically enhanced in the SC state. The enhancement of intensity was considered to be contributed by Fano resonance. In YBCO, the integrated intensities of Ba A_{1g} mode increase several times below T_c, but only in the XX channel. In Bi-2212, an intensity anomaly when using an excitation laser of 633 nm is more obvious than 458 nm. A resonant study of *R*BCO-123 (*R* = Yb, Er, Sm, Nd) revealed that the O(4) mode in X'X' channel and the O(2)–O(3) mode in X'Y' channel become stronger in the SC state when excitation energy is larger than 2.1 eV, while they are suppressed using an excitation laser energy less than 2.1 eV.[110] The characteristic energy does not depend on carrier concentration. Similar resonance phenomena were also observed in YBCO thin films deposited on $SrTiO_3$ substrate.[109] Sherman *et al.* proposed an EPC picture to explain the above results.[107] Generally, Raman intensity of B_{1g} mode is determined by interband EPC and the coupling strength depends on the ratio of EPC matrix elements to the interband gap. The smaller the gap is, the larger the coupling will be. The SC transition effectively extends the phase volume of electrons in the intermediate scattering process, due to a small gap and a large density of states at Fermi level in the SC state. This explains why the scattering intensity is raised across the SC transition. The intensity anomaly of A_{1g} mode was analyzed within the framework of weak-coupling BCS theory.[108] The excitation energy dependence of O(4) mode intensity mentioned above,[110] could be also understood within this picture. The change of force constants induced by the SC transition also seems to be a plausible explanation for the intensity anomaly at T_c.[55]

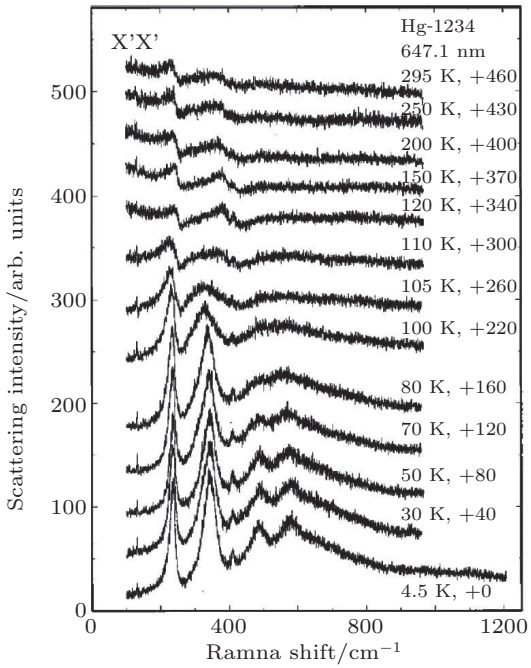


Fig. 11. Raman spectra at various temperatures in Hg-1234 with $T_c \sim 123$ K.[103]

4. Electron–phonon coupling in iron-based superconductors

There are not so many Raman studies of EPC in iron-based superconductors as in cuprates. In the 1111 system, no phonon asymmetry was reported so far. Phonon anomalies are absent at structural, spin-density-wave (SDW), and SC transitions. It implies that EPC and phonon renormalization are weak in the system.[111–113] Similarly, in the 111 system, without structural and magnetic phase transitions, the observations are basically close to those in 1111 system.[114] While in the 11 and 122 systems, Fano lineshapes and renormalization are observed. We discuss this in more detail in the following.

4.1. Fano effect in Fe superconductors

Fano effect was explicitly reported in Co-doped $BaFe_2As_2$,[115] as shown in Fig. 12. An asymmetry of A_{1g} phonon lineshape appears in the SDW state. The asymmetry becomes more clear with increased doping, indicating an enhancement of EPC. The changes in frequency and width are too large to be explained by a c-axis change of only about 0.3%. From the EPC point of view, the phonon softening and broadening are a consequence of the increase in the density of states at the Fermi level induced by carrier doping. This is quite similar to the case of cuprates, in which phonon asymmetry strongly depends on doping level. This conclusion seems to be supported also by calculations.[116] Furthermore, it was found that a spin-ordered state also favors EPC. The maximum EPC strength λ even reaches up to 0.35 in 122. However, the value is still not large enough to estimate T_c.

