Electron-phonon interaction in the lamellar cobaltate Na\textsubscript{2}CoO\textsubscript{2}

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(Dated: February 3, 2008)

We study theoretically and experimentally the dependence of the electron-phonon interaction in Na\textsubscript{2}CoO\textsubscript{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.

PACS numbers: 74.70.-b, 74.25.Kc, 78.30.-j, 71.38.-k

Introduction. 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 \(T_c=4.6\text{K}\) in water intercalated sodium cobaltate, Na\textsubscript{2}CoO\textsubscript{2} \(\cdot\) \(y\)H\textsubscript{2}O \textsuperscript{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\textsubscript{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 \(x \approx 0.5\), and unusual magnetism for \(x \geq 0.6\) \textsuperscript{2}. There is also increasing experimental and theoretical evidence for unexpected strong correlation effects as the cobaltates approach the band insulating limit at \(x=1\) \textsuperscript{4, 5, 6, 7, 8, 9, 10, 11, 12}. In Na\textsubscript{2}CoO\textsubscript{2} the Na ions reside between the CoO\textsubscript{2} layers, with Co ions forming a triangular lattice, and donate \(x\) electrons to the partially filled Co-\(d(t_{2g})\) orbitals. Due to the presence of a trigonal crystalline electric field, the \(t_{2g}\) levels split into the higher lying \(a_{1g}\) singlet and the two lower lying \(e'_{g}\) states \textsuperscript{12}. Angle-Resolved Photo-Emission Spectroscopy (ARPES) \textsuperscript{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 \(\Gamma\) point and has mostly \(a_{1g}\) character. It has been argued that such an effect may arise due to strong electronic correlations \textsuperscript{8, 15}, or Na induced disorder \textsuperscript{10}, however, no consensus in the literature has been reached yet (see, for example, \textsuperscript{16, 17, 18, 19}).

Despite of intensive studies of the electronic and magnetic properties little is known about the phonon excitations and their doping evolution in Na\textsubscript{2}CoO\textsubscript{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\textsubscript{2}CoO\textsubscript{2} has been discussed in the context of its relevance to superconductivity and charge ordering on the basis of a \(t-V\) model \textsuperscript{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. Initially, the effect of renormalization of the optical phonons by the conduction electrons in layered cuprates has been considered in Ref. \textsuperscript{21}.

In this Rapid Communication we investigate the electron-phonon interaction in the Na\textsubscript{2}CoO\textsubscript{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 \( g_{eff} = 3 \text{meV} \). 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. Our results shed light on the possible role of the electron-phonon interaction in this compound.

**Experiment details.** Raman scattering experiments have been performed in quasi-backscattering geometry on freshly cleaved single crystal surfaces. The sample have been fully characterized using basic thermodynamic as well as spectroscopic techniques. After cleavage the crystals were rapidly cooled down in He exchange gas to prevent degradation. In \( \text{Na}_x \text{CoO}_2 \) in-plane \( E_{1g} \) and out-of-plane \( A_{1g} \) oxygen modes have been observed in Raman scattering and the corresponding oxygen displacements are depicted in Fig. 1(a) and (b). The Co site is not Raman-active. Modes of the Na sites have not been identified unambiguously. 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 cross-over from one to the other crystallographic phases (shaded areas) given by a different occupation of the Na sites leads for the \( A_{1g} \) modes to small additional frequency shifts and for the \( E_{1g} \) modes to new modes with a larger energy off-set. The latter are omitted for clarity. 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* LDA (Local Density Approximation) calculations using projection procedure for \( x = 0.33 \). The free-electron Hamiltonian of the \( t_{2g} \)-band model in a hole representation is given by

\[
H_0 = - \sum_{\mathbf{k},\sigma} (e^\sigma - \mu) n_{\mathbf{k}\sigma} - \sum_{\mathbf{k},\alpha,\sigma} \sum_{\mathbf{r}} \alpha^\beta d^\dagger_{\mathbf{k}\alpha\sigma} d_{\mathbf{k}\beta\sigma}, \tag{1}
\]

where \( n_{\mathbf{k}\sigma} = d^\dagger_{\mathbf{k}\sigma} d_{\mathbf{k}\sigma} \) is the annihilation (creation) operator for the \( t_{2g} \)-hole with spin \( \sigma \), orbital index \( \alpha \), and momentum \( \mathbf{k} \), \( t_{\mathbf{k}\alpha\beta} \) is the hopping matrix element, and \( e^\alpha \) is the single-electron energy. To obtain the dispersion we diagonalize the Hamiltonian calculating the chemical potential \( \mu \) self-consistently. Due to the non-zero inter-orbital 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 \( e^\alpha_k \) with the new orbital index \( \alpha' \).

