PHYSICAL REVIEW B **78**, 224518 (2008)

Phonon dynamics in $Sr_{0.6}K_{0.4}Fe_2As_2$ and $Ca_{0.6}Na_{0.4}Fe_2As_2$ from neutron scattering and lattice-dynamical calculations

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(Received 24 October 2008; revised manuscript received 17 November 2008; published 19 December 2008)

We report inelastic neutron scattering measurements of the phonon density of states of superconducting $Sr_{0.6}K_{0.4}Fe_2As_2$ ($T_c=32$ K) and $Ca_{0.6}Na_{0.4}Fe_2As_2$ ($T_c=21$ K). Compared with the parent compound $BaFe_2As_2$ doping affects mainly the lower and intermediate frequency part of the vibrations. Mass effects and lattice contraction cannot solely explain these changes. Softening of phonon modes below 10 meV has been observed in both samples on cooling from 300 to 140 K. In the Ca-doped compound the softening amounts to about 1 meV while for the Sr-doped compound the softening is about 0.5 meV. There is no appreciable change in the phonon density of states on crossing T_c.

DOI: 10.1103/PhysRevB.78.224518 PACS number(s): 74.25.Kc, 78.70.Nx, 63.20.−e

The recent discovery of superconductivity in Fe-As layered structure compounds has attracted a huge attention[1–20] in the scientific community. In general terms these systems can be classified in two different families. The first is derived from the parent compounds *R*FeAsO (*R*=La, Sm, Ce, Pr, Nd, and Gd) for which—depending on the lanthanide ion—T_c can be as high as 55 K on doping[2] with F at the O site. The highest T_c (56.5 K) for iron-based superconductors so far was achieved in $Gd_{1-x}Th_xOFeAs$ without F doping.[3] The second type of family is derived from parent compounds of the composition MFe_2As_2 (where M=Ba, Sr, Eu, and Ca). T_c values as high as 38 K are found[4] on partial substitution of Ba by K atoms. The difference between the two families is that whereas in MFe_2As_2 the Fe-As layers are separated by M atoms, in *R*FeAsO the separation is achieved by *R*-O layers.

Extensive efforts have been undertaken to raise T_c and to understand the mechanism of superconductivity in these compounds. Electronic structure calculations[9] show that in FeAs compounds the electronic bands around the Fermi level are formed mainly by Fe and As states, while the bands of La-O or M atoms are far from the Fermi level. It is therefore natural to believe that superconductivity in these compounds is due to the structural and electronic properties of the Fe-As layers. Recent inelastic neutron scattering measurements carried out on $BaFe_2As_2$ and $Ba_{0.6}K_{0.4}Fe_2As_2$ (Refs. 10 and 11) evidenced the presence of magnetic excitations. Therefore, spin fluctuations are believed to play an important role for the mechanism of superconductivity. In particular it has been calculated that phonons couple selectively to the spin system.[12] Therefore, despite the fact that simple electron-phonon coupling mechanisms[13] seem to be rather unlikely, it is important to investigate the phonon spectrum experimentally in order to clarify the role of phonon in the pairing mechanism of the electron in the superconducting phase. Electron-phonon coupling may be inferred via the changes in phonon lifetime and phonon energies.

Due to the greater polarizability of As ion in comparison to divalent oxygen, these compounds are expected to be more compressible than copper-based systems. The application of pressure is known to have important effects on the physical properties of these systems. In particular it has been shown that the structural and magnetic phase transitions that are known in these compounds[5–8] are suppressed under high pressure. These observations suggested that the structural phase transition to a collapsed tetragonal phase in MFe_2As_2 is related to superconductivity. The application of pressure results in a steep increase[5] in T_c in F-doped LaOFeAs from 27 to 43 K at 4 GPa. In $CaFe_2As_2$ pressure-induced superconductivity[6,7] was detected at a pressure onset of 0.35 GPa while superconductivity for $BaFe_2As_2$ and $SrFe_2As_2$ appears[8] at significantly higher pressures (3.8 and 3.2 GPa, respectively). An important effort on measuring and simulating the excitations in these systems has recently been done; in particular, the results of polarized Raman spectra of nonsuperconducting $CaFe_2As_2$ and $SrFe_2As_2$ and superconducting $Sr_{0.6}K_{0.4}Fe_2As_2$ have been communicated.[15] The lattice dynamics of $LaFeAsO_{1-x}F_x$, $PrFeAsO_{1-y}$, and NdFeAsO has also been investigated via inelastic x-ray scattering and first-principles calculations.[16] It has also been discovered that the experimental phonon data on LaFeAsO are not fully explained by *ab initio* calculations.[17]

