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Phonon density of states and the search for a resonance mode in $LaFeAsO_{0.85}F_{0.15}$ (T_c=26 K)

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Abstract. While the high transition temperatures suggest that the conventional BCS phononmediated mechanism may not provide the main pairing mechanism in the recently discovered $RFeAsO_{1-x}F_x$ (1111-type) superconductors, there is, as yet, no consensus, despite extensive experimental and theoretical study. We report here the results of an inelastic neutron scattering investigation of an overdoped polycrystalline sample of $LaFeAsO_{1-x}F_x$ with x=0.15 (T_c=26 K). Four excitation peaks were observed at 13.6±1.5, 24.2±0.8, 32.2±0.5, and 41.4±1.0 meV. They were identified as phonon modes based on their wavevector and temperature dependence. The peak positions agree well with first-principles calculations of phonon density of states as well as experimental data on both the insulating parent and optimally doped $LaFeAsO_{1-x}F_x$ compounds. No evidence for the presence of a resonance mode was found. We found that the phonon density of states of the x=0.15 sample remains unchanged below T_c and is similar to samples with other fluorine concentrations. This suggests that a standard electron-phonon pairing mechanism cannot explain the high transition temperatures observed in these materials.

1. Introduction

The recent discovery [1] of high transition temperature superconductivity in $RFeAsO_{1-x}F_x$ (1111-type), where R is La or a rare earth element, has once again generated intense interest in the field of superconductivity. Since then, four other types of iron-based compounds have been found to also superconduct with doping or under high pressure. They are: BFe_2As_2 (122-type with B=Ba, Sr, Ca) [2], AFeAs (111-type with A=alkali metal) [3], $\alpha-$FeSe(Te) (11-type) [4], and $K_{0.8}Fe_2As_2$ (isostructural to $BaFe_2As_2$) [5]. These new Fe-based superconductors are the only alternative to the cuprate superconductors for exploring high temperature superconductivity. Despite many experimental and theoretical studies, there is still no consensus on the pairing mechanism that leads to high superconducting transition temperatures in these Fe-based materials. The high transition temperatures might suggest that a phonon-mediated mechanism cannot be the main pairing mechanism, and firstprinciples calculations based on density functional theory [6] also appear to support the notion that an exotic mechanism may play a role. However there is some evidence, including the strong relationship found between the

magnetic state of iron sublattice [7] and T_c [8] to the FeAs configuration that indicates the lattice (i.e. phonons) could also be involved.

The magnetic and superconducting phase diagrams of the cuprate and the Fe-based superconducting materials have some striking similarities. In both systems, the parent material exhibits antiferromagnetic order and superconductivity appears when the antiferromagnetically ordered phase has been suppressed by chemical doping, although the long range AF ordered phase in the Fe-based parent materials is metallic rather than Mott insulating as it is with cuprates. One prominent neutron scattering feature of cuprate superconductors is the presence of a resonance excitation mode [9]. The resonance mode is localized in both wavevector and energy. It is believed that this resonance mode is intimately related to superconductivity in cuprates as its intensity increases below T_c and its energy scales with T_c as a function of doping [10]. Resonance modes have also been observed [11] in Co, Ni, and K-doped $BaFe_2As_2$ as well as in the FeSe(Te) superconductors, indicating that magnetic interactions play a similar role in the superconductivity of the Fe-based materials as they do in the cuprates. Recent experiments have also found [12] a resonance mode in an optimally doped $LaFeAsO_{0.92}F_{0.08}$ polycrystalline sample with T_c=29 K. In order to investigate whether the resonance energy in Fe-based superconductors scales with T_c as it does in the cuprates, further experiments on Fe-based superconductors with different doping levels across the phase diagram are required.

Among all families of the Fe-based superconductors, the RFeAs$O_{1-x}F_x$ superconductors exhibit the highest superconducting transition temperatures (55 K for R=Sm [13]). Therefore a complete investigation of the RFeAs$O_{1-x}F_x$ properties, including the phonon and magnetic excitations, is required to gain a comprehensive understanding of high superconducting transition temperatures found in these materials. For most of the rare earths, the excitation spectra of the RFeAs$O_{1-x}F_x$ materials is complicated due to the presence of the crystal field excitations of the rare earth 4f electrons. RFeAs$O_{1-x}F_x$ compounds with R=La (no 4f electrons) provide an alternative to study the phonon and magnetic excitation spectra in these Fe-based superconductors without the complication of the crystal field excitations. We report here the wavevector and energy dependence of the excitations in an overdoped $LaFeAsO_{0.85}F_{0.15}$ polycrystalline sample with T_c=26 K at temperatures below and above T_c.

