

Softening of bond-stretching phonon mode in $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ at the metal-insulator transition

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(Received 29 October 2010; revised manuscript received 8 April 2011; published 30 June 2011)

The dispersion of phonons in $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ along the $(3 + q, 0, 0)$ direction in reciprocal space was determined for $x = 0, 0.30, 0.37$, and 0.52 using high-resolution inelastic x-ray scattering. The observed phonon energies near Γ were in good agreement with published optical data. It was found that two high-energy modes strongly soften near $q = 0.25$ when the system becomes metallic ($x > 0.35$). There was no softening in the insulating phase, even at $x = 0.30$, very near the metal-insulator transition. This suggests that the phonon softening is linked to the appearance of the Fermi surface.

DOI: [10.1103/PhysRevB.83.224525](https://doi.org/10.1103/PhysRevB.83.224525)

PACS number(s): 63.20.D-, 63.20.kd, 74.25.Kc, 71.30.+h

I. INTRODUCTION

The electron-phonon interaction plays a central role in conventional BCS-Eliashberg superconductors. While, in most cases, the phonons contributing to superconductivity are low-frequency modes, there are two examples of superconductors in which a high-frequency optical mode predominantly contributes to pairing: $\text{Ba}(\text{Pb,Bi})\text{O}_3$, with a superconducting transition temperature of $T_c = 11$ K,¹ and $(\text{Ba,K})\text{BiO}_3$, with $T_c = 30$ K.² In these materials the Bi-O breathing-mode phonon couples strongly to the electronic system.^{3,4} Another example is MgB_2 , $T_c = 39$ K,⁵ where coupling to the high-energy boron optical mode plays a main role in superconductivity.⁶

Strong electron-phonon coupling often manifests itself as phonon softening. For MgB_2 , inelastic x-ray scattering (IXS) has demonstrated that the dispersion of the E_{2g} phonon strongly softens and broadens near the zone center.^{7,8} Subsequent work with carbon-substituted MgB_2 revealed that the amount of phonon softening correlates with T_c ,⁹ providing direct proof that this phonon is responsible for the high- T_c superconductivity in MgB_2 .

By contrast, in high- T_c superconducting cuprates (HTSCs), the phonon contribution to superconductivity remains unclear, although the high-frequency oxygen mode (Cu-O bond-stretching mode) shows strong softening as the momentum transfer is changed.¹⁰⁻¹⁷ The main subject of discussion for the cuprates is the rapid drop in the bond-stretching phonon frequency and increase in linewidth, in the $(q,0,0)$ direction at $q > 0.25$, which is not reproduced by local density approximation calculations.^{14,18,19} The carrier doping dependence of the phonon softening is also an unresolved problem. That is, why the softening gradually develops with hole doping and saturates at high doping levels¹⁵ is not well understood.

The latter problem motivated us to investigate the doping dependence of phonon softening in other superconducting systems near a metal-insulator (MI) transition. Phonon softening near an MI transition is usually observed when the temperature is lowered. However, there are few studies of phonon softening as a function of carrier doping by chemical substitution, particularly in systems where there is a doping-induced MI transition. From this viewpoint, $(\text{Ba,K})\text{BiO}_3$ (BKBO) is a

good candidate for study. BKBO has a perovskite structure, similar to the cuprates. As K is doped into the system, BKBO changes from a charge-density-wave (CDW) insulator to a metal that superconducts ($T_c \sim 30$ K) at $x \sim 0.35$. The crystal structure also changes with x from a monoclinic to an orthorhombic structure (at $x \sim 0.1$) and then to a cubic structure (at $x \sim 0.35$).²⁰

The phonon dispersion of BKBO has previously been investigated only by neutron scattering measurements for K concentrations of $x = 0.02$ and 0.40 .^{21,22} That work showed that no phonon softening was observed in the insulator, while dramatic softening of the Bi-O stretching and breathing modes appeared in the metallic (superconducting) phase. It is thus interesting to consider how the softening changes as one increases x through the MI phase transition, for example, whether the phonon softens only near the MI transition or whether the phonon softening correlates with T_c as the electron-phonon coupling changes with K concentration. However, the earlier work^{21,22} was limited to only two K concentrations.

