

Phonon Dynamics in Parent and Superconducting FeAs Compounds

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(Received April 12, 2010)

The comparison of the phonon spectra of the parent MFe_2As_2 ($M = Ba, Ca, Sr$) and $MFeAsF$ ($M = Ca, Sr$) compounds show a very strong renormalization in the lower and intermediate frequency part of the vibrations. Mass effects and lattice contraction cannot solely explain these changes. Further, the measurement of temperature dependence of phonon density of states across the magnetic/structural phase transition temperature indicates that softening of low energy modes in the paramagnetic state of these compounds may be due to the interaction of phonons with electron or short-range spin fluctuations in the normal state of the superconducting compound as well as in the parent compound. Detailed lattice dynamical calculations are carried out to analyze the experimental data.

PACS numbers: 74.25.Kc, 78.70.Nx, 63.20.-e

I. INTRODUCTION

The discovery of superconductivity in fluorine-doped $RFeAsO$ ($R =$ rare earth) and K -doped $BaFe_2As_2$ has stimulated enormous interest [1–26] in the field of condensed matter physics. It is important to note that these compounds have high superconducting transition temperatures without requiring the presence of copper oxide layers. There are mainly three types of FeAs-based superconductors such as $RFeAsO$ (R : Rare earth elements), MFe_2As_2 (M : alkaline-earth elements), and $MFeAsF$. The structural, magnetic, electronic properties of all the compounds have been extensively investigated to understand the mechanism of superconductivity. In particular, for all these compounds, strong anomalies have been found [3] in the specific heat, resistivity and magnetic susceptibility in the temperature range of 110 to 180 K. These anomalies are now known to be a prerequisite for superconductivity in FeAs compounds. Electron or hole doping suppresses these phase transitions and induces superconductivity at lower temperatures. At present, it remains unclear whether the change in the electron concentration by doping is essential for achieving superconductivity

or whether the suppression of the phase transition into a magnetically ordered state is the main effect.

Theoretical electronic structure calculations[7] have been reported. These calculations show that in FeAs compounds electronic bands around the Fermi level are formed mainly by Fe-As layers, whereas the bands of R-O layers are far from the Fermi level. Therefore, superconductivity in these compounds is believed to be mainly due to the structural and electronic states of the Fe-As layers.

The application of hydrostatic pressure in F-doped LaFeAsO results [4] in an increase in T_c from 25 K to 43 K. Pressure-induced superconductivity has been reported [5, 6] in MFe_2As_2 ($M = Ba, Ca$ and Sr). These measurements indicate that a T_c of 12 K, 27 K and 29 K can be achieved at high pressures in the parent compounds $CaFe_2As_2$, $SrFe_2As_2$ and $BaFe_2As_2$, respectively. Neutron powder diffraction measurements for $CaFe_2As_2$ have shown [4] that superconductivity occurs in a new phase, which is not magnetically ordered. It appears at pressures more than 3.5 kbar at $T=50$ K. The new phase remains tetragonal but the lattice parameters change dramatically at the transition. The structural transition [4, 8] causes a decrease in the c-axis lattice parameter by $\sim 10\%$ and an increase in the a-axis parameter by $\sim 2\%$.

The mechanism of superconductivity, and in particular, the role of lattice dynamics in superconducting pair formation in these newly discovered compounds is still to be settled. Meanwhile, it is necessary to study phonon dynamics carefully in these materials. This has motivated us to carry out measurements of the temperature dependence of the phonon dynamics in parent and superconducting FeAs compounds.

II. THEORY AND EXPERIMENT

The phonon frequencies as a function of wave vectors in the entire Brillouin zone have been calculated for MFe_2As_2 ($M = Ba, Ca, Sr$) and $MFeAsF$ ($M = Ca, Sr$) using quantum-mechanical ab-initio methods and semiempirical interatomic potentials. The details about the calculations are given in our previous publications [16–21]. Both magnetic (including the low-temperature magnetic ordering) and non-magnetic calculations are done to highlight the effect of magnetism on phonon spectra.

