

## Surface acoustic wave generation and detection in the quantum paraelectric regime of SrTiO<sub>3</sub>-based heterostructures

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Strontium titanate (STO), apart from being a ubiquitous substrate for complex-oxide heterostructures, possesses a multitude of strongly coupled electronic and mechanical properties. Surface acoustic wave (SAW) generation and detection offers insight into electromechanical couplings that are sensitive to quantum paraelectricity and other structural phase transitions. Propagating SAWs can interact with STO-based electronic nanostructures, in particular LaAlO<sub>3</sub>/SrTiO<sub>3</sub> (LAO/STO). Here, we report the generation and detection of SAW within LAO/STO heterointerfaces at cryogenic temperatures ( $T \geq 2$  K) using superconducting interdigitated transducers. The temperature dependence shows an increase in the SAW quality factor that saturates at  $T \approx 8$  K. The effect of backgate tuning on the SAW resonance frequency shows the possible acoustic coupling with the ferroelastic domain wall evolution. This method of generating SAWs provides a pathway towards the dynamic tuning of ferroelastic domain structures, which are expected to influence the electronic properties of complex-oxide nanostructures. Devices that incorporate SAWs may in turn help to elucidate the role of ferroelastic domain structures in mediating electronic behavior.

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**Introduction.** Strontium titanate (STO) holds a unique place among the growing family of complex-oxide heterostructures and nanostructures [1]. Apart from possessing a wealth of physical phenomena—ferroelectricity [2,3], ferroelasticity [4,5], superconductivity [6–8], high spin-to-charge interconversion [9], large third-order optical susceptibility [10]—STO also exhibits fascinating transport properties at interfaces and within conductive nanostructures [11,12]. These latter properties arise when STO is capped with a thin layer, often but not exclusively LaAlO<sub>3</sub> (LAO), which results in electron doping near the STO interface [13]. Conductive nanostructures of many types have been created by “sketching” with a conductive atomic force microscope (c-AFM) tip [14] or an ultralow-voltage focused electron beam [15]. The properties of these devices are profoundly affected by the intrinsic behavior of STO and are in many aspects not well understood.

One of the least well-understood property interrelationships concerns the coupling between electronic, ferroelectric, and ferroelastic degrees of freedom. STO is centrosymmetric at room temperature with ABO<sub>3</sub> cubic perovskite structure. Upon cooling below  $\sim 105$  K [16,17], STO undergoes a cubic-to-tetragonal antiferrodistortive (AFD) phase transition [18]. Further cooling to  $\sim 35$  K [19] gives rise to an incipient ferroelectric or “quantum paraelectric” phase transition (QPE) in which the dielectric constant  $\epsilon$  diverges before saturating below 10 K [20]. At cryogenic temperature ( $T < 10$  K),

STO shows giant piezoelectricity even larger than the best well-known piezoelectric material such as quartz [21]. At microscopic scales, the coupling between polar phases in STO and ferroelastic domains [22] is quite strong and can be directly observed using scanning single electron transistor microscopy [5]. Piezoelectric distortions were found to be the result of reorienting tetragonal domains, whose in-plane and out-of-plane lattice constants differ by  $\sim 10^{-3}$ .

Surface acoustic waves (SAWs), also known as Rayleigh waves [23], arise from linear piezoelectric coupling, and propagate parallel to the sample surface with its depth comparable to the SAW wavelength. SAW propagation is sensitive to both mechanical and electrical changes at the sample surface, making it surface sensitive and useful for radio-frequency (rf) signal processing. A common technique to generate and detect SAWs is to apply rf signals to a pair of metallic interdigitated transducers (IDTs). However, the complexity and subtlety of the STO structure with multiple phases make SAW generation and detection difficult to achieve. With STO, dc fields have been used to break cubic symmetry and generate polarization above 150 K [24,25] via an electrostrictive effect. To generate SAWs, an extra piezoelectric layer, lead zirconate titanate (PZT), was deposited on top of LAO [26], and SAWs were observed down to  $T = 110$  K. Below this temperature, the signal disappeared and SAW generation and detection has not to our knowledge been reported in STO or LAO/STO or at temperatures lower than  $T = 105$  K.

