

Electron-phonon interaction in the lamellar cobaltate Na_xCoO_2

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We study theoretically and experimentally the dependence of the electron-phonon interaction in Na_xCoO_2 on the sodium concentration x . For the two oxygen phonon modes found in Raman experiments, A_{1g} and E_{1g} , we calculate the matrix elements of the electron-phonon interaction. Analyzing the feedback effect of the conduction electrons on the phonon frequency we compare the calculated and experimentally observed doping dependence of the A_{1g} mode. Furthermore, due to the momentum dependence of the electron-phonon coupling for the E_{1g} symmetry we find no renormalization of the corresponding phonon frequency which agrees with experiment. Our results shed light on the possible importance of the electron-phonon interaction in this system.

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The origin of the unconventional superconductivity in low-dimensional perovskite systems attracts much attention and belongs to the most challenging questions of condensed matter physics. The best known example among these materials are high- T_c cuprate superconductors. There, one of the possible scenarios for the Cooper-pair formation is the so-called spin fluctuation mechanism. At the same time, due to the complexity of the transition metal oxides, other energy scales are present, and their role in the formation of superconductivity remains under debate. This, in particular, concerns the electron-phonon interaction. For example, its relevance for superconductivity in layered cuprates and the anomalous normal state has been discussed in, e.g., Ref. 1. Despite some progress, a complete understanding of the physics of electron-phonon coupling in perovskites is still lacking because of the crystallographic complexities of these materials.

The discovery of superconductivity with $T_c=4.6$ K in water intercalated sodium cobaltate, $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$,² is of great interest on its own and also because of similarities with layered cuprates. The sodium cobaltate has a quasi-two-dimensional layered structure with CoO_2 layers and rich phase diagram as a function of the Na concentration, which includes superconductivity at $x \approx 0.3$, an insulating phase at $x \sim 0.5$, and unusual magnetism for $x \geq 0.6$.³ There is also increasing experimental and theoretical evidence for unexpected strong correlation effects as the cobaltates approach the band insulating limit at $x=1$.⁴⁻¹¹ In Na_xCoO_2 the Na ions reside between the CoO_2 layers, with Co ions forming a triangular lattice, and donate x electrons to the partially filled $\text{Co}-d(t_{2g})$ orbitals. Due to the presence of a trigonal crystal-line electric field, the t_{2g} levels split into the higher lying a_{1g} state and the two lower lying e'_g states.¹² Angle-resolved photoemission spectroscopy (ARPES)^{13,14} reveals a doping dependent evolution of the Fermi surface, which shows no sign of the e'_g hole pockets for $0.3 \leq x \leq 0.8$. The observed Fermi surface is centered around the Γ point and has mostly a_{1g}

character. It has been argued that such an effect may arise due to strong electronic correlations,^{8,15} or Na-induced disorder,¹⁶ however, no consensus in the literature has been reached yet (see, for example, Refs. 17-19).

Despite intensive studies of the electronic and magnetic properties, little is known about the phonon excitations and their doping evolution in Na_xCoO_2 . At the same time, due to the relatively low superconducting transition temperature the possible relevance of phonons for superconductivity cannot be neglected. For example, the role of the electron-phonon coupling in Na_xCoO_2 has been discussed in the context of its relevance to superconductivity and charge ordering on the basis of a $t-V$ model.²⁰ In addition, due to some similarity with high- T_c cuprates the understanding of the phonon renormalization in the sodium cobaltates is of great importance. For high-temperature superconductors the effect of renormalization of the optical phonons by the conduction electrons has been considered in Refs. 21 and 22.

In this Rapid Communication we investigate the electron-phonon interaction in the Na_xCoO_2 as a function of doping concentration and its superconducting relative by means of Raman spectroscopy. We observe two oxygen phonon modes at small wave vectors with A_{1g} and E_{1g} symmetries. Then we derive the diagonal and off-diagonal electron-phonon matrix elements for these modes. Calculating the renormalization of the phonon frequencies by conduction electrons we compare our results with the doping dependent evolution of the A_{1g} mode and obtain the electron-phonon coupling constant $g_{off}^{A_{1g}}=3$ meV. Due to the structure of the electron-phonon matrix element for the E_{1g} mode we obtain no doping dependence of the corresponding phonon frequency in good agreement with experiment.