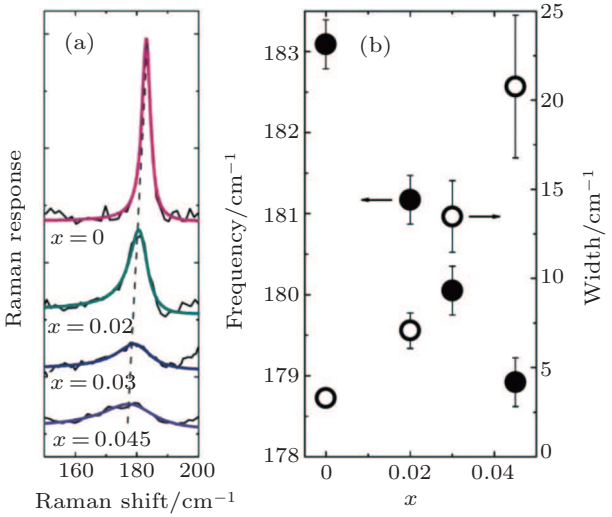


Fig. 12. (a) The evolution of A_{1g} phonons with doping in $Ba(Fe_{1-x}Co_x)_2As_2$. The solid lines are Fano fitting results. (b) Doping dependence of frequencies and widths of the A_{1g} phonon.[115]

4.2. Renormalization of self-energy induced by SDW and SC transitions

So far, there is no consensus on whether or not phonon self-energy renormalization occurs at the structural, SDW, or SC transitions in Fe-based compounds. Litvinvhuk *et al.* reported that no anomaly was found in $Sr_{1-x}K_xFe_2As_2$ ($x = 0$, 0.4),[117] similar to the results in 1111 system. While in $CaFe_2As_2$, a new A_{1g} mode appears below the SDW transition temperature and the B_{1g} mode exhibits an anomalous broadening and a hardening of $\sim$4 cm^{-1}.[118] This can be well understood in terms of a phonon renormalization due to the SDW gap rather than a simple lattice change. The SDW gap was probed in optical absorption spectra.[119] Phonon renormalization caused by the SDW gap was also in parent compound $BaFe_2As_2$[120] and hole-doped $R_{1-x}K_xFe_2As_2$ (R = Ba, Sr).[121] Figure 13 shows the behavior of A_{1g} mode in $Ba_{0.72}K_{0.28}Fe_2As_2$ and $BaFe_2As_2$ at SDW transition temperature. The A_{1g} renormalization effect is weaker in K-doped samples than in the parent compound. Similar but weaker results were obtained in the B_{1g} channel. This was supported by first-principles calculations, which indicate that EPC related to As ions is stronger.[122–124] Besides, a small softening of 0.3 cm^{-1} and a kink in width appear at T_c, which was also reported in $Sr_{0.85}K_{0.15}Fe_2As_2$ ($T_c = 29$ K). This was considered to be phonon self-energy renormalization associated with the opening of the SC gap, which implies that the SC gap 2Δ should be smaller than the A_{1g} phonon frequency, i.e., $2\Delta \leq 23$ meV. Actually, the renormalization induced by the SC gap was also observed in the B_{1g} channel in $Pr_{0.12}Ca_{0.88}Fe_2As_2$.[125]

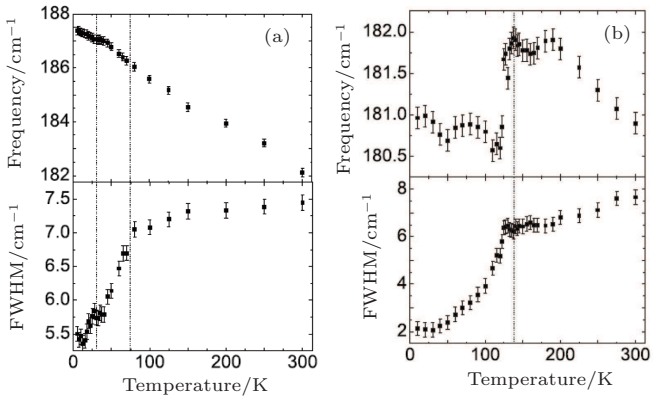


Fig. 13. The evolution of frequencies and widths of A_{1g} mode with temperature in (a) underdoped $Ba_{0.72}K_{0.28}Fe_2As_2$ ($T_c = 29$ K, $T_s = 75$ K) and (b) parent compound $BaFe_2As_2$ ($T_s = 138$ K).[121]

