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Elastic Softening of Surface Acoustic Wave Caused by Vacancy Orbital in Silicon Wafer

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We have performed surface acoustic wave (SAW) measurements to examine vacancies in a surface layer of a boron-doped silicon wafer currently used in semiconductor industry. A SAW with a frequency of $f_s = 517$ MHz was optimally generated by an interdigital transducer with a comb gap of $w = 2.5 \,\mu\text{m}$ on a piezoelectric ZnO film deposited on the (001) silicon surface. The SAW propagating along the [100] axis with a velocity of $v_s = 4.967 \,\text{km/s}$ is in agreement with the Rayleigh wave, which shows an ellipsoidal trajectory motion in the displacement components u_x and u_z within a penetration depth of $\lambda_p = 3.5 \,\mu\text{m}$. The elastic constant C_s of the SAW revealed the softening of $\Delta C_s/C_s = 1.9 \times 10^{-4}$ below 2 K down to 23 mK. Applied magnetic fields of up to 2 T completely suppress the softening. The quadrupole susceptibilities based on the coupling between the electric quadrupoles O_u , O_v , and O_{zx} of the vacancy orbital consisting of $\Gamma_8 - \Gamma_7$ states and the symmetry strains ε_u , ε_v , and ε_{zx} associated with the SAW account for the softening and its field dependence on C_s . We deduced a low vacancy concentration $N = 3.1 \times 10^{12}/\text{cm}^3$ in the surface layer within $\lambda_p = 3.5 \,\mu\text{m}$ of the silicon wafer. This result promises an innovative technology for vacancy evaluation in the fabrication of high-density semiconductor devices in industry.

1. Introduction

Acoustic waves propagating on a free surface of substrate matter were investigated for the first time by Rayleigh. One hundred years later, electronic technology has widely adopted a surface acoustic wave (SAW) in miniature electronic modules of ladder-type delay lines, resonators, and band-pass filters using an interdigital transducer (IDT) with high-Q resonance and low insertion loss. The SAW is regarded as a diagnostic tool for investigating 4f-electronic states of rare-earth compounds, 6-80 quantized Hall effects, 9,100 topological insulators, 110 and graphene. 120 The observation of vacancies in the surface layer of substrate silicon wafers currently used in semiconductor industry is a prominent issue in the diagnostic application of the SAW.

Silicon wafers contain two different types of intrinsic point defects, which consist of a vacancy denoting an unoccupied lattice site and a silicon interstitial corresponding to a silicon atom at an irregular lattice site. The low-temperature softening of the elastic constants of boron-doped silicon observed by bulk ultrasonic waves was successfully described in terms of the quadrupole susceptibilities of a vacancy orbital with a Γ_8 quartet ground state and a Γ_7 doublet excited state at 1 K. ^{13–16}) Since the fabrication of semiconductor devices for information technology is carried out using the active region beneath the surface of the substrate silicon wafer, the evaluation of vacancy concentration in the surface layer of the silicon wafer is keenly required. No research on vacancy evaluation in the surface layer of silicon wafers has been reported so far to the best of our knowledge.

In the present study, we show low-temperature measurements of the elastic constant of the SAW propagating on a boron-doped silicon wafer. The excitation of the SAW using an IDT and low-temperature SAW measurement are described in Sect. 2. The low-temperature elastic softening of the

SAW and its magnetic field dependence are presented in Sect. 3. The equations of motion for specifying the SAW on the silicon surface and the quadrupole susceptibilities for the vacancy orbital for describing the softening are shown in Sect. 4. We give our discussion in Sect. 5 and our conclusions in Sect. 6.

2. Experimental Procedure

We fabricated an IDT on a boron-doped silicon wafer of $\sharp 9018$, which was sliced from a Czochralski (CZ) silicon ingot with a diameter of $300\,\mathrm{mm}$. The wafer shows a resistivity range of $21\text{--}23\,\Omega$ ·cm at room temperature indicating a boron concentration of about $0.7\times10^{15}/\mathrm{cm}^3$. An infrared spectrometer detected an absorption peak at $1107\,\mathrm{cm}^{-1}$ of interstitial atomic oxygen with a concentration of $1.0\times10^{18}/\mathrm{cm}^3$ in the wafer. Using a laser cutter, we reduce the diameter of the wafer concerned from 300 to $200\,\mathrm{mm}$. Because we placed an order for the optimized treatment of $200\,\mathrm{mm}$ wafers with inland semiconductor facilities, all steps for the IDT fabrication for the SAW measurement are needed to process the wafers with the same diameter of $200\,\mathrm{mm}$.

A piezoelectric ZnO film was deposited on the (001) surface of the wafer using an AC-sputtering method. The hexagonal c-axis of the ZnO film is aligned perpendicular to the surface of the wafer. Aluminum (Al) comb-shaped electrodes were fabricated on the ZnO film. There were two cut-in patterns of 5-tooth-comb fingers with a separation gap of $w=2.5\,\mu\mathrm{m}$ and a finger length of 2.025 mm as the design rule 800w. Ordinary semiconductor processing by photoresist lithography and reactive ion etching was used to pattern the IDT.

The sample of $\sharp 9018-8$ with a rectangular shape of $10 \times 20 \times 0.775 \, \text{mm}^3$ was diced out from the center of the wafer of $\sharp 9018$. A schematic view of the IDT is shown in

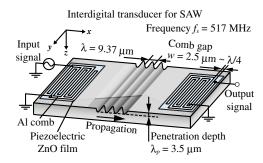


Fig. 1. (Color online) Schematic view of the IDT for the SAW measurements on the silicon wafer.

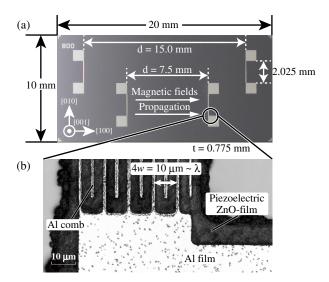


Fig. 2. (Color online) (a) Top view of the IDT fabricated on a silicon sample with the (001) surface diced out to $10 \times 20 \times 0.775 \,\mathrm{mm}^3$. A pair of IDTs with a distance of $d=7.5 \,\mathrm{mm}$ fabricated on the lower side was employed in the present SAW measurements. The magnetic fields were applied along the [100] axis parallel to the propagation direction of the SAW. (b) Expanded view of the Al comb electrode deposited on the piezoelectric ZnO film photographed by a laser microscope.

Fig. 1. The SAW propagates along the x-direction on the z surface of the silicon wafer. The IDT consisting of Al electrodes with a comb gap of $w=2.5\,\mu\mathrm{m}$ fabricated on the piezoelectric ZnO film was designed to optimally generate the SAW with a wavelength of $\lambda\sim 4w$. The opposite IDT receives the traveled SAW.

Figure 2(a) shows a top view of the sample on which two pairs of IDTs were placed. One IDT consists of a pair of sender and receiver electrodes. They are separately placed facing each other with distances of d=15.0 mm in the upper line and d=7.5 mm in the lower line. The present SAW measurements were carried out on the lower line with d=7.5 mm. Figure 2(b) shows an expanded view of the Al comb electrode of the IDT photographed by a laser microscope.

The low-temperature elastic softening of the SAW was measured using a dilution refrigerator (Oxford Instruments Kelvinox 400HA), which was equipped with coaxial lines of CuNi tubes for efficient transmission of high-frequency electronic signals of the SAW.¹⁷⁾ The sample of #9018-8 for the SAW measurements was placed in thermal contact with a Ag plate with 1.5 mm in thickness. The Ag plate was attached to a Cu rod of an extended tail from a mixing chamber for

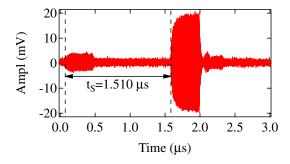


Fig. 3. (Color online) Pulsed shape SAW traveled from the sender IDT to the receiver IDT, which is $d=7.5\,\mathrm{mm}$ away from the sender, on the (001) surface of the silicon wafer. A time of flight $t_\mathrm{s}=1.510\,\mathrm{\mu s}$ gives a group velocity $v_\mathrm{s}=d/t_\mathrm{s}=4.967\,\mathrm{km/s}$ of the SAW.

thermal grounding using a Ag wire with a diameter of 1.0 mm. The adoption of the Ag plate has the advantage of minimizing low-temperature nuclear specific heat enhancement in applying magnetic fields. ^{18,19} The temperature of the sample was measured using a calibrated RuO₂ resistance. A superconducting magnet with a bore of 77 mm was equipped to the dilution refrigerator for the generation of magnetic fields of up to 9 T.

The SAW with a pulsed shape of $0.3\,\mu s$ width at a repetition rate of 1 kHz was generated. The velocity change $\Delta v_s/v_s$ of the SAW as a function of temperature and magnetic field was determined by measuring by phase delay of the pulsed shape SAW. The relatively high frequency of $f_s = 517\,\mathrm{MHz}$ in the SAW measurements has the advantage of a high resolution better than $\Delta v_s/v_s = 10^{-6}$. This excellent resolution leads to the reliable detection of low vacancy concentrations in the silicon wafer.

