Resonant ultrasound studies of the layered perovskite system $Ca_{2-x}Sr_xRuO_4$

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The elastic response of the layered perovskite system $Ca_{2-x}Sr_xRuO_4$ ($0.2 \leq x \leq 2$) has been studied as a function of temperature and doping concentration x using resonant ultrasound spectroscopy. The elastic constants c_{11} and c_{44} have been obtained for three polycrystalline samples ($x=1.0$, 0.5, and 0.3) and show a softening trend with increasing Ca-content. In addition, the temperature-dependence of the elastic response of five single-crystals ($x=2.0$, 1.9, 0.5, 0.3, and 0.2) has been measured. For $2.0 \geq x \geq 0.5$, a dramatic softening over a wide temperature range is observed upon cooling, which is attributed to the rotational instability of RuO_6 octahedra (for $x=2.0$ and 1.9) and the static rotation of the octahedra (for $x=0.5$). For the Ca-rich samples ($x=0.3$ and 0.2), the softening occurs in a very narrow temperature range, corresponding to the structural phase transition from high-temperature tetragonal to low-temperature orthorhombic symmetry.
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I. INTRODUCTION

Sr_2RuO_4, the $n=1$ member of the Ruddlesden–Popper series $Sr_{n+1}Ru_nO_{3n+1}$, is the only copper-free superconductor that shares the layered perovskite structure with the high-T_c superconductor $La_{2-x}Ba_xCuO_4$. The discovery of exotic superconductivity in this compound by Maeno *et al.*[1] in 1994 has generated extensive research on all aspects of Sr_2RuO_4 (Refs. [2–7]) and its doped varieties. The $Ca_{2-x}Sr_xRuO_4$ series, obtained by the isovalent substitution of Ca for Sr, has undoubtedly received the most attention. It is a Mott transition system, connecting the Mott insulator Ca_2RuO_4 with the spin-triplet superconductor Sr_2RuO_4 and exhibits a variety of physical properties for different x values. In 2000, the first experimental phase diagram of $Ca_{2-x}Sr_xRuO_4$ was reported by Nakatsuji *et al.*[8] and is schematically illustrated in Fig. 1. The phase diagram is known for its rich behavior, including an antiferromagnetic transition, a metal-insulator transition, a structural transition, and a ferromagnetic instability near $x \sim 0.5$. Unlike high-T_c cuprates, chemical doping of Sr_2RuO_4 eliminates superconductivity, as it is extremely sensitive to impurities.[3] Later, the magnetic phase diagram of $Ca_{2-x}Sr_xRuO_4$ (Ref. [9]) was deduced from first-principles calculations and qualitatively explained the experimental phase diagram. Therefore, this is a prototype system for studying interplay between structural, electronic, and magnetic properties.

In an attempt to obtain a better understanding of the lattice dynamics of $Ca_{2-x}Sr_xRuO_4$, we have initiated a study of the elastic response of several members of the series. The elastic response is amongst a material's most important physical properties, with the elastic moduli directly related to the inter-atomic bonding and to quantities such as the velocity of sound. Measurements of the temperature-dependence of the elastic moduli can yield valuable information to fully understand the thermodynamics of a material, especially in the vicinity of phase transitions. So far, there are only a few reports[10–14] on the elastic properties of Sr_2RuO_4, and there are no reports on the elastic properties of other compositions in the $Ca_{2-x}Sr_xRuO_4$ series. The full elastic tensor of Sr_2RuO_4 single-crystal was measured at room temperature by Paglione *et al.*[12] Transverse ultrasonic measurements were performed on a single-crystal of Sr_2RuO_4 by Okuda *et al.*[14] across the superconducting transition temperature $T_c (\sim 1.40$ K), using the pulse-echo technique. Upon cooling, a jump-like decrease in T_c was found in the transverse elastic modulus c_{66}, which was attributed to the coupling between the strain and the two-dimensional superconducting order parameter (OP) with broken time-reversal symmetry.