The parent and superconducting polycrystalline samples of MFe_2As_2 and $MFeAsF$ were prepared by heating stoichiometric mixtures of the corresponding purified elements. Structural analysis from x-ray powder diffraction indicates that the samples contain about 1% of FeAs as an impurity phase. The inelastic neutron scattering experiments were performed using the IN4C and IN6 time of flight spectrometers at the Institut Laue Langevin (ILL), France, as in our previous works. Measurements were made on about 10 grams of polycrystalline samples. We have used incident neutron wavelengths of 1.2 Å (56.8 meV) and 5.12 Å (3.12 meV) for measurements at IN4 and IN6, respectively. The high incident neutron wavelength of 5.12 Å at IN6 only allows measurements to be performed in neutron-energy gain. Hence data cannot be measured down to very low temperatures. The low incident neutron wavelength of 1.2 Å at IN4C allows the measurements of phonon spec-

tra in the energy loss mode. Hence data can be measured at very low temperatures of 2 K. The incoherent approximation [27] has been used for extracting neutron-weighted phonon density of states from the measured scattering function $S(Q, E)$.

III. RESULTS AND DISCUSSION

We have carried out extensive measurements of the phonon density of states for parent and superconducting MFe_2As_2 and $MFeAsF$ compounds. The results obtained from our studies are summarized below:

Parent and superconducting MFe_2As_2 compounds

A. Comparison of phonon spectra among various MFe_2As_2 ($M = Ba, Ca, Sr$) compounds

All the 122 compounds have nearly the same value of the lattice parameter a (~ 3.89 Å), while the lattice parameter c in Ca, Sr and Ba compounds is 11.758 Å, 12.37 Å and 13.04 Å, respectively. We note that the lattice constant c decreases monotonically from $BaFe_2As_2$ to $SrFe_2As_2$ to $CaFe_2As_2$, as one would expect when given the ionic radii: $Ca^{2+} < Sr^{2+} < Ba^{2+}$.

The high-resolution phonon density of states measured for the AFe_2As_2 ($A = Ba, Ca, Sr$) compounds using IN6 at 300 K are shown in Fig. 1(a). The phonon density of states for Ba, Sr and Ca compounds show pronounced differences below 22 meV. It seems there is no effect of change in A (Ba, Ca, Sr) atom on the 25-meV peak. However, as expected, the Fe-As stretching modes around 32 meV shift towards lower energies in the order of Ba, Sr and Ca compounds due to the increase in the Fe-As bond lengths. Considering the large difference in the masses of Ba ($m = 137.34$ amu), Sr ($m = 87.62$ amu) and Ca ($m = 40.08$ amu), the mass effect and the contraction of the unit cell should result in shifting of the phonon modes in the Ca compound to higher energies in comparison to the Ba and Sr cases. However, we find that the peaks at about 12 meV and 20 meV in the Ba and Sr compounds, respectively, are shifted to 10 meV and 19 meV in the Ca compound.

In order to understand this difference, we have calculated the partial density of states (Fig. 2) of various atoms in $BaFe_2As_2$, $SrFe_2As_2$ and $CaFe_2As_2$, where the magnetic interactions are considered. We find that the vibrational modes due to A (Ba, Sr, Ca) atoms in AFe_2As_2 scale approximately in the ratio as expected from the masses of the Ba, Sr and Ca atoms. Furthermore, as expected, the Fe and As vibrations in Ba and Sr compounds are similar. However, we observe that there is a substantial difference in the vibrations of Fe and As atoms in Ca compounds in comparison to Ba and Sr compounds. The calculated Fe and As vibrations are found to soften in the range below 20 meV by about 1 to 2 meV in the Ca compound compared to the Ba and Sr compounds. This is contrary to our expectation. The softening of the rest of the Fe and As vibrations is found to be responsible for the softening of the phonon spectra (Fig. 2) below 22 meV as seen in $CaFe_2As_2$. As expected the Fe-As stretching modes around 32 meV in the Ca compound shift towards