3. Results

The IDT sends an incident pulsed shape SAW of 0.3 µs in width with a carrier wave of frequency $f_s = 517 \,\mathrm{MHz}$. The pulsed shape SAW travels along the [100] direction on the (001) surface to the opposite IDT receiver. As is shown in Fig. 3, a time of flight of $t_s = 1.510 \,\mu s$ for the signal received gives a group velocity of $v_s = d/t_s = 4.967 \,\mathrm{km/s}$ at 4.2 K. This corresponds to a wavelength $\lambda_s = v_s/f_s = 9.61 \,\mu\text{m}$, which is fairly consistent with the wavelength $4w = 10 \,\mu m$ expected from the IDT design. Furthermore, the experimentally determined group velocity $v_s = 4.967 \,\mathrm{km/s}$ of the SAW on the silicon wafer is in agreement with the Rayleigh wave velocity $v = 4.844 \,\mathrm{km/s}$, which is theoretically calculated using the equations of motion for the SAW in Sect. 4.1. The velocity $v_s = 4.967 \,\mathrm{km/s}$ and density $\rho = 2.330 \,\mathrm{g/cm^3}$ of the silicon wafer give an elastic constant $C_s = \rho v_s^2 = 5.748 \times$ $10^{10} \,\mathrm{J/m^3}$ for the SAW at low temperatures.

Figure 4 shows the temperature dependence of the elastic constant C_s of the SAW propagating along the [100] axis on the (001) surface of the silicon wafer. Appreciable softening with a relative change $\Delta C_s/C_s = 1.9 \times 10^{-4}$ below 2 K down to 23 mK was observed. As is shown in the theoretical expression of Eq. (39) in Sect. 4.2, the amount of softening of C_s is proportional to the vacancy concentration N in the surface layer of the silicon wafer. In Fig. 4, we schematically picture the dangling bonds of ϕ_1 , ϕ_2 , ϕ_3 , and ϕ_4 at the vacancy site with a cubic site symmetry of T_d in the silicon

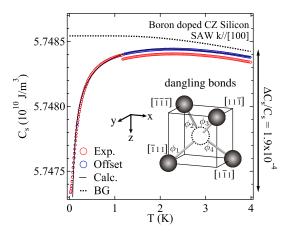


Fig. 4. (Color online) Low-temperature softening of the elastic constant C_s of the boron-doped silicon wafer of $\sharp 9018$ -8. The SAW propagating along the [100] direction with the (001) surface was used for the measurements. The discontinuity at 1.17 K is an extrinsic phase shift due to the superconducting transition of the Al comb electrodes used in the IDT. The solid line is a fit in terms of the quadrupole susceptibilities. A schematic view of the four dangling bonds ϕ_i (i=1,2,3,4) of a vacancy is also shown.

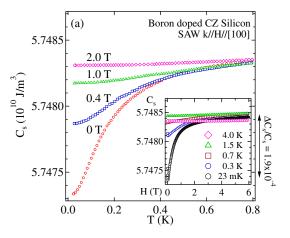
lattice, which form the vacancy orbital and bring about the softening of C_s .

There exists a distinct discontinuity in C_s at 1.17 K of Fig. 4. This is caused by a superconducting transition of the Al film used in the comb electrodes of the IDT. The slight modification in the effective impedance of the Al comb electrodes brings about an extrinsic phase shift at 1.17 K in superposing on the phase delay of the traveled SAW. Nevertheless, the softening of C_s is properly analyzed by the quadrupole susceptibility of the vacancy orbital in Sect. 4.2 in merely offsetting the extrinsic phase shift at 1.17 K. The solid line in Fig. 4 is a fit using a theoretical expression Eq. (39), which is described in terms of the quadrupole susceptibility of Eq. (34) in Sect. 4.2.

In Fig. 5(a), we show the low-temperature dependence of the C_s of the SAW below 0.8 K down to 23 mK at applied magnetic fields of up to 2.0 T along the [100] axis parallel to the propagation direction. The appreciable softening was observed at zero magnetic field, while the softening was considerably reduced at applied magnetic fields of 0.4 and 1.0 T. The higher magnetic field of 2.0 T fully suppresses the low-temperature softening of C_s .

The inset of Fig. 5(a) shows the magnetic field dependence of C_s of the SAW up to 6T at low temperatures. The appreciable increase in the amount of $\Delta C_s/C_s=1.9\times10^{-4}$ at a base temperature of 23 mK with increasing magnetic fields from zero to 6T in the inset of Fig. 5(a) is well consistent with the amount of softening of $\Delta C_s/C_s=1.9\times10^{-4}$ below 2 K down to 23 mK at zero magnetic field shown in Fig. 4(a). The increased temperature of 0.3 K reduces the field dependence to $\Delta C_s/C_s=0.7\times10^{-4}$. Further increase in temperatures to 0.7, 1.5, and 4.0 K completely smears out the field dependence of C_s . We show the results of the theoretical calculation by the solid and dashed lines in Fig. 5(b). Details of the calculation of C_s at the magnetic fields in terms of the quadrupole susceptibilities are shown in Sect. 4.2.

Phase shifts associated with the critical field $H_{\rm c}\sim 0.02\,{\rm T}$ of the superconducting phase in the Al electrodes were



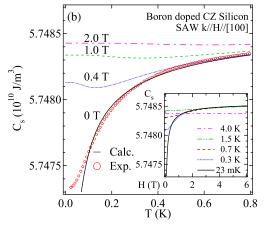


Fig. 5. (Color online) (a) Temperature dependence of the elastic constant C_s of the SAW on the boron-doped silicon wafer of $\sharp 9018-8$ below 0.8 K down to 23 mK at applied magnetic fields along the [100] axis. The inset shows the magnetic field dependence of C_s at low temperatures. Anomalies associated with the superconducting critical field of the Al comb electrodes were observed at 0.02 T. (b) Theoretical calculation of temperature dependence of C_s in terms of the quadrupole susceptibilities. The inset shows the magnetic field dependence of the calculation of C_s at low temperatures.

observed in the inset of Fig. 5(a). The alternative adoption of Cu electrodes of a normal metal instead of Al electrodes can be favorable for avoiding the extrinsic phase shift due to the superconducting transition in Al electrodes.

4. Theory

4.1 Equations of motion for SAW

We shall treat equations of motion for the SAW propagating along the *x*-direction on the *z* surface of the substrate silicon wafer.^{20,21)} We set a free space for z < 0 and a solid region for z > 0. This boundary condition gives constraint for stress components of σ_{iz} (i = x, y, z) acting on the free *z* surface as

$$\sigma_{xz} = \sigma_{zx} = C_{44}\varepsilon_{zx} = C_{44}\left(\frac{\partial u_x}{\partial z} + \frac{\partial u_z}{\partial x}\right) = 0,$$

$$\sigma_{yz} = C_{44}\varepsilon_{yz} = C_{44}\left(\frac{\partial u_z}{\partial y} + \frac{\partial u_y}{\partial z}\right) = 0,$$

$$\sigma_{zz} = C_{12}(\varepsilon_{xx} + \varepsilon_{yy}) + C_{11}\varepsilon_{zz}$$

$$= C_{12}\left(\frac{\partial u_x}{\partial x} + \frac{\partial u_y}{\partial y}\right) + C_{11}\frac{\partial u_z}{\partial z} = 0.$$
(1)

Here, u_i (i = x, y, z) denote the displacement vector components, and C_{11} , C_{12} , and C_{44} denote the bulk elastic constants of the silicon lattice with a cubic symmetry. The elastic strains ε_{ij} (i, j = x, y, z) are defined as²²)

$$\varepsilon_{xx} = \frac{\partial u_x}{\partial x},$$

$$\varepsilon_{yy} = \frac{\partial u_y}{\partial y},$$

$$\varepsilon_{zz} = \frac{\partial u_z}{\partial z},$$

$$\varepsilon_{yz} = \frac{\partial u_z}{\partial y} + \frac{\partial u_y}{\partial z},$$

$$\varepsilon_{zx} = \frac{\partial u_x}{\partial z} + \frac{\partial u_z}{\partial x},$$

$$\varepsilon_{xy} = \frac{\partial u_y}{\partial x} + \frac{\partial u_z}{\partial y}.$$
(2)

The SAW propagating along the x-direction on the z surface leads to a constraint that the displacement vector components u_x and u_z are relevant and u_y is irrelevant. The SAW propagating along the x-direction keeps the angles for the x-y and y-z axes orthogonal. The equations of motion for the SAW consisting of the displacement components u_x and u_z in each infinitesimal volume are described as

$$\rho \frac{\partial^{2} u_{x}}{\partial t^{2}} = C_{11} \frac{\partial^{2} u_{x}}{\partial x^{2}} + C_{44} \frac{\partial^{2} u_{x}}{\partial z^{2}} + (C_{12} + C_{44}) \frac{\partial^{2} u_{z}}{\partial z \partial x},$$

$$\rho \frac{\partial^{2} u_{z}}{\partial t^{2}} = C_{11} \frac{\partial^{2} u_{z}}{\partial z^{2}} + C_{44} \frac{\partial^{2} u_{z}}{\partial x^{2}} + (C_{12} + C_{44}) \frac{\partial^{2} u_{x}}{\partial x \partial z}.$$
(3)

The SAW with the displacement components u_x and u_z propagates along the x-axis with a phase velocity v and a wavenumber $k=2\pi f/v$ for a frequency f. Furthermore, the SAW penetrates into the silicon surface along the z-axis for z>0. Therefore, u_x and u_z are given as

$$u_{x} = \sum_{i=1}^{2} U_{i} \exp[-kq_{i}z + ik(x - vt)],$$

$$u_{z} = \sum_{i=1}^{2} W_{i} \exp[-kq_{i}z + ik(x - vt)].$$
(4)

Here, $\sum_{i=1}^{2}$ means the sum over two solutions of penetration factors q_i (i = 1, 2) for the z-axis. U_i and W_i denote amplitude of u_x and u_z of the SAW, respectively.