In this paper, we present results, for the first time, on the elastic response of doped $Ca_{2-x}Sr_xRuO_4$ with $0.2 \leq x \leq 2$ as a function of temperature between 5 and 300 K. The elastic constants c_{11} and c_{44} have been obtained at room temperature

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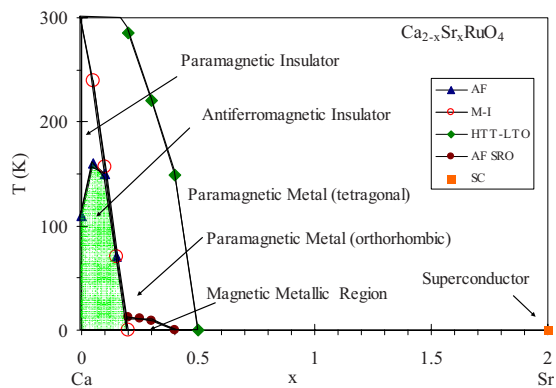b)Present address: Louisiana State University, Baton Rouge, Louisiana 70803.

FIG. 1. (Color online) Phase diagram of $Ca_{2-x}Sr_xRuO_4$, based on data from Ref. 8.

FIG. 2. (Color online) A typical resonance spectrum measured by RUS.

for three polycrystalline samples ($x=1.0$, 0.5, and 0.3) and show a softening trend with increasing Ca-content. In addition, we have measured the temperature dependence of the elastic response of five single-crystals ($x=2.0$, 1.9, 0.5, 0.3, and 0.2), probing the high-temperature tetragonal to low-temperature orthorhombic phase transition.

II. EXPERIMENTAL DETAILS

Polycrystalline rods were prepared using a standard solid-state reaction with $CaCO_3$ (99.9995% Alfa Aesar), $SrCO_3$ (99.9995% Alfa Aesar), and RuO_2 (99.99% Alfa Aesar) powders as starting materials. These powders are mixed with the appropriate molar ratio, depending on the composition of the polycrystal to be made. After the mixture is well ground in a vibratory micro mill for about 1 h, the mixture is pressed into cylinder-shaped rods with a typical diameter of 5 mm and 120 mm length. The resulting polycrystalline rods are subsequently sintered at 950 °C in air for 24 h.

Single-crystals of $Ca_{2-x}Sr_xRuO_4$ are grown via the floating zone technique. A major advantage of this method is that it is crucible-free, avoiding contamination from the crucible material. A polycrystalline rod is used as the feed rod for the single-crystal growth in an NEC SC-M15HD image furnace. The seed rod can be a polycrystal or a single-crystal. During the growth, both the feed rod and the seed rod rotate in the opposite direction with the same rotation rate and the single crystal is grown in a mixed oxygen/argon atmosphere.

The current study focuses on the elastic response of the $Ca_{2-x}Sr_xRuO_4$ system, measured using resonant ultrasound spectroscopy (RUS).[15–18] RUS is a powerful method for determining the elastic moduli of a solid, based on measurements of the mechanical resonances of a sample. Whereas these "free vibrations" can easily be calculated for a sample with known elastic tensor and well-defined shape and dimensions, the RUS experiment works "backwards," and the sample's resonances are measured under nearly free boundary conditions. An iteration procedure is used to "match" the measured lines with the calculated spectrum. For samples with irregular shape or symmetry lower than orthorhombic, the procedure for calculating the elastic moduli from the resonances can be quite challenging, but RUS measurements can give important information even when it is not possible to obtain an absolute value for the elastic constants: any de-
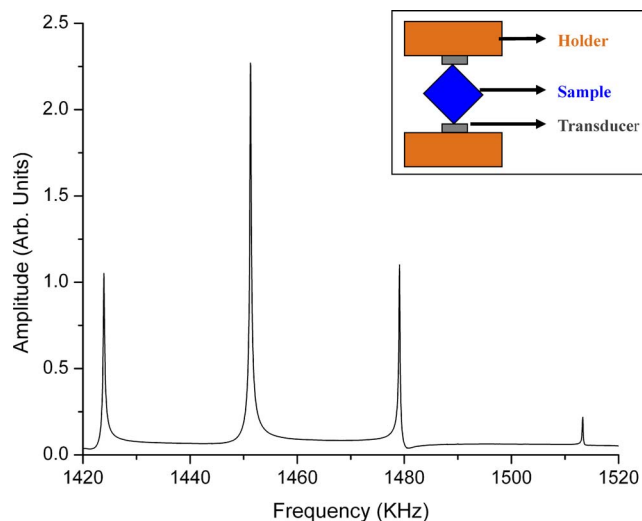
viation from "normal" thermodynamic behavior will be reflected in the temperature-dependence of the resonant frequencies, and these can be measured regardless of sample shape or symmetry. In a RUS measurement, two transducers are used, one to excite the resonances and the other to detect the response of the sample. The frequency is swept within a typical range 200–2000 KHz, and a large response will be detected when the frequency corresponds to one of the sample's eigenfrequencies. Figure 2 shows a typical resonance spectrum measured using RUS and the inset shows a schematic drawing of the sample and transducer set-up.

