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Resonant second-harmonic phase spectroscopy of the buried interfaces of Column IV semiconductors

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Abstract. The second-harmonic interferometric spectroscopy (SHIS) is proposed as a new spectroscopic technique to study the resonant electron response of solid state nanostructures. The combination of the second-harmonic (SH) amplitude and phase spectra is shown to be more sensitive to resonant parameters of the electron density of states in comparison with the conventional SH intensity measurements. The resonant anisotropic SH response of buried oxidized Si(111) and Ge(111) surfaces is studied using SHIS in the vicinity of the E_2 critical points. Both spectra of the phase and amplitude of the SH field with different lineshapes for Si and Ge surfaces are obtained.

Introduction

The nonlinear-optical response of the solid-state nanostructures is an effective finger-print of the properties of the quantum-confined electron subsystem of low-dimensional structures. The second-harmonic (SH) generation stands out among other nonlinear-optical techniques as inherently sensitive to nanostructures fabricated from centrosymmetric materials, since the SH generation is forbidden in the bulk of media with inversion symmetry in the dipole approximation. It means that the SH spectroscopy can be used for a study of the features of the electron density of states in nanostructures and in subsurface and interface layers of centrosymmetric semiconductors, where electron motion is also confined, that was successfully demonstrated in numerous works [1]. Recently the technique of a single-beam SH phase measurements [2] was modified in order to measure simultaneously the spectra of the phase and amplitude of the SH radiation of nanostructures, including the buried semiconductor interfaces [3]. The combination of the SH phase and amplitude spectra, extracted from the SH interferometric spectroscopy (SHIS) data, is more sensitive to parameters of resonances and it circumvents the sign uncertainty of the real part of quadratic susceptibility inhering in the conventional spectroscopy of the SH intensity. In this paper, the spectral dependences of the phase and amplitude of the SH field from the buried oxidized Si(111) and Ge(111) interfaces are measured in the vicinity of the E_2 critical point (CP) of Si and E_1 and E_2 CP of Ge using SHIS. The different types of spectral lineshapes of the SH resonances for Si and for Ge are observed.

Results and discussion

The samples are natively oxidized Ge(111) and slightly vicinal Si(111) wafers. The p-polarized output of a tunable nanosecond parametric generator/amplifier laser system operating in the interval of 490–690 nm is used. A 1 mm-thick plate of fused quartz coated with a 30 nm-thick indium-tin oxide ITO) film is chosen as a reference for SHIS. The total

SH intensity $I^{2\omega}$ detected is a result of the coherent superposition of the SH fields from the reference, $E_r^{2\omega}$, and from the sample, $E_s^{2\omega}$. The dependence of $I^{2\omega}$ on the distance l between the reference and the sample (the SH interferogram) is given as

$$I^{2\omega}(l) = \frac{c}{8\pi}\left|E_r^{2\omega} + E_s^{2\omega}\right|^2 = I_r^{2\omega} + I_s^{2\omega} + 2\alpha\sqrt{I_r^{2\omega} I_s^{2\omega}}\cos\left(\frac{4\pi\Delta n}{\lambda_\omega} l + \Phi_{rs}\right), \quad (1)$$

with $\Delta n = n_{2\omega} - n_\omega$ describing the air dispersion and $\alpha < 1$ stands for the laser coherence. Additional position-independent phase shift $\Phi_{rs}(\lambda_\omega, \lambda_{2\omega})$ between $E_r^{2\omega}$ and $E_s^{2\omega}$ is associated with spectral properties of a quadratic susceptibility $\chi^{(2)}$ of the reference and the sample and Green's corrections for SH radiation propagation and reflection.

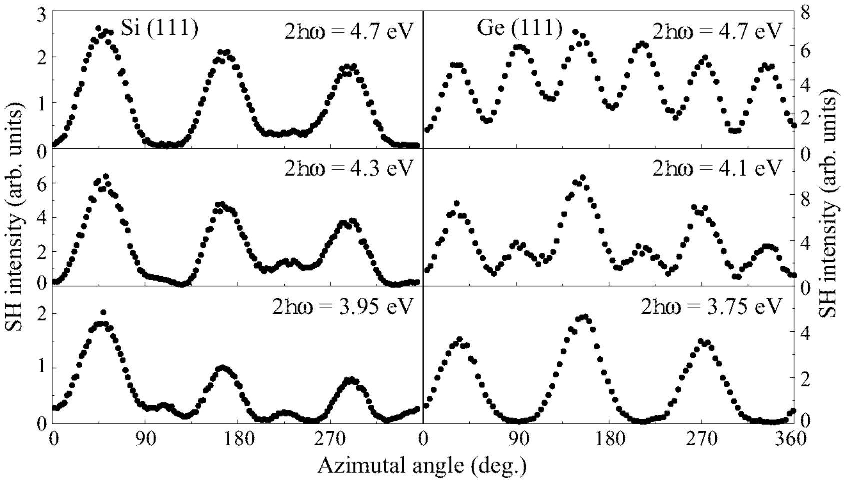

Fig. 1. The SH rotational azimuthal dependences a slightly vicinal Si(111) (left panel) and Ge(111) (right panel) oxidized surfaces measured for different SH photon energies in the *p*-in, *p*-out geometry.