Substituting u_x and u_z of Eq. (4) into Eq. (3), we obtain simultaneous equations

$$(C_{11} - \rho v^2 - C_{44}q_i^2)U_i + (C_{12} + C_{44})q_i(iW_i) = 0,$$

$$(C_{12} + C_{44})q_iU_i - (C_{44} - \rho v^2 - C_{11}q_i^2)(iW_i) = 0.$$
(5)

The second line of Eq. (5) gives a relation between the amplitude of U_i and W_i as

$$iW_i = \frac{(C_{12} + C_{44})q_i}{C_{44} - \rho v^2 - C_{11}q_i^2} U_i = \gamma_i U_i.$$
 (6)

Here, γ_i is a ratio denoting the difference in the amplitude and phase between U_i and W_i :

$$\gamma_i = \frac{(C_{12} + C_{44})q_i}{C_{44} - \rho v^2 - C_{11}q_i^2}.$$
 (7)

Nontrivial solutions of Eq. (5) require an equation for the determinant:

$$\begin{vmatrix} C_{11} - \rho v^2 - C_{44} q_i^2 & (C_{12} + C_{44}) q_i \\ (C_{12} + C_{44}) q_i & -C_{44} + \rho v^2 + C_{11} q_i^2 \end{vmatrix} = 0.$$
 (8)

Equation (8) gives an algebraic equation of q_i as

$$C_{11}C_{44}q_i^4 - \{C_{11}(C_{11} - \rho v^2) + C_{44}(C_{44} - \rho v^2) - (C_{12} + C_{44})^2\}q_i^2 + (C_{11} - \rho v^2)(C_{44} - \rho v^2) = 0.$$
 (9)

Now we shall go back to the boundary conditions of the stress tensor $\sigma_{iz}=0$ acting on the free z surface at z=0 of Eq. (1). Substituting Eq. (4) into Eq. (1), we obtain simultaneous equations for U_1 and U_2 as

$$U_{1}(\gamma_{1} - q_{1}) \exp[-kq_{1}z]$$

$$+ U_{2}(\gamma_{2} - q_{2}) \exp[-kq_{2}z] = 0,$$

$$U_{1}(C_{12} + C_{11}\gamma_{1}q_{1}) \exp[-kq_{1}z]$$

$$+ U_{2}(C_{12} + C_{11}\gamma_{2}q_{2}) \exp[-kq_{2}z] = 0.$$
(10)

Nontrivial solutions of Eq. (10) require that the determinant should satisfy

$$\begin{vmatrix} \gamma_1 - q_1 & \gamma_2 - q_2 \\ C_{12} + C_{11}\gamma_1 q_1 & C_{12} + C_{11}\gamma_2 q_2 \end{vmatrix} = 0.$$
 (11)

Using the γ_i of Eq. (7) and the q_i of Eq. (9), we obtain an algebraic equation from the determinant of Eq. (11):

$$\left(1 - \frac{C_{11}}{C_{44}}R\right)\left(1 - \frac{C_{12}^2}{C_{11}^2} - R\right)^2 = R^2(1 - R). \tag{12}$$

Here, we introduce the ratio of the elastic constant $C_s = \rho v^2$ of the SAW to the bulk elastic constant C_{11} as $R = C_s/C_{11}$.

Substituting the elastic constants $C_{11}=15.681$, $C_{12}=6.181$, and $C_{44}=8.063$ in $10^{10}\,\mathrm{J/m^3}$ units at $4.2\,\mathrm{K^{15}}$) and the mass density $\rho=2.330\,\mathrm{g/cm^3}$ of the silicon into Eq. (12), we obtain three solutions for SAW velocities of v=4.844, 8.341, and 11.873 in km/s units. Among them, the lowest velocity of $v=4.844\,\mathrm{km/s}$ corresponds to the Rayleigh wave, while other higher velocities signify the pseudo SAW.²³⁾ The group velocity $v_{\rm s}=4.967\,\mathrm{km/s}$ determined by the time of flight of the pulsed shape SAW in Fig. 3 is in fair agreement with the lowest velocity $v=4.844\,\mathrm{km/s}$ of the Rayleigh wave in the calculation of Eq. (12).

Substituting the solution $v = 4.844 \,\mathrm{km/s}$ and the bulk elastic constants of the silicon crystal into Eq. (9), we obtain the penetration factor q_i of the Rayleigh wave. Because the SAW is expected to penetrate into the solid for z > 0 during decay, we adopt the solution q with a positive value in the real part, q_{Re} , and a negative one in the imaginary part, q_{Im} , as

$$q = q_{Re} + iq_{Im} = 0.4311 - 0.5216i.$$
 (13)

This solution gives the ratio γ as

$$\gamma = \gamma_{\text{Re}} + i\gamma_{\text{Im}} = -0.4311 - 1.1120i. \tag{14}$$

The corresponding u_x and u_z of the SAW of Eq. (4) are given as

$$u_x = U \exp[-kqz + ik(x - vt)],$$

$$u_z = -i\gamma u_x = 1.19 \exp[i\alpha]U \exp[-kqz + ik(x - vt)].$$
(15)

Here, we have an angle of $\alpha=158.81^\circ$ for the phase shift of $-i\gamma$. The displacement components of u_x and u_z vary at the positions x and z and the time t with a phase velocity $v=4.844\,\mathrm{km/s}$ along the x-direction. The real parts of u_x and u_z are written as

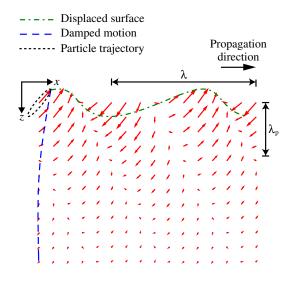


Fig. 6. (Color online) Instantaneous displacement components u_x and u_z of the SAW with a wavelength $\lambda=9.37\,\mu\mathrm{m}$ propagating along the x-axis on the z surface of the silicon wafer. The dot-dashed line denotes the displaced surface of the SAW. The dotted line denotes the ellipsoidal particle trajectory of u_z and u_x when penetrating into the z-axis on the silicon wafer with a penetration depth $\lambda_p=3.5\,\mu\mathrm{m}$. The damped motion of u_x and u_z is shown by the dashed line.

$$u_x^{(Re)} = U \exp[-kq_{Re}z] \cos[k(x - vt) - kq_{Im}z],$$

$$u_z^{(Re)} = 1.19U \exp[-kq_{Re}z] \cos[k(x - vt) - kq_{Im}z + 158.81^{\circ}].$$
(16)

The ellipsoidal trajectory of the instantaneous amplitude and directions of u_x and u_z in Eq. (16) is shown by arrows in Fig. 6. The dot-dashed line in Fig. 6 shows the displaced surface of u_x and u_z along the x-direction oscillating with a wavelength $\lambda=9.37\,\mu\mathrm{m}$ in the present SAW of $f_s=517\,\mathrm{MHz}$. As shown in Fig. 6, the amplitude of u_x and u_z decreases to 1/e when penetrating in the z-direction at a depth $\lambda_p=\lambda/(2\pi q_\mathrm{Re})=3.5\,\mu\mathrm{m}$ accompanied by a oscillation with a wavelength of $\lambda_z=2\pi/(k|q_\mathrm{Im}|)=\lambda/|q_\mathrm{Im}|=18.0\,\mu\mathrm{m}$.

In order to analyze the softening of the SAW in terms of the quadrupole-strain interaction in Sect. 4.2, we shall treat the symmetry strains ε_B , ε_u , and ε_v defined as

$$\varepsilon_{\rm B} = \varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz} = \frac{\partial u_x}{\partial x} + \frac{\partial u_y}{\partial y} + \frac{\partial u_z}{\partial z},$$

$$\varepsilon_{\rm u} = \frac{2\varepsilon_{zz} - \varepsilon_{xx} - \varepsilon_{yy}}{\sqrt{3}} = \frac{1}{\sqrt{3}} \left(2\frac{\partial u_z}{\partial z} - \frac{\partial u_x}{\partial x} - \frac{\partial u_y}{\partial y} \right),$$

$$\varepsilon_{\rm v} = \varepsilon_{xx} - \varepsilon_{yy} = \frac{\partial u_x}{\partial x} - \frac{\partial u_y}{\partial y}.$$
(17)

Substituting Eq. (15) into Eqs. (2) and (17), we obtain the symmetry strains

$$\varepsilon_{\rm B} = ik(\gamma q + 1)u_{x},$$

$$\varepsilon_{\rm u} = \frac{ik}{\sqrt{3}}(2\gamma q - 1)u_{x},$$

$$\varepsilon_{\rm v} = iku_{x},$$

$$\varepsilon_{yz} = 0,$$

$$\varepsilon_{zx} = k(\gamma - q)u_{x},$$

$$\varepsilon_{xy} = 0.$$
(18)

Consequently, the Rayleigh wave of Eq. (15) propagating along the x-direction on the z surface of the silicon wafer

consists of the four symmetry strains ε_B , ε_u , ε_v , and ε_{zx} . The real parts of Eq. (18) are written as

$$\varepsilon_{\rm B}^{\rm (Re)} = A_{\rm B}kU \exp[-kq_{\rm Re}z] \cos[k(x-vt) - kq_{\rm Im}z + \theta_{\rm B}],$$

$$\varepsilon_{\rm u}^{\rm (Re)} = A_{\rm u}kU \exp[-kq_{\rm Re}z] \cos[k(x-vt) - kq_{\rm Im}z + \theta_{\rm u}],$$

$$\varepsilon_{\rm v}^{\rm (Re)} = A_{\rm v}kU \exp[-kq_{\rm Re}z] \cos[k(x-vt) - kq_{\rm Im}z + \theta_{\rm v}],$$

$$\varepsilon_{\rm zx}^{\rm (Re)} = A_{\rm zx}kU \exp[-kq_{\rm Re}z] \cos[k(x-vt) - kq_{\rm Im}z + \theta_{\rm zx}].$$
(19)

The numerical values q of Eq. (13) and γ of Eq. (14) give the sharing parameters in the amplitude $A_{\Gamma_{\gamma}}$ (phase shift $\theta_{\Gamma_{\gamma}}$) as $A_{\rm B}=0.346$ ($\theta_{\rm B}=42.61^{\circ}$), $A_{\rm u}=1.491$ ($\theta_{\rm u}=281.37^{\circ}$), $A_{\rm v}=1.000$ ($\theta_{\rm v}=90.00^{\circ}$), and $A_{zx}=1.045$ ($\theta_{zx}=214.41^{\circ}$).