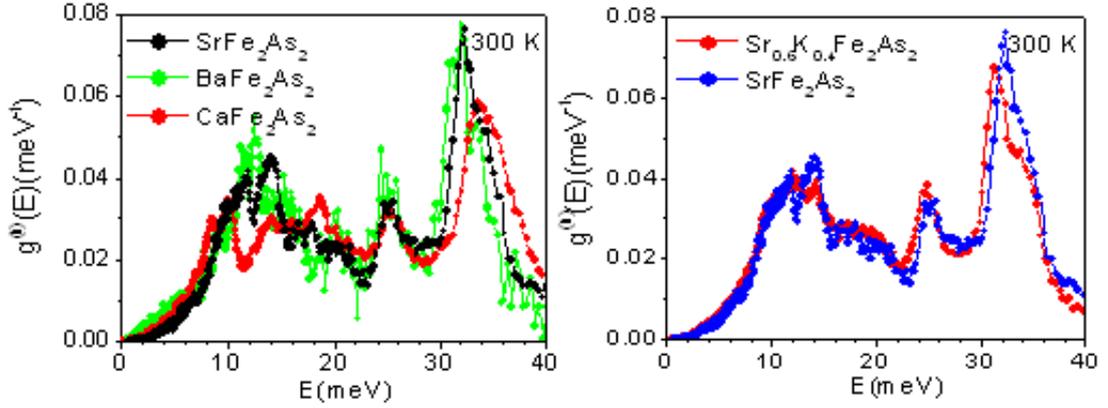


FIG. 1: (a) The comparison of the experimental phonon spectra for (a) $M\text{Fe}_2\text{As}_2$ ($M = \text{Sr}, \text{Ba}, \text{Ca}$) and (b) SrFe_2As_2 and $\text{Sr}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$. The phonon spectra are measured with an incident neutron wavelength of 5.12 \AA using the IN6 spectrometer at the ILL. All the phonon spectra are normalized to unity.

higher energies.

The phonon spectra for both the parent and superconducting Sr have been shown in Fig. 1(b). Both compounds have nearly the same value of the lattice parameter a ($\sim 3.89 \text{ \AA}$), while the lattice parameter c in SrFe_2As_2 and $\text{Sr}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ is 12.37 \AA and 12.923 \AA , respectively. The doped system has slightly longer Sr-As and Fe-As bond lengths that would result in a softening of the phonon modes above 22 meV .

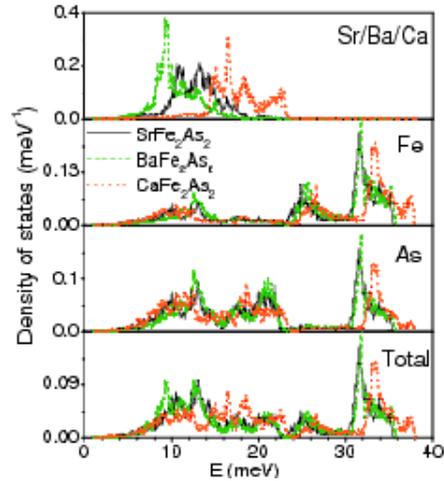


FIG. 2: Calculated partial density of states for the various atoms in $M\text{Fe}_2\text{As}_2$ ($M = \text{Sr}, \text{Ba}, \text{Ca}$) using ab-initio magnetic calculations. The spectra are normalized to unity.

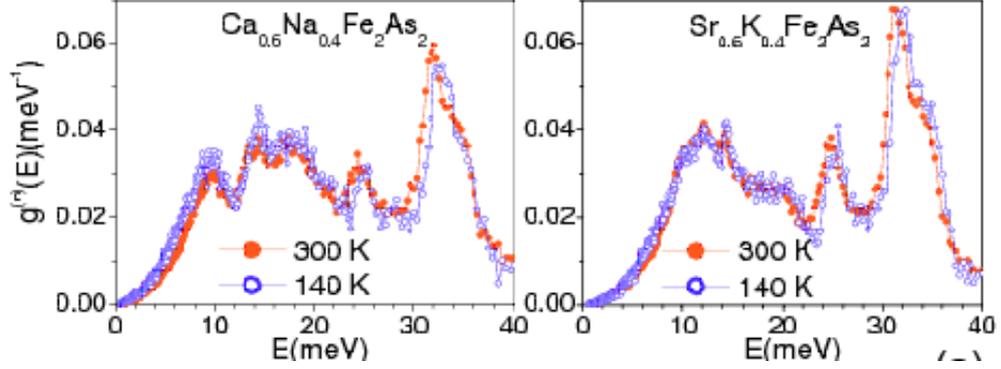


FIG. 3: The experimental phonon spectra [18] of $\text{Sr}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ and $\text{Ca}_{0.6}\text{Na}_{0.4}\text{Fe}_2\text{As}_2$ measured with incident neutron energy of 3.12 meV using IN6 spectrometer at ILL.