A systematic study of phonon renormalization in the 122 system is shown in Fig. 14.[126] In the three parent compounds, clear kinks in both frequency and width can be seen at SDW transition temperatures. This was explained as phonon renormalization that is due to the opening of the SDW gap. The larger and discontinuous jump in $CaFe_2As_2$ is characteristic of a first-order structural transition rather than the SDW transition. In SC 122 samples, the hardening and softening of B_{1g} mode caused by SC transition could be evaluated after subtracting contributions from the SDW transition. The values give us an EPC constant $\lambda_{B_{1g}} \approx 0.02$, using the Allen formula $\Gamma = 2\pi\lambda N(0)\omega^2$,[104] where the phonon width Γ is related to EPC, ω is the phonon frequency, λ is the EPC strength, and $N(0)$ is the density of states at the Fermi surface. Phonon renormalization provides an alternative way to get EPC strength through the formula $\lambda = -K\,\mathrm{Re}[(\sin u)/u]$ with $K = \omega^{SC}/\omega^{N} - 1$ and $u \equiv \pi + 2i\cosh^{-1}(\omega^{N}/2\Delta)$, where ω^{SC} and ω^{N} are the phonon frequencies in the SC state and normal state, respectively. This gives $\lambda_{B_{1g}} \approx 0.01$. The EPC strengths estimated by both methods are far smaller than 1, just as required by strong-coupling theories if one would explain the high T_c by a pure EPC picture.[122] This seems to imply that EPC may play a minor role in Fe-based superconductors.

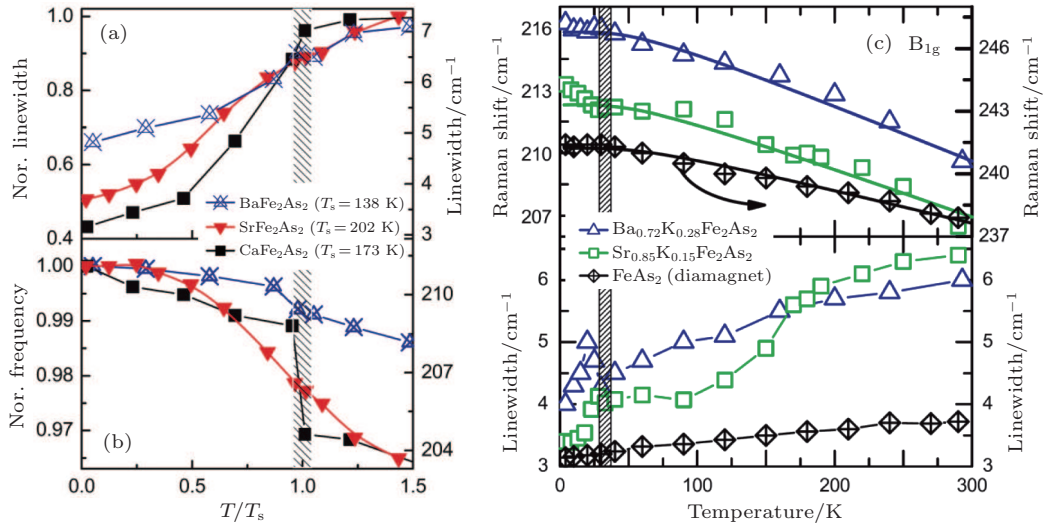


Fig. 14. Temperature dependence of normalized (a) linewidth and (b) frequency in the three 122 parent compounds. (c) Raman frequencies and widths of B_{1g} mode in two parent compounds and reference compound $FeAs_2$.[126]

Phonon anomalies across SDW and structural transitions were also reported in the 11 system. The B_{1g}mode in $Fe_{1.02}Te$ shows an anomalous kink in frequency near the transition temperatures.[127,128] With the substitution of Se for Te, both the SDW transition and the kink are suppressed, which suggests that the kink is a reflection of the opening of the SDW gap (Fig. 15). Actually, the suppression was also found when doping excess Fe into parent compounds.[129,130] Similarly, phonon renormalization is weak in 11 compounds with excess Fe. Phonon anomalies caused by the SDW transition indicate the existence of EPC in the 11 system.

