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Enhancement of second-harmonic generation from silicon stripes under external cylindrical strain

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The enhanced second-harmonic (SH) generation from Si (111) stripes induced by external cylindrical strain is investigated. The dependence of the intensity of the strain-induced SH on the sample azimuth shows that the Si under cylindrical strain has $3m$ symmetry, which is similar to that of the Si (111) surface. Further studies indicate that the intensity of the enhanced SH is a quadratic function of the cylindrical strain within the magnitude that the Si stripe can bear. For the p-polarized and s-polarized SH, the intensities are, respectively, enhanced by 47.9% and 13% at $\epsilon_0=2.93\times10^{-4}$. The enhancement of SH is due to the contributions from the strain-induced second-order nonlinear susceptibility $\chi^{(2)}_{\text{strain}}$ to the bulk dipole. © 2009 Optical Society of America

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According to the electric dipole approximation, second-harmonic generation (SHG) should be forbidden in bulk Si because of its centrosymmetry [1]. However, both the built-in electric field and internal strain at the interfaces of Si devices can break the centrosymmetry of Si, thus generating so-called electric-field-induced second harmonic (EFISH) and strain-induced SH (SISH), respectively. So the SHG from the interfaces should include two mechanisms, EFISHG and SISHG. It is very helpful to distinguish these two mechanisms, because people are willing to apply the sensitive SHG method for *in situ* research into both the electric properties and the mechanical characteristics of the buried interfaces. To differentiate these two mechanisms, both EFISHG and SISHG must be intensively and quantitatively studied. Over the past years, intensive investigation in EFISHG has been carried out [2–4]. However, SISHG from Si surfaces or interfaces has not attracted enough attention, even though the strain often plays an especially important role [5–7] in Si devices. There are several references about SISHG from Si/SiO$_2$ interfaces, but in these papers authors are concerned mainly with the effect of the thickness of SiO$_2$ layers on the SH intensity, and the internal mechanical strain at Si/SiO$_2$ interfaces has not been scaled quantitatively [8–10].

In this Letter, we report observations of the enhanced SHG from the Si surface induced by external strain that can be scaled, and we quantitatively study the dependence of the intensity of SISHG on the external strain.

A naturally oxidized Si (111) wafer of 0.5 mm thickness was cut into narrow stripes along the direction [$1\bar{1}0$], and cylindrical strain was induced in the Si stripe by moving the sharp edge of the strain device [11,12] in the [111] direction (that is, the z axis), shown in the right-hand part of Fig. 1(a). The tension strain along the [$1\bar{1}0$] direction of the Si stripe (that is, the x axis) depends on the deformation $J(r)$, where r is the distance from the center of the stripe and $J(r)=J_0(a-r)(2a^2+2ar-r^2)/2a^3$ [11]. Consequently the component of strain along the x direction is inhomogeneous. The maximum strain ϵ_0 is induced at the center of the Si stripe, which is independent of the elasticity of the material and can be calculated as $\epsilon_{xx}=\epsilon_0=3hJ_0/2a^2$ [11]. As for the rest of the strain

