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Iron-specific phonon density of states in the superconductors $\mathbf{LaFeAsO_{1-x}F_x}$ and $\mathbf{La_{1-x}Ca_xFePO}$

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We measured element-specific Fe-phonon densities of states (Fe-PDOSs) of $LaFeAsO_{1-x}F_x$ ($x=0,0.11$) and $La_{1-x}Ca_xFePO$ ($x=0.13$) by nuclear resonant inelastic scattering of synchrotron radiation. Since the bands crossing the Fermi edge are mainly Fe 3*d* bands, Fe is the key element for iron pnictide superconductivity, and therefore element-specific Fe-PDOS offers crucial information on the phonon contribution in iron pnictide superconductors. The measured Fe-PDOSs of superconductor $LaFeAsO_{0.89}F_{0.11}$ ($T_c=26$ K) and that of non-superconductor LaFeAsO have similar structures both below T_c (15 K) and above T_c (298 K); therefore, fluorine doping does not have a notable effect on the Fe-PDOS. The measured Fe-PDOS of $LaFeAsO_{1-x}F_x$ agrees well with Fe-PDOS spectrum determined from a previous first-principles calculation and shows structural resemblance with a calculated Éliashberg function $\alpha^2F(\omega)$, which indicates small electron-phonon coupling. Therefore, our results indicate that phonons are not the main contributors to the high-T_c superconductivity of $LaFeAsO_{1-x}F_x$.

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I. INTRODUCTION

It was recently discovered that some iron- and nickel-based pnictides [$LaFeAsO_{1-x}F_x$,[1] LaNiPO,[2] and $LaFeAsO_{1-x}F_x$ (Ref. 3)] exhibit superconductivity. After this discovery, many studies have been performed on iron pnictide compounds. Further, it was found that replacing La with other rare-earth elements [Pr,[4] Nd,[5] Sm,[6] and Gd (Refs. 7 and 8)] raises the transition temperature (T_c) to above 50 K and increasing the pressure raises the T_c.[9–11] In addition, oxygen-deficient ($ReFeAsO_{1-x}$, Re: rare-earth metal[8,12,13]) and oxygen-free bilayer [$Ba_{1-x}K_xFe_2As_2$ (Ref. 14)] superconductors were synthesized. In addition to cuprate superconductors, this new family of iron pnictide compounds provides another platform to explore high-T_c superconductivity. In the cuprate superconductors, superconductivity arises by doping a magnetically ordered parent compound even though the parent compound itself is not a superconductor. Neutron-scattering,[15] Mössbauer spectroscopy,[16,17] and muon spin rotation[17] measurements have revealed the magnetic order of Fe moments in LaFeAsO, and upon doping the magnetic order is suppressed and superconductivity is observed.[15,16] This is the most striking similarity between cuprate and iron pnictide superconductors. However, in the latter, the relevance of the electron correlation effect is controversial[18–20] and multiband superconductivity composed mainly of five Fe 3*d* bands near the Fermi level is predicted from first-principles calculations,[21–26] whereas cuprates have only one relevant band.

The pairing mechanism that leads to the high T_c in iron pnictide superconductors is currently the most critical and controversial issue. First-principles calculations predict that the phonon mechanism is not the main mechanism that induces high-T_c superconductivity in iron pnictides.[20,27,28] Further, optical spectroscopy[29] also suggests that the electron-phonon interaction does not contribute to the main pairing mechanism. On the other hand, the strong electron-phonon coupling of the Fe-breathing mode of $LaFeAsO_{1-x}F_x$ has been reported,[30] and it is suggested that a possible phonon-mediated mechanism on the basis of the presently available calculations should not be discarded.[31] Therefore, the experimental study of the phonon states in iron pnictides is important. To determine whether the electron-phonon interaction is the main pairing factor in conventional superconductors or not, structural similarity between the phonon density of states (PDOS) and the Éliashberg function $\alpha^2F(\omega)$ obtained by tunneling measurements is important.[32] Thus far, Raman, infrared spectroscopy, and neutron inelastic measurements and x-ray inelastic scattering[29,31,33–39] have been performed to investigate the phonons. In particular, information about element-specific Fe-phonon densities of states (Fe-PDOSs) is highly desired because the bands crossing the Fermi edge are mainly Fe 3*d* bands, as expected and confirmed by photoemission spectroscopy.[40–42] Therefore, to determine the cause of pairing in iron pnictide superconductors, Fe is the key element. The nuclear resonant inelastic scattering of synchrotron radiation[43,44] is one of the most effective methods to obtain element (isotope)-specific phonon energy spectra. This method has been improved further to determine site-specific phonon densities of states.[45] Further, this method is very suitable and effective for the experimental studies on iron pnictide superconductors. In the case of powdered iron pnictide samples, methods such as neutron inelastic scattering and x-ray inelastic scattering do not give true phonon densities of states. This is because these methods give scattering-intensity-weighted phonon densities of states. On the other hand, the nuclear resonant inelastic-scattering method gives true phonon densities of states concerning Fe atoms.