In this paper, we explore these compounds using the techniques of inelastic neutron scattering and lattice-dynamical model calculations. Attention will be focused on electron-phonon coupling that should be detectable in the changes upon doping as well as in the temperature evolution of the spectra. The results presented here concerned the supercon-

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ducting phase of $Sr_{0.6}K_{0.4}Fe_2As_2$ and $Ca_{0.6}Na_{0.4}Fe_2As_2$. A large part of the discussion will be based on our earlier investigation[19] of the parent compound $BaFe_2As_2$, which we will use as a reference system.

The polycrystalline samples of $Sr_{0.6}K_{0.4}Fe_2As_2$ ($T_c = 32$ K) and $Ca_{0.6}Na_{0.4}Fe_2As_2$ ($T_c = 21$ K) were prepared[4,20] by heating stoichiometric mixtures of the corresponding purified elements. All samples were prepared in batches of 3–4.5 g and heated and annealed several times in sealed niobium tubes under an atmosphere of purified argon. After each annealing step, the mixtures were homogenized in an agate mortar and pressed into pellets before the last annealing step. The samples were heated to 1073–1173 K ($Sr_{0.6}K_{0.4}Fe_2As_2$) and 973–1073 K ($Ca_{0.6}Na_{0.4}Fe_2As_2$) in the different annealing steps and kept at these temperatures for 30–48 h. In the first step, the mixtures were heated very slowly in the temperature range from 573 to 873 K and kept at this temperature for 12 h in order to prevent undesirable reactions. The phase purity for the polycrystalline samples was checked by x-ray powder diffraction. Rietveld refinements were performed using the TOPAS ACADEMIC package.[21] For $Sr_{0.6}K_{0.4}Fe_2As_2$ sample we find that Fe impurity is about 6%, while for $Ca_{0.6}Na_{0.4}Fe_2As_2$ unknown impurity phases are estimated to be less than 10%. Both samples have body-centered-tetragonal cell and space group is determined to be $I4/mmm$.The a and c lattice parameters and the z parameter for the As atom are 3.886 Å (3.854 Å), 12.923 Å (12.066 Å), and 0.3577 (0.3669), respectively, for $Sr_{0.6}K_{0.4}Fe_2As_2$ ($Ca_{0.6}Na_{0.4}Fe_2As_2$). The refined Sr:K ratio of $Sr_{0.6}K_{0.4}Fe_2As_2$ is 0.59(1):41(1); the Ca:Na ratio of $Ca_{0.6}Na_{0.4}Fe_2As_2$ is 0.44(8):0.56(8). The superconducting character of the samples was checked by ac-susceptibility measurements under zero-field-cooled (shielding) and field-cooled (Meissner) cycles of finely ground powder samples using a superconducting quantum interference device (SQUID) magnetometer (MPMS-XL5, Quantum Design, Inc.) at 1.5 T. The samples showed superconducting transitions at 32 and 21 K for $Sr_{0.6}K_{0.4}Fe_2As_2$ and $Ca_{0.6}Na_{0.4}Fe_2As_2$, respectively.

The inelastic neutron scattering experiments were performed using the IN4C and IN6 time-of-flight spectrometers at the Institut Laue Langevin (ILL), France. The measurements were performed on about 8 g of polycrystalline samples. On IN4C, the measurements were done in the neutron-energy-loss mode using an incident neutron wavelength of 1.18 Å at a temperature below (2.5 K) and above (50 K) the superconducting transition. In this configuration the elastic energy resolution of the spectrometer is about 3 meV. The detector bank covered scattering angles from 13° to 120°. For the IN6 measurements, an incident neutron wavelength of 5.1 Å was used. The measurements were performed in the neutron-energy-gain mode and the data were collected at 140 and 300 K. In this configuration the energy resolution of the spectrometer is 0.07 meV at elastic position so that a higher resolution is achieved in the low-frequency range with regard to IN4C data. The angular range of the spectrometer covers 10°–113°. The scattering from a standard vanadium sample was used to calibrate the detectors both on IN4C and IN6. In the incoherent one-phonon approximation the measured scattering function $S(Q,E)$, as observed in the neutron experiments, is related to the phonon density of states[22] as follows:

$$g^{(n)}(E) = A\left\langle \frac{e^{2W_k(Q)}}{Q^2} \frac{E}{n(E,T) + \frac{1}{2} \pm \frac{1}{2}} S(Q,E) \right\rangle, \tag{1}$$

$$g^{n}(E) = B\sum_k \left\{ \frac{4\pi b_k^2}{m_k} \right\} g_k(E), \tag{2}$$

where the + or − signs correspond to energy loss or gain of the neutrons, respectively, and where $n(E,T) = [\exp(E/k_B T) - 1]^{-1}$. A and B are normalization constants and b_k, m_k, and $g_k(E)$ are the neutron scattering length, mass, and partial density of states of the kth atom in the unit cell, respectively. The quantity within $\langle \cdots \rangle$ represents suitable average over all Q values at a given energy. $2W(Q)$ is the Debye-Waller factor. The weighting factors $\frac{4\pi b_k^2}{m_k}$ for various atoms in the units of barns/amu are Sr: 0.071; K: 0.050; Na: 0.143; Ca: 0.071; Fe: 0.208; and As: 0.073.

The experimental $S(Q,E)$ measured for $Sr_{0.6}K_{0.4}Fe_2As_2$ and $Ca_{0.6}Na_{0.4}Fe_2As_2$ using the IN4C spectrometer are shown in Fig. 1. In contrast to recent reports on similar measurements[11] carried out on powder samples of $Ba_{0.6}K_{0.4}Fe_2As$ using the MERLIN spectrometer at ISIS, our measurements do not show any clear signature from resonant spin excitation. However, the (Q,E) range attainable using IN4C is reduced compared to that of MERLIN at low-Q values. This added to a lower signal-to-noise ratio on IN4C (MERLIN is known to posses a particularly high ratio) can prevent detecting this weak feature. Therefore, while it would be tempted to conclude that such excitation is absent in the data reported in this paper, we feel that further investigations might be necessary for being certain.

The generalized phonon density of states obtained from the $S(Q,E)$ for both compounds at 2.5 and 50 K are shown in Fig. 2. One clearly observes a strong similarity of the generalized density of states measured above and below the superconducting transition temperature for both compounds. This suggests that the formation of Cooper pairs have only a very minor influence on the overall vibration spectrum.

The lattice-dynamical calculation procedure follows closely the one that we previously reported for $BaFe_2As_2$.[19] The structure parameters that we used were those reported in Ref. 20. The semiempirical interatomic potential consists of Coulombic and short-range terms plus a van der Waals attractive interaction between the oxygen atoms. The polarizability of the As atom was accounted for in the framework of the shell model. The parameters of the interatomic potential are the effective charge and the radius of the atoms. The choice of the radii parameter corresponding to Sr/K and Ca/Na was done such that the potential satisfies the conditions of static and dynamic equilibrium.[23,24] The radii parameters for Sr/K and Ca/Na site atoms are $R(\mathrm{Sr/K}) = 2.11$ Å and $R(\mathrm{Ca/Na}) = 2.08$ Å. All other parameters of the potential are the same as those reported for $BaFe_2As_2$. The calculations have been carried out using the current version of the software DISPR developed at Trombay.