Introducing a function of $f_{\Gamma_{\gamma}}(z, x, t; \theta_{\Gamma_{\gamma}})$ depending on x, z, and t with the phase shift $\theta_{\Gamma_{\gamma}}$ as

$$f_{\Gamma_{\gamma}}(z, x, t; \theta_{\Gamma_{\gamma}}) = \exp[-kq_{\text{Re}}z] \cos[k(x - vt) - kq_{\text{Im}}z + \theta_{\Gamma_{\gamma}}],$$
(20)

we can write the symmetry strains ε_B , ε_u , ε_v , and ε_{zx} with the amplitude A_{Γ_v} and phase shifts of θ_{Γ_v} of Eq. (19) as

$$\varepsilon_{\Gamma_{\nu}}^{(\text{Re})} = A_{\Gamma_{\nu}} f_{\Gamma_{\nu}}(z, x, t; \theta_{\Gamma_{\nu}}) \delta. \tag{21}$$

Here, an external perturbation parameter $\delta = kU = 2\pi U/\lambda$, proportional to the injection amplitude of the pulsed shape SAW, is introduced.

Figure 7(a) shows the instantaneous amplitude of the symmetry strains $\varepsilon_{\rm B}$, $\varepsilon_{\rm u}$, $\varepsilon_{\rm v}$, and ε_{zx} of Eq. (19) along x-axis at z=0 and t=0. The symmetry strains $\varepsilon_{\rm u}$ with the phase shift $\theta_{\rm u}=281.37^{\circ}$ and $\varepsilon_{\rm v}$ with $\theta_{\rm v}=90.00^{\circ}$ in Fig. 7(a) oscillate in keeping the almost antiphase relation $|\theta_{\rm u}-\theta_{\rm v}|=191.37^{\circ}$. Furthermore, the symmetry strains $\varepsilon_{\rm B}$ with $\theta_{\rm B}=42.61^{\circ}$ and ε_{zx} with $\theta_{zx}=214.41^{\circ}$ also share almost the antiphase relation $|\theta_{\rm B}-\theta_{zx}|=171.80^{\circ}$. This is caused by the fact that the SAW travels along the x-direction like a plane wave but considerably decays along the z-direction. The SAW located within the surface layer of $\lambda_{\rm p}$ is reminiscent of a stationary wave confined in an end-closed waveguide.

The instantaneous amplitude of the symmetry strains $\varepsilon_{\rm B}$, $\varepsilon_{\rm u}$, $\varepsilon_{\rm v}$, and ε_{zx} along the penetrating z-direction for z>0 is shown in Fig. 7(b) for kx=1.79 and t=0. The amplitude of an individual strain in Fig. 7(b) considerably decays along the z-direction as deep as $k\lambda_{\rm p}=2\pi\lambda_{\rm p}/\lambda=1/q_{\rm Re}=2.3$. The almost antiphase sharings between the symmetry strains $\varepsilon_{\rm u}$ and $\varepsilon_{\rm v}$ and between the strains $\varepsilon_{\rm B}$ and ε_{zx} are again observed in Fig. 7(b).

The total elastic energy $U_{\text{total}}(z,x,t;\delta)$ depending on x,z, and t associated with the SAW is written by the sum of the elastic energies $U_{\text{B}},\,U_{\text{u}},\,U_{\text{v}},\,$ and $U_{zx},\,$ which are caused by the symmetry strains $\varepsilon_{\text{B}},\,\,\varepsilon_{\text{u}},\,\,\varepsilon_{\text{v}},\,\,$ and $\varepsilon_{zx},\,\,$ respectively. Taking account of the symmetry strain $\varepsilon_{\Gamma_{\gamma}}$ with the individual amplitude $A_{\Gamma_{\gamma}}$ and the phase shift $\theta_{\Gamma_{\gamma}},\,\,$ we obtain the total elastic energy

$$\begin{split} U_{\text{total}}(z, x, t; \delta) &= \sum_{\Gamma_{\gamma}} \frac{1}{2} C_{\Gamma_{\gamma}} (\varepsilon_{\Gamma_{\gamma}}^{(\text{Re})})^2 \\ &= \sum_{\Gamma_{\gamma}} \frac{1}{2} C_{\Gamma_{\gamma}} \{A_{\Gamma_{\gamma}} f_{\Gamma_{\gamma}}(z, x, t; \theta_{\Gamma_{\gamma}})\}^2 \delta^2 \\ &= \frac{1}{2} C_{\text{B}} (A_{\text{B}} f_{\text{B}}(z, x, t; \theta_{\text{B}}))^2 \delta^2 \end{split}$$

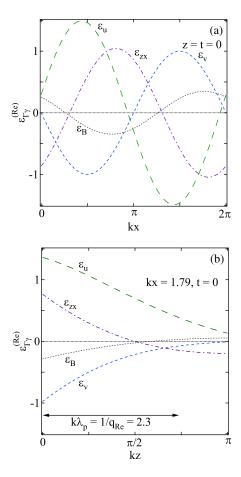


Fig. 7. (Color online) (a) Instantaneous amplitude for the symmetry strains $\varepsilon_{\rm B}$, $\varepsilon_{\rm u}$, $\varepsilon_{\rm v}$, and ε_{zx} induced by the SAW at the surface z=0 and time t=0. The strains $\varepsilon_{\rm B}$ and ε_{zx} , and $\varepsilon_{\rm u}$ and $\varepsilon_{\rm v}$ oscillate in an almost antiphase manner relative to each other. This implies the stationary wave character of the SAW during decay along the z-axis with the penetration depth $\lambda_{\rm p}$. (b) The instantaneous amplitude for the symmetry strains $\varepsilon_{\rm B}$, $\varepsilon_{\rm u}$, $\varepsilon_{\rm v}$, and ε_{zx} induced by the SAW along the z>0 direction for kx=1.79 and t=0.

$$+ \frac{1}{2} C_{\mathrm{T}} (A_{\mathrm{u}} f_{\mathrm{u}}(z, x, t; \theta_{\mathrm{u}}))^{2} \delta^{2}$$

$$+ \frac{1}{2} C_{\mathrm{T}} (A_{\mathrm{v}} f_{\mathrm{v}}(z, x, t; \theta_{\mathrm{v}}))^{2} \delta^{2}$$

$$+ \frac{1}{2} C_{44} (A_{zx} f_{zx}(z, x, t; \theta_{zx}))^{2} \delta^{2}.$$
 (22)

Here, we abbreviate the elastic constant as $C_T = (C_{11} - C_{12})/2$. $\sum_{\Gamma_{\gamma}}$ denotes the sum over the symmetry strains ε_B , ε_U , ε_V , and ε_{zx} . In Fig. 8(a), the instantaneous feature of the total elastic energy $U_{\text{total}}(z,x,t;\delta)$ along the x-direction at z=0 and t=0 for the perturbation parameter $\delta=kU=1$ is shown together with the instantaneous energies caused by the symmetry strains. The total elastic energy $U_{\text{total}}(z,x,t;\delta)$ of the SAW at t=0 in Fig. 8(a) shows its maximum for kx=1.79.