In this Letter, we demonstrate direct SAW generation and detection on LAO/STO surfaces at cryogenic temperatures using superconducting IDTs. The linearly coupled SAW

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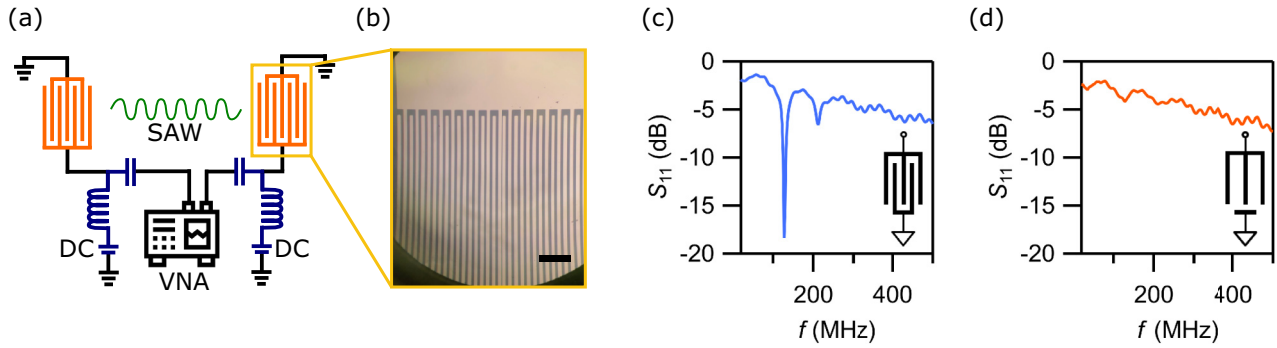


FIG. 1. Surface acoustic wave (SAW) generated and detected by interdigitated transducers (IDTs). (a) Schematic diagram of the experiment setup. The orange parts are IDTs. Blue circuits denote the bias tee inserted between the vector network analyzer (VNA) port and IDT. (b) Optical image of an IDT patterned with NbTiN. The black scale bar is 20  $\mu\text{m}$ . (c)  $S_{11}$  from an experiment device with normal IDT comb structure in a pair. The resonances are observable. (d) Reflection signal  $S_{11}$  from a control device without one side of the IDT comb structure. There is no resonance observed from this control device.

shows an ultralow phase velocity, indicating softening of the STO crystal at low temperature and is consistent with earlier reports of large piezoelectric and electrostriction coefficients [21]. The temperature at which the quality factor  $Q$  of the SAW resonator saturates coincides with the quantum-paraelectric transition temperature ( $T_{QPE}$ ), showing that  $Q$  is coupled to the dielectric constant and can be used to identify the onset of the quantum paraelectric phase. The resonance frequency can be tuned with a backgate voltage. The tunability with negative backgate voltages, but not positive voltages, is consistent with the tuning of ferroelastic domains [5], providing evidence of coupling between ferroelastic domains and surface phonons. An applied dc bias confirms the electrostrictive effect from STO by showing the quadratic tuning behavior.

**Experiment.** LaAlO<sub>3</sub> epitaxial films were grown on TiO<sub>2</sub>-terminated STO (001) substrates by pulsed laser deposition [13]. The thickness of LAO is fixed to 3.4 u.c., and is precisely controlled by *in situ* high-pressure reflection high-energy electron diffraction (RHEED) [27,28], close to the critical thickness of metal-insulator transition [29] (for details of sample growth, see the Supplemental Material [30]). To form a uniform-type single electrode IDT, an 80-nm-thick film of NbTiN is deposited on top of LAO/STO. IDT fingers are oriented along the (010) direction. Superconducting NbTiN is chosen as the IDT material for three principal reasons: (i) to help with impedance matching; (ii) to maximize the transmission; and (iii) to minimize ohmic losses and heating. A metallization ratio  $m$ , defined as the finger width divided by the finger spacing,  $m \equiv w/(w + d)$ , is fixed such that  $m = 0.5$  in all devices. SAW-related experiments are carried out in a physical property measurement system (PPMS) at temperatures  $T \geq 2$  K. Each IDT is grounded on one side, and the other side is connected to an input port of a vector network analyzer (VNA) to enable two-port scattering parameter measurements [Fig. 1(a)]. Between the IDT and the VNA, a bias tee is inserted on each side to allow a dc bias  $V_{\text{bias}}$  to be applied between the IDT fingers. SAWs are generated by an IDT, transmitted along the (100) direction, and detected by the second IDT pair. To reduce contributions from bulk acoustic waves, the LAO/STO sample bottom surface is roughened and coated with silver epoxy as a “soft conductor” [31]. The