We have performed nuclear resonant inelastic-scattering measurements for $LaFeAsO_{1-x}F_x$ ($x=0, 0.11$) and have obtained temperature-dependent element-specific phonon densities of states. We have also measured element-specific phonon densities of states of superconducting $La_{1-x}Ca_xFePO$ ($x=0.13$). In this paper, we discuss the temperature dependence of Fe-PDOS and the change upon doping by comparing the Fe-PDOS and the Éliashberg function calculated from first-principles band calculations.[28]

II. EXPERIMENT

$LaFeAsO_{1-x}F_x$ ($x=0, 0.11$) and $La_{1-x}Ca_xFePO$ ($x=0.13$) samples were synthesized by methods described in Refs. 3 and 46, respectively. The quality of the samples was examined by x-ray diffraction, and the samples were found to consist of almost single phases with small amounts of FeAs and $LaAsO_4$ in LaFeAsO and LaAs in $LaFeAsO_{0.89}F_{0.11}$. From the previous Mössbauer-effect measurements,[16,47] it is also confirmed that the measured samples are almost in a single phase and the impurity in each sample is less than 10%. Electrical resistivity measurements of the superconductors $LaFeAsO_{0.89}F_{0.11}$ and $La_{0.87}Ca_{0.13}FePO$ gave T_c values 26 and 5.4 K, respectively. On the other hand, LaFeAsO did not undergo a superconducting transition but showed a resistivity anomaly[3] and structural phase transition[15,48] at around 150 K followed by magnetic ordering.[16] Samples of these compounds (~25 mg) were mixed with BN and polyethylene powder, and the mixtures were pressed to form pellets with a diameter of 10 mm.

Nuclear resonant inelastic-scattering experiments were performed at the nuclear resonant scattering beamline (BL09XU) and the Japan Atomic Energy Agency beamline (BL11XU) of SPring-8. The electron-beam current of the storage ring was 100 mA at 8 GeV. A double-crystal Si(111) premonochromator was used to handle the high-heat load of undulator radiation, and the radiation was monochromatized to the bandwidth of 2.5 meV (full width at half maximum) with a nested high-resolution monochromator consisting of asymmetric Si(511) and asymmetric Si(975) channel-cut crystals. The energy of the radiation was varied around the first nuclear resonant excitation energy of ^{57}Fe (14.413 keV). The intensity of the incident beam was monitored with an ionization chamber and a beam flux monitor. We have measured element-specific phonon energy spectra of Fe in the samples as a function of the incident x-ray energy by counting the number of delayed photons, which are generated by nuclear resonant scattering. To count the number of delayed photons, we used a multielement Si avalanche photodiode (APD) detector. Measurements were performed on $La_{0.87}Ca_{0.13}FePO$ at 298 K, on $LaFeAsO_{0.89}F_{0.11}$ at 15, 40, and 298 K, and on LaFeAsO at 15 and 298 K using a liquid-He-flow cryostat.