Employing the power-reduction formula and linear combination of trigonometric functions, we can reduce the total energy $U_{\text{total}}(z,x,t;\delta)$ in Eq. (22) to the time-dependent and time-independent parts of the elastic energies as

$$U_{\text{total}}(z, x, t; \delta) = \frac{1}{2} C_{\text{s}}^{0}(|A_{\text{s}}| f_{\text{s}}(z, x, t; \theta_{\text{s}}))^{2} \delta^{2} + \frac{1}{2} K_{0}(z) \delta^{2}.$$
 (23)

The former time-dependent energy means the part propagating along the *x*-direction, which is described in terms of the function

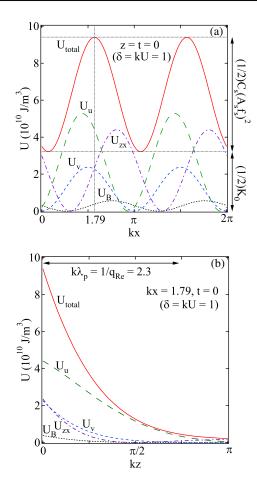


Fig. 8. (Color online) (a) Instantaneous elastic energy U_{total} of the SAW of Eq. (22) for $\delta = kU = 1$ along the x-axis at the surface of z = 0 and for time t = 0 together with the elastic energies U_{B} , U_{u} , U_{v} , and U_{zx} . The $U_{\text{total}}(x,z,t;\delta)$ of the SAW is described by the sum of the time-dependent and time-independent parts. (b) The instantaneous elastic energy U_{total} of the SAW penetrating in the z-direction for kx = 1.79 and t = 0 is shown together with the elastic energies U_{B} , U_{u} , U_{v} , and U_{zx} for $\delta = kU = 1$. The elastic energy shows a rapid decay with a penetration depth of $k\lambda_p = 2.3$.

$$f_{s}(z, x, t; \theta_{s})$$

$$= \exp[-kq_{Re}z]\cos[k(x - vt) + kq_{Im}z + \theta_{s}], \qquad (24)$$

and the amplitude

$$|A_{\rm s}| = \sqrt{\frac{4\sqrt{g_{\rm c}^2 + g_{\rm s}^2}}{C_{\rm s}^0}}. (25)$$

The phase shift θ_s of the SAW is represented by

$$\cos[2\theta_{s}] = \frac{g_{c}}{\sqrt{g_{c}^{2} + g_{s}^{2}}},$$

$$\sin[2\theta_{s}] = \frac{g_{s}}{\sqrt{g_{c}^{2} + g_{s}^{2}}},$$

$$g = \frac{1}{4} \sum_{\Gamma_{\gamma}} C_{\Gamma_{\gamma}} A_{\Gamma_{\gamma}}^{2},$$

$$g_{c} = \frac{1}{4} \sum_{\Gamma_{\gamma}} C_{\Gamma_{\gamma}} A_{\Gamma_{\gamma}}^{2} \cos 2\theta_{\Gamma_{\gamma}},$$

$$g_{s} = \frac{1}{4} \sum_{\Gamma_{\gamma}} C_{\Gamma_{\gamma}} A_{\Gamma_{\gamma}}^{2} \sin 2\theta_{\Gamma_{\gamma}}.$$
(26)

The latter time-independent energy denotes the non-propagating part along the penetrating z-direction. This is described by the function of

$$K_0(z) = 2(g - \sqrt{g_c^2 + g_s^2}) \exp[-2kq_{Re}z].$$
 (27)

The time-dependent elastic energy for the propagating part of the SAW is described by the square of $A_s f_s(z,x,t;\theta_s)\delta$ for the amplitude $|A_s|=1.505$ and the phase $\theta_s=77.47^\circ$. The time-independent elastic energy for the stationary wave is written as $(1/2)K_0(z)\delta^2$ for $K_0(z)=6.428\times 10^{10}\,\mathrm{J/m^3}$ at z=0. The time-dependent and time-independent parts of the instantaneous energy of Eq. (23) for $\delta=kU=1$ at z=0 and t=0 are also pictured in Fig. 8(a). The Rayleigh wave shares out the amplitudes and phase shifts among the symmetry strains of Eq. (19) to minimize the time-dependent elastic energy of $(1/2)C_s\{A_sf_s(z,x,t;\theta_s)\}^2\delta^2$.

Figure 8(b) shows the instantaneous feature of the total energy of $U_{\rm total}(z,x,t)$ and its symmetry parts of $U_{\rm B}$, $U_{\rm u}$, $U_{\rm v}$, and $U_{\rm zx}$ in penetrating into the z-direction for z>0. Here, we adopt the position of kx=1.79 and t=0 for $\delta=kU=1$. The instantaneous energy of the SAW rapidly decays within the surface layer of the penetration depth of $k\lambda_{\rm p}=2.3$. The diagnostic application of the SAW guarantees for the vacancy evaluation within a surface layer as deep as $\lambda_{\rm p}=3.5\,\mu{\rm m}$ of the silicon wafer. This prominent feature of the SAW measurement is definitely distinguished from the bulk ultrasonic measurement to evaluate the vacancy concentration of the inside of the wafer.

4.2 Quadrupole susceptibility

The vacancy orbital of a charge state V^+ in the borondoped silicon wafer accommodates three electrons owing to charge neutrality, where one electron is released from the vacancy orbital to an acceptor state of the boron dopant. Four sp^3 dangling bonds of ϕ_i (i = 1, 2, 3, 4) shown in Fig. 4 at a cubic site symmetry of T_d are lifted to a singlet a_1 state with an energy of -3γ and a triplet t_2 state with γ owing to the transfer energy of $-\gamma = \langle \phi_j | H_0 | \phi_i \rangle$ $(\gamma > 0)$.²⁴⁾ The energy gap of $4\gamma \sim 1.7 \,\text{eV} \sim 2.0 \times 10^4 \,\text{K}$ between the singlet a_1 state and the triplet t_2 state was theoretically estimated.^{25,26)} The former singlet a_1 state located beneath the top of the valence band accommodates two electrons with an antiparallel spin orientation, while the latter triplet t_2 state located near the chemical potential possesses an electron with spin S = 1/2. The softening of the elastic constants of the silicon wafer measured by the bulk ultrasonic waves was successfully described in terms of the quadrupole susceptibility of the vacancy orbital. 13,27,28)

The magnetic field dependence of the elastic softening is responsible for the magnetism of the vacancy orbital accommodating three (odd) number electrons. The magnetism of the vacancy orbital is caused by the spin-orbit interaction of $H_{\rm SO}=\xi \boldsymbol{L}\cdot\boldsymbol{S}$ ($\xi<0$) of the electron with orbital L=1 and spin S=1/2 in the triplet t_2 state. This $H_{\rm SO}$ gives rise to the Γ_8 quartet ground state with the energy of $\xi/2$ and the Γ_7 excited state with the energy of $-\xi$ as 29

$$|\Gamma_7\rangle \left\{ |\alpha\rangle = \sqrt{\frac{2}{3}} \left| 1, -\frac{1}{2} \right\rangle - \frac{1}{\sqrt{3}} \left| 0, \frac{1}{2} \right\rangle \right. \\ |\beta\rangle = \sqrt{\frac{2}{3}} \left| -1, \frac{1}{2} \right\rangle - \frac{1}{\sqrt{3}} \left| 0, -\frac{1}{2} \right\rangle ,$$

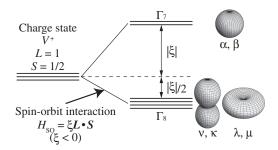


Fig. 9. The triplet t_2 state with L=1 and S=1/2 of the charge state V^+ of the vacancy orbital in boron-doped silicon forms the Γ_8 quartet ground state $(\nu, \kappa, \lambda, \text{ and } \mu)$ and Γ_7 doublet excited state $(\alpha \text{ and } \beta)$ due to the spinorbit interaction $H_{SO} = \xi \boldsymbol{L} \cdot \boldsymbol{S}$ ($\xi < 0$). The $\Gamma_8 - \Gamma_7$ states ν and κ , λ and μ , and α and β on the right-hand side have polar, donut, and spherical charge distribution, respectively.

$$|\Gamma_{8}\rangle\begin{cases} |\nu\rangle = \frac{1}{\sqrt{3}} \left| 1, -\frac{1}{2} \right\rangle + \sqrt{\frac{2}{3}} \left| 0, \frac{1}{2} \right\rangle \\ |\kappa\rangle = \frac{1}{\sqrt{3}} \left| -1, \frac{1}{2} \right\rangle + \sqrt{\frac{2}{3}} \left| 0, -\frac{1}{2} \right\rangle \\ |\lambda\rangle = \left| 1, \frac{1}{2} \right\rangle \\ |\mu\rangle = \left| -1, -\frac{1}{2} \right\rangle \end{cases}$$
(28)

Here, $|L_z,S_z\rangle$ means the state with angular momentum components over $L_z=\pm 1,0$ for L=1 and spin components over $S_z=\pm 1/2$ for S=1/2. The quadrupole susceptibilities for the vacancy orbital with Γ_8 (0 K) and Γ_7 (1 K) for the spin–orbit coupling of $\xi=-2/3$ K well described the softening of the elastic constants measured by the bulk ultrasonic waves. ^{14–16}

The spatial charge distribution distinguishes among three Kramers doublets of the vacancy orbital of Eq. (28). As shown in Fig. 9, the Kramers doublet of ν and κ in the Γ_8 quartet shows a polar charge distribution elongating along the z-axis, while the Kramers doublet of λ and μ bears a donuttype distribution spreading in the x-y plane. The Γ_7 Kramers doublet of α and β has a spherical charge distribution. The characteristic charge distribution in the $\Gamma_8-\Gamma_7$ states of the vacancy orbital is responsible for the coupling between the electric quadrupoles and the symmetry strains associated with the SAW.

It is known that the Γ_8 quartet has three magnetic dipoles, five electric quadrupoles, and seven magnetic octupoles. The electric quadrupoles couple to the elastic strains associated with ultrasonic waves. Actually, the considerable elastic softening with decreasing temperature was observed in $\text{CeB}_6^{31,32}$ and $\text{Ce}_3\text{Pd}_{20}\text{Ge}_6^{33}$ with the Γ_8 quartet ground state. The coupling between the electric quadrupoles of the Γ_8 quartet ground state of the vacancy orbital and the elastic strains of the SAW is relevant for the description of the softening of C_8 .