bottom conducting electrode is also used to apply a voltage  $V_{\text{bg}}$  from the back of the STO substrate.

By applying a signal with power  $P = -10$  dBm to the IDT, a clear resonant feature can be seen at 127.5 MHz in the reflection spectrum  $S_{11}$  [Fig. 1(c)], defined as the center frequency  $f_c$ . By contrast, in a control device in which one side of the two comb structures in the IDT is missing there is no resonance [Fig. 1(d)], demonstrating that the resonance feature is a result of the paired comb structures patterned with NbTiN, and not due to bulk acoustic wave transmission or an electrical resonance from the cable or other parts of the instrument. Meanwhile, the SAW phase velocity is obtained from the measured  $f_c$  by  $v = f_c \lambda$ . The wavelength  $\lambda$  is determined by the distance between a pair of nearest IDT fingers with the same polarity. Here, we have  $\lambda = 8$   $\mu\text{m}$ , giving a SAW velocity on LAO/STO of  $v = 1020$  m/s. The IDT comb structure generates SAWs by converting the electrical energy to elastic energy, causing a resonance dip in the reflection signal.

The total quality factor  $Q$  is defined as

$$Q \equiv f_c/B, \quad (1)$$

where  $f_c$  is the center resonance frequency and  $B$  is the half-power ( $-3$  dB) bandwidth. The resonance spectrum shows a quality factor  $Q = 17.5$ , which is consistent with previous reports [25] on STO-based acoustic resonators without resonance-enhanced structures (e.g., Bragg mirrors). Theoretically the bandwidth  $B$  can be determined from the IDT geometry according to Ref. [32],

$$B \approx 0.9 f_c / N_p, \quad (2)$$

where  $N_p = 16$  is the number of comb pairs in the IDT. Here, the calculated bandwidth is 7.2 MHz is close to the expected value of 7.3 MHz. The  $<2\%$  difference can come from the imperfect edge of IDT geometry related to the maskless photolithography precision.

The transmission spectrum  $S_{21}$  [Fig. 2(a)] shows a resonance peak at a frequency  $f_c$  that coincides with the reflection dip in  $S_{11}$ , supporting the scenario that energy is transmitted efficiently via SAW from the transmitting IDT to the receiving IDT. When we sweep the temperature, the resonance peak disappears sharply at temperatures larger than 13.7 K,