Experiment details. Raman scattering experiments have been performed in quasibackscattering geometry on freshly cleaved single crystal surfaces. The samples have been fully characterized using basic thermodynamic as well as spectroscopic techniques.²³⁻²⁸ After cleavage the crystals were rap-

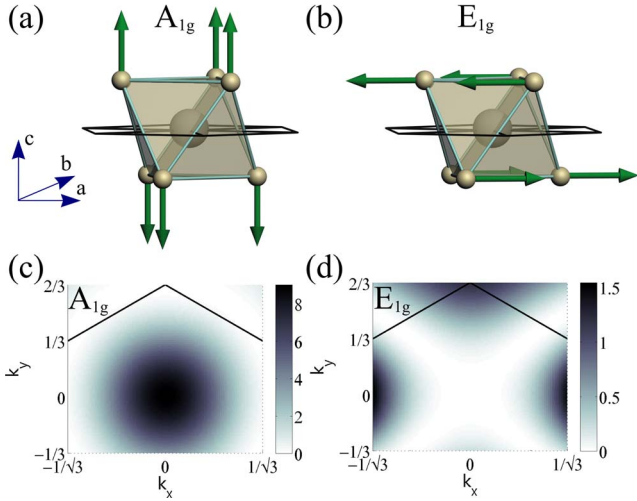


FIG. 1. (Color online) Schematic illustration of the (a) A_{1g} and (b) E_{1g} phonon modes. The arrows indicate the oxygen displacement in the CoO_6 octahedra. On the left-hand side of (a) we indicate the crystallographic a , b , and c directions. (c), (d) The calculated momentum dependence of the electron-phonon structure factors $[F_{\mathbf{q}}^l]^2$ in the first Brillouin zone for the A_{1g} and E_{1g} phonon modes, respectively.

idly cooled down in He exchange gas to prevent degradation. In Na_xCoO_2 in-plane E_{1g} and out-of-plane A_{1g} oxygen modes have been observed in Raman scattering.^{24,29,30} The corresponding oxygen displacements are depicted in Figs. 1(a) and 1(b). The Co site is not Raman active. Modes of the Na sites have not been identified unambiguously.²⁹ This is probably related to disorder on the partially filled sites. The two dimensionality with respect to structure and bonding leads to a decoupling of the Na and the CoO_2 layers. The observed doping dependence of the A_{1g} and E_{1g} oxygen phonon frequencies are shown in Fig. 2. The crossover from one to the other crystallographic phases (shaded areas) given by an ordering of the differently occupied Na sites leads for the A_{1g} modes to small frequency jumps superimposed on the global frequency shift due to doping. In the frequency regime of the E_{1g} phonon new modes with a larger energy offset are induced that are omitted here for clarity. We also restrict ourselves to the low-temperature data, as the temperature dependence of the oxygen phonon modes is negligible ($\leq 1-2\%$) and is approximately independent of doping. Summarizing the experimental data, the two phonon modes display a markedly different doping dependence.

Tight-binding model. To describe the electronic subsystem we use a tight-binding t_{2g} -band model with parameters (in-plane hoppings and the single-electron energies) derived previously from the *ab initio* local density approximation (LDA) calculations using projection procedure for $x=0.33$.⁸

The free-electron Hamiltonian of the t_{2g} -band model in a hole representation is given by

$$H_0 = - \sum_{\mathbf{k}, \alpha, \sigma} (\epsilon^\alpha - \mu) n_{\mathbf{k}\alpha\sigma} - \sum_{\mathbf{k}, \sigma} \sum_{\alpha, \beta} t_{\mathbf{k}}^{\alpha\beta} d_{\mathbf{k}\alpha\sigma}^\dagger d_{\mathbf{k}\beta\sigma}, \quad (1)$$

where $n_{\mathbf{k}\alpha\sigma} = d_{\mathbf{k}\alpha\sigma}^\dagger d_{\mathbf{k}\alpha\sigma}$, $d_{\mathbf{k}\alpha\sigma}$ ($d_{\mathbf{k}\alpha\sigma}^\dagger$) is the annihilation (creation) operator for the t_{2g} hole with spin σ , orbital index α ,