The RUS data reported here were carried out as a function of temperature (5–300 K) using a custom designed probe that was inserted in a commercial Quantum Design Physical Properties Measurement System (PPMS). For our study, three $Ca_{2-x}Sr_xRuO_4$ polycrystals with $x=1.0$, 0.5, and 0.3 were cut and polished into rectangular parallelepipeds (RPs) and their elastic constants were determined from measurements of the resonant frequencies. In addition, the temperature-dependence of the resonant frequencies were measured on $Ca_{2-x}Sr_xRuO_4$ single-crystals with five different compositions, i.e., $x=2.0$, 1.9, 0.5, 0.3, and 0.2. The shape and small size of these samples prevented crystallographic orientation, but valuable information can be obtained from the temperature-dependence of the frequencies, as explained below. RUS measurements were carried out upon both cooling and warming, and no difference was observed. The results presented here are based on the cooling data.

III. RESULTS AND DISCUSSION

A. $Ca_{2-x}Sr_xRuO_4$ polycrystals

Polycrystalline materials are elastically isotropic and have two independent elastic moduli: the longitudinal modulus c_{11} and the shear modulus c_{44}. Table I lists the experimental values for the elastic moduli obtained at room temperature using RUS for $Ca_{2-x}Sr_xRuO_4$ polycrystals with $x=1.0$, 0.5, and 0.3. Also listed in Table I are the calculated polycrystalline moduli for the parent compound Sr_2RuO_4 ($x=2$), using the Voigt approximation.[19] The latter allows

TABLE I. Elastic constants of $Ca_{2-x}Sr_xRuO_4$ polycrystals at room temperature. The calculated values are corrected for porosity, as explained in the text.

Composition	C_{11} (10^{11} Pa)		C_{44} (10^{11} Pa)	
	Exp.	Corrected	Exp.	Corrected
Sr_2RuO_4	2.1883[a]		0.6679[a]	
$CaSrRuO_4$	1.008	1.946	0.307	0.534
$Ca_{1.5}Sr_{0.5}RuO_4$	0.459	0.905	0.176	0.350
$Ca_{1.7}Sr_{0.3}RuO_4$	0.364	0.722	0.155	0.320

[a]Calculated by using single-crystal values in Ref. 12.

computation of the shear and bulk modulus of a polycrystalline solid based on the values of elastic moduli of the single-crystal, under the assumption that the stress is uniform everywhere within the sample. The general expressions for the Voigt approximation are as follows.

Bulk modulus,

$$B = \frac{1}{9}(C_{11} + C_{22} + C_{33}) + \frac{2}{9}(c_{12} + c_{23} + c_{13}). \quad (1)$$

Shear modulus:

$$G = \frac{1}{15}(c_{11} + c_{22} + c_{33}) - \frac{1}{15}(c_{12} + c_{23} + c_{13})$$
$$+ \frac{1}{5}(c_{44} + c_{55} + c_{66}). \quad (2)$$

The c_{11} and c_{44} values of Sr_2RuO_4 polycrystal are calculated from the data reported in Ref. 12, using the above Voigt equations together with the polycrystalline relations $c_{44} = G$ and $c_{11} = B + (4G/3)$.

Before comparing the elastic moduli found for the various compositions, we need to point out that the density of the polycrystals is only about 70% of the theoretical density. As the elastic moduli of a solid depend on its density, a meaningful comparison of the experimental values requires that they are corrected to zero porosity. Following the model of Ledbetter and Datta[20] for spherical inclusions in a matrix and Chandra Sekhar et al.'s[21] treatment of a porous ceramic as a composite material containing spherical voids, the moduli for zero porosity $Ca_{2-x}Sr_xRuO_4$ polycrystals are estimated, and included in Table I. Figure 3 illustrates how the values of the elastic constants decrease with decreasing Sr-content.
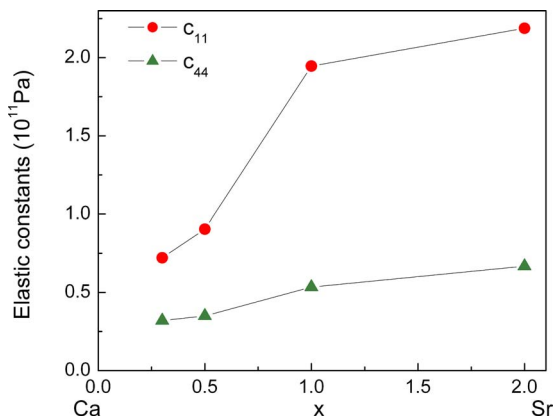