2. Experiments

The experiments were performed on a polycrystalline sample of $LaFeAsO_{0.85}F_{0.15}$ prepared by solid state reaction at ambient pressure from La, As, Fe, Fe_2O_3 and FeF_2. LaAs was first synthesized starting from pure elements in an evacuated sealed glass tube at a maximum temperature of 550°C. The final sample was synthesized by mixing LaAs, Fe, Fe_2O_3 and FeF_2 powders in stoichiometric proportions, using uniaxial pressing to make powders into a pellet. To avoid loss of fluorine, the heat treatment of the pellet was performed in a tantalum crucible welded under argon, at 1250°C for 72 hours, followed by cooling. Resistivity measurements indicated a superconducting transition temperature of 26 K. The sample was analyzed by powder x-ray diffraction in a Guinier camera, with Si as internal standard. The powder pattern showed the sample to be almost pure, with the space group P4/nmm ($a = 4.028(1)$ Å and $c = 8.720(1)$ Å). However, two faint lines were detected at low angles and attributed to LaOF.

The inelastic neutron scattering spectra were measured using the thermal time-of-flight neutron spectrometer IN4 at the Institut Laue Langevin (ILL), France. A crystal monochromator is used to set the incident energy and a time-of-flight technique is employed to analyse the scattered neutron energies, through proper phasing of a Fermi chopper with the acquisition electronics of the detectors. The spectrometer is equipped with a wide angular range detector assembly designed for powder work. The incident neutron wavelength of 1.1 Å (E_i=67.6 meV) was chosen to allow measurements with an energy transfer of up to 60 meV (neutron energy loss).

About 2 g of the polycrystalline $LaFeAsO_{0.85}F_{0.15}$ sample was placed inside a thin aluminium platelike mount. The inelastic spectra were collected in the temperature range 2 K to 50 K. The scattering from the empty aluminium mount was also measured to be used as background.

3. Results and discussion

The inelastic spectrum of the $LaFeAsO_{0.85}F_{0.15}$ sample was measured at several temperatures below and above T_c (=26 K). The observed scattering from the empty mount was subtracted from the signal. Figure 1 shows the background subtracted inelastic intensity as a function of wavevector and energy transfer at 2 K with an incident energy of E_i=67.6 meV. There are four excitation peaks that appear as bands of scattering at constant energy transfers: 13, 24, 32, and 41 meV. As the intensity of all four excitations increases with Q, we have identified them as phonon modes.