The applied magnetic fields of \boldsymbol{H} lift the Kramers doublets of the vacancy orbital. This is caused by the Zeeman energy as

$$H_{\text{Zeeman}} = \mu_{\text{B}}(\mathbf{L} + 2\mathbf{S}) \cdot \mathbf{H}. \tag{29}$$

Here, $\mu_{\rm B}=e\hbar/(2mc)\sim 0.67\,{\rm K/T}$ is the Bohr magneton. Among the two Kramers doublets of the Γ_8 quartet ground

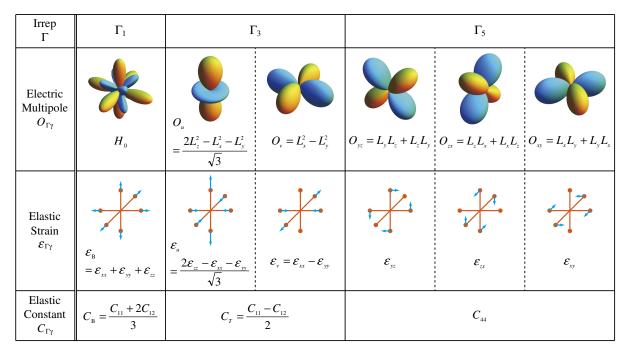


Fig. 10. (Color online) Electric multipoles, counterpart symmetry strains, and corresponding elastic constants. The quadrupole-strain interaction for the vacancy orbital brings about the softening of the elastic constant C_s . H_0 means the electric hexadecapole described by the Γ_1 -irreducible representation (irrep).

state, the ν and κ states with magnetic moments of $\pm (2/3)\mu_{\rm B}$ and λ and μ states with $\pm 2\mu_{\rm B}$ are distinct from each other at magnetic fields along the z-axis. The Γ_7 Kramers doublet state α and β have different magnetic moments of $\pm (1/3)\mu_{\rm B}$. In the present experiments, we applied the magnetic fields along the x-axis parallel to the propagating direction of the SAW. The corresponding Zeeman energy $H_{\rm Zeeman} = \mu_{\rm B}(L_x + 2S_x)H_x$ is responsible for the magnetic field effects of the elastic constant $C_{\rm s}$ of the present SAW measurements on the silicon wafer.

The coupling between the electric monopole and the volume strain $\varepsilon_{\rm B}$ with the full symmetry $\Gamma_{\rm 1}$ is relevant for the valence instability system like SmB₆.³⁴⁾ Because the charge state V^+ of the boron-doped silicon is stable, the coupling between the electric monopole and $\varepsilon_{\rm B}$ seems to be irrelevant. The coupling between the higher order electric hexadecapole $H_0 = 35L_z^4 - \{30L(L+1) - 25\}L_z^2 + 3L^2(L+1)^2 - 6L(L+1) + (5/2)(L_+^4 + L_-^4)$ and the volume strain $\varepsilon_{\rm B}$ is principally possible in the highly degenerate $\Gamma_{\rm 8}$ – $\Gamma_{\rm 7}$ states. The present analysis, however, excludes it because of its negligible contribution to the softening. We restrict ourselves to the coupling between the symmetry breaking strains associated with the SAW and the appropriate electric quadrupoles of the vacancy orbital.

The softening of C_s of the SAW down to the base temperature of 23 mK in Fig. 4 is accounted for in terms of the quadrupole susceptibility for the vacancy orbital, which is located in the surface layer within the penetration depth of λ_p . The scalar potential energy V of the vacancy orbital is modulated by the elastic strains of the SAW as $V = V_0 + \sum_{\Gamma_{\gamma}} (\partial V/\partial \varepsilon_{\Gamma_{\gamma}}^{(\text{Re})}) \varepsilon_{\Gamma_{\gamma}}^{(\text{Re})}$. Here, V_0 denotes an unperturbed potential energy. The derivative of $\partial V/\partial \varepsilon_{\Gamma_{\gamma}}^{(\text{Re})}$ is represented by the electric quadrupole $O_{\Gamma_{\gamma}}$, which is written by the symmetry square of the Cartesian coordinates x, y, and z, and the quadrupole-strain coupling constant g_{Γ} . Therefore, the

perturbation energy due to the elastic strains of the SAW is written by the quadrupole-strain interaction H_{OS} as $^{35,36)}$

$$H_{\rm QS} = g_{\Gamma_3} (O_{\rm u} \varepsilon_{\rm u}^{\rm (Re)} + O_{\rm v} \varepsilon_{\rm v}^{\rm (Re)}) + g_{\Gamma_5} O_{\rm zx} \varepsilon_{\rm zx}^{\rm (Re)}. \tag{30}$$

The eigenfunctions of the $\Gamma_8-\Gamma_7$ states of the vacancy orbital of Eq. (28) are represented by the quantum numbers of L_z and S_z . Taking account of the Wigner–Eckert theorem,³⁷⁾ we can write the electric quadrupoles using the symmetry square of the orbital angular momentum operators of L_x , L_y , and L_z . We adopt the electric quadrupoles of $O_{\rm u}=(2L_z^2-L_x^2-L_y^2)/\sqrt{3}$ and $O_{\rm v}=L_x^2-L_y^2$ with the Γ_3 -irreducible representation and $O_{zx}=L_zL_x+L_xL_z$ with the Γ_5 -irreducible representation. In Fig. 10, we list the electric quadrupoles (multipoles) and the counterpart symmetry strains in the quadupole-strain interaction of the cubic system.

The perturbation energy $E_i(\varepsilon_{\Gamma_\gamma}^{(\mathrm{Re})})$ for the ith state $(i = \alpha, \beta, \nu, \kappa, \lambda, \mu)$ of the vacancy orbital consisting of the Γ_8 – Γ_7 states is calculated using the perturbation Hamiltonian $H_{\mathrm{OS}} = g_\Gamma O_{\Gamma_\gamma} \varepsilon_{\Gamma_\gamma}^{(\mathrm{Re})}$ for the appropriate symmetry strain of $\varepsilon_{\Gamma_\gamma}^{(\mathrm{Re})}$ as

$$E_{i}(\varepsilon_{\Gamma_{\gamma}}^{(\text{Re})}) = E_{i}^{0} + g_{\Gamma}\langle i|O_{\Gamma_{\gamma}}|i\rangle\varepsilon_{\Gamma_{\gamma}}^{(\text{Re})} + g_{\Gamma}^{2}\sum_{il\neq ij}\frac{|\langle i|O_{\Gamma_{\gamma}}|j\rangle|^{2}}{E_{i}^{0} - E_{j}^{0}}(\varepsilon_{\Gamma_{\gamma}}^{(\text{Re})})^{2}.$$
 (31)

Here, E_i^0 denotes the unperturbed energy of the *i*th state of the vacancy orbital of Eq. (28).

The total free energy of the system consisting of the vacancy orbital with the vacancy concentration N in the surface layer of the silicon wafer is written as

$$F(\varepsilon_{\Gamma_{\gamma}}^{(\text{Re})}) = \frac{1}{2} C_{\Gamma_{\gamma}}^{0} (\varepsilon_{\Gamma_{\gamma}}^{(\text{Re})})^{2} - Nk_{\text{B}}T \ln \left[\sum_{i} \exp \left[-\frac{E_{i}(\varepsilon_{\Gamma_{\gamma}}^{(\text{Re})})}{k_{\text{B}}T} \right] \right]. \quad (32)$$

The second derivative of the free energy with respect to the symmetry strain $\varepsilon_{\Gamma_\gamma}^{(\mathrm{Re})}$ leads to the elastic constant C_{Γ_γ} as

$$C_{\Gamma_{\gamma}} = \frac{\partial^2 F}{\partial \varepsilon_{\Gamma_{\gamma}}^{(\text{Re})2}} = C_{\Gamma_{\gamma}}^0 - Ng_{\Gamma}^2 \chi(O_{\Gamma_{\gamma}}). \tag{33}$$

Here, the quadrupole susceptibility $-\chi(O_{\Gamma_\gamma})$ for the electric quadrupole O_{Γ_γ} is defined as ^{7,8,35)}

$$\chi(O_{\Gamma_{\gamma}}) = \sum_{E_{i}^{j,j} \neq E_{0}^{0}} \frac{n_{j} - n_{i}}{E_{i}^{0} - E_{j}^{0}} |\langle i|O_{\Gamma_{\gamma}}|j\rangle|^{2}
+ \frac{1}{k_{\mathrm{B}}T} \left[\sum_{i} |\langle i|O_{\Gamma_{\gamma}}|i\rangle|^{2} n_{i} - \left(\sum_{i} |\langle i|O_{\Gamma_{\gamma}}|i\rangle|n_{i} \right)^{2} \right].$$
(34)

Here, $n_i = Z^{-1} \exp[-E_i^0/k_BT]$ is the Boltzmann occupation probability with the partition function $Z = \sum_i \exp[-E_i^0/k_BT]$. On the analogy of the magnetic susceptibility, the

first term on the right-hand side of Eq. (34) denotes the van Vleck term associated with off-diagonal processes and the second term is the Curie term associated with the diagonal processes. The former van Vleck term leads to the temperature-independent softening, while the latter Curie term brings about the steep softening proportional to the reciprocal temperature. The quadrupole interaction among the vacancy orbitals at different sites is excluded in the present analysis, because the strong quadrupole-strain interaction dominates the low-temperature softening of the system and the intersite quadrupole interaction scarcely contributes to the softening. ¹⁵⁾