In the present paper we investigate the phonon dispersion of BKBO single crystals with $x = 0, 0.30, 0.37$, and 0.52 to clarify the phonon behavior near the MI transition. Despite great effort, we were not able to grow single-phase samples with dimensions larger than about 0.5 mm, however, our technique of measuring phonon dispersion, IXS, allows us to perform these measurements on such small samples. Samples with $x = 0$ and 0.30 are in the insulating phase, while those with $x = 0.37$ and 0.52 are metallic. So far, there has been no report of phonon dispersion for $x = 0.30$, near the MI transition, and for $x = 0.52$, a heavily doped composition of the metallic phase.

II. EXPERIMENTS

Single crystals of BaBiO_3 were grown by a flux method using Bi_2O_3 and BaCO_3 , and K-doped crystals, by the electrochemical technique using Bi_2O_3 , $\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$ and KOH .²³⁻²⁵ Our sample sizes were typically $\sim 0.3 \times 0.3 \times 0.3$ mm³ (< 0.2 mg); while effort was made to find larger crystals, they were always multidomain and/or nonuniform in doping. It was particularly difficult to grow homogeneous

samples with $x \sim 0.3$, and this may explain why there are relatively few experimental investigations of samples with $x \sim 0.3$ to 0.35. The lattice constants of the samples were measured using a four-circle x-ray diffractometer, and the K concentration was determined assuming a linear relationship between the lattice parameter and x in $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$.²⁰ Superconducting transition temperatures were determined from the magnetic susceptibility measured by a superconductor quantum interference device (SQUID) magnetometer. The K content estimated from the lattice parameter was in good agreement with the value expected from the measured T_c .²⁰

IXS spectra were measured at the BL35XU of SPring-8.²⁶ We measured spectra along the (100) direction of the simple perovskite lattice, that is, from Γ to X points, $(3+q,0,0)$, where $0 < q < 0.5$, at room temperature. Depending on the setup, the energy resolution varied from 1.5 to 6 meV according to the choice of x-ray energy and analyzer Bragg reflection [1.5/3/6-meV resolution at Si (11 11 11)/(999)/(888) at 21.7/17.8/15.8 keV]. The incident beam at the sample position was about $60 \times 100 \mu\text{m}^2$ in the full width at half-maximum (FWHM). Because of the strong x-ray absorption by heavy elements, the x-ray penetration depth in BKBO is small ($< 20 \mu\text{m}$), for example, about 40% that for $(\text{La,Sr})_2\text{CuO}_4$ which resulted in a relatively weak phonon signal. For analysis of IXS spectra, the pseudo-Voigt functions were used.

III. RESULTS

Figure 1 shows the IXS spectrum of BaBiO_3 at various q values. The spectra are well reproduced by fitting curves consisting of an elastic scattering peak and six phonon peaks. Since the measurement direction is $(3+q,0,0)$ in the present experiment, the intensity of longitudinal optical (LO) phonons is enhanced, while transverse optical (TO) phonons are almost undetectable. Since the intensity at an energy higher than 80 meV is very low, we consider the features in this energy range unreliable and ignore them. Based on the published optical data,^{27,28} we assign the six peaks from low to high frequency as the acoustic mode, external mode (relative displacement of Ba and BiO octahedron), Bi mode, Bi-O bending mode, Bi-O stretching mode, and Bi-O breathing mode. Although there are only four longitudinal modes, including the acoustic mode, for a cubic perovskite structure, the lattice distortion coupled with CDW creates additional phonon branches. For example, the Bi-O breathing mode at the zone boundary becomes a zone center mode that is Raman active. The Bi-Bi vibration mode that is originally an acoustic mode turns to an infrared active mode because of inequivalent neighboring Bi charges.²⁷

Figure 2 illustrates a comparison of our data near the Γ point ($q \sim 0.1$) with published optical data including both far-infrared and Raman scattering measurements.²⁷⁻³¹ As shown in the figure, our IXS data agree well with the optical data over the whole composition range, except for the peak at 60 meV. This is strong support for the reliability of our measurement. As for the 60-meV peak, we tentatively attribute it to the LO bond-stretching mode, although its energy is different from the far-infrared data (69 meV). Another possibility is a TO mode, which, while it should be weak in our IXS data, is known to be at 60 meV from far-infrared measurements.