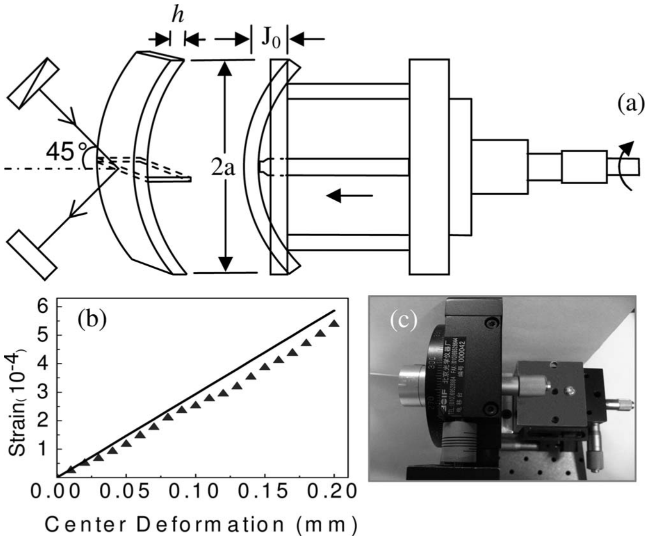

Fig. 1. (a) Left, schematic diagram of the experimental setup for SHG measurements. The two rectangles are the polarizer and analyzer, respectively. Right, schematic structure of the uniaxial strain equipment. J_0, h, and $2a$ are the central deformation, thickness, and length of the sample, respectively. (b) Strain along stripes of thickness h calculated from the central deformation J_0. Data points are experimentally obtained by measuring the radius of curvature. (c) Object assemblage of the uniaxial strain device and experimental setup.

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components, only the one perpendicular to the surface of the stripe will be nonzero, and it can be calculated by the equation $\epsilon_{zz}=\epsilon_0[2(S_{11}+2S_{12})+(6S_{12}-S_{44})]/[8(S_{11}+2S_{12})+2S_{44}]$ [12], where S_{11}, S_{12}, and S_{44} are the elastic compliances of the Si material. The inversion symmetry of Si crystal would be destroyed in the volume near the center of the stripe because the applied strain is inhomogeneous [13,14].

The SHG experimental setup is shown in the left-hand part of Fig. 1(a). A 120 fs unamplified Ti:sapphire femtosecond laser was used. The *s*-polarized fundamental light with the wavelength of 802 nm was focused onto the sample at an angle of 45° with respect to the *z* axis. The reflected *s*-polarized and *p*-polarized SHG signals were selected by a polarization analyzer, filtered by the saturated $CuSO_4$ solution, and then detected by a photomultiplier tube and a lock-in amplifier. In experiments, we first demonstrated the dependence of strain ϵ_0 on the center deformation by the deflection of a laser beam method [15], shown in Fig. 1(b). Note that the strain increases linearly with the rising of the center deformation.

In the dipole approximation [16], the surface dipole, bulk quadrupole, and dc electronic-field-induced and strain-induced bulk dipole are the main contributions to the SHG from Si surface. The bulk symmetry of Si material belongs to $m3m$ point group, and the strain-induced bulk dipolar contribution should be zero. However, application of inhomogeneous strain to the Si crystal can decrease its inversion symmetry [13,14]. Therefore the bulk dipolar contribution is not zero any longer under the cylindrical strain, and it will enhance the SH intensity. Then, by rotating the Si sample around its surface normal to change the azimuth, we measured the intensity of the *p*-polarized and *s*-polarized SH reflecting from the Si (111) face under *s*-polarized excitation as a function of the azimuth of the sample under different external mechanical forces.

According to the phenomenological theory of SHG, the intensities of *p*-polarized and *s*-polarized SH generated from Si surface can, respectively, be written as [9,14,16]