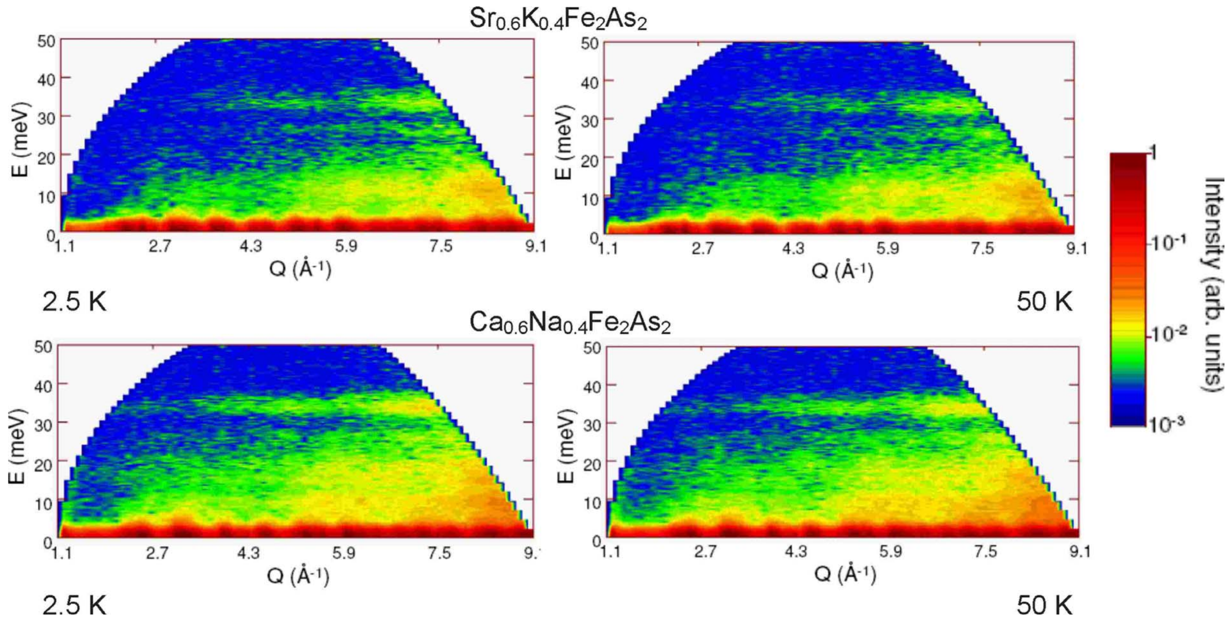

FIG. 1. (Color online) The experimental $S(Q,E)$ plots for $Ca_{0.6}Na_{0.4}Fe_2As_2$ and $Sr_{0.6}K_{0.4}Fe_2As_2$ at 2.5 and 50 K measured using the IN4C spectrometer at the ILL with an incident neutron wavelength of 1.18 Å. The values of $S(Q,E)$ are normalized to the mass of sample in the beam. For clarity, a logarithmic representation is used for the intensities.

To simulate the nonstochiometry arising from the doping the calculations are carried out for a $2\times2\times2$ supercell, where 40% of the $M(\mathrm{Sr,Ca})$ atoms are randomly replaced by either K or Na atoms in SrK and CaNa compounds, respectively. The calculated phonon spectra are also shown in Fig. 2. While the calculated spectra compare reasonably well with the experimental data from IN4C at high frequency, there are many discrepancies at energies around 20 meV in Sr and Ca compounds. This reflects the difficulties already encountered when simulating the dynamics of these systems with one or more extra peak in this energy range. Even more sophisticated techniques such as *ab initio* phonon calculations fail to perfectly reproduce the experimental phonon spectra[12,17] of LaFeAsO and $BaFe_2As_2$ (Refs. 12 and 17) in this energy range. As an example, the three-peak structure observed in the experimental[17] phonon density of states of LaFeAsO between 20 to 50 meV is not reproduced by *ab initio* calculations which predict only one peak. Despite these disagreements, the calculations are useful for identifying the dynamical contributions to frequency distribution arising from the various species of atoms in the SrK and CaNa compounds. The partial densities of states (Fig. 3) give the dynamical contributions to the frequency distribution arising from the various species of atoms. The Sr/K and Ca/Na atoms mainly contribute in the 0–25 meV range, while the As and Fe atoms contribute in the whole 0–40 meV range. Above 30 meV the contributions are mainly due to Fe-As stretching modes. Below 15 meV the contributions arise