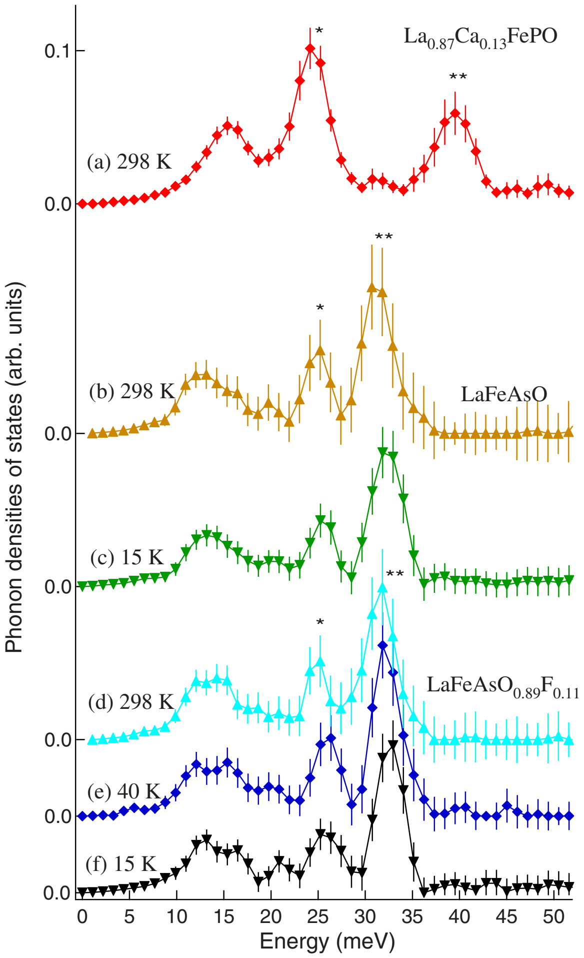

FIG. 1. (Color online) Element specific Fe-phonon densities of states of $La_{0.87}Ca_{0.13}FePO$ [(a) 298 K], LaFeAsO [(b) 298 and (c) 15 K], and $LaFeAsO_{0.89}F_{0.11}$ [(d) 298, (e) 40, and (f) 15 K] obtained from the corresponding spectra of nuclear resonant inelastic scattering of synchrotron radiation.

III. RESULTS AND DISCUSSION

The measured nuclear resonant inelastic-scattering spectra were converted to element-specific Fe-PDOS according to the method described in Ref. 49 and shown in Fig. 1. Since the impurity in each sample is less than 10%, the Fe-PDOSs obtained were only due to the Fe atoms in iron pnictide without resorting to the site-specific nuclear resonant scattering method.[45] In $LaFeAsO_{1-x}F_x$ Fe-PDOSs, no drastic spectrum change was observed upon fluorine doping at the temperatures used.

The $La_{0.87}Ca_{0.13}FePO$ spectrum shows three main peaks (Fig. 1). The positions of low (16 meV) and middle (25 meV, labeled with * in Fig. 1) energy peaks are near the corresponding low (12 meV) and middle (25 meV) energy peaks of $LaFeAsO_{1-x}F_x$ spectra measured at 298 K, respectively, but the high-energy peak at about 41 meV (labeled with ** in Fig. 1) is at a much higher energy than that in $LaFeAsO_{1-x}F_x$ (~31 meV). It is known that vibrational frequency is proportional to the square of the mass ratio (1.56; As: 74.92 and P: 30.97), assuming that the coupling constants are identical. Since this value is close to the frequency ratio (1.28), the highest peak is thought to be due to coupling between Fe and pnicogen. This assignment is supported by first-principles band calculations.[28] It is noted that, in the case of PDOS spectra, the observed peak usually does not correspond to one phonon mode as observed in Raman or IR spectra. In conventional phonon-mediated superconductors with the same electron densities at the Fermi level and the same strength of electron-phonon interactions, higher phonon energy leads to a higher T_c. Therefore, the obtained energy shift shows the opposite result to that expected from the conventional phonon Bardeen-Cooper-Schrieffer theory by assuming that all other parameters are the same. However, because the peak intensities of $\mathrm{LaFeAsO}_{1-x}\mathrm{F}_x$ are larger than those of $\mathrm{La}_{0.87}\mathrm{Ca}_{0.13}\mathrm{FePO}$, the coupling between Fe and pnicogen in $\mathrm{LaFeAsO}_{1-x}\mathrm{F}_x$ may be stronger than that of $\mathrm{La}_{0.87}\mathrm{Ca}_{0.13}\mathrm{FePO}$.