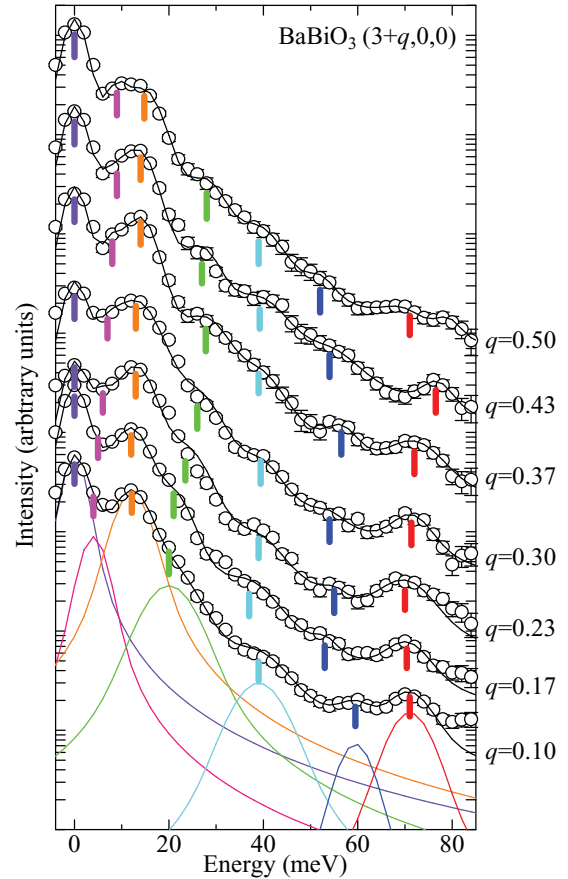


FIG. 1. (Color online) A typical IXS spectrum for the BaBiO_3 system at various q values. Open circles show experimental data with their statistical error bars. Lines below the spectrum show the pseudo-Voigt fitting for elastic scattering and each phonon mode indicated by a short bar. Solid black lines show the total fitting of the experimental data.

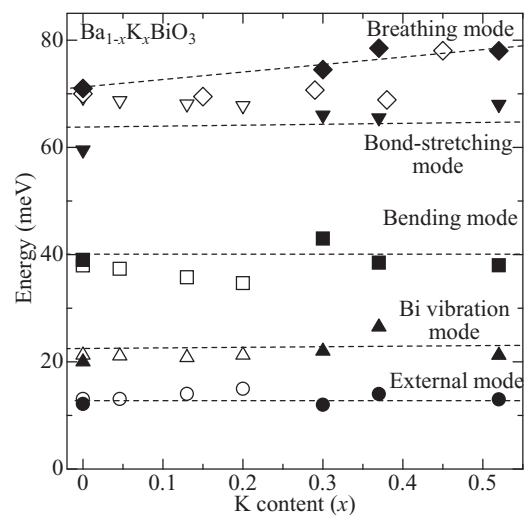


FIG. 2. Potassium content dependence of phonon energies for various modes at $q \sim 0.1$ in $(3+q,0,0)$ momentum (filled symbols). Open symbols are the other experimental data for LO phonon modes from Raman and optical spectroscopies.²⁸⁻³⁰ Dashed horizontal lines are guides for the eye.

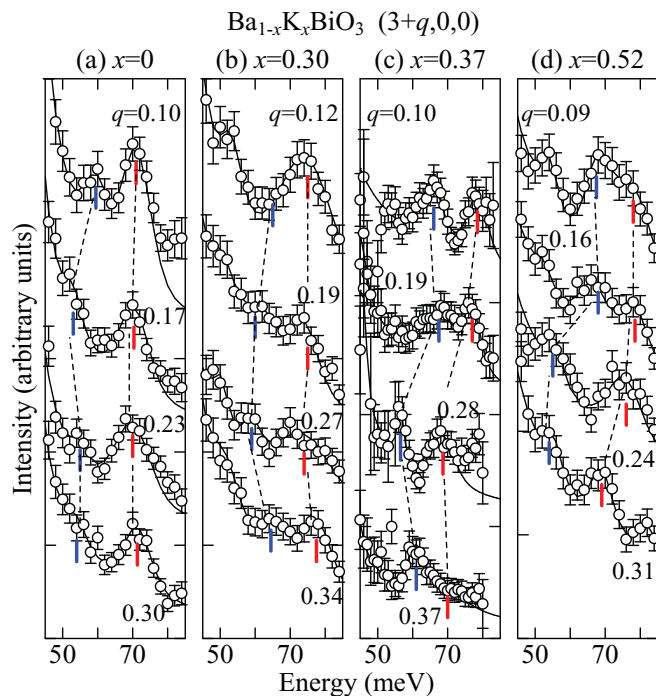


FIG. 3. (Color online) High-energy part of the IXS spectra of the BKBO system for the four compositions. The number beside the data shows the q value in $(3+q,0,0)$ momentum. Open symbols show experimental data and solid lines show the fitting result. Short vertical bars on the data indicate the positions of the phonon peaks. Dashed vertical lines are guides for the eye, to track the phonon dispersions. Data were plotted by a shift in the vertical axis to distinguish the different q patterns.