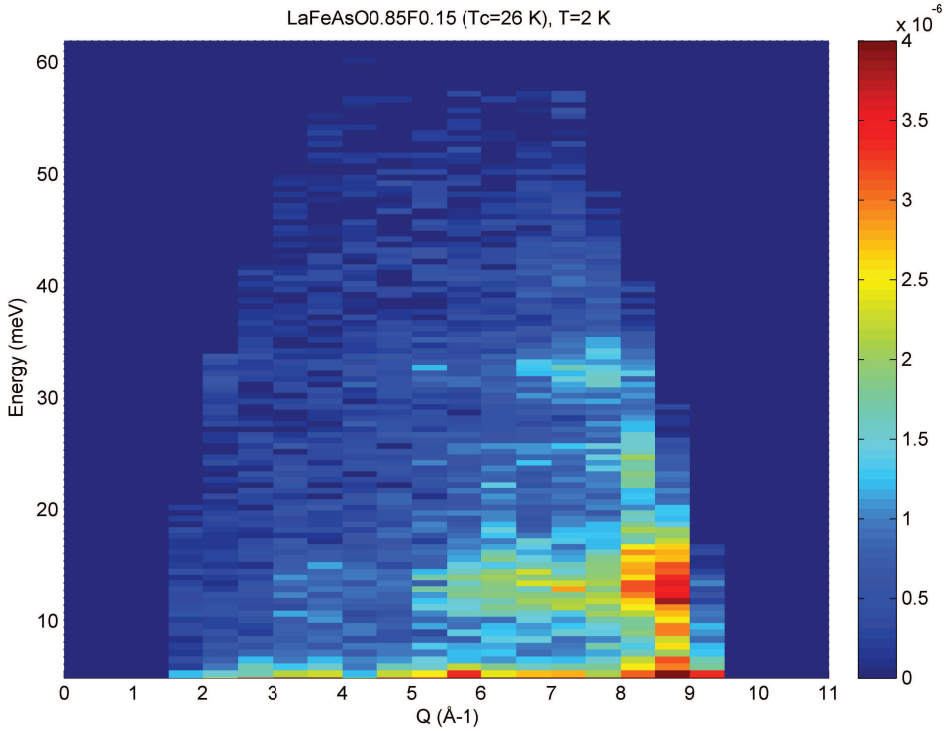

Figure 1. Observed scattering as a function of wavevector and energy transfer at 2 K. The data from the empty sample mount has been used to subtract the background. Four bands of excitations are observed at constant energy transfers: 13, 24, 32, and 41 meV. The intensity of all four excitations increases with Q indicating they are phonon modes. Similar data have been observed at temperatures higher than T_c. We find no evidence for presence of resonance characterized by an increase in intensity below T_c at any Q within our reach.

Within the limit of the single-phonon scattering and large wavevector transfer, the intensity from neutron down-scattering by phonons is given by [14]:

$$I(\mathbf{Q},\omega) = \frac{k_f}{k_i}\left(N\sum_{i=1}^{n} b_i^2 \frac{\hbar Q^2}{2M_i} e^{-2W_i(Q)} Z_i(\omega)\right)\frac{[n(\omega)+1]}{\hbar\omega} \quad (1)$$

Where k_f and k_i are final and incident neutron wavevectors, respectively; N is the number of unit cells; the sum runs over all atoms in the unit cell; b_i, M_i, $W_i(Q)$, are the scattering length, mass, and the Debye-Waller factor of the atom at site i in the unit cell, respectively; $Z_i(\omega)$ is the contribution to the phonon density of states from the atom at site i. The generalized phonon density of states defined by

$$g(\omega) = \sum_{i=1}^{n} \frac{b_i^2}{2M_i} e^{-2W_i(Q)} Z_i(\omega) / \sum_{i=1}^{n} \frac{b_i^2}{2M_i} e^{-2W_i(Q)} \quad (2)$$

can be extracted from the observed inelastic intensity integrated over all measured Q's:

$$I(\omega) = \int dQ\ I(\mathbf{Q},\omega) = Ag(\omega)\frac{[n(\omega)+1]}{\hbar\omega} \quad (3)$$

The observed integrated intensities $I(\omega)$ at 2 K (below T_c) and 50 K (above T_c) are shown in figure 2(a). In figure 2(b), we show $g(\omega)$ calculated from equation (3) at two temperatures. It is clear that no noticeable change is observed below T_c. We were able to fit four Gaussian peaks to the data centered at 13.6±1.5, 24.2±0.8, 32.2±0.5, and 41.4±1.0 meV. The peak positions for all four modes agree reasonably well with the first principles calculations of the phonon density of states for LaFeAs(O,F) [6,15] as well as with the previously reported phonon modes in the insulating parent LaFeAsO and optimally doped $LaFeAsO_{0.9}F_{0.1}$ [16]. The consistency between our results and those of first principles calculations that predict weak electron–phonon (e–p) coupling suggests that the e–p coupling is indeed weak and therefore that the standard e–p mediated coupling cannot explain the high transition temperatures observed in $LaFeAsO_{1-x}F_x$ superconductors. This view is further supported by the observation of similar phonon density of states for insulating and superconducting materials from optimally doped to overdoped with different T_c. If e–p coupling were indeed responsible for the superconductivity in this system, one would expect it to change as superconductivity appeared and the transition temperature evolved, and thus lead to a composition dependence in the phonon density of states.