B. Temperature dependence of phonon spectra for MFe_2As_2

The high-resolution data collected for superconducting $\text{Ca}_{0.6}\text{Na}_{0.4}\text{Fe}_2\text{As}_2$ and $\text{Sr}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ using the IN6 spectrometer show softening of phonon modes (Fig. 3) below 10 meV as we decrease the temperature from 300 K to 140 K. Softening of low-energy phonons is larger for Ca compound (about 1 meV) in comparison to that in the Sr compound (about 0.5 meV). These phonon modes below 10 meV arise from the atomic vibrations of Ca/Na or Sr/K atoms. The higher softening in the CaNa compound may indicate that electron-phonon coupling is stronger than for SrK. The phonon spectrum for CaFe_2As_2 also shows the softening of the low-energy modes by about 1 meV when decreasing the temperature from 300 K to 180 K. There is no appreciable change in the phonon density of states across the structural and anti-ferromagnetic phase transition at 172 K. As the tetragonal to orthorhombic phase transition is suppressed in the superconducting compounds, the structure phase transition does not appear to be relevant for the observed phonon softening. The measurement of temperature dependence of phonon spectra in SrFe_2As_2 shows that the low-energy phonon modes do not vary with the change in temperature from 2 K to 300 K. These results indicate that the softening of low-energy phonon modes in these compounds may be due to the interaction of phonons with electron or short-range spin fluctuations in the normal state of the superconducting compound as well as in the parent compound.

Parent and superconducting MFeAsF ($\text{M} = \text{Ca}, \text{Sr}$) compounds

We have measured the composition as well as temperature dependence of the phonon density-of-states in FeAs compounds with composition $\text{CaFe}_{1-x}\text{Co}_x\text{AsF}$ ($x = 0, 0.06, 0.12$). The electronic structure calculations [13] for these compounds show that bands near the Fermi level are mainly formed by Fe 3d states, which are quite different from other 122 and 1111 FeAs compounds, where both Fe and As are believed to be related to superconductivity. The difference in electronic structure for fluorine-based compounds may cause phonon spectra to behave differently as a function of composition and temperature in comparison

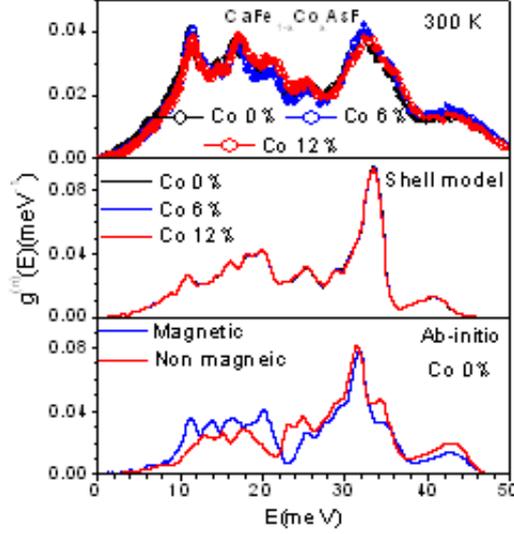


FIG. 4: Comparison of experimental phonon spectra for $\text{CaFe}_{1-x}\text{Co}_x\text{AsF}$ ($x = 0, 0.06, 0.12$). The phonon spectra are measured with incident neutron wavelength of 5.12 \AA using the IN6 spectrometer at ILL. The calculated phonon spectra using the shell model and ab-initio are also shown. The calculated spectra have been convoluted with a Gaussian of FWHM of 5% of the energy transfer in order to describe the effect of energy resolution in the experiment.

with our phonon studies on parent and superconducting MFe_2As_2 ($M = \text{Ba}, \text{Ca}, \text{Sr}$). The composition as well as the temperature dependence of phonon spectra for $\text{CaFe}_{1-x}\text{Co}_x\text{AsF}$ ($x = 0, 0.06, 0.12$) compounds have been measured using time of flight IN4C and IN6 spectrometers at ILL, France.