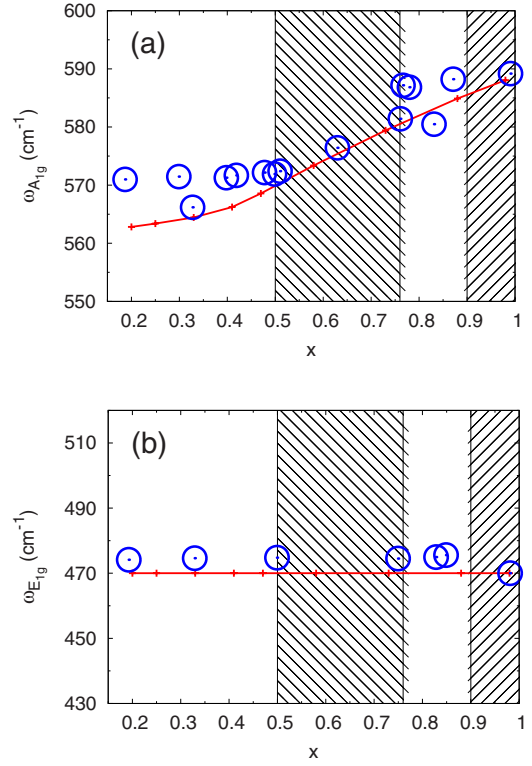


FIG. 2. (Color online) The experimental Raman data at 10 K is shown by (blue) circles. The calculated doping dependence of the (a) A_{1g} and (b) E_{1g} phonon modes is shown by red crosses, the solid curve is a guide to the eye. The slight scattering of the experimental points around $x=0.5$, $x=0.76$, and $x=0.9$ (shown by the hatched areas) is the result of the different crystallographic phases around these points (Ref. 28). These structural modifications only weakly affect the electron-phonon coupling as it mainly involves Na ordering. Measurements of A_{1g} mode at 290 K (not shown) follow a similar trend as the 10 K data.

and momentum \mathbf{k} , $t_{\mathbf{k}}^{\alpha\beta}$ is the hopping matrix element, and ϵ^α is the single-electron energy. To obtain the dispersion we diagonalize the Hamiltonian (1) calculating the chemical potential μ self-consistently. Due to the nonzero interorbital hopping matrix elements, a_{1g} and e'_g bands are hybridized. However, only one of the hybridized bands crosses the Fermi level. We refer to the diagonalized bands as $\epsilon_{\mathbf{k}}^{\alpha'}$ with the new orbital index α' .

Electron-phonon interaction. In analogy to previous considerations for cuprates,^{31,32} we derive the electron-phonon matrix elements, $g_{\mathbf{k}\mathbf{q}}$, for the A_{1g} and E_{1g} phonon modes depicted in Figs. 1(a) and 1(b). Namely, to obtain the main contribution to the diagonal (intraband) part of the electron-phonon interaction we expand the Coulomb energy between Co and oxygen, $H_C = \frac{ee^*}{\epsilon} \sum_{i, \alpha', \sigma, \gamma} c_{i\alpha'\sigma}^\dagger c_{i\alpha'\sigma} \left(\frac{1}{|\mathbf{R}_i - \mathbf{r}_{i,\gamma}|} + \frac{1}{|\mathbf{R}_i - \mathbf{r}_{i,-\gamma}|} \right)$, in the small displacements of the oxygen ions. Here, e is the electron charge, $e^* = -2e$ is the oxygen ion charge, ϵ is the dielectric constant, \mathbf{R}_i are the Co ion positions, $\mathbf{r}_{i,\gamma}$ are the vector positions of the vibrating oxygens, and index γ ($-\gamma$) labels the three oxygen positions within CoO_6 unit cell above (below) the Co layer. Here, $c_{i\alpha'\sigma}^\dagger$ refers to the diagonal form of the Hamiltonian (1). After introducing the creation (anni-

hilation) operator $b_{-\mathbf{q}}^\dagger$ ($b_{\mathbf{q}}$) for the phonon with momentum \mathbf{q} , we arrive to the following form of the electron-phonon interaction

$$H_{el-ph}^{diag} = \sum_{\mathbf{k}, \mathbf{q}, \alpha', \sigma} g_{\mathbf{q}}^{\alpha' \alpha'} c_{\mathbf{k}\alpha' \sigma}^\dagger c_{\mathbf{k}-\mathbf{q}\alpha' \sigma} (b_{\mathbf{q}} + b_{-\mathbf{q}}^\dagger). \quad (2)$$