$$I_{s\rightarrow p}(2\omega)\sim|a_{s,p}+c_{s,p}\cos(3\phi)|^2, \tag{1}$$

$$I_{s\rightarrow s}(2\omega)\sim|b_{s,s}\sin 3\phi|^2, \tag{2}$$

where the azimuthal angle ϕ is measured from the $[1\bar{1}0]$ direction, and $a_{s,p}$, $b_{s,s}$, and $c_{s,p}$ for strained Si are different from the corresponding unstrained counterparts, and they should include the additional strain-induced electric dipolar $\chi^{(2)}_{\text{strain}}$ compared with the unstrained situation. As a result, the intensities of *p*-polarized and *s*-polarized SHs should be enhanced. The results are shown in Figs. 2 and 3, respectively. These curves agree with formulas (1) and (2) very well. Without the external strain, the SHG from the Si (111) surface is still considerable. But curves B and C show that the external strain enhances the intensity of the SH greatly. However, in our experiments the external strain does not change the azimuthal dependence of the SH intensity, which still has the characteristic of $3m$ symmetry. That means that the small strain (about 10^{-4}) induced by the uniaxial device does not greatly affect the $3m$ symmetry of the Si (111) surface, but evidently enhances the SH intensity. So the nonvanishing independent components of the $\chi^{(2)}_{\text{strain}}$ tensor are $\chi^{(2),\text{strain}}_{zzz}$, $\chi^{(2),\text{strain}}_{zxx}=\chi^{(2),\text{strain}}_{zyy}$, $\chi^{(2),\text{strain}}_{yyy}=\chi^{(2),\text{strain}}_{xxy}$.

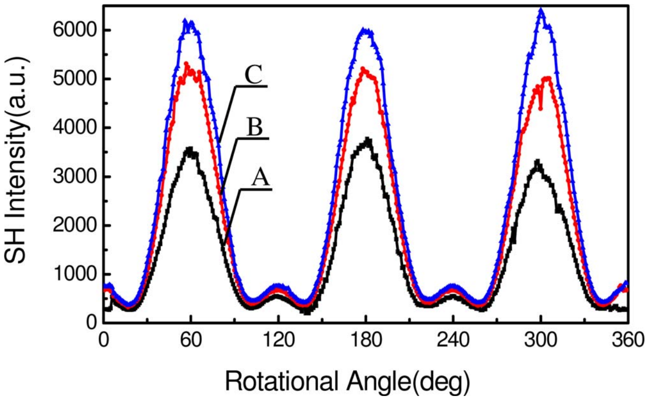

Fig. 2. (Color online) The *p*-polarized SHG intensity from the unstrained and strained Si (111) surface for *s*-polarized excitation as a function of the sample rotation angle ϕ. Curve A is obtained from the unstrained Si surface. The corresponding strain values with curves B and C are 2.93 $\times 10^{-4}$ and 5.86×10^{-4}, respectively.

Furthermore, we quantitatively investigate the dependence of the SH intensity on the strain in order to more profoundly understand the effect of strain on SHG from Si. The experimental results are shown in Fig. 4. The dependences of *p*-polarized and *s*-polarized SH intensity on the external strain are measured at the azimuth, of 60° and 30° (where the SH intensity has the maximum), respectively. Note that the reflected SH intensity increases quadratically with the external strain till the Si wafer is broken.

The *p*-polarized SH electric field reflected from strained Si with the excitation of an *s*-polarized field

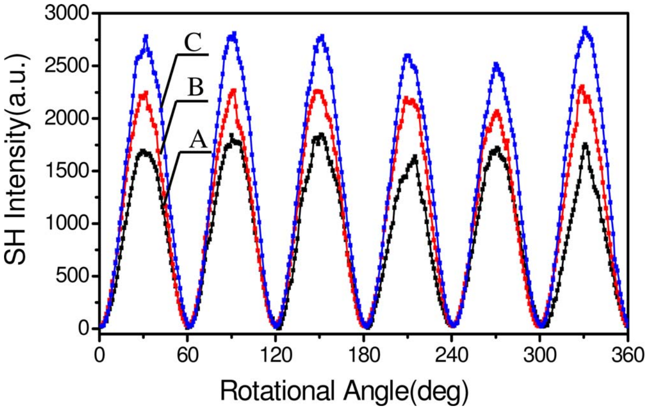

Fig. 3. (Color online) The *s*-polarized SHG intensity from unstrained and strained Si (111) surfaces for *s*-polarized excitation as a function of the sample rotation angle ϕ. Curve A is obtained from the unstrained Si surface. The corresponding strain values with curves B and C are 2.93 $\times 10^{-4}$ and 5.86×10^{-4}, respectively.