FIG. 3. (Color online) Elastic constants (corrected for porosity) versus composition x in $Ca_{2-x}Sr_xRuO_4$ polycrystals. The error for c_{11} is less than 1.5% and less than 0.3% for c_{44}.
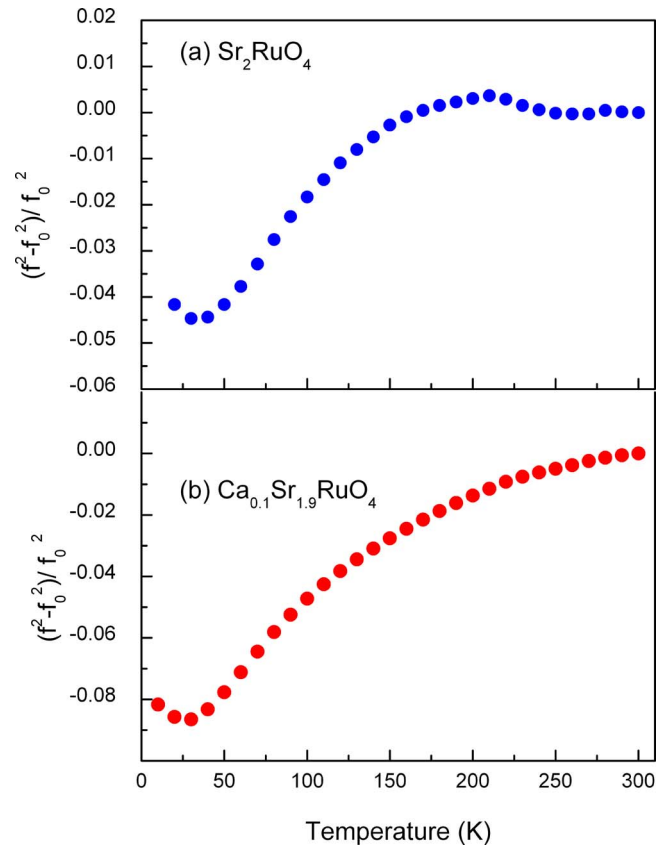
FIG. 4. (Color online) Relative change in representative squared resonant frequencies vs temperature for Sr_2RuO_4 (a) and $Ca_{0.1}Sr_{1.9}RuO_4$ (b).

Since Ca atoms are smaller than Sr atoms, the replacement of Sr by Ca atoms in Sr_2RuO_4 will cause both randomness in SrO layers and lattice distortion in RuO_2 layers, and thus induce a decrease in elastic constants.

B. $Ca_{2-x}Sr_xRuO_4$ single-crystals

The resonant frequencies of five $Ca_{2-x}Sr_xRuO_4$ single crystals ($x = 2.0$, 1.9, 0.5, 0.3, and 0.2) were measured between 5 and 300 K (the $x = 0.2$ composition was measured up to 350 K). The squared resonant frequencies are directly proportional to the elastic moduli[16,17] and any irregularity in the elastic response is therefore reflected in the temperature-dependence of the resonant frequencies.

The temperature dependence of representative squared resonant frequencies for the five single-crystals is plotted in Figs. 4 and 5. A significant softening is apparent in the behavior of all five crystals, but whereas this softening is rather gradual and spans more than 100 K in the compounds with $x > 0.5$, it is more abrupt in the samples with high Ca-content ($x \leq 0.5$).