For the sake of simplicity we assume the diagonal electron-phonon interaction is independent on the orbital index α' . Thus, for the A_{1g} and E_{1g} optical Raman-active phonon oxygen modes one finds $g_{\mathbf{q}}^{A_{1g}} = g_{diag}^{A_{1g}} F_{\mathbf{q}}^{A_{1g}}$, $g_{\mathbf{q}}^{E_{1g}} = g_{diag}^{E_{1g}} F_{\mathbf{q}}^{E_{1g}}$, where the structure factors of the electron-phonon interaction are

$$F_{\mathbf{q}}^{A_{1g}} = \cos \frac{q_1 - q_2}{3} + \cos \frac{q_1 + q_3}{3} + \cos \frac{q_2 + q_3}{3}, \quad (3)$$

$$F_{\mathbf{q}}^{E_{1g}} = \cos \frac{q_1 - q_2}{3} - \frac{1}{2} \left(\cos \frac{q_1 + q_3}{3} + \cos \frac{q_2 + q_3}{3} \right). \quad (4)$$

Here, $q_1 = (\sqrt{3}/2)q_x - \frac{1}{2}q_y$, $q_2 = q_y$, $q_3 = (\sqrt{3}/2)q_x + \frac{1}{2}q_y$, in units of $2\pi/a$ with a being the in-plane lattice constant, $g_{diag}^\Gamma = -\frac{ee^*}{\epsilon} \frac{2L_\Gamma}{\sqrt{d^2 + l^2}} \sqrt{\frac{\hbar}{2M\omega_\Gamma}}$, where ω_Γ is the corresponding bare phonon frequency ($\Gamma = A_{1g}, E_{1g}$), $L_{A_{1g}} = d = a/\sqrt{6}$ is the distance between the Co and the oxygen plane, $L_{E_{1g}} = l = a/\sqrt{3}$ is the planar distance between Co and oxygen, and M is the oxygen mass. Assuming that in the band insulator, $\text{Na}_{x=1}\text{CoO}_2$, the renormalization of the phonons by the conduction electrons is absent, we use $\omega_{A_{1g}} = 589 \text{ cm}^{-1}$ and $\omega_{E_{1g}} = 470 \text{ cm}^{-1}$. These values are close to those obtained by the first principles calculations.³³ The resulting momentum dependence of the structure factors for both modes is shown in Figs. 1(c) and 1(d). Interestingly, one sees that while $g_{\mathbf{q}}$ for the A_{1g} mode shows a maximum at the Brillouin zone center, the corresponding $g_{\mathbf{q}}$ for the E_{1g} mode vanishes there. Therefore, for $\mathbf{q} = 0$ the electron-phonon coupling for the E_{1g} channel is zero. Taking $\epsilon \sim 20$ we estimate $g_{diag}^{A_{1g}} \approx 0.05 \text{ eV}$.

The off-diagonal (interband) contribution to the electron-phonon interaction arises mainly from the modulation of the interorbital Co-Co hopping matrix element via oxygen. Assuming the linear terms in the expansion of the nearest neighbors hopping matrix element $t_{ij}^{\alpha\beta}(u_\gamma) = t_{ij}^{\alpha\beta} + V^{\alpha\beta} u_\gamma$ over the oxygen displacements u_γ , one obtains

$$H_{el-ph}^{off} = \sum_{\mathbf{k}, \mathbf{q}, \alpha' \neq \beta', \sigma} g_{\mathbf{kq}}^{\alpha' \beta'} c_{\mathbf{k}\alpha' \sigma}^\dagger c_{\mathbf{k}-\mathbf{q}\beta' \sigma} (b_{\mathbf{q}} + b_{-\mathbf{q}}^\dagger), \quad (5)$$

where $g_{\mathbf{kq}}^{\alpha' \beta'} = g_{off}^\Gamma F_{\mathbf{q}}^\Gamma [\gamma(\mathbf{k}) + \gamma(\mathbf{k} + \mathbf{q})]$ with $\gamma(\mathbf{k}) = \cos k_2 + \cos k_3 + \cos k_1$ being the Co lattice structure factor. Again one could see that for $\mathbf{q} = 0$ the off-diagonal electron-phonon coupling for the E_{1g} channel is zero due to the momentum dependence of the electron-phonon structure factor, $F_{\mathbf{q}}^{E_{1g}}$. Therefore, in Raman experiments which probes $\mathbf{q} = 0$ response this mode shows no doping dependence due to the coupling to the electronic subsystem. This is also confirmed by the fact that the observed phonon mode energy for all doping levels lies close to the value obtained by *ab initio* calculations.³³ The only Raman-active optical phonon mode

which will couple to the conduction electrons at $\mathbf{q} = 0$ is the A_{1g} mode.