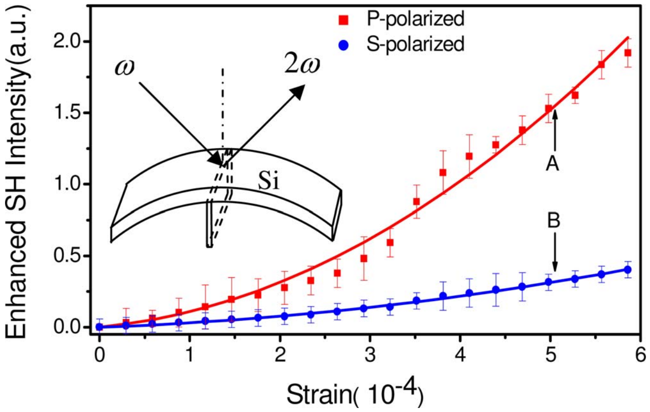

Fig. 4. (Color online) Intensity of the p-polarized and s-polarized SH signals from reflected Si surface as a function of the strain. The solid curves A and B are fitted, and the fitting functions can be expressed as $I_{s\to p}=4.8773\times 10^6\epsilon_0^2+0.0006\times 10^6\epsilon_0$ for the p-polarized SH signal and $I_{s\to s}=8.0749\times 10^5\epsilon_0^2+0.0022\times 10^5\epsilon_0$ for the s-polarized SH signal, respectively.

[9,14,16] is found to be proportional to $\chi_{zyy}^{(2),\text{strain}}$, that is,

$$E_{s\to p}(2\omega)\propto i\chi_{zyy}^{(2),\text{strain}}+a\zeta, \tag{3}$$

where $\chi_{zyy}^{(2),\text{strain}}$ is the strain-induced component of the second-order bulk contribution and ζ is the bulk quadrupole contribution that is independent of the strain, a constant number containing the Fresnel factors for reflection. From Eq. (3), the enhanced SH intensity should be proportional to $|\chi_{zyy}^{(2),\text{strain}}|^2$, which results from the inhomogeneous lattice strain [14]. In the presence of strain Si–Si bonds are distorted. This mechanism can be explained by a microscopic model that describes the interface stress in the subsurface layers of semiconductor crystals such as Si, and $\chi_{zyy}^{(2),\text{strain}}$ can be calculated as follows [9,14,17]:

$$\chi_{zyy}^{(2),\text{strain}}(\text{esu})=1.03\times 10^{-6}\epsilon_0. \tag{4}$$

Therefore, strain-enhanced p-polarized SH intensity $I_{s\to p}(2\omega)$ should be proportional to $|\epsilon_0|^2$, which agrees with the experimental data well. In experiments, the SH intensity is enhanced by 47.9% at $\epsilon_0=2.93\times 10^{-4}$, which is also comparable with the theoretical enhancement of 57%.

The other component is $\chi_{yyy}^{(2),\text{strain}}$, according to its relation with $\chi_{zyy}^{(2),\text{strain}}$ as described in [14], and $\chi_{yyy}^{(2),\text{strain}}(\text{esu})=-0.78\times 10^{-6}\epsilon_0$. Similarly, the s-polarized SH intensity is enhanced by 13% in the experiments, and it is enhanced by 13.7% in theory at $\epsilon_0=2.93\times 10^{-4}$.

In conclusion, this Letter provides a method of measurement of small strain. The experimental results of the SH intensity versus the azimuth angle of Si indicate that external cylindrical strain can reduce the bulk symmetry of Si crystal from $m3m$ to $3m$. Consequently, a strain-induced $\chi_{\text{strain}}^{(2)}$ will contribute to the considerable enhancement of SH from the Si sample. The experiments of SH intensity as a function of the external strain testify that the reflected SH intensity increases quadratically with the external cylindrical strain within the magnitude that the Si stripe can bear. These studies will be helpful to distinguish the contribution of the mechanical strain from that of the surface electric field to SHG from Si surfaces and interfaces, even though further investigations are still required in order to realize this goal.

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