We also searched for a neutron resonance peak characterized by an increase in intensity below T_c. A comparison of the data at 2 K (figure 1) and 50 K indicated no increase in intensity below T_c at any wavevector and energy transfer within our reach. The resonance peak observed [11] in Co, Ni, and K doped $BaFe_2As_2$ as well as in FeSe(Te) superconductors by neutron scattering, scales with T_c: $E_{res} = 4.9\,k_B T_c$. Recent experiments have also found [12] the resonance mode at 13 meV in an optimally doped $LaFeAsO_{0.92}F_{0.08}$ polycrystalline sample with T_c=29 K indicating a scaling ratio of 5.2. For our sample with a T_c of 26 K, the resonance peak would be expected within energy range of 10-12 meV. However, as indicated we did not detect any increase in intensity in this region below T_c. This could be due to the low sensitivity (small sample volume) of our experiment or because of the fact that the measurements were performed with a large E_i and hence the low-Q region (< 1.5 Å^{-1}) in the first Brillouin zone was not accessible in this energy range. However, the second Brillouin zone was accessible, but here the failure to observe a resonance could be due to reduced sensitivity as a result of the form factor [12]. We note that no resonance peak was observed in another overdoped $LaFeAsO_{0.87}F_{0.13}$ [17]. Further experiments are required to establish whether the resonance mode is absent in the overdoped region of Fe-based superconductors.

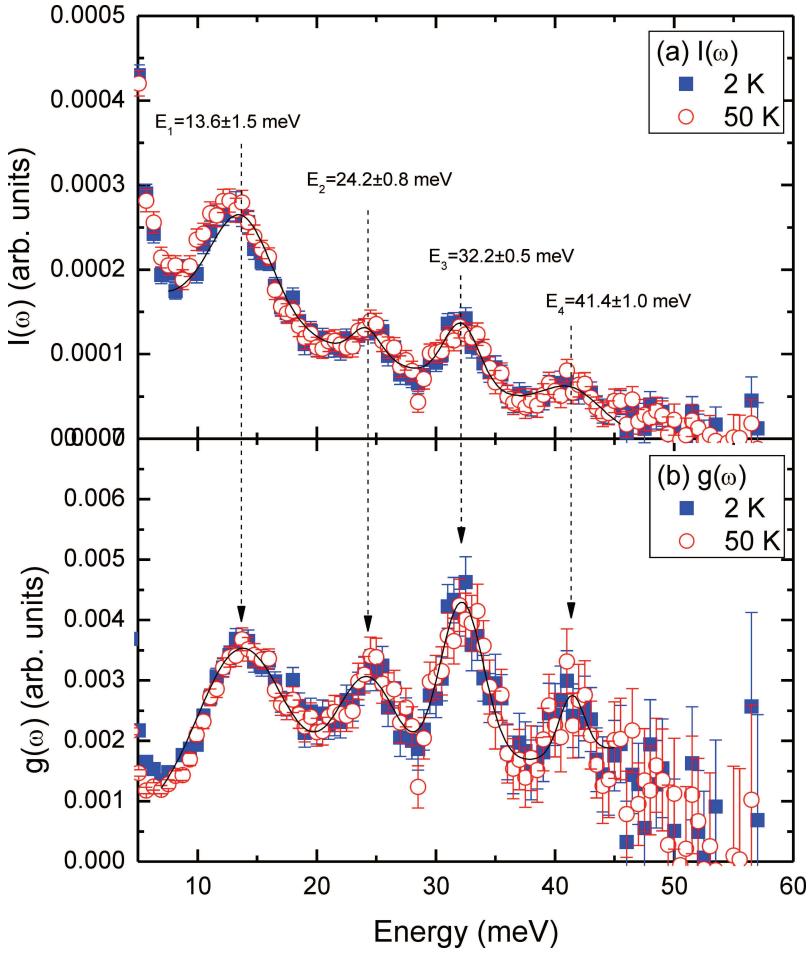

Figure 2. The observed intensity integrated over all wavevectors, $I(\omega) = \int dQ I(\mathbf{Q}, \omega)$, at 2 and 50 K. (b) The generalized phonon density of states at 2 and 50 K, obtained by multiplying the data in (a) by $\frac{\hbar\omega}{[n(\omega)+1]}$. See text for details. No noticeable change is observed below T_c.

4. Conclusions

We have measured the inelastic spectrum of an overdoped $LaFeAsO_{0.85}F_{0.15}$ with T_c=26 K. We found four excitation peaks at 13.6±1.5, 24.2±0.8, 32.2±0.5, and 41.4±1.0 meV. Based on their wavevector and temperature dependence we have identified all four peaks as phonon modes. The peak positions agree well with the first principle theoretical calculations of phonon density of states as well as experimental data on both the insulating parent and optimally doped $LaFeAsO_{1-x}F_x$ compounds. This suggests a standard electron-phonon pairing mechanism cannot explain the high transition temperatures observed in these materials. We have found no evidence for the presence of a resonance peak.

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