We now focus on higher frequency phonons. In Fig. 3, the high-energy part of the spectra ($\hbar\omega > 45$ meV) is shown at several q values for all measured crystals. For insulating samples with $x = 0$ and 0.30, the q dependence of the phonon frequency is weak, while phonons in metallic samples with $x = 0.37$ and 0.52 exhibit large changes between the second and the third q vectors (at $q \sim 0.2$).

The observed phonon frequencies at all q values are summarized in Fig. 4. Anomalous softening of the bond-stretching and the breathing modes is observed for $x = 0.37$ and 0.52, while almost no softening is seen for $x = 0$ and 0.30. The neutron scattering results of Braden^{21,22} are also plotted for comparison. The agreement with the neutron data is good for $x = 0$ but not for $x = 0.37$. This discrepancy between our data and the neutron result might be due to a difference in K content of the samples. Our IXS results are in better agreement with the optical data. Although the phonon frequencies are different, the tendency toward phonon softening is the same in both neutron scattering and IXS experiments.

It should be noted that the breathing mode as well as the Bi-Bi vibration mode survives even in the metallic phase, although these modes should not be present in a cubic perovskite structure without the CDW distortion. The present results suggest that local lattice distortions remain even in the nominally cubic phase. In fact, we observed strong superlattice peaks during the experiments, as if the CDW distortion remained. Also, there have been several reports

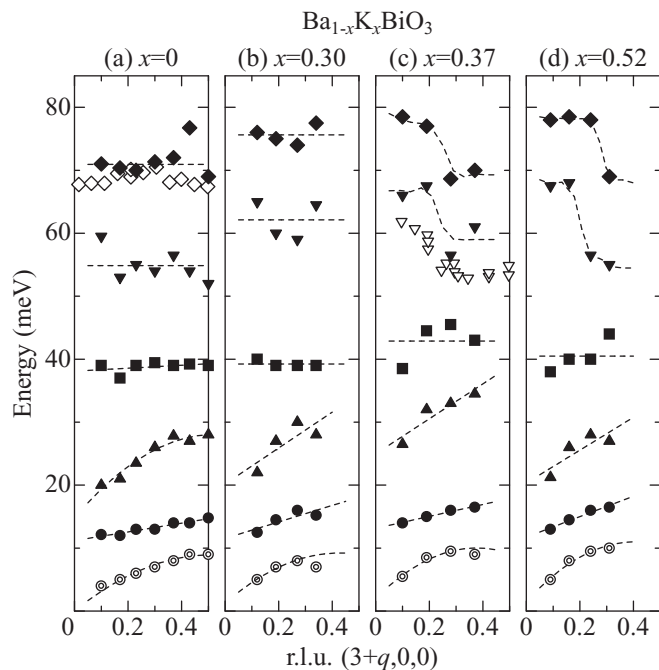


FIG. 4. Phonon branches of the BKBO system in the (100) direction for four values of x (filled symbols and double circles) and data from neutron scattering studies^{21,22} for comparison (open symbols). Dashed lines are guides for the eye.

on the short-range structure distortion suggested by x-ray absorption fine structure,³² ultrasonic measurements,³³ and Raman scattering³⁴ in the metallic phase. Recently, even a neutron diffraction measurement has detected a long-range structural distortion in superconducting BKBO characterized by a tilt of the BiO octahedra.³⁵

IV. DISCUSSION

An important result of the present study is that no softening is observed in the phonon dispersion for $x = 0.3$, very close to the MI transition in the insulator phase. Often (e.g., in a conventional CDW material) phonon softening occurs as the temperature is swept through the MI transition, with the softening being greatest at the phase transition and then recovering. In the present case, we change the doping at a fixed temperature (300 K), and the crystal structure changes from orthorhombic to cubic at $x \sim 0.35$, accompanied by a change from a CDW insulator to a metal. (The change from monoclinic to orthorhombic at $x = 0.1$ does not seem to affect the phonon dispersion.) Making an analogy between T - and x -sweep experiments, we expected phonon softening at x just before and after the MI transition. The absence of phonon softening at $x = 0.3$ may be because the MI transition is too sharp as a function of x (or T) to follow the transient behavior of the phonon. Since even near $x = 0.35$ the MI transition temperature is much higher than room temperature, we cannot observe softening at 300 K. This fact implies that the CDW gap is still large at $x = 0.30$, as suggested by optical absorption data (~ 0.5 eV at $x = 0.28$ and 2 eV at $x = 0$).³⁶

In contrast, clear phonon softening in BKBO is observed in the metallic phase with a large Fermi surface (FS). Once the

FS is formed after the phase transition, it changes only weakly with x , which might result in the x -independent ($x > 0.35$) softening seen in the present work.