A. Comparison of phonon spectra among various $\text{CaFe}_{1-x}\text{Co}_x\text{AsF}$ ($x = 0, 0.06, 0.12$) compounds

The high-resolution density of states measurements for $\text{CaFe}_{1-x}\text{Co}_x\text{AsF}$ compounds ($x = 0, 0.06, 0.12$) measured at 300 K using the IN6 spectrometer are shown in Figure 4. The structural and magnetic transition in these compounds is below 134 K. The measurements on IN6 can only be performed with incident neutron energy of 3.12 meV in the neutron-energy gain mode, which does not give enough intensity at low temperatures. We have carried out density of states measurements only at 300 K. These measurements, carried out with very high elastic energy resolution of about $200 \mu\text{eV}$ (inelastic focusing mode), show that low-energy phonon modes below 12 meV harden on doping of 6% of Co at the Fe site (Fig. 4). A further increase in the Co concentration to 12% does not seem to affect the low-energy phonon spectra. However, the intermediate energy phonon modes from 18 meV to 25 meV also harden in addition to the acoustic modes on 12% Co doping in comparison to the parent and 6% Co doped compounds.

The calculated partial density of states (Fig. 7, below) of various atoms in

$\text{CaFe}_{1-x}\text{Co}_x\text{AsF}$ compounds ($x=0, 0.06, 0.12$) show that at low energies below 12 meV, the contribution to phonon spectra is mainly from the $\text{Fe}_{1-x}\text{Co}_x$ or As sub lattice. The shell model calculation, which does not consider the effect of electron-phonon coupling influence on the phonon spectra, shows that the partial substitution of Co at the Fe site has little effect on the phonon spectra.

Electronic structure calculations [13] show that for CaFeAsF bands near the Fermi level are mainly formed by Fe 3d states. The substitutions of Co at the Fe site would result in an increase in electrons in the doped compound in comparison to the parent compound. Indeed, there are seven electrons in the d-shell of the Co^{+2} cation, whereas the valence d-shell of Fe^{+2} consists of six electrons. Thus, the partial substitution of Co by Fe would be reflected by an increase in the electronic density of states of the $\text{Fe}_{1-x}\text{Co}_x$ sub-lattice at the Fermi level. The changes in the electronic system upon doping may be responsible for the hardening of the low-energy modes. The hardening can thus be taken as an indication for coupling between the electrons and the phonons (Fig. 4). Further increase in Co doping to 12% would further perturb the electronic density of states of $\text{Fe}_{1-x}\text{Co}_x$ at the Fermi level. The additional increase in strength of the electronic density of states may cause intermediate energy phonons to interact with the electrons and in turn result in the hardening of intermediate energy modes (Fig. 4).

The peak in the phonon spectra resulting from the stretching vibrations of Fe-As modes remains centered at about 32 meV in all the $\text{CaFe}_{1-x}\text{Co}_x\text{AsF}$ compounds ($x=0, 0.06, 0.12$). This indicates that Fe-As bond lengths are not affected by partial substitution of Fe by Co atoms. This is what we also observe from the analysis of neutron diffraction [22] on these compounds. However, the peak at about 32 meV is found to be slightly broader in Co-doped compounds in comparison of the parent compound. The slight distribution of Fe/Co-As bond lengths would broaden the 32 meV peak in Co doped compounds.

B. Temperature dependence of phonon spectra for SrFeAsF

SrFeAsF is the parent compound of the Sm-doped counterpart $\text{Sr}_{0.5}\text{Sm}_{0.5}\text{FeAsF}$ [23] having a very high T_c of 56 K. Our neutron diffraction measurements show that the parent compound SrFeAsF undergoes [24] a tetragonal to orthorhombic phase transition at 180 K followed by the magnetic phase transition at 133 K. Figure 5 shows the temperature dependence of the measured phonon spectra of SrFeAsF at 160 K, 200 K and 300 K using IN6, and at 2 K, 160 K and 200 K using IN4C. Phonon modes are expected to shift towards higher energies with decrease in the unit cell volume with the temperature. The phonon spectrum from 22 meV to 35 meV, including the peak at 32 meV due to stretching modes of Fe-As, reacts to the temperature change and moves slightly to higher energies with a decrease in the temperature. However our measurements show that the temperature variation across the tetragonal to orthorhombic phase transition or magnetic phase transition has little effect on the phonon dynamics of the parent compounds.