Starting with the parent compound, Sr_2RuO_4, a gradual softening of about 5% is observed upon cooling, reaching a minimum around 40 K, as shown in Fig. 4(a). While it may be tempting to attribute this softening to the superconductivity in this material, the two are most likely unrelated, since a similar—albeit more significant—softening is observed in the slightly Ca-doped (and non-superconducting) $Ca_{0.1}Sr_{1.9}RuO_4$ [Fig. 4(b)]. In contrast to La_2CuO_4, where

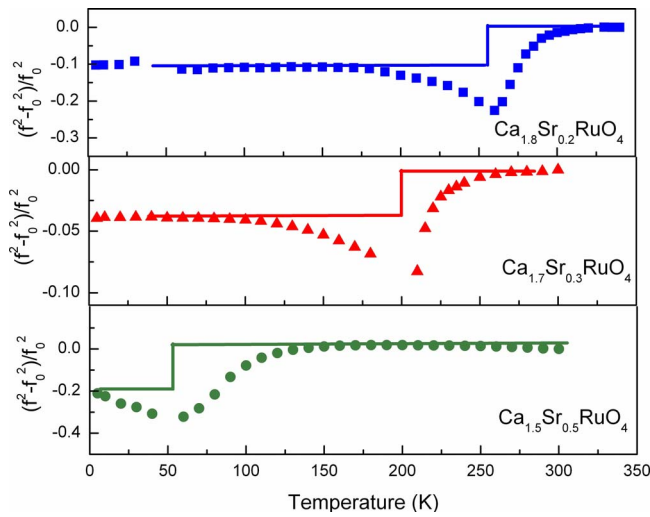FIG. 5. (Color online) Relative change in representative squared resonant frequencies vs temperature for $Ca_{1.5}Sr_{0.5}RuO_4$ (a), $Ca_{1.7}Sr_{0.3}RuO_4$ (b), and $Ca_{1.8}Sr_{0.2}RuO_4$ (c).
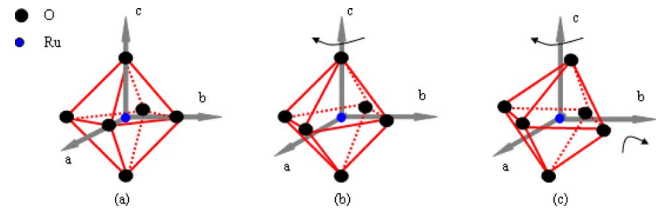

FIG. 6. (Color online) The RuO_6 octahedron in different tilt and rotational distorted configurations: (a) no distortion, (b) rotation around c-axis, and (c) combined tilt and rotational distortion.

tilting of the Cu–O octahedra leads to a tetragonal-to-orthorhombic phase transition, neutron diffraction on single-crystals of Sr_2RuO_4 confirms that its K_2NiF_4-type structure remains undistorted.[22] Thus the observed softening of resonant frequencies in Sr_2RuO_4 upon cooling cannot be attributed to a structural transition. Instead, we believe that the softening is due to a rotational instability of RuO_6 octahedra around the c-axis. Even though neither tilting nor rotation of RuO_6 is revealed in Sr_2RuO_4, large atomic displacement parameters[22] (ADPs) indicate that Sr_2RuO_4 is close to a rotational instability. Such instability has indeed been observed in the phonon dispersion relation of Sr_2RuO_4, using inelastic neutron scattering.[23] The rotation of the RuO_6 octahedra corresponds to a Σ_3 mode, whose second optical branch along [110] direction is found to soften continuously in the Brillouin zone.[23] This softening interacts with the first optical branch and the acoustic branch of Σ_3, leading to a sharp drop in the acoustic branch near the zone boundary. This steep drop in the dispersion relation of the Σ_3 mode is an indication of a rotational instability, which we believe to be the cause of the observed softening of the resonant frequencies in Sr_2RuO_4 and $Ca_{0.1}Sr_{1.9}RuO_4$ upon cooling. With increasing Ca-concentrations, the instability develops into a static rotation of RuO_6 that distorts the $I4/mmm$ symmetry and results in $I4_1/acd$ symmetry when the Sr-content decreases below $x = 1.5$.[24]

Plotted in Fig. 5(a) is the temperature-dependence of a representative squared resonant frequency for $Ca_{1.5}Sr_{0.5}RuO_4$ ($x = 0.5$). The softening in this sample is more pronounced, and happens in a narrower temperature region than the softening observed in Sr_2RuO_4 and $Ca_{0.1}Sr_{1.9}RuO_4$. This indicates that a different mechanism is most likely responsible for the sudden drop in resonant frequency, observed below 150 K in $Ca_{1.5}Sr_{0.5}RuO_4$. $Ca_{1.5}Sr_{0.5}RuO_4$ is known to be at the verge of a structural phase transition, from the tetragonal $I4_1/acd$ symmetry to orthorhombic $Pbca$ symmetry. As the Ca-content increases, a tilt of RuO_6 octahedra develops in addition to the already present rotation. This tilt around an

axis parallel to the ab plane deepens the lattice distortion and leads to the structural phase transition in the Ca-rich compounds ($x < 0.5$). Figure 6 illustrates the RuO_6 octahedron in different tilt and rotational configurations.