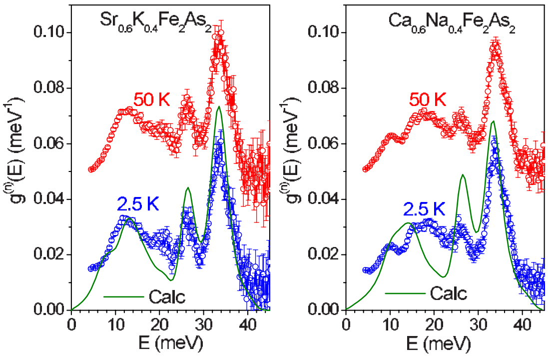

FIG. 2. (Color online) Comparison between the calculated and experimental phonon spectra of $Sr_{0.6}K_{0.4}Fe_2As_2$ and $Ca_{0.6}Na_{0.4}Fe_2As_2$. The measurements are carried out with incident neutron wavelength of 1.18 Å using the IN4C spectrometer at the ILL. For better visibility the experimental phonon spectra at 50 K are shifted along the y axis by 0.04 meV^{-1}. The calculated spectra have been convoluted with a Gaussian of full width at half maximum (FWHM) of 3 meV in order to describe the effect of energy resolution in the experiment.

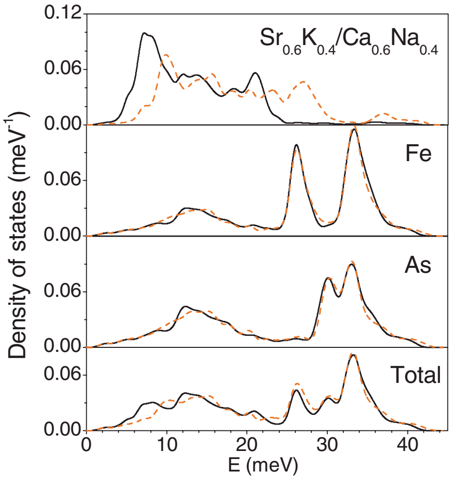

FIG. 3. (Color online) Calculated partial density of states. Solid and dashed lines correspond to $Sr_{0.6}K_{0.4}Fe_2As_2$ and $Ca_{0.6}Na_{0.4}Fe_2As_2$, respectively.