According to the theoretical prediction, the electron-phonon coupling constant λ , which has the largest contribution from the breathing mode, decreases with increasing x ,⁴ because the FS shrinks as the band filling is reduced. This could explain why the T_c for $x = 0.52$ is lower than that for $x = 0.37$. However, the present results do not show a clear x (or T_c) dependence of phonon softening in the metallic phase. Therefore, at least in the present case, the amount of softening of the bond-stretching phonon is not a good measure for the strength of the electron-phonon coupling that determines T_c .

The reason for this unexpected result is not clear. One possibility is that the phonon change is too small to be observed with the present experimental resolution. According to Shirai's calculation,⁴ λ changes from 0.88 ($x = 0.4$) to 0.53 ($x = 0.5$), from which T_c was estimated as 25 K ($x = 0.4$) and 10 K ($x = 0.5$), on the basis of the Allen-Dynes equation.³⁷ These values are not far from the experimental data (30 K for $x = 0.4$ and 17 K for $x = 0.5$).²⁰ Compared to such a drastic change in T_c with λ , the phonon frequency is expected to show a weaker λ dependence. The phonon renormalization due to electron-phonon coupling is generalized as $\omega_{\text{ph}}^2 \sim \Omega_{\text{ph}}^2 / (1 + \lambda)$,³⁸ where ω_{ph} is the renormalized phonon frequency and Ω_{ph} is the bare phonon frequency.³⁹ This expression gives about a 10% change in phonon frequency when λ changes from 0.88 ($x = 0.4$) to 0.53 ($x = 0.5$), which is much smaller than the 60% reduction in T_c . While the above relation between λ and the renormalized phonon frequency is a very rough approximation, the important idea in the present context is that the fractional changes in phonon frequency may be small compared to the changes in λ .

Finally, we note that the present result brings the unusual phonon behavior of the HTSCs into sharp relief. Softening of the bond-stretching phonon has been reported in cuprates with a layered perovskite structure,^{10–17} however, the doping dependence in cuprates¹⁵ is very different. In BKBO, the electronic change with doping is small in the metallic phase, where a rigid-band picture is applicable as the first

approximation. In this situation, as we see in the present study, even near the MI transition, a 10% change in band filling⁴⁰ gives only a little change in phonon softening. By contrast, in underdoped cuprates (e.g., at $0 < x < 0.15$ of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$), the system is in the so-called "pseudogap" state,⁴¹ where the FS rapidly develops with carrier doping from a Mott insulator.⁴² Therefore, it is likely that such a radical change in FS topology accompanied by a huge enhancement of the electronic density of states is responsible for the gradual increase in softening of the Cu-O bond-stretching phonon with carrier doping. The importance of FS topology in the electron-phonon coupling of cuprates was also pointed out in Ref. 17. Further detailed information about the FS will promote more precise calculations of the phonon behaviors in cuprates.

V. SUMMARY

We have investigated the phonon dispersion of $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ in the $(3+q, 0, 0)$ direction for $x = 0, 0.30, 0.37$, and 0.52 using IXS. In contrast to the nearly doping-independent behavior of the low-frequency modes, the higher frequency modes related to Bi-O bond-stretching vibrations showed a clear and abrupt change with K content. In the insulating phase, $x < 0.35$, mode dispersions were almost flat, while radical softening was observed toward the zone boundary in the metallic phase, $x > 0.35$. The degree of softening is relatively unaffected by the K content ($x > 0.35$). This doping dependence of the phonon softening suggests that the present softening is different from the softening often associated with a structural phase transition and, more probably, is electronic in origin.

ACKNOWLEDGMENTS

The authors thank T. Oguchi for a useful discussion. This work was partially supported by a Grant-in-Aid for Scientific Research (Grant No. 19204038) from the Ministry of Education, Culture, Sports, Science and Technology of Japan. H.K. appreciates the support from the Japan Society for the Promotion of Science (JSPS) scholarship and its research grants.

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