The comparison between the experimental phonon spectra and ab-initio calculations for CaFeAsF and SrFeAsF are shown in Figs. 4 and 5, respectively. The observed features in the experimental data are well reproduced computationally when magnetism is considered. Both the peak positions and intensities are significantly improved, especially for the low-

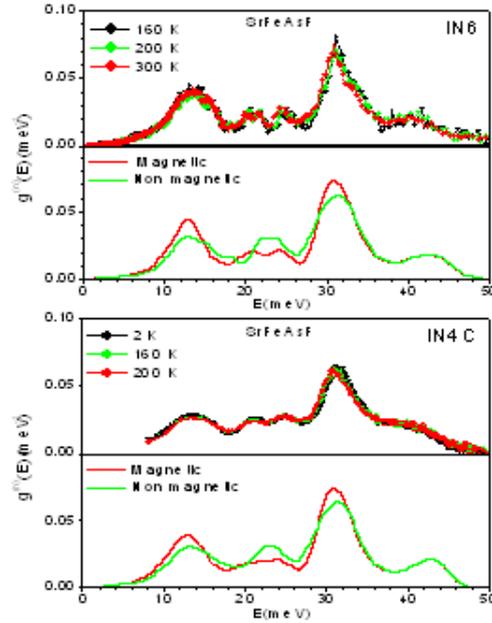


FIG. 5: The temperature dependence of experimental phonon spectra for SrFeAsF. The phonon spectra are measured with incident neutron wavelength of 5.12 Å and 1.2 Å using the IN6 and IN4C spectrometers at ILL, respectively. The calculated phonon spectra using ab-initio magnetic lattice dynamics are also shown. The calculated spectra have been convoluted with a Gaussian of FWHM of 10% of the energy transfer in order to describe the effect of energy resolution in the experiment carried out using the IN6 spectrometer. For comparison with the experiment carried out using IN4C spectrometer, the calculated spectra have been convoluted with a Gaussian of FWHM of 3 meV.

frequency and mid-frequency ranges. In SrFeAsF, the modes within the range 20-30 meV involving Fe and As (Fig. 6) are well resolved and match better the observations when magnetic interactions are included. Similarly, the low-lying 0-12 meV frequency range is well described in CaFeAsF through a correct E^2 -dependence in the magnetic calculations. This is also the case for the intensity profile in 22 to 26 meV. Furthermore for both the systems, the stretching modes due to F (shoulder around 45 meV) (Fig. 6) are found in the correct frequency position, indicating a good description of both structural and chemical interactions involving the lightest element F.

C. Comparison of phonon spectra among various MFeAsF (M = Ca, Sr) compounds

Now we compare the high-resolution phonon spectra of SrFeAsF and CaFeAsF measured using the IN6 spectrometer. The comparison of the phonon spectra of SrFeAsF and CaFeAsF (Fig. 7) show pronounced differences in the full spectral range. At room temperature both compounds crystallize in a tetragonal structure (space group P4/nmm). The unit cell volume of CaFeAsF ($a=3.8859$ Å, $c=8.595$ Å) is about 10% smaller in comparison

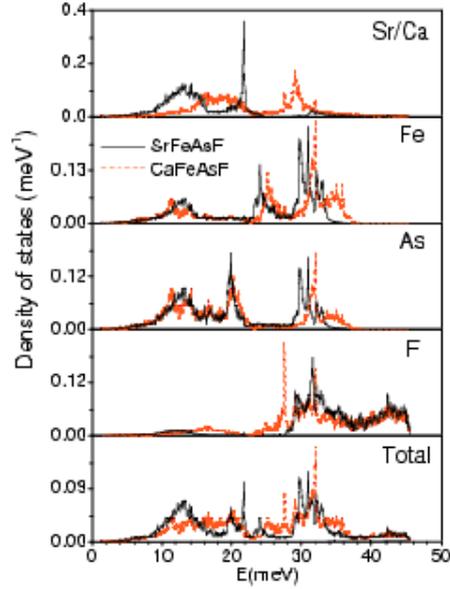


FIG. 6: Calculated partial density of states for the various atoms in $M\text{FeAsF}$ ($M = \text{Ca}, \text{Sr}$) using ab-initio magnetic calculations. The spectra are normalized to unity.

to SrFeAsF ($a = 3.9996 \text{ \AA}$, $c = 8.9618 \text{ \AA}$). It is not possible to attribute these changes to a simple change in unit cell volume and mass renormalization of the modes involving the Sr ($m = 87.62 \text{ amu}$) and Ca ($m = 40.08 \text{ amu}$) atoms. Qualitatively, our data particularly show that the peaks at 10 meV and 16 meV in CaFeAsF are combined together in one peak centered at about 14 meV in SrFeAsF . Consequently a gap is opened up in the phonon density of states of SrFeAsF at about 18 meV. Another important change in the phonon spectra is the intensity of the peak at 32 meV. The peak has very high intensity in SrFeAsF compared to CaFeAsF . Further, the peak centered at about 45 meV in CaFeAsF is shifted to about 40 meV in SrFeAsF .