No significant change in the Fe-PDOS spectra upon fluorine doping was observed in $\mathrm{LaFeAsO}_{1-x}\mathrm{F}_x$. Although LaFeAsO undergoes a structural phase transition at around 150 K,[15,48] the change is so small that the Fe-PDOS is thought to be unchanged within experimental error. As shown in Fig. 2, the Fe-PDOS spectra of $\mathrm{LaFeAsO}_{1-x}\mathrm{F}_x$ agree very well with those obtained from the first-principles band calculations,[28] which also calculated the electron-phonon coupling constant (λ) as 0.21, giving a value of T_c below 0.5 K and the Éliashberg function giving a T_c value of 0.8 K. Further, another first-principles band calculation gives the value of λ as 0.2.[27] A comparison of the obtained Fe-PDOS and the calculated Éliashberg function[28] is also shown in Fig. 2. The similarity between our experimentally obtained Fe-PDOS and the calculated spectrum shows the reliability of the calculation and implies that the phonon mechanism alone cannot explain the high-T_c value of the superconductivity in iron pnictides. On the other hand, the strong electron-phonon coupling of the Fe-breathing mode (29.4 meV) was pointed out.[30] It is known that electron-phonon coupling should result in a large phonon linewidth and, in fact, a broadening (~25 meV along Γ to A) was observed in the E_{2g} mode in phonon-mediated superconductor MgB_2.[50,51] Compared to the energy of the E_{2g} mode (above 60 meV), the energy of the Fe-breathing mode is relatively low. Therefore, if phonons are the main mechanism of superconductivity in iron pnictides, strong electron-phonon coupling is also expected, although their T_c is slightly different. In our Fe-PDOS spectra, phonon density around the expected value of 29.4 meV is small. It should be noted that the observed peak positions in Fe-PDOS spectra are lower than the respective positions of the calculated Fe-PDOS spectrum[28] as shown in Fig. 2. Therefore, the true energy of the breathing mode may be slightly shifted (possibly around 26 meV). If the predicted strong electron-phonon coupling exists, a broadening of the linewidth and/or the reduction in the relative intensity due to the broadening may be expected. Even if the broadening is caused by a single-mode phonon, the dispersion of the phonon is expected to cause a change in the Fe-PDOS spectra. However, no apparent change in the Fe-PDOS of superconducting $\mathrm{LaFeAsO}_{0.89}\mathrm{F}_{0.11}$ and the parent compound LaFeAsO was observed compared to the Fe-PDOS calculated without considering the Fermi-surface nesting effect, i.e., with a standard electron-phonon coupling method as discussed above. Further, apparent broadening and reduction in the relative intensity were not observed at other peaks (~12 and ~32 meV) as well. Since the energy resolution of our monochromator is 2.5 meV and the width of the peak at 25 meV in the calculated Fe-PDOS spectrum is less than 10 meV, the resolution is sufficient to observe the same order broadening of the peak as that observed in MgB_2 superconductor. The excellent structural agreement between our results and the calculations with the standard electron-phonon coupling method suggests that phonons are not the main contributors to the high-T_c superconductivity. However, it should be noted that it is impossible to completely exclude phonon contributions to superconductivity with the resolution of our monochromator. From the experimental viewpoint, comparing our observed Fe-PDOS to experimentally obtained glue bosonic spectra, which can be obtained with tunneling or optical measurements, provides clear insight that the fundamental mechanism of the iron pnictide superconductors is due to phonons or other mechanisms. In fact, bosonic spectra, which were obtained by optical spectroscopy[29] with some assumptions, are entirely different from the PDOS.