**Electron-phonon interaction.** In analogy to previous considerations for cuprates, \( \text{Na}_x \text{CoO}_2 \), we derive the electron-phonon matrix elements, \( g_{\mathbf{q}\alpha\beta} \), for the \( A_{1g} \) and \( E_{1g} \) phonon modes depicted in Fig. 1(a) and (b). Namely, to obtain the main contribution to the diagonal (intra-band) part of the electron-phonon interaction we expand the Coulomb energy between Co and oxygen, \( H_C = \frac{e^2}{4\pi\epsilon_0} \sum_{\mathbf{r}} c^\dagger_{\mathbf{r}\alpha\sigma} c_{\mathbf{r}\alpha\sigma} \left( \frac{1}{\mathbf{R}_i - \mathbf{r}} + \frac{1}{\mathbf{R}_i - \mathbf{r} - \mathbf{q}} \right) \), in the small displacements of the oxygen ions. Here, \( e^\dagger \) is the electron charge, \( e^\ast = -2e \) is the oxygen ion charge, \( \epsilon \) 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 \( \gamma (\bar{\gamma}) \) labels the three oxygen positions within CoO\(_6\) unit cell above (below) the Co layer. Here, \( c_{\mathbf{r}\alpha\sigma}^\dagger \) refers to the diagonal form of the Hamiltonian. After introducing the creation (annihilation) operator \( b^\dagger_{-\mathbf{q}}(\mathbf{b}_{\mathbf{q}}) \) for the phonon with momentum \( \mathbf{q} \), we arrive to the following form of the electron-phonon interaction

\[
H_{el-\mathbf{p}h}^{diag} = \sum_{\mathbf{q},\alpha'\alpha} g^\alpha_{\mathbf{q}\alpha'} c^\dagger_{\alpha'\sigma} c_{\mathbf{k}-\mathbf{q}\alpha'(\mathbf{b}_\mathbf{q} + b^\dagger_{-\mathbf{q}}). \tag{2}
\]

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

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

\[ F_{E_{1g}}^{A_{1g}} = \cos \frac{q_1 - q_2}{3} - \frac{1}{2} \left[ \cos \frac{q_1 + q_3}{3} + \cos \frac{q_2 + q_3}{3} \right], \quad (4) \]

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.

\( g_{\text{diag}}^{\Gamma} = -\frac{\omega_0^*}{\sqrt{\omega_0^2 + \omega_0^*} \sqrt{\hbar M}} \)

where \( \omega_\Gamma \) 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, \( Na_{x=1}CoO_2 \), 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 [22]. The resulting momentum dependence of the structure factors for the both modes is shown in Fig. 1(c) and (d). Interestingly, one sees that while the \( g_q \) for the \( A_{1g} \) mode shows a maximum at the BZ center, the corresponding \( g_q \) for the \( E_{1g} \) mode vanishes there. Therefore, for \( q = 0 \) the electron-phonon coupling for the \( E_{1g} \) channel is zero. Taking \( \epsilon \sim 20 \) we estimate \( g_{\text{diag}}^{A_{1g}} \approx 0.05 \text{eV} \).

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

\[ H_{e-ph}^{\text{off}} = \sum_{k, q, \alpha, \beta, \gamma} g_{kq}^{\alpha \beta \gamma} \epsilon_{\alpha \epsilon} c_{\alpha k}^\dagger c_{\beta q}^{\gamma} (b_q + b_q^\dagger). \quad (5) \]

where \( g_{kq}^{\alpha \beta \gamma} = g_{\text{off}}^{\alpha \beta} F_{E_{1g}}^{\alpha \beta} (\gamma k) + (k + q) \) with \( \gamma(k) = \cos k_3 + \cos k_3 + \cos k_1 \) being the Co lattice structure factor. Again one could see that for \( q = 0 \) the off-diagonal electron-phonon coupling for the \( E_{1g} \) channel is zero due to the momentum dependence of the electron-phonon structure factor, \( F_{E_{1g}} \). Therefore, in Raman experiments which probes \( q = 0 \) 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 \textit{ab-initio} calculations [22]. The only Raman-active optical phonon mode which will couple to the conduction electrons at \( q = 0 \) is the \( A_{1g} \) mode.

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

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

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

\[ \Pi(q, \omega) = -2 \sum_{\alpha, \beta} \sum_{k} \left(g_{kq}^{\alpha \beta} \right)^2 \frac{f(\omega_{k+q}^{\alpha \beta}) - f(\omega_{k}^{\beta \gamma})}{\omega - \epsilon_{k+q}^{\alpha \beta} + \epsilon_{k}^{\beta \gamma} + i\delta}. \quad (7) \]

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 \( q \rightarrow 0 \) 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 \( g_{kq}^{\alpha \beta} \) while intra-band 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 \( \Pi(q \rightarrow 0, \omega) \) for various \( x \) values. We obtain the value of the off-diagonal electron-phonon interaction \( g_{\text{off}}^{A_{1g}} \approx 3 \text{meV} \) 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 one and Re$\Pi(q,0) = 0$. Away from this point Re$\Pi$ will be simply determined by the number of holes in the system. This explains the linear 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. 

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 $k_B T_c = 1.14 \omega_{A1g} \exp\left(-1/N(E_F)\gamma_{A1g}\right)^{T_c} \sim 7$ K for the $N(E_F) \approx 4.0$ states/eV [12]. This estimate is in agreement with the observed $T_c$ in water intercalated Na$_x$CoO$_2$, 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 the may be introduced by the water molecules. Very recently the so called Suhl-Kondo resonance was suggested as an origin of the Cooper paring in the Na$_x$CoO$_2 \cdot y$H$_2$O [37]. In any case a study of the isotope effect on $T_c$ is highly desirable.

We would like to thank P. Fulde, B. Keimer, I. Mazin, K. Morawetz, E. Schneider, A. Yaresko, and G. Zwicknagl for useful discussions. We thank P. Scheib and A. Boothroyd for contributions to our experimental study. M.M.K. acknowledges support from INTAS (YS Grant 05-109-4891) and RFBR (Grants 06-02-90537-BNTS). I.E. acknowledges support from Volkswagen Foundation. The experimental studies have been supported by DFG and ESF-HFM.

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