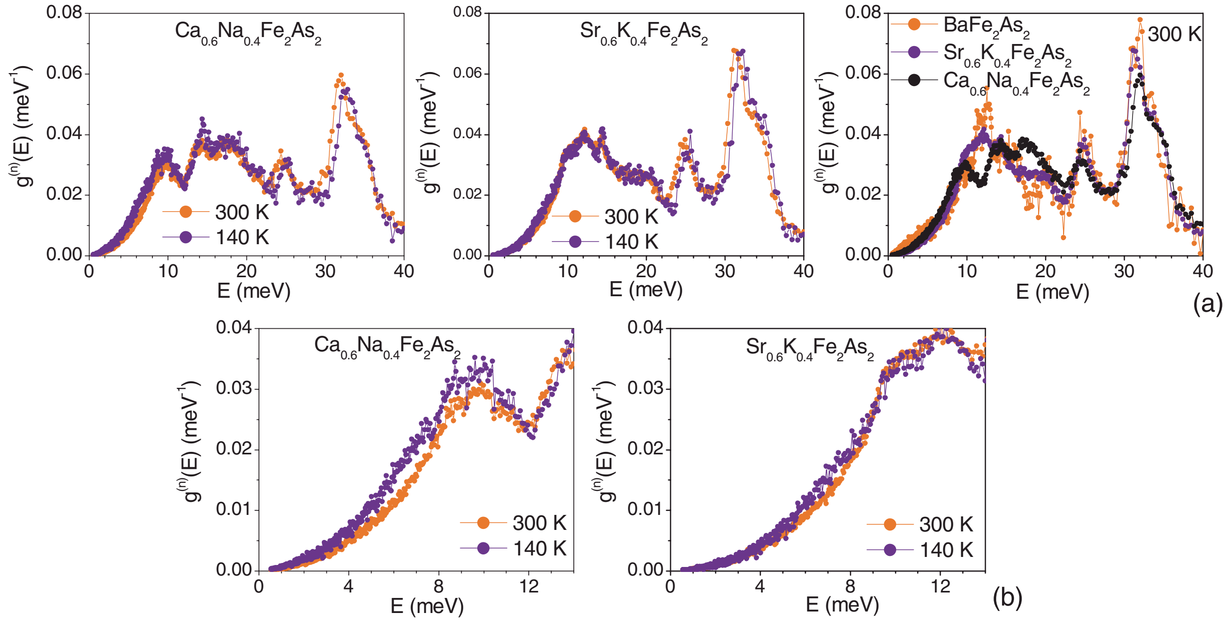

FIG. 4. (Color online) (a) The experimental phonon spectra of $Sr_{0.6}K_{0.4}Fe_2As_2$, $Ca_{0.6}Na_{0.4}Fe_2As_2$, and $BaFe_2As_2$ measured with incident neutron wavelength of 5.12 Å using IN6 spectrometer at ILL. The experimental phonon data for $BaFe_2As_2$ are taken from Ref. 12. All the phonon spectra are normalized to unity. (b) Zoom of the low-energy part.

mainly from Ca/Na and Sr/K atoms. The first low-energy peak in the partial density of states (Fig. 3) of Ca/Na and Sr/K is at about 8 meV.

The superconducting properties of MgB_2 with a T_c of 38 K, i.e., similar to that of the pnictide superconductors, have been explained in the framework of standard electron-phonon coupling. In this compound, the phonons mediating the pairing are of very high energy. Their involvement in superconductivity is reflected in the strong change they experience on doping.[25] In the present case the high-frequency band at 34 meV reacts relatively weak to doping and the small changes observed are more likely related to minor variations in the bond lengths. Therefore, despite the fact that a BCS-like gap has been found[18] in the superconductor $SmFeAsO_{0.85}F_{0.15}$ by Andreev spectroscopy, the high T_c of the pnictides seems incompatible with standard electron-phonon coupling alone.

We have completed the phonon density-of-states measurements at low temperatures with measurements of higher resolution in the low-frequency range (0–20 meV) using the IN6 spectrometer with an incident wavelength of 5.1 Å (Fig. 4). Due to their nature (up scattering) these measurements are limited to higher temperatures. The phonon densities of states for Ba, CaNa, and SrK compounds (Fig. 4) measured at 300 K using IN6 show pronounced differences in the lower half of the spectral range. It is not evident to attribute these changes to a simple mass renormalization of the modes involving Ba(m=137.34 amu), Sr(m=87.62 amu), and Ca(m=40.08 amu). Qualitatively our data show in particular that the peak found at about 21 meV in Ba and Sr compounds has been shifted to a lower energy of about 17.5 meV in the Ca compound. If we compare the unit-cell dimensions of CaNa, Ba, and SrK compounds, then we notice that all three compounds feature nearly the same values for the lattice parameter a, while the lattice parameter c in the CaNa compound is about 10% shorter in comparison to that of the Ba and SrK compounds. This implies that the M-As (M =Ca/Na) and Fe-As bond lengths are slightly reduced in the CaNa compound with regard to the other. This contraction of the unit cell and/or in the bond lengths should normally result in shifting the modes, if at all, to higher energies. The fact that the contrary is observed indicates that the bonding scheme is different for the CaNa compound. The buffer layers are thus not just charge reservoirs. The unit-cell contraction is accompanied by a substantial change in the free structural parameter of As (from z=0.3577 to z=0.3669). It is known from density-functional calculations that this parameter has a crucial influence on the binding and in particular on the phonon frequencies[12] in $BaFe_2As_2$. It may be speculated that it equally has substantial responsibility for the changes observed here. A clarification of this point has to await *ab initio* calculations for the doped compounds.

Apart from the doping dependence the temperature dependence of the phonon spectra may give valuable insight into the dynamics of a superconductor. Usually phonon modes are found to shift toward higher energies with a decrease in the unit-cell volume with decreasing temperature. This is actually what we observe for the high-frequency band centered on 34 meV when going from 300 to 140 K. It can be seen that at 140 K the band around 25 and 34 meV are narrower and shifted to higher energies in comparison to the data at 300 K. Contrary to the high-frequency bands the low-energy phonon modes up to about 10 meV soften (Fig. 4) for both the CaNa and SrK compounds as we decrease the temperature from 300 to 140 K. In the CaNa-doped compound the softening amounts to about 1 meV while for the SrK-doped compound the softening is about 0.5 meV. A similar softening of low-frequency modes has, e.g., been observed in superconducting[26] $LuNi_2B_2C$.