Figure 1 shows the dependences of the intensity $I_s^{2\omega}$ of the p-polarized SH light on the azimuthal angle ψ of the rotation of the sample around its normal for a slightly vicinal Si(111) (left panel) and Ge(111) (right panel) oxidized surfaces measured for different fundamental wavelengths. The SH azimuthal dependences produced by the interference of the one- and three-fold anisotropic SH components with the rotationally isotropic SH term:

$$I_s^{2\omega}(\psi) = \left|C_0 + C_1 e^{i\phi_1}\cos(\psi - \psi_0) + C_3 e^{i\phi_3}\cos[3(\psi - \psi_0)]\right|^2. \quad (2)$$

The one-fold component, C_1, comes entirely from the vicinal surface, while the three-fold, C_3, and isotropic, C_0, terms are generated by both surface dipole and bulk quadrupole sources. For Si(111)-SiO$_2$ interface $I_s^{2\omega}(\psi)$ dependences measured for different fundamental wavelengths are similar one to each other. This indicates that both anisotropic C_1, C_3 and isotropic C_0 components have almost the same spectral dependences, while their relative phases, ϕ_1 and ϕ_3, are spectral-independent. In the case of the Ge(111) surface, the vicinal contribution C_1 is negligible and the SH azimuthal dependences comes from the interference of the three-fold anisotropic and isotropic SH contributions. The $I_s^{2\omega}(\psi)$ dependence for Ge(111) surface varies from the almost three-fold shape at the "red" end of the tuning region to the six-fold shape at the "blue" end. It means that not only amplitudes of the isotropic and anisotropic contributions are spectral dependent, but the relative phase ϕ_3 has also a pronounced spectral dependence, changing approximately on $\pi/2$ on the region of the fundamental wavelength tuning.

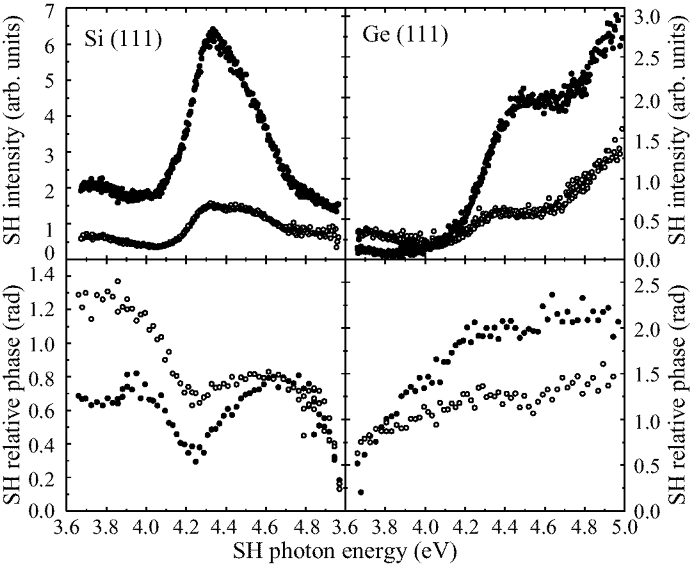

Fig. 2. The SH intensity (top panels) and the SH wave phase (bottom panels) as functions of the SH photon energy measured for Si(111) (left) and Ge(111) (right) buried interfaces in different points of the SH azimuthal dependence.

Figure 2 shows the spectra of intensity (top panel) and the phase (bottom panel) of the SH wave measured in the maximum of the p-in, p-out SH azimuthal anisotropy, where both isotropic and anisotropic SH components contribute to the SH signal (filled circles), and in the position, 30° shifted from the maximum, where the SH response originates entirely from the isotropic term (open circles). For the Si(111) surface, both SH intensity spectra peak at approximately 4.3 eV close to the resonance of the E_2 CP and have an Lorenz-like line shape. The spectral dependences of the SH phase, extracted from the set of the SH interferograms, increase within the interval of 4.2–4.6 eV, which is typical for near-resonance phase behavior near E_2 CP; however outside this energy region the spectral derivative of the SH phase changes the sign, what unambiguously indicates the influence of the lower (E_0'/E_1) and higher (E_1') resonances of Si interband transitions. In the case of the Ge(111) surface, the SH intensity spectra show a strong increase in the spectral range of 4.0–4.6 eV. However, the maxima of the spectra are sufficiently shifted from the interval of 4.1–4.3 eV, where both one-photon E_1 and two-photon E_2 resonances are located and where the SH phase spectra have the maxima of the spectral derivative in this region. Such a nonsymmetric line shape could be attributed with the 2D type of CP.

Summarizing, the general scheme of the SH interferometric spectroscopy is presented. Using this technique, the phase and amplitude of the SH radiation from the buried Si(111) and Ge(111) surfaces are measured in the interval of SH photon energies from 3.6 eV to 5 eV, and contributions from interband transitions located at E_2 critical point of Si and Ge are separated.

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