The calculated partial density of states (Fig. 6) show that Ca and Sr vibrations in SrFeAsF and CaFeAsF scale approximately with the ratio expected due to the mass effect. We find that there is a significant difference in the atomic vibrations of F. The partial density of states of F atoms in both compounds extends up to full spectral range of 45 meV. The structure of these compounds has FeAs_4 and $\text{F}(\text{Sr}/\text{Ca})_4$ tetrahedral units. The mass normalization shifts Sr vibrations down to lower energies, which, in turn, affects the F vibrations. In particular, a peak at 15 meV in the F partial density of states in CaFeAsF is shifted to 12 meV in SrFeAsF .

The reason that mass and unit cell contraction effects are not able to explain the difference in phonon spectra may be due to the fact that the unit cell contraction in these compounds is accompanied by a substantial change [25, 26] in the free structural parameter of the As atom.

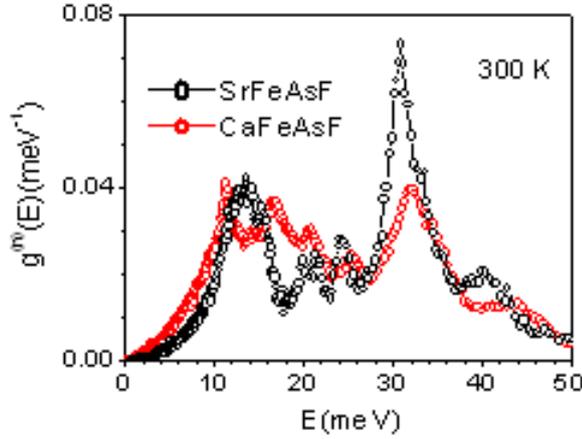


FIG. 7: The comparison of the experimental phonon spectra for SrFeAsF and CaFeAsF. The phonon spectra are measured with an incident neutron wavelength of 5.12 Å using the IN6 spectrometer at the ILL. The experimental phonon data for CaFeAsF are taken from Ref. [21]. All the phonon spectra are normalized to unity.

IV. CONCLUSIONS

We have carried out systematic studies of the temperature dependence of phonon spectra for the parent compounds MFe_2As_2 and $MFeAsF$. We found that in case of Sr parent compounds, the low-energy phonon modes do not vary with the change of temperature from 2 K to 300 K. This is contrary to our observation of phonon softening in $CaFe_2As_2$ - and $Ca_{0.6}Na_{0.4}Fe_2As_2$ - based compounds. The low instability pressure in Ca compounds coupled with the strong spin fluctuations may be responsible for the large softening of low energy phonon modes in both the parent and superconducting Ca compounds. Further, the comparison of the phonon spectra in different systems shows that as far as dynamics is concerned, the phonon spectra of Ba and Sr systems mimic each other. However, there are substantial differences when we compare the phonon spectra of Ba and Sr compounds with the Ca system. The mass and lattice contraction effects alone cannot explain these changes. These differences may be due to the fact that the unit cell contraction is accompanied by a substantial change in the free structural parameter of the As atom. The phonon spectra of $CaFe_{1-x}Co_xAsF$ compounds ($x=0, 0.06, 0.12$) strongly react to the amount of Co substitution in the parent compound. The renormalization of phonon modes is believed to be due to electron-phonon coupling. Our results then support coupling of electrons and phonons in Co-doped CaFeAsF compounds.

The comparison of experimental data and the calculations shows that magnetic interactions/ordering have to be considered in order to reproduce correctly the measured density of states. Further we have shown that in MFe_2As_2 and $MFeAsF$ compounds, the contributions from both Fe and As are similar, whereas the vibrations from M are quite different, which indicates that bonding of the M atom is quite different in MFe_2As_2 and

MFeAsF. This is due to the additional M-F interactions in MFeAsF, and consequently, the interaction between the M and Fe-As layer is also quite different compared to that in MFe₂As₂.

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