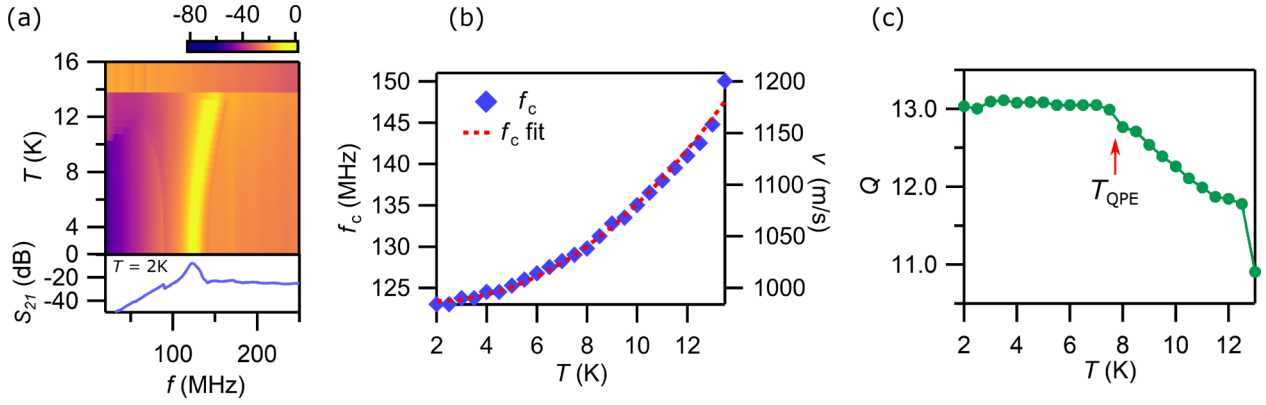


FIG. 2. Temperature dependence of resonance. (a) The upper intensity plot shows transmission signals with respect to the temperature between 2 and 16 K. The lower figure is a transmission signal line cut through 2 K temperature showing the resonance peak. (b) Temperature dependence of resonance center frequency and calculated SAW phase velocity (blue diamonds). The red dashed line is a quadratic fit. (c) Quality factor  $Q = f_c/B$  plotted with respect to the temperature. The red arrow shows the STO quantum paraelectric saturation temperature.

corresponding to the temperature  $T_c$  above which the NbTiN is no longer superconducting (for details of NbTiN superconductivity, see the Supplemental Material [30]). The NbTiN normal resistance 1.57 k $\Omega$  gives an impedance mismatch which causes most of the power to be reflected and dissipated both internally in the IDT and externally into the transmission line leading to a sharp cutoff on the transmission signal.

Notably, the resonance frequency is temperature dependent. A quadratic scaling is observed between the center frequency and temperature, with a lower  $f_c$  at a lower temperature. To understand this scaling, we may consider a Helmholtz free energy of phonons,  $F$ , description of its equilibrium state,

$$F(t, \psi) = a(t) + b(t)\psi^2 + c(t)\psi^4 + \dots, \quad (3)$$

where  $\psi$  is the order parameter, and  $t = (T - T_c)/T_c$  is the reduced temperature. When we only consider the equilibrium states, we obtain

$$b(t)\psi + 2c(t)\psi^3 = 0,$$

$$|\psi| \approx (b_1/2c_0)|t|^{1/2}.$$

The asymptotic expression for  $F$  becomes

$$F(t, \psi) \approx a_0 - \frac{b_1^2}{4c_0}t^2 + \dots \quad (4)$$

Therefore, the free energy is expected to scale quadratically with temperature. The resonance frequency  $f_c$ , depending linearly on  $F$ , scales quadratically with temperature [Fig. 2(b)].

With a constant IDT geometry, such that the wavelength  $\lambda$  is kept fixed, a smaller  $f_c$  corresponds to a lower SAW phase velocity  $v$ . We observe that lowering the temperature reduces the SAW velocity. This trend contrasts with results on most piezoelectric materials such as PZT, in which SAW velocities have been reported to increase as temperature is lowered, because of the decreasing thermal fluctuations and increasing stiffness of the sample [33].

The piezoelectric coefficient  $d_{311}$ , defined as  $S_{ij} = d_{ijk}E_k$ , and the electrostriction coefficient  $R_{311}$ , defined as  $S_{ij} = R_{ijk}E_k^2$ , where  $S$  is the strain and  $E$  is a field, increases

with decreasing temperature, especially for  $T < 10$  K [21]. This means with a certain electric field, the piezoelectric and electrostrictive response can induce more strain in the material, consistent with the mode softening and more efficient electromechanical energy conversion. This behavior is aligned with the fact that we observe that the SAW frequency decreases upon cooling, while the SAW velocity is lowered and implies a softer crystal. The quality factor, plotted versus temperature in Fig. 2(c), first increases as temperature is decreased, and then saturates at  $T \approx 8$  K. The saturation temperature coincides with the STO quantum paraelectric phase transition ( $T_{QPE}$ ) where the dielectric permittivity  $\epsilon$  saturates, described by Barrett's formula [20]. This correspondence indicates that SAW is sensitive to the quantum paraelectric phase transition and its  $Q$  factor is related to the  $\epsilon$  variance. When  $T > T_c$ ,  $Q$  drops quickly to near zero due to the increasing resistance  $R$  for the IDT.

