

Temperature dependence of elastic properties of NiO single crystal studied by resonant ultrasound spectroscopy

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1. Introduction

Nickel oxide (NiO) is a typical antiferromagnet known for more than half a century. The magnetic phase transition temperature, called the Néel point (T_N), is 523 K. NiO is antiferromagnetic at room temperature and paramagnetic above T_N .¹⁾ Antiferromagnets have attracted great attentions for a high-density memory device, therefore, basic properties of NiO have been keenly studied.

However, the elastic constants of NiO remain unclear. Elastic constants are important to design structures and analyze stress and strain. They are also one of the indices for nondestructive evaluation because they reflect defects and residual stress. Furthermore, elastic constants are crucial parameters for the thermal dynamics of condensed matter materials; they relate to the curvature of the interatomic forces, representing the stability of the material and relating to many other thermal parameters. Although the elastic constants of NiO have been measured by various methods, they vary widely in the literature:²⁻⁵⁾ For example, reported Young's modulus at room temperature varies between 90 and 165 GPa.^{2,6)} Furthermore, temperature dependence of the elastic constants across T_N is also interesting and contradictory. Some components of elastic constants increase more than 100% above T_N , however, reported temperature dependences show different behaviors quantitatively and qualitatively.

Such differences may be due to the magnetostrictive effect of NiO. In the paramagnetic state, NiO shows the NaCl-type structure without the spins ordering.⁷⁾ In the antiferromagnetic state, the spins are parallel on one of four-types $\{111\}$ planes and antiparallel on the neighboring $\{111\}$ planes. The interaction between these spins induces the compressive spontaneous magnetostriction of $\sim 2 \times 10^{-5}$ in the corresponding $\langle 111 \rangle$ direction,⁸⁻¹⁰⁾ resulting in rhombohedral structure. This strain forms domains in NiO called as the twin wall (T-wall). In a $\{111\}$ plane, spins parallelly align three directions $[211]$, $[1\bar{2}1]$, or $[11\bar{2}]$, making the spin wall (S-wall).¹¹⁻¹⁴⁾ Since these domains differ from specimen to specimen, the reported values of elastic constants may change.

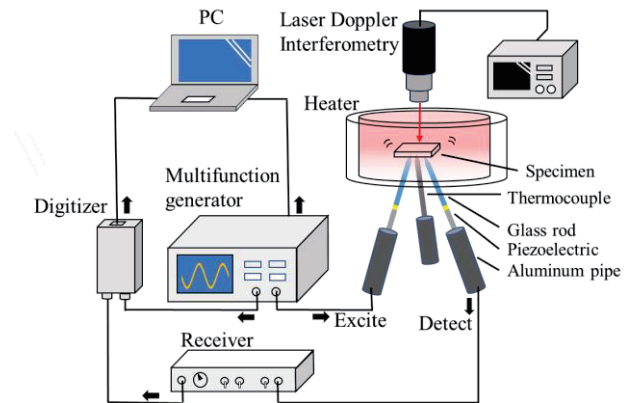


Fig. 1 Schematic of the tripod RUS measurement system with laser Doppler interferometry.

In this study, we clarify the elastic constants and their relationship to magnetic structures of NiO by measuring the temperature dependence of resonance frequencies and acoustic damping of various vibration modes using resonant ultrasound spectroscopy (RUS). We use the tripod RUS measurement method combining the laser Doppler interferometer (LDI) system to accurately measure the resonance frequencies and to confirm their vibration modes.^{15,16)} To carefully determine the elastic constants, we use four specimens whose sizes, face planes, and synthesis conditions are different.

2. Experiment

The RUS method can determine all of the elastic constants and acoustic damping from a single specimen.¹⁷⁾ We show the experimental setup in **Fig. 1**, which composes the tripod RUS measurement system with a heater and an LDI system. In this method, we can excite ideal free vibration because of no acoustic coupling agent and no external force except the gravity. Furthermore, the contact area between the piezoelectric transducer and the specimen is minimized, enabling us to accurately measure the acoustic damping in the specimen. Here, we fabricate needle-type transducers (pinducers) using piezoelectric rods, glass rods, and aluminum pipes.