In general it is not easy to make a connection between temperature-induced softening and electron-phonon coupling. In the present case the behavior of the system under pressure may give an indication. Superconductivity in the parent MFe_2As_2 compounds emerges when there is suppression of the structural phase transition and its concomitant magnetic ordering. The large volume collapse[7] of $CaFe_2As_2$ at a temperature corresponding to its pressure-induced superconductivity indicates that the phase transition is electronic in origin. This strong correlation of electronic properties and structure would be compatible with a softening of low-energy phonon modes in CaNa and SrK compounds due to electron-phonon coupling (lowering of temperature corresponding structurally to an increase in pressure). The higher softening in the CaNa compound may indicate that electron-phonon coupling is stronger than for SrK. Since the phonon softening is observed only in the normal state of both superconducting samples, it is unlikely to be directly associated to superconductivity, and this is despite the isotope effect observed in pnictide superconductors.[14] As the tetragonal to orthorhombic phase transition is suppressed in the superconducting compounds, the structure phase transition also appears to be not relevant to the observed phonon softening.

In summary, we have reported an experimental phonon study of the $Sr_{0.6}K_{0.4}Fe_2As_2$ and $Ca_{0.6}Na_{0.4}Fe_2As_2$ superconducting compounds using inelastic neutron scattering and lattice dynamics calculations. We show that doping affects mainly the lower and intermediate frequency part of the vibrations. In particular the region around 20 meV that had already retained our attention in the parent $BaFe_2As_2$ shows a very strong renormalization. Mass effects and lattice contraction solely cannot explain these changes. Therefore, the type of buffer ion influences the bonding in the Fe-As layers. The buffers thus cannot be considered as a mere charge reservoir. The high-frequency band reacts moderately to the doping. In both compounds the low-energy phonon modes soften with temperature. This softening, which is stronger in the CaNa compound, might be due to electron-phonon coupling effects. No anomalous effects are observed in the phonon spectra when passing the superconducting transition temperature. All this indicates that while electron-phonon coupling is present it cannot be solely responsible for the electron pairing.

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[1] Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, J. Am. Chem. Soc. **130**, 3296 (2008).

[2] Z.-A. Ren, W. Lu, J. Yang, W. Yi, X.-L. Shen, Z.-C. Li, G.-C. Che, X.-L. Dong, L.-L. Sun, F. Zhou, and Z.-X. Zhao, Chin. Phys. Lett. **25**, 2215 (2008).

[3] C. Wang, L. Li, S. Chi, Z. Zhu, Z. Ren, Y. Li, Y. Wang, X. Lin, Y. Luo, S. Jiang, X. Xu, G. Cao, and Z. Xu, Europhys. Lett. **83**, 67006 (2008).

[4] M. Rotter, M. Tegel, and D. Johrendt, Phys. Rev. Lett. **101**, 107006 (2008).

[5] H. Takahashi, K. Igawa, K. Arii, Y. Kamihara, M. Hirano, and H. Hosono, Nature (London) **453**, 376 (2008).

[6] M. S. Torikachvili, S. L. Bud'ko, N. Ni, and P. C. Canfield, Phys. Rev. Lett. **101**, 057006 (2008).

[7] A. Kreyssig, M. A. Green, Y. Lee, G. D. Samolyuk, P. Zajdel, J. W. Lynn, S. L. Bud'ko, M. S. Torikachvili, N. Ni, S. Nandi, J. Leão, S. J. Poulton, D. N. Argyriou, B. N. Harmon, P. C. Canfield, R. J. McQueeney, and A. I. Goldman, Phys. Rev. B **78**, 184517 (2008).

[8] P. L. Alireza, J. Gillett, Y. T. Chris Ko, S. E. Sebastian, and G. G. Lonzarich, J. Phys.: Condens. Matter **21**, 12208 (2009).

[9] D. J. Singh, Phys. Rev. B **78**, 094511 (2008).

[10] R. A. Ewings, T. G. Perring, R. I. Bewley, T. Guidi, M. J. Pitcher, D. R. Parker, S. J. Clarke, and A. T. Boothroyd, Phys. Rev. B **78**, 220501(R) (2008).

