FEATURES OF THE RAMAN SCATTERING IN QUASIPERIODIC NANOSTRUCTURES OF POROUS SILICON

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The possibility of intensity enhancement of Raman scattering of near infrared light on the photon band gap edge in periodic multilayer structures produced from porous silicon consisting of porous silicon layers sequences with a high and low refractive index was shown theoretically and experimentally. The obtained results show perspectivity of use of these structures as a matrix for enhancement of Raman scattering efficiency.

Introduction

Today crystalline silicon (c-Si) is a basic material of the up-to-date microelectronics, however, high isotropy of its linear optical properties essentially restricts possibilities of its use in a photonics. Formation of anisotropic silicon micro- and nanostructures which possess the significant optical anisotropy necessary for light control can be a solution to the problem [1]. Multilayer structures (in particular periodic ones), fabricated on the basis of porous silicon (por-Si) can be very perspective in this case. These structures possess unique optical properties significantly different from those for c-Si that is caused by presence of quasi-periodic variations of dielectric constants. It allows to create high quality optical materials with desirable properties via control of their structural parameters.

At present the Raman scattering [2,3], nonlinear optical properties [4,5] and photon crystal properties [5,6] of por-Si layers formed by electrochemical etching are well explored and these data show new mechanisms of nonlinear optical interactions and efficiency amplification of the process in such low-dimension silicon structures. Multilayer structures based on por-Si can be used for fabrication of photon crystals (PC) integrated with silicon technology of microelectronics that will allow to control light streams inside the chip and radiative modes on the edge of a photon band gap (PBG) or in the area of the microresonators defective modes.

Essential modification of optical properties of nanostructures based on por-Si is caused by effects of local electric fields which appear at distribution of light in such heterogeneous dielectric structures. More appreciable influence of local electric fields can be expected for these structures for Raman scattering and other nonlinear optical processes. However until recently not enough attention was given to studying of the Raman scattering phenomenon in the lamellar structures made of por-Si and in particular to studying of influence of these silicon nanostructures parameters on efficiency of a Raman scattering. The investigation of Raman scattering in silicon structures attracts attention due to the recent works regarding the Raman laser on silicon [7], compatible to planar semiconductor technology.

Experimental setup

Samples of multilayer porous silicon structures were fabricated by electrochemical etching of (100)-oriented p-type c-Si wafers with a specific resistivity of ~ 20 mOhm·cm in an electrolyte based on fluoric acid (HF 48 %) with ethanol (C_2H_5OH), taken in the ratio 1:1. During the etching there was an alternation of etching current density for formation of lamellar structure with periodic alternation of layers with various porosity. Consequently obtained structures consisted of sequences of mesoporous silicon layers with the typical size of silicon nanocrystals about ten nanometers. Before the beginning of electrochemical etching the c-Si wafer was immersed in a solution of fluoric acid for removal of natural oxide on its surface. After preparation lamellar structures based on por-Si were dried on ambient air. Structural parameters and labels of explored samples are given in Table 1.

Table 1. Structure parameters of the multilayer structures based on PS

Sample	Current density, mA/cm²	Refractive index of layers	Thickness of layers, nm	Quantity of layer's pairs	Center of PB, cm^{-1} (μm)
PC-1	j_1=10 j_2=40	n_1 = 2.53 n_2 = 2.32	d_1 = 100 d_2 = 110	15	8495 (1.177)
PC-2	j_1=10 j_2=40	n_1 = 2.53 n_2 = 2.32	d_1 = 96 d_2 = 110	15	10000 (1.00)
PC-3	j_1=20 j_2=30	n_1 = 2.49 n_2 = 2.39	d_1 = 96 d_2 = 114	30	9780 (1.022)

The reflection spectra of explored samples in near infrared range were measured by using a FTIR spectrometer Bruker IFS-66 v/S. Nonpolarized light of near infrared range were used for measuring of reflection spectra of lamellar structures based on por-Si. The Raman spectra were measured by using a FTIR spectrometer Bruker IFS-66 v/S with a FRA-106 unit for the Raman scattering detection under excitation with a cw Nd:YAG laser at 1.06 μm. The measurements of Raman spectra were performed in backscattering geometry with a spectral resolution of 2 cm^{-1}. Linear polarized excitation light was focused in spot with diameter about 1.5 mm at a laser power of 100 mW. Absence of the essential heating of samples was controlled by invariance of ratio of Stokes and anti-Stokes intensities with the change of excitation light intensity. All experiments were carried out at room temperature.

Results and discussion

The experiments have shown that multilayer structures based on por-Si possess properties of the high quality one-dimensional PC in near infrared range (see Fig. 1).