To verify the linear dispersion of the SAW ( $v = f\lambda$ ), two different pairs of IDTs with different electrode spacing are compared, keeping the metallization ratio fixed such that  $m = 0.5$ . The IDT geometry is as shown in Fig. 3(a). The IDT finger widths  $w = d = 2$   $\mu\text{m}$  and  $w = d = 3$   $\mu\text{m}$  correspond to wavelengths  $\lambda = 8$   $\mu\text{m}$  and  $\lambda = 12$   $\mu\text{m}$ , respectively. The measured resonance frequency  $f_c$ , labeled with black arrows in Figs. 3(b) and 3(c), shows the expected inverse linear scaling with wavelength, providing further confirmation of the SAW origin of the resonance feature.

The transmission resonances in both devices show an appreciable hardening as a function of backgate voltage  $V_{bg}$  [Figs. 3(b) and 3(c)]. The rise in acoustic velocity is associated with induced ferroelectric displacement which breaks the inversion symmetry of the crystal structure and couples to the strain field  $S$ . This phenomenon can be modeled using a Landau-Ginsburg-Devonshire (LGD) free-energy expression, expanding in powers of the displacement  $D$  up to the second order [Eq. (5)] [25,34–36]. For paraelectric STO, the dielectric electromechanical response is described within LGD theory [37] by

$$F - F_0 = -pSD + \frac{1}{2}\chi^{-1}D^2 + \frac{1}{2}GSD^2 + \dots, \quad (5)$$

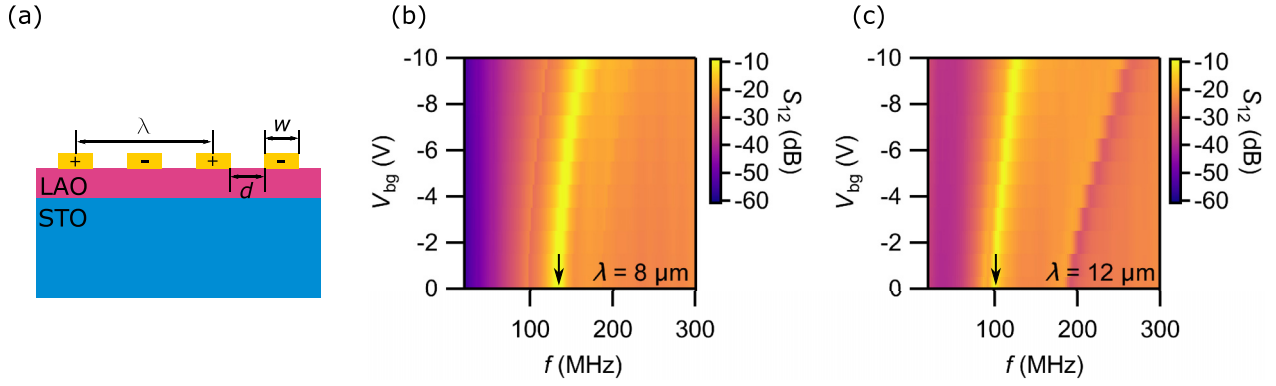


FIG. 3. Resonance frequency shift due to different wavelengths and negative backgate voltages. (a) Schematic diagram of NbTiN IDT geometry, where  $w$  is the finger width and  $d$  is the gap distance. Wavelength  $\lambda$  is determined by the center distance between two nearest same polarity fingers. (b) Transmission spectrum of device A ( $\lambda = 8 \mu\text{m}$ ) as a function of  $V_{bg}$ . The black arrow denotes the resonance frequency. (c) Transmission spectrum of device B ( $\lambda = 12 \mu\text{m}$ ) as a function of  $V_{bg}$ . The black arrow denotes the resonance frequency. All data taken at  $T = 2 \text{ K}$ .

where  $p$  is the piezoelectric tensor,  $\chi$  is the dielectric permittivity tensor, and  $G$  is the electrostrictive tensor.