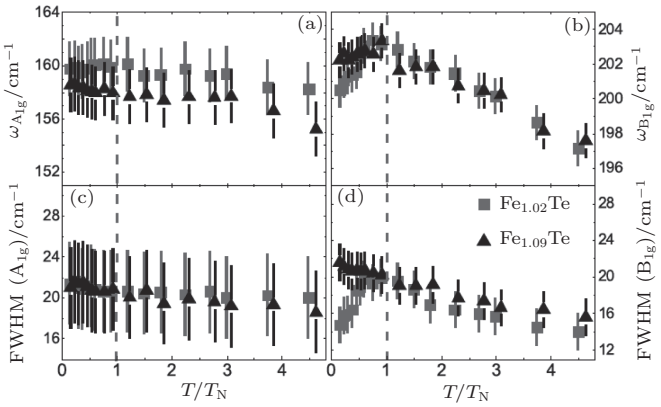


Fig. 15. Temperature dependence of (a, b) frequencies and (c, d) widths of A_{1g} and B_{1g} phonons in FeTe parent compounds with different Fe content.[127]

EPC was observed in the newly discovered $K_xFe_{2-y}Se_2$ system. Raman measurements by Zhang *et al.* reveal that the Ag mode at 180 cm^{-1} exhibits a hardening of ~ 1.5 cm^{-1} when entering the SC state,[131] as shown in Fig. 16. The indication of renormalization suggests an SC gap of $2\Delta \leq 22.5$ meV (180 cm^{-1}), which is close to the SC gap of about 20 meV measured by angle-resolved photoemission.[132] In addition, the rapid decrease of integrated intensities of Se A_g mode at 66 cm^{-1} with temperature, is similar to that of A_{1g} modes in cuprate superconductors (Fig. 11), which has been considered to be evidence of EPC.

So far, we find two kinds of renormalization phenomena in Fe-based superconductors. One is associated with the opening of the SDW gap, as displayed in 11 and 122 systems. This is evidence for EPC and further calculations demonstrate that EPC favors a spin-ordered state. Some experimental indications of spin–lattice coupling exist in the compounds. This implies a tight binding of spin, lattice, and charge degrees of freedom in the two systems. Though EPC may play a minor role in pairing, it has an important impact on superconductivity.[115,116] The other one is the renormalization induced by the SC transition. Up to now, the effect was reported only in 122 and $K_{0.8}Fe_{1.6}Se_2$ systems. The reason it is weak in other Fe-based systems may be that the mismatch between SC gap and relevant phonons is relatively large. As mentioned above, phonon renormalization gives $\lambda_{B_{1g}} \approx 0.02$.[126] For comparison, EPC strengths of 0.2 and 0.12 were estimated by femtosecond time-resolved photoemission[133] and pump–probe optical reflectivity,[134] respectively. First-principles calculations produce $\lambda \sim 0.23$,[123] 0.35,[116] and 0.37.[135] Raman scattering gives a smaller EPC strength because it measures partial phonon branches at the Brillouin zone center. Even using the calculated EPC strengths, one cannot satisfactorily explain the T_c's.

The experimental and calculated EPC strengths in Fe-based systems are summarized in Table 1. For comparison, EPC strengths of cuprate, MgB_2, and A15 Nb_3Sn superconductors are also listed in the table. In the 11 system, the experimental EPC strength is ~ 0.16,[137,138] and the calculated values are 0.17,[138] 0.15–0.39,[139] and 0.3.[140] Like

the case of 122 system, the strengths are too small to explain the T_c's. There is no consensus on EPC in 1111 system. The EPC strengths were estimated to be 0.25 and 1.53 by resistivity in non-SC and SC $PrFeAsO_{1-x}F_x$ polycrystals,[141] which seems to support a BCS explanation on $T_c \sim 50$ K. However, optical pump–probe femtosecond spectroscopy measured $\lambda \sim 0.18$,[142] and first-principles calculations gave $\lambda \sim 0.21$ in 1111 system,[122] which predicts a T_c of ~ 0.8 K. Further exploration of EPC in the 1111 system is still waiting for high-quality single crystals to become available. A similar case is found in 111 system. There is a big difference between EPC strengths presented by ARPES and band calculations, which are 1.53 and 0.29, respectively.