For $x = 0.5$, the transition is believed to have quantum-critical character.[8] A quantum-critical phase transition takes place at the absolute zero of temperature, where crossing the phase boundary means that the quantum ground state of the system changes in some fundamental way.[25] As shown in the phase diagram (Fig. 1), the temperature of the transition increases rapidly with increasing Ca-content. Figures 5(a)–5(c) illustrate how the gradual softening below 100 K, indicating the proximity of a phase-transition in $Ca_{1.5}Sr_{0.5}RuO_4$, develops into a sharp decrease in a very narrow temperature range in $Ca_{1.7}Sr_{0.3}RuO_4$ ($x = 0.3$) and $Ca_{1.8}Sr_{0.2}RuO_4$ ($x = 0.2$). The minimum in the resonant frequencies corresponds to a transition temperature T_c of 50 K for $Ca_{1.5}Sr_{0.5}RuO_4$, 210 K for $Ca_{1.7}Sr_{0.3}RuO_4$, and 260 K for $Ca_{1.8}Sr_{0.2}RuO_4$, respectively. Note that $T_c > 0$ K for $Ca_{1.5}Sr_{0.5}RuO_4$, indicating that the actual Ca content in our sample is slightly greater than 1.5. Nevertheless, these transition temperatures agree well with the phase diagram of $Ca_{2-x}Sr_xRuO_4$ and illustrate how RUS measurements probe structural phase transitions. In addition to the softening, a significant broadening of the resonance peaks is observed in the vicinity of the transition, indicating considerable ultrasonic attenuation.

The sharp softening of resonant frequencies across the structural transition is due to the coupling between the elastic strain and the order parameter, i.e., the tilt angle of the RuO_6 octahedra. The Landau theory of structural phase transitions predicts a step-like decrease in the elastic moduli when approaching T_c from high temperatures, as observed in $Ca_{1.7}Sr_{0.3}RuO_4$ and $Ca_{1.8}Sr_{0.2}RuO_4$, when the coupling is linear in strain and quadratic in order parameter.[26] The deviation from true step-like behavior can be attributed to the thermal fluctuations or some imperfections in the crystals. A similar behavior of the resonant frequencies was also revealed across the tetragonal-to-orthorhombic transition in the cuprates $La_{2-x}Sr_xCuO_4$ (Ref. 27) and in the pyrochlore $Cd_2Re_2O_7$.[28]

Whereas the above results clearly illustrate the ability of RUS to qualitatively probe instabilities and phase transitions in a crystal lattice, a full quantitative analysis requires measurements on oriented single-crystals. Since the resonant frequencies represent a mixture of the elastic moduli, i.e., a given frequency typically depends on a combination of elastic constants, it is not possible to evaluate the temperature-dependence of individual moduli at this time. Efforts to grow

larger single-crystals that can be oriented are currently under way, and a quantitative study of the elastic response of these crystals is planned for the future.

IV. CONCLUSIONS

In summary, we report the elastic constants c_{11} and c_{44} for three $Ca_{2-x}Sr_xRuO_4$ polycrystals (x=1.0, 0.5, and 0.3). It is found that the replacement of Sr by smaller Ca atoms causes a decrease in elastic constants. The temperature-dependence of the resonant frequencies for five $Ca_{2-x}Sr_xRuO_4$ single-crystals (x=2.0, 1.9, 0.5, 0.3, and 0.2) all show softening, due to rotation and/or tilts of the Ru–O tetrahedral, and clearly illustrate the ability of RUS to qualitatively probe instabilities and phase transitions in a crystal lattice. The gradual softening of resonant frequencies over a large temperature span in single-crystals Sr_2RuO_4 (x=2) and $Ca_{0.1}Sr_{1.9}RuO_4$ (x=1.9) is caused by a rotational instability in the lattice. The sharp softening of resonant frequencies over a very narrow temperature range for $Ca_{1.7}Sr_{0.3}RuO_4$ (x=0.3) and $Ca_{1.8}Sr_{0.2}RuO_4$ (x=0.2) corresponds to the tetragonal to orthorhombic structural phase transition, which involves both rotation and tilting of RuO_6 octahedra.

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