[11] A. D. Christianson, E. A. Goremychkin, R. Osborn, S. Rosenkranz, M. D. Lumsden, C. D. Malliakas, L. S. Todorov, H. Claus, D. Y. Chung, M. G. Kanatzidis, R. I. Bewley, and T. Guidi, arXiv:0807.3932 (unpublished).

[12] M. Zbiri, H. Schober, M. R. Johnson, S. Rols, R. Mittal, Y. Su, M. Rotter, and D. Johrendt, arXiv:0807.4429 (unpublished).

[13] L. Boeri, O. V. Dolgov, and A. A. Golubov, Phys. Rev. Lett. **101**, 026403 (2008).

[14] R. H. Liu, T. Wu, G. Wu, H. Chen, X. F. Wang, Y. L. Xie, J. J. Yin, Y. J. Yan, Q. J. Li, B. C. Shi, W. S. Chu, Z. Y. Wu, and X. H. Chen, arXiv:0810.2694 (unpublished).

[15] A. P. Litvinchuk, V. G. Hadjiev, M. N. Iliev, B. Lv, A. M. Guloy, and C. W. Chu, Phys. Rev. B **78**, 060503 (2008); K.-Y. Choi, D. Wulferding, P. Lemmens, N. Ni, S. L. Budko, and P. C. Canfield, arXiv:0810.2040 (unpublished).

[16] T. Fukuda, A. Q. R. Baron, S. Shamoto, M. Ishikado, H. Nakamura, M. Machida, H. Uchiyama, S. Tsutsui, A. Iyo, H. Kito, J. Mizuki, M. Arai, H. Eisaki, and H. Hosono, J. Phys. Soc. Jpn. **77**, 103715 (2008); M. Le Tacon, M. Krisch, A. Bosak, J. W. G. Bos, and S. Margadonna, Phys. Rev. B **78**, 140505(R) (2008).

[17] A. D. Christianson, M. D. Lumsden, O. Delaire, M. B. Stone, D. L. Abernathy, M. A. McGuire, A. S. Sefat, R. Jin, B. C. Sales, D. Mandrus, E. D. Mun, P. C. Canfield, J. Y. Y. Lin, M. Lucas, M. Kresch, J. B. Keith, B. Fultz, E. A. Goremychkin, and R. J. McQueeney, Phys. Rev. Lett. **101**, 157004 (2008).

[18] T. Y. Chen, Z. Tesanovic, R. H. Liu, X. H. Chen, and C. L. Chien, Nature (London) **453**, 1224 (2008).

[19] G. Catelani, X. S. Wu, and P. W. Adams, Phys. Rev. B **78**, 104515 (2008).

[20] K. Sasmal, B. Lv, B. Lorenz, A. M. Guloy, F. Chen, Y. Y. Xue, and C. W. Chu, Phys. Rev. Lett. **101**, 107007 (2008); G. Wu, H. Chen, T. Wu, Y. L. Xie, Y. J. Yan, R. H. Liu, X. F. Wang, J. J. Ying, and X. H. Chen, J. Phys.: Condens. Matter **20**, 422201 (2008).

[21] A. A. Coelho, TOPAS ACADEMIC 4.1, 2007.

[22] D. L. Price and K. Skold, in *Neutron Scattering*, edited by K. Skold and D. L. Price (Academic, Orlando, 1986), Vol. A; J. M. Carpenter and D. L. Price, Phys. Rev. Lett. **54**, 441 (1985); S. Rols, H. Jobic, and H. Schober, C. R. Phys. **8**, 777 (2007).

[23] R. Mittal, S. L. Chaplot, and N. Choudhury, Prog. Mater. Sci. **51**, 211 (2006).

[24] S. L. Chaplot, N. Choudhury, S. Ghose, M. N. Rao, R. Mittal, and K. N. Prabhatasree, Eur. J. Mineral. **14**, 291 (2002).

[25] B. Renker, K. B. Bohnen, R. Heid, D. Ernst, H. Schober, M. Koza, P. Adelmann, P. Schweiss, and T. Wolf, Phys. Rev. Lett. **88**, 067001 (2002).

[26] F. Gompf, W. Reichardt, H. Schober, B. Renker, and M. Buchgeister, Phys. Rev. B **55**, 9058 (1997).