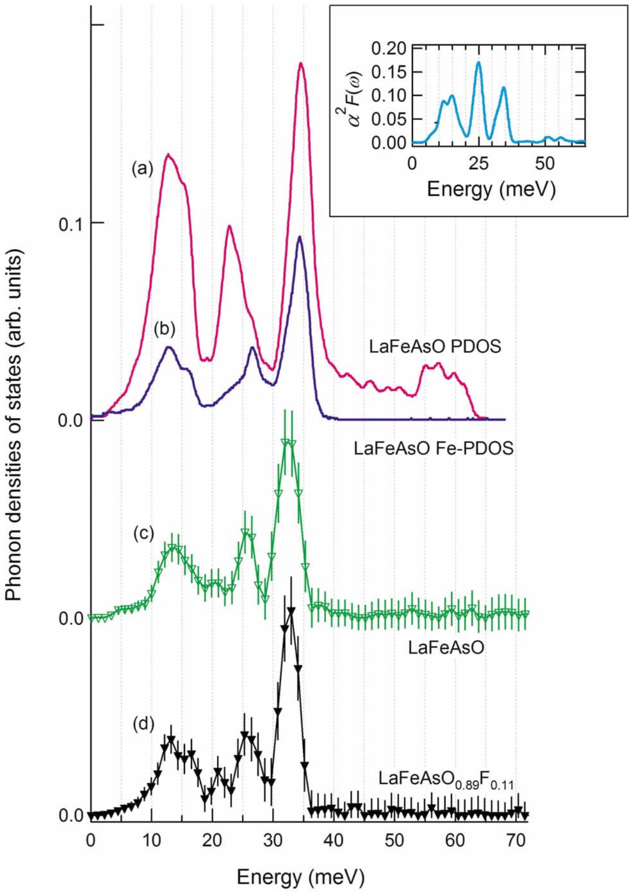

FIG. 2. (Color online) Experimentally observed and calculated phonon densities of states. (a) Phonon density of states and (b) partial Fe-phonon density of states of LaFeAsO calculated by Cao *et al.* (Ref. 23) and element-specific Fe-phonon densities of states of (c) LaFeAsO and (d) $\mathrm{LaFeAsO}_{0.89}\mathrm{F}_{0.11}$ measured at 15 K. Inset is the Éliashberg function $\alpha^2F(\omega)$ calculated from first-principles band calculations (Ref. 28).

TABLE I. Temperature dependence of the energy shifts of peak positions observed in the Fe-PDOS spectra of $La_{0.87}Ca_{0.13}FePO$, LaFeAsO, and $LaFeAsO_{0.89}F_{0.11}$. Middle-energy peak in this table denotes the peak position labeled with * in Fig. 1 and high-energy peak denotes the position labeled with ** in Fig. 1.

Temperature (K)	Peak positions (meV)		
	$LaFeAsO_{0.89}F_{0.11}$	LaFeAsO	$La_{0.87}Ca_{0.13}FePO$
	(Middle)	(Middle)	(Middle)
298	24.9(4)	24.9(5)	25.3(1)
40	25.9(4)		
15	25.9(5)	25.6(4)	
	(High)	(High)	(High)
298	31.6(4)	31.3(4)	40.5(3)
40	32.3(2)		
15	32.7(2)	32.5(2)	

We have found the energy shifts of the middle and highest peaks in $LaFeAsO_{1-x}F_x$ Fe-PDOS accompanying the temperature changes. These changes are listed in Table I, in which the peak positions were obtained from Gaussian fits to the peaks. These indicate an anharmonic effect around the Fe atoms. In fact, the energy shifts of A_{1g} (As) and B_{1g} (Fe) modes of $K_{0.4}Sr_{0.6}Fe_2As_2$ were observed in Raman spectra and were attributed to the anharmonic effect.[38] However, the temperature-dependent IR active E_u mode[36] was found to be due to a harmonic effect,[52] and the temperature-dependent study of neutron-weighted PDOS indicated a rather harmonic system for the parent compound $BaFe_2As_2$.[39]

Although the substitution of La with Sm raises the T_c value up to 54.6 K in iron pnictides,[6] it has not exactly been proved that phonon-mediated superconductors do not attain such high-T_c value. The structural resemblance and the same temperature dependence of Fe-PDOS of nonsuperconducting LaFeAsO and superconducting $LaFeAsO_{0.89}F_{0.11}$ give the impression that phonons are not the main mechanism for the high-T_c value in iron pnictide superconductors. However, if strong electron-phonon interactions, which our method failed to detect and which cause the structural phase transition in LaFeAsO, are present between Fe and As high-T_c superconductivity may arise in $LaFeAsO_{0.89}F_{0.11}$. This may be due to the suppression of both the structural phase transition and antiferromagnetic ordering by fluorine doping. In fact, each fine structure of $LaFeAsO_{0.89}F_{0.11}$, LaFeAsO, and calculated Fe-PDOS is slightly different as seen in Fig. 2. Serious discrepancies in the Fe moment between experiments[15,16] and band calculations[23,25,53] require further investigation of the electronic states of iron pnictides both experimentally and theoretically.

IV. SUMMARY

In summary, we have measured element-specific Fe-PDOSs of $LaFeAsO_{1-x}F_x$ ($x=0,0.11$) and $La_{1-x}Ca_xFePO$ ($x=0.13$) by nuclear resonant inelastic scattering of synchrotron radiation. Observed Fe-PDOS of $LaFeAsO_{1-x}F_x$ agree well with the calculated Fe-PDOS from the first-principles band calculations.[28] The excellent agreement with the small λ [0.2 (Ref. 27) and 0.21 (Ref. 28)] and the same temperature dependence and similar structures of $LaFeAsO_{1-x}F_x$ ($x=0,0.11$) indicate that the high T_c in iron pnictide superconductors is not due mainly to phonons.

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