In the following we consider the renormalization of the A_{1g} phonon. The corresponding Dyson equation reads

$$D^{-1}(\mathbf{q}, \omega) = D_0^{-1}(\omega) - \Pi(\mathbf{q}, \omega), \quad (6)$$

where $D_0(\omega) = \frac{2\omega_\Gamma}{\omega^2 - \omega_\Gamma^2 + i\delta}$ is the momentum-independent bare phonon propagator. The polarization operator is given by

$$\Pi(\mathbf{q}, \omega) = -2 \sum_{\alpha', \beta'} \sum_{\mathbf{k}} (g_{\mathbf{kq}}^{\alpha' \beta'})^2 \frac{f(\epsilon_{\mathbf{k}+\mathbf{q}}^{\alpha'}) - f(\epsilon_{\mathbf{k}}^{\beta'})}{\omega - \epsilon_{\mathbf{k}+\mathbf{q}}^{\alpha'} + \epsilon_{\mathbf{k}}^{\beta'} + i\delta}, \quad (7)$$

where $f(\epsilon)$ is the Fermi function. To find the renormalization of the bare phonon frequency and to compare the results to the Raman experiments we set $\mathbf{q} \rightarrow 0$ limit and solve Eq. (6) numerically as a function of the doping concentration. The main contribution to the renormalization of the optical phonon modes comes from the interband transitions, i.e., terms with $g_{\mathbf{kq}}^{\alpha' \neq \beta'}$ while intraband transitions renormalize the acoustic phonon modes. The results of our numerical calculations are shown in Fig. 2. The doping evolution has been deduced by calculating $\Pi(\mathbf{q} \rightarrow 0, \omega)$ for various x values. We obtain the value of the off-diagonal electron-phonon interaction $g_{off}^{A_{1g}} \approx 3 \text{ meV}$ by comparison to the experimental data points. This value is an order of magnitude smaller than the diagonal contribution to the electron-phonon interaction. One sees that the phonon renormalization changes smoothly as a function of doping. The renormalization effects tend to vanish close to the band insulator regime at $x=1$ because all of the Fermi functions entering Eq. (7) are equal to one and $\text{Re } \Pi(\mathbf{q}, 0) = 0$. Away from this point $\text{Re } \Pi$ will be determined by the number of holes in the system, which explains the doping dependence of the renormalization effects. We also note that the electronic correlation effects seem to play a minor role for the renormalization of the Raman-active optical phonon modes. The situation may differ, however, for the acoustic phonons where the density of the states at the Fermi level plays the most important role.³⁴

Of course, our estimated value for the electron-phonon interaction corresponds to the interband scattering which is not directly related to the superconductivity. Therefore, we use our estimated value for the diagonal part of the electron-phonon interaction and obtain using the BCS formula $k_B T_c = 1.14 \omega_{A_{1g}} \exp[-1/N(E_F)g^{A_{1g}}]$, $T_c \sim 7 \text{ K}$ for the $N(E_F) \approx 4.0 \text{ states/eV}$.¹² This estimate is in agreement with the observed T_c in water intercalated Na_xCoO_2 , which points towards potential relevance of the electron-phonon interaction for the superconductivity in this compound. At the same time, the smooth evolution of the phonon frequencies as a function of the doping concentration on the one hand, and the rich phase diagram of the lamellar cobaltates on the other, requires further understanding the role of the phonons in the formation of superconductivity. One interesting possibility to explore is the possible enhancement of T_c due to resonance levels that may be introduced by the water

molecules.^{35,36} Very recently the so-called Suhl-Kondo resonance was suggested as an origin of the Cooper pairing in the $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$.³⁷ In any case a study of the isotope effect on T_c is highly desirable.

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