In order to explain the appreciable softening of C_s caused by the Curie term of Eq. (34), we present the matrix elements $\langle i|O_{\Gamma_y}|j\rangle$ of the electric quadrupoles O_u , O_v , and O_{zx} for the Γ_8 – Γ_7 states of the vacancy orbital in Eq. (28) at zero magnetic field as

$$\langle i|O_{\rm u}|j\rangle = \frac{1}{\sqrt{3}} \begin{pmatrix} \alpha & \beta & \nu & \kappa & \lambda & \mu \\ 0 & 0 & \sqrt{2} & 0 & 0 & 0 \\ 0 & 0 & 0 & \sqrt{2} & 0 & 0 \\ 0 & 0 & 0 & \sqrt{2} & 0 & 0 \\ 0 & \sqrt{2} & 0 & -1 & 0 & 0 & 0 \\ 0 & \sqrt{2} & 0 & -1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \end{pmatrix}, \quad \langle i|O_{\rm v}|j\rangle = \frac{1}{\sqrt{3}} \begin{pmatrix} \alpha & \beta & \nu & \kappa & \lambda & \mu \\ 0 & 0 & 0 & 0 & \sqrt{2} & 0 \\ 0 & 0 & 0 & \sqrt{2} & 0 & 1 \\ 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & \sqrt{2} & 0 & 1 & 0 & 0 \\ 0 & \sqrt{2} & 0 & 1 & 0 & 0 & 0 \\ 0 & \sqrt{2} & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & -\sqrt{\frac{3}{2}} & 0 & 0 & 1 & 0 \\ 0 & \sqrt{\frac{3}{2}} & -\frac{1}{\sqrt{2}} & 0 \\ 0 & 0 & -\sqrt{\frac{3}{2}} & 0 & 0 & 1 & 0 \\ 0 & \sqrt{\frac{3}{2}} & 0 & 0 & 0 & -1 \\ 0 & \frac{1}{\sqrt{2}} & 0 & 1 & 0 & 0 & 0 \\ 0 & \frac{1}{\sqrt{2}} & 0 & -1 & 0 & 0 & 0 \\ 0 & \frac{1}{\sqrt{2}} & 0 & -1 & 0 & 0 & 0 \\ 0 & \frac{1}{\sqrt{2}} & 0 & -1 & 0 & 0 & 0 \\ 0 & \frac{1}{\sqrt{2}} & 0 & -1 & 0 & 0 & 0 \\ \end{pmatrix}$$
 (35)

The electric quadrupoles $O_{\rm u}$, $O_{\rm v}$, and $O_{\rm zx}$ commonly have diagonal elements for the v, κ , λ , and μ of the Γ_8 quartet ground state, but no elements for the α and β of the Γ_7 doublet excited state. The lifting of the Γ_8 quartet ground state to two Kramers doublets due to the quadrupole-strain interaction in Eq. (30) brings about the softening of the elastic constant due to the Curie term in Eq. (34). The off-diagonal elements of $O_{\rm u}$, $O_{\rm v}$, and $O_{\rm zx}$ between the Γ_8 quartet ground state and Γ_7 doublet exited state in Eq. (35) lead to the van Vleck term in Eq. (34).

For accurate calculations of the low-temperature softening of C_s , we explain the perturbation Hamiltonian of Eq. (30) using the time-dependent symmetry strains of Eq. (21) induced by the SAW as

$$H_{QS} = \sum_{\Gamma_{\gamma}} g_{\Gamma} O_{\Gamma_{\gamma}} \varepsilon_{\Gamma_{\gamma}}^{(Re)}$$

$$= \sum_{\Gamma_{\gamma}} g_{\Gamma} O_{\Gamma_{\gamma}} A_{\Gamma_{\gamma}} f_{\Gamma_{\gamma}}(z, x, t; \theta_{\Gamma_{\gamma}}) \delta$$

$$= g_{\Gamma_{3}} O_{u} A_{u} f_{u}(z, x, t; \theta_{u}) \delta + g_{\Gamma_{3}} O_{v} A_{v} f_{v}(z, x, t; \theta_{v}) \delta$$

$$+ g_{\Gamma_{5}} O_{zx} A_{zx} f_{zx}(z, x, t; \theta_{zx}) \delta. \tag{36}$$

Taking account of the quadrupole-strain interaction of Eq. (36), the perturbation energy for the vacancy orbital is calculated up to the second-order processes of the external perturbation δ using Eq. (31). Here, the matrix elements of the electric quadrupoles of Eq. (35) are employed.

The total elastic energy U_{total} of Eqs. (22) and (23) for the SAW is renormalized by the quadrupole-strain interaction of Eq. (36) as

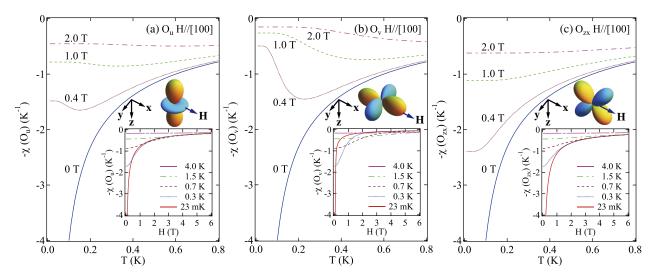


Fig. 11. (Color online) Temperature dependences of the quadrupole susceptibilities $-\chi(O_{\rm u})$ in (a), $-\chi(O_{\rm v})$ in (b), and $-\chi(O_{\rm zx})$ in (c) under applied the magnetic fields of $\mathbf{H} = (H_x, 0, 0)$ parallel to the propagation direction of the SAW. The anisotropic charge distribution of the electric quadrupoles is also presented. The inset shows the magnetic field dependences of the quadrupole susceptibilities at low temperatures.

$$U_{\text{total}} = \frac{1}{2} C_{\text{s}} (A_{\text{s}} f_{\text{s}}(z, x, t; \theta_{\text{s}}))^{2} \delta^{2} + \frac{1}{2} K_{0}(z) \delta^{2}$$

$$= \frac{1}{2} C_{\text{B}} (A_{\text{B}} f_{\text{B}}(z, x, t; \theta_{\text{B}}))^{2} \delta^{2}$$

$$+ \frac{1}{2} (C_{\text{T}} - N g_{\Gamma_{3}}^{2} \chi(O_{\text{u}})) (A_{\text{u}} f_{\text{u}}(z, x, t; \theta_{\text{u}}))^{2} \delta^{2}$$

$$+ \frac{1}{2} (C_{\text{T}} - N g_{\Gamma_{3}}^{2} \chi(O_{\text{v}})) (A_{\text{v}} f_{\text{v}}(z, x, t; \theta_{\text{v}}))^{2} \delta^{2}$$

$$+ \frac{1}{2} (C_{44} - N g_{\Gamma_{5}}^{2} \chi(O_{zx})) (A_{zx} f_{zx}(z, x, t; \theta_{zx}))^{2} \delta^{2}.$$
(37)

The second derivative of the elastic energy of the SAW of Eq. (37) with respect to the external perturbation δ gives the softening and its magnetic field dependence on the renormalized elastic constant C_s of the SAW as

$$\frac{\partial^{2} U_{\text{total}}}{\partial \delta^{2}} = C_{s} (A_{s} f_{s}(z, x, t; \theta_{s}))^{2} + K_{0}(z)$$

$$= C_{B} (A_{B} f_{B}(z, x, t; \theta_{B}))^{2}$$

$$+ (C_{T} - Ng_{\Gamma_{3}}^{2} \chi(O_{u}))(A_{u} f_{u}(z, x, t; \theta_{u}))^{2}$$

$$+ (C_{T} - Ng_{\Gamma_{3}}^{2} \chi(O_{v}))(A_{v} f_{v}(z, x, t; \theta_{v}))^{2}$$

$$+ (C_{44} - Ng_{\Gamma_{5}}^{2} \chi(O_{zx}))(A_{zx} f_{zx}(z, x, t; \theta_{zx}))^{2}. \quad (38)$$

The temperature dependence of C_s shown in Fig. 4 and the magnetic field dependence of C_s shown in Fig. 5(a) are explained in terms of the quadrupole susceptibilities $-\chi(O_u)$, $-\chi(O_v)$, and $-\chi(O_{zx})$ as

$$C_{s} = C_{s}^{0} - \frac{N}{(A_{s}f_{s}(z, x, t; \theta_{s}))^{2}} [g_{\Gamma_{3}}^{2} (A_{u}f_{u}(z, x, t; \theta_{u}))^{2} \chi(O_{u})$$

$$+ g_{\Gamma_{3}}^{2} (A_{v}f_{v}(z, x, t; \theta_{v}))^{2} \chi(O_{v})$$

$$+ g_{\Gamma_{s}}^{2} (A_{zx}f_{zx}(z, x, t; \theta_{zx}))^{2} \chi(O_{zx})].$$
(39)

Here, the quadrupole susceptibilities of $-\chi(O_{\rm u})$, $-\chi(O_{\rm v})$, and $-\chi(O_{\rm zx})$ as functions of temperature and magnetic field along the *x*-direction are shown in Figs. 11(a)–11(c), respectively. $C_{\rm s}^0$ is the background elastic constant where the quadrupole-strain interaction is ignored as

$$C_s^0(A_s f_s(z, x, t; \theta_s))^2$$

$$= C_B(A_B f_B(z, x, t; \theta_B))^2 + C_T(A_u f_u(z, x, t; \theta_u))^2$$

$$+ C_T(A_v f_v(z, x, t; \theta_v))^2 + C_{44}(A_{zx} f_{zx}(z, x, t; \theta_{zx}))^2$$

$$- K_0(z). \tag{40}$$

 C_s^0 is expected to faintly increase with decreasing temperature because of anharmonic vibrations of the Rayleigh wave.