Surprisingly, the dependence of  $f_c$  on  $V_{bg}$  is much stronger when  $V_{bg} < 0$  compared to  $V_{bg} > 0$  (Fig. 4). This dependence contrasts with pure electrical tuning of the dielectric constant through  $V_{bg}$ , in which the tuning effect is symmetric across zero bias [38]. The LAO thickness is below the critical thickness for spontaneous formation of a conductive LAO/STO interface [29]. The interface remains insulating during the experiment, even for the maximum backgate voltage that has been applied (for details of sample characterization under backgate voltages, see the Supplemental Material [30]), and thus carrier screening or other effects associated with a gate-induced insulator-to-metal transition can be ruled out. One is left with explanations that are tied to the formation and evolution of ferroelastic domains. The motion of such domains under backgate bias is consistent with prior imaging from Honig *et al.* [5] which showed that under large negative backgate voltage, tetragonal ferroelastic domains are observed, leading to the anomalously large piezoelectricity at low temperature. The formation and drifting of the ferroelastic

domain under negative backgate voltages play an important role in coupling with the SAW.

To investigate how the magnitude of the SAW coupling depends on the static bias across the IDT, we incorporate a bias tee between the VNA port and IDT connection to apply a dc bias  $V_{bias}$  between neighboring IDT fingers with opposite polarity, and we measure the change in the resonance amplitude as a function of  $V_{bias}$ . The result [Fig. 5(a)] shows a quadratic relationship for  $S_{11}$  amplitude. The quadratic dependence can be understood as an electrostriction effect in which an electric field couples to strain up to the second order [Eq. (5)]. The first-order electric field induced by  $V_{bias}$  breaks the inversion symmetry of STO, yielding a linear coupling. The scaling indicates a built-in STO polarization that can be modulated with an applied bias across the IDT. For comparison, port 2 is not subject to a dc bias, and as a result no tuning of the  $S_{22}$  amplitude [Fig. 5(a)] is observed. At  $V_{bias} \approx -1 \text{ V}$ , the applied  $V_{bias}$  cancels the built-in polarization and the resonance amplitude is minimized. Similar tuning is observed for the transmission signals  $S_{12}$  and  $S_{21}$ , as expected.

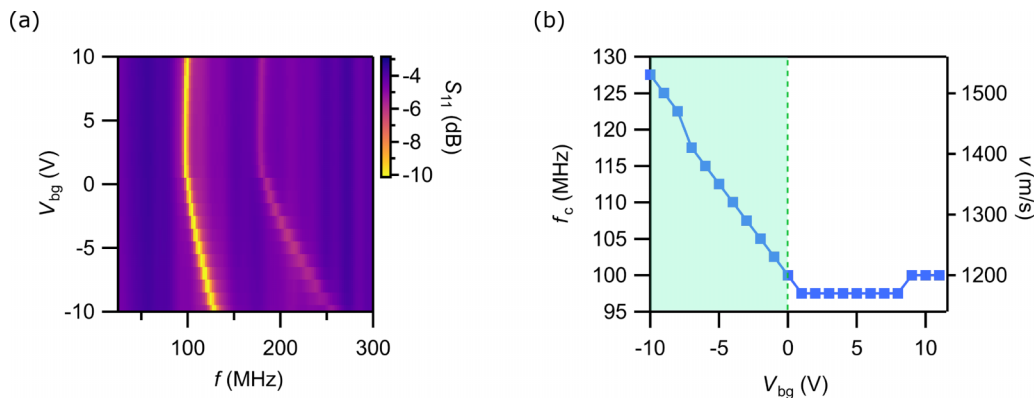


FIG. 4. Positive and negative  $V_{bg}$  dependence of reflection spectrum and resonance frequency. (a) Reflection spectrum as a function of  $V_{bg}$ . There is a resonance dip and a second harmonic dip observed in the spectrum. (b) Resonance center frequency  $f_c$  as a function of  $V_{bg}$ . The green region highlights where the resonance frequency can be tuned with negative  $V_{bg}$ .



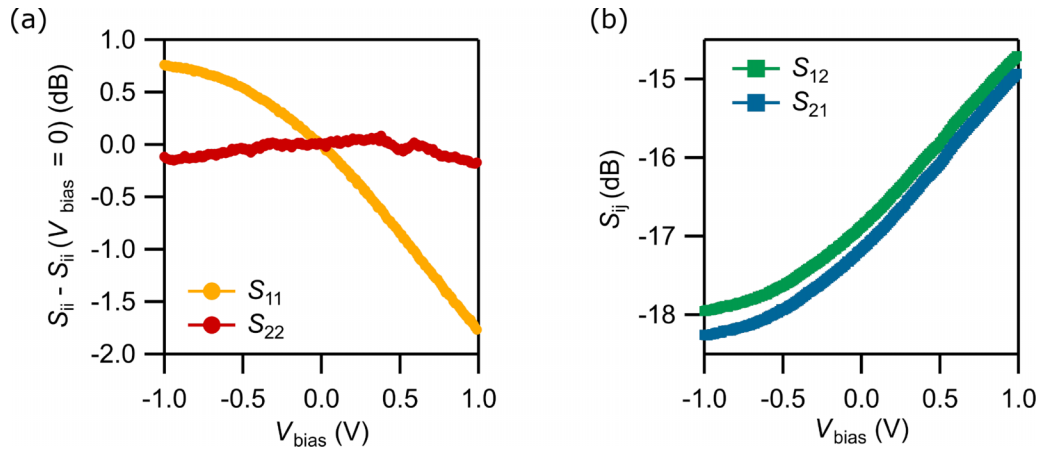


FIG. 5. Reflection scattering parameters ( $S_{11}$ ,  $S_{22}$ ) measured as a function of  $V_{\text{bias}}$  applied across the IDT fingers in port 1, with zero bias at port 2. (a) Reflection amplitude for ports 1 ( $S_{11}$ ) and 2 ( $S_{22}$ ), referenced against the zero-bias value  $S_{ii}$  ( $V_{\text{bias}} = 0$ ). (b) Transmission amplitudes ( $S_{12}$ ,  $S_{21}$ ), measured as a function of  $V_{\text{bias}}$ .

**Discussion and conclusion.** Although 1.2-nm LAO is much smaller than the SAW wavelength and will hardly interfere with the SAW propagation, the LAO layer, which is intrinsically polarized, contributes an important role in providing the piezoelectricity to the SAW generation. The other possible contribution to this piezoelectricity is the polar domain twin walls of STO ferroelasticity [39–43].

With an acoustic speed five orders lower than the speed of light, a relatively short acoustic wavelength, and high degree of surface sensitivity, SAW generation, propagation, and detection can be regarded as useful building blocks for manipulating electronic and lattice degrees of freedom in complex-oxide heterostructures and nanostructures. Specifically, SAWs have the potential to contribute to quantum information processing architectures [32] both in superconducting qubits [44–46] and electron spin-based quantum computing architectures [47–49]. Coupling superconducting qubits with SAW can help control and measure quantum states [50]. Furthermore, SAW generates moving potential wells with a mesoscopic scale which transport electron charges with spin information propagating at the speed of sound in a confined one-dimensional channel, helping to meet the architectural challenges of long-range transport of spin information [51–53]. At the same time, SAW manipulation of electronic properties may help provide insight into the nature of correlated electronic phases such as superconductivity.

In conclusion, we have demonstrated the direct generation and detection of SAW on LAO/STO at cryogenic temperature using superconducting IDTs. Spurious contributions arising from possible bulk acoustic wave components and electronic resonances from the instrument are carefully ruled out. The

SAW shows an ultralow phase velocity which reveals the coupling to the high permittivity of the STO at low temperatures. The SAW quality factor saturates in the quantum paraelectric phase regime, corroborating the related  $Q$  factor with dielectric permittivity. This method can thus be used to probe behavior near the quantum phase transition. The behavior is consistent with a linear electromechanical coupling that is tightly coupled with the ferroelastic domain evolution. Our work encountered the barrier of probing the temperature dependence of QPE transition at the cryogenic temperature on STO, also generating a platform that can leverage SAW as a sensitive detector of structural phase transition. This is an instance where a temperature-dependent quantum paraelectric phase transition (QPE) has been measured without relying on transport measurements. It further opens up the possibilities of coupling the SAW with transport phenomena in the superconducting or paired regime of LAO/STO nanostructures.

Data generated or analyzed during this study are included in this published article (and its Supplemental Material).

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The authors declare no conflict of interest.

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