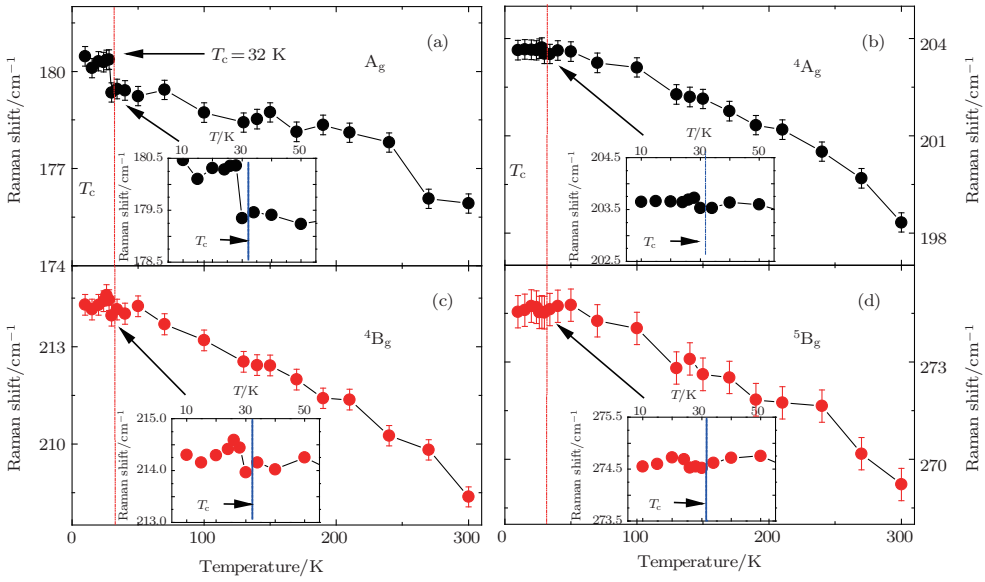


Fig. 16. Temperature dependence of Raman frequencies of (a) A_g, (b) ^4A_g, (c) ^4B_g, and (d) ^5B_g modes in $K_{0.8}Fe_{1.6}Se_2$.[131]

Table 1. Experimental and calculated EPC strengths in Fe-based and other superconductors.

System	Experimental λ	Calculated λ
11	0.16–0.01,[136] 0.16[137]	0.17[138], 0.15–0.39,[139] 0.3[140]
111	1.38[143]	0.29[144]
1111	0.25(NSC),[141] 1.53(SC),[141] 0.18[142]	0.21[122]
122	0.02,[126] 0.2,[133] 0.12[134]	0.23,[123] 0.35,[116] 0.37[135]
122*	–	0.19–0.34[139]
YBCO-123	0.016–0.020[47]	0.27[148]
MgB_2	0.3[158]	1.01[159]
Nb_3Sn	1.80[160]	1.19[161]

5. Conclusion

The consequences of EPC observed in cuprate superconductors, such as asymmetric phonon lineshape, softening or hardening, broadening, intensity changes, etc., are exactly demonstrated in Fe-based systems. The weaker EPC in Fe-based systems may be due to stronger screening effects. The measured EPC strength is roughly in agreement with the calculated one in Fe-based systems, but it seems not to be a dominant factor for pairing. On the other hand, due to the strong coupling among electrons, spins, and phonons, EPC's effect on superconductivity remains to be revealed.

The situation is more complicated in cuprate superconductors. A lot of calculations demonstrated that EPC alone is not enough to explain such high T_c's.[145–148] Some calculations[149,150] even indicated that EPC works against pairing. On the experimental side, both ARPES[151] and STM[152] suggest that EPC is a key factor for pairing, which is supported by some theoretical calculations.[153,154] Egami[155] proposed that EPC in cuprates is a more complicated spin–charge–phonon coupling rather than BCS-like, due to spin-dependent charge transfer mediated by phonons in strongly correlated systems. A d-wave pairing can be realized in an EPC system with the aid of spin fluctuations.[156] After summarizing many experiments, Kulic proposed that EPC is just the pairing glue in cuprates.[157] Therefore, although a clear microscopic picture is absent, EPC is substantially involved in pairing in some way in both high-T_c superconductors. As their parent compounds are spin-ordered, we conclude that a delicate coupling between electrons, spins, and phonons may be crucial to high-T_c superconductivity.

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