The tripod RUS consists of three supporting needles, where we use two pinducers and one thermocouple to measure the specimen temperature. We apply continuous sinusoidal wave to a pinducer

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and detect the specimen's vibration from the other pinducer. By sweeping the frequency of the applied wave, the vibration amplitude takes local maximums at resonance frequencies of the specimen. These resonance frequencies depend on dimensions, mass density, shape, crystal-axis orientation, and elastic constants of the specimen. Therefore, we can calculate the resonance frequencies using estimated elastic constants with known dimensions, mass density, specimen shape, crystal-axis orientation. By comparing the calculated and measured resonance frequencies, the elastic constants of the specimen can be determined using an inverse calculation. In this inverse calculation, we must identify the resonance modes and compare the frequencies between the same modes in the calculation.¹⁴⁾ Therefore, we use the LDI method to observe the vibration distribution of the specimen surface at each resonance mode. We compare the measured vibration distribution with the calculated one. The LDI measurement also enables us to precisely detect resonance vibration of the specimen without noise from the detection-pinducer resonance.

3. Results and Discussion

Figure 2 shows the resonance spectra of NiO measured by pinducer and LDI at 298 and 655 K. The signal-to-noise ratio of the LDI-detection measurement is larger than that of pinducer-detection measurement. Moreover, the intensity of the LDI spectra changes with measurement point, and some resonance modes cannot be measured at different measurement points, insisting on selectivity for eigen resonance modes.

Measured resonance frequencies and Q values largely increase above T_N . For example, the first resonance frequency changes from 86 to 129 kHz as the temperature increases. The number of observable resonance frequencies is also different; we determine the resonance frequencies about 30 and 50 peaks at 298 and 655K, respectively. From the measured resonance frequencies and dimensions, we calculate the elastic constant at each temperature. In addition, the spectrum measured at 298 K has broad resonance peaks, indicating a large energy loss. For example, as the temperature increases, Q value of the first resonance peak increases by $\sim 1600\%$. These results indicate that the antiferromagnetic ordering strongly affects elastic constants of NiO.

4. Conclusion

In this study, we measure the elastic properties of NiO using the tripod RUS method with the LDI system. We succeed in accurately identifying the vibration modes by comparing the measured and calculated vibration distributions. Resonance

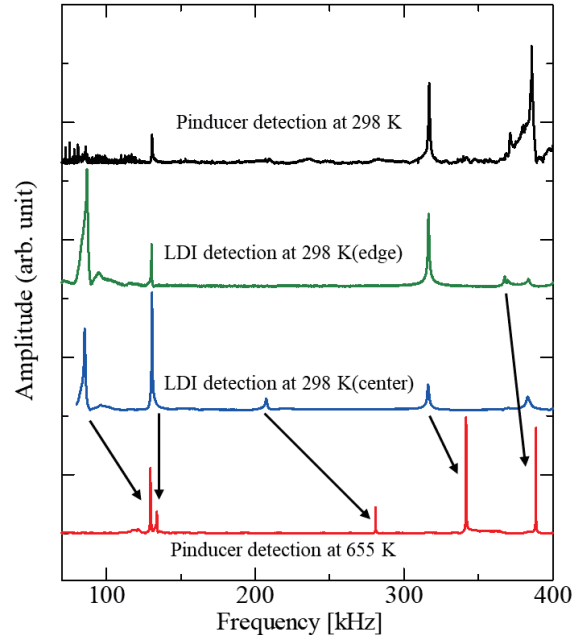


Fig. 2 Resonance spectra of NiO measured by the two methods; black line is measured by pinducer at 298 K. Green and blue lines are measured by the LDI system irradiated to the edge and center, respectively at 298 K. Red line is measured by pinducer at 655 K.

frequencies significantly increase above T_N , indicating that antiferromagnetic ordering strongly affects the elastic constants of NiO. Q values also increase above T_N . We will further analyze and discuss their relationship to the magnetic structure.

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