5. Discussion

In order to fit the softening and its magnetic field dependence on C_s by Eq. (39), we need numerical values of the quadrupole-strain coupling constants of g_{Γ_3} and g_{Γ_5} in Eq. (30). Recently, the authors' group has reported the sum rule between the number of the vacancies consumed in the void formation and the number of residual vacancies proportional to the low-temperature softening of the C_{44} of the bulk transverse ultrasonic wave. ¹⁶⁾ This sum rule leads to a strong coupling constant of $g_{\Gamma_5} = 2.8 \times 10^5 \,\mathrm{K}$ between the electric quadrupole of O_{zx} and the counterpart symmetry strain of ε_{zx} in Eq. (30). Namely, the deformation energy for the external strain of $\varepsilon_{zx} = 1$ corresponds to $g_{\Gamma_5}\langle \Gamma_8 | O_{zx} | \Gamma_8 \rangle = 2.8 \times 10^5 / \sqrt{3} \,\mathrm{K} = 1.6 \times 10^5 \,\mathrm{K} = 14 \,\mathrm{eV}.$ Furthermore, the ratio of $g_{\Gamma_5}/g_{\Gamma_3}=1.6$ was deduced by measuring of the relative amount of the low-temperature softening of both elastic constants C_{44} and $(C_{11} - C_{12})/2.$ ¹⁵⁾ Using the coupling constants $g_{\Gamma_5} = 2.8 \times 10^5 \,\mathrm{K}$ and $g_{\Gamma_3} =$ $1.8 \times 10^5 \,\mathrm{K}$ and the amplitude $A_{\Gamma_{\nu}}$ and the phase $\theta_{\Gamma_{\nu}}$ in the function $f_{\Gamma_{\nu}}(z,x,t;\theta_{\Gamma_{\nu}})$ of Eq. (19) in Sect. 4.1, we have made a fit for the softening of C_s by the solid line in Fig. 4. We have deduced the vacancy concentration $N = 3.1 \times$ $10^{12}/\text{cm}^3$ in the surface layer within $\lambda_p = 3.5\,\mu\text{m}$ of the present silicon wafer. In the fitting, we simply offset the extrinsic phase shift at a superconducting transition temperature of 1.17 K of the Al electrode to continue on both sides. Furthermore, we adopted the temperature-dependent background of the elastic constant in Eq. (40) described by $C_s^0 = C^0 - a/(\exp[b/T] - 1)$ with the parameters $C^0 =$ $5.749 \times 10^{10} \,\text{J/m}^3$, $a = 6.30 \times 10^6 \,\text{J/m}^3$, and $b = 7.3 \,\text{K}$. The dashed line in Fig. 4 shows the background.

The magnetic fields applied parallel to the propagating xdirection of the SAW reduce the low-temperature softening of C_s . As shown in Figs. 5(a) and 5(b), the softening of C_s is completely suppressed at magnetic fields of up to 2.0 T. This is due to the lifting of the Kramers doublets of the Γ_8 - Γ_7 states by the Zeeman energy in Eq. (29). The magnetic field dependences of the quadrupole susceptibilities $-\chi(O_{\rm u})$, $-\chi(O_{\rm v})$, and $-\chi(O_{\rm zx})$ under the fields along the x-direction of up to 6T for various temperatures are shown in Figs. 11(a)–11(c), respectively. The configurations between the electric quadrupole and the applied field direction along the x-direction are also shown in Fig. 11. Furthermore, the insets of Figs. 11(a)-11(c) show the magnetic field dependence of the quadrupole susceptibilities of $-\chi(O_u)$, $-\chi(O_{\rm v})$, and $-\chi(O_{\rm zx})$, respectively, up to 6 T. The magnetic fields along the x- and z-directions give the same effects on the unperturbed Γ_8 - Γ_7 states of the vacancy orbital at the cubic site symmetry. When we treat the quadrupole-strain interaction of Eqs. (30) and (36) for the SAW propagating along the x-direction on the z surface, the present magnetic field along the x-direction affects the softening differently from that along the z-direction. The solid and dashed lines in Fig. 5(b) are fits for the softening and its field dependence on $C_{\rm s}$ in terms of the quadrupole susceptibilities in Eq. (34). The results of the theoretical calculations of C_s in Fig. 5(b) using the quadrupole susceptibilities are in fair agreement with those of the SAW experiments on C_s in Fig. 5(a).

Despite the fairly good consistency between the results of the SAW experiments and calculations based on the vacancy orbital with the Γ_8 – Γ_7 states, the softening of C_s below 0.1 K in the zero magnetic field in Fig. 5(b) slightly deviates from the theory. Because of the strong quadrupole-strain coupling constants $g_{\Gamma_3} = 1.8 \times 10^5 \,\mathrm{K}$ for the symmetry strains ε_{u} and $\varepsilon_{\rm v}$ and $g_{\Gamma_5} = 2.8 \times 10^5 \, {\rm K}$ for ε_{zx} , even a tiny strain of $\varepsilon \sim 10^{-6}$ may lift the Γ_8 quartet ground state to two Kramers doublets with an order of energy of 0.1 K. Although we have attempted to eliminate unwanted strains, tiny strains might still remain in the silicon sample. This may give rise to the lifting of the Γ_8 ground state, which plausibly accounts for the slight deviation in the softening of C_s from the theory. The precise measurement of the low-temperature softening of $C_{\rm s}$ as a function of externally applied stress acting on silicon samples is reserved for future work to verify the strainsensitive feature of the vacancy orbital with the Γ_8 ground state.

6. Conclusions

Focusing on boron-doped silicon wafers currently available for the fabrication of semiconductor devices, we have measured SAWs propagating on the surface of the wafer. An IDT with a comb gap of $w=2.5\,\mu\mathrm{m}$ optimally generated a SAW of $f_s=517\,\mathrm{MHz}$. A pulsed shape SAW propagates along the x-direction with a group velocity $v_s=4.967\,\mathrm{km/s}$ and a wavelength $\lambda_s=v_s/f_s=9.61\,\mu\mathrm{m}\sim 4w=10\,\mu\mathrm{m}$. The equations of motion for the SAW on the z surface of the silicon wafer give the Rayleigh wave with a velocity $v=4.844\,\mathrm{km/s}$, which is in agreement with the observed SAW velocity $v_s=4.967\,\mathrm{km/s}$. The SAW with the ellipsoidal trajectory of the displacement components u_x and u_z induces the time-dependent symmetry strains ε_B , ε_u , ε_v , and ε_{zx} with the different amplitude and phase shifts. The

almost anti-phase relations between ε_B and ε_{zx} and between ε_u and ε_v indicate the stationary wave character of the SAW during decay along the penetrating *z*-direction.

The remarkable softening of $\Delta C_s/C_s = 1.9 \times 10^{-4}$ below 2 K down to a base temperature of 23 mK is caused by the coupling between the SAW and the vacancy orbital with the Γ_8 ground state and Γ_7 excited state located at 1 K. Magnetic fields of up to 2T fully suppress the softening of the SAW due to the lifting of the Γ_8 – Γ_7 states at the magnetic fields. The low-temperature softening and its magnetic field dependence are well described in terms of the quadrupole susceptibilities based on the coupling between the symmetry strains $\varepsilon_{\rm u}$, $\varepsilon_{\rm v}$, and ε_{zx} of the SAW and the appropriate electric quadrupoles. Taking account of the strong quadrupole-strain interactions of $g_{\Gamma_5} = 2.8 \times 10^5 \,\mathrm{K}$ and $g_{\Gamma_3} = 1.8 \times 10^5 \,\mathrm{K}$ for the vacancy orbital, we deduced the low vacancy concentration $N = 3.1 \times 10^{12} / \text{cm}^3$ in the surface layer within a penetration depth $\lambda_p = 3.5 \,\mu m$ of the silicon wafer. The present result promises a highly sensitive diagnostic tool of the SAW where the softening of $\Delta C_s/C_s = 1.0 \times 10^{-4}$ corresponds to a vacancy concentration $N = 1.6 \times 10^{12} / \text{cm}^3$ in the surface layer. The advantage of the SAW is clearly seen as compared with sensitivity of the bulk transverse ultrasonic wave measurement where the softening of $\Delta C_{44}/C_{44} =$ 1.0×10^{-4} corresponds to $N = 1.5 \times 10^{13}/\text{cm}^3$ in the bulk region of the wafer.¹⁶⁾

The complementary metal oxide semiconductor is widely adopted in the fabrication of various large-scale integrated (LSI) devices of center processing units, dynamic random access memories, NAND-type flash memories, and charge couple devices to name a few. 38,39) The active region beneath the surface of the silicon wafer in the depth range of 1-3 μm is particularly utilized in LSI device fabrication. The vacancies play important roles in the proper control of a diffusion of dopants, precipitation of bulk micro-defects, and gettering of metal impurities in device fabrication. Therefore, the vacancy evaluation in the surface layer of silicon wafers is crucially required for the fabrication of the high-density LSI devices beyond 19 nm in size in order to obtain a high yield. The continuous pursuit of the high-quality wafers of polished, annealed, and epitaxial types with 450 mm as well as 300 mm in diameter demands the fine control of the vacancy concentration in the surface layer. The low-temperature measurement of the SAW demonstrated in the present study surely suggests an innovative technology for determining the vacancy concentration in the surface layer, which is one of the figure merits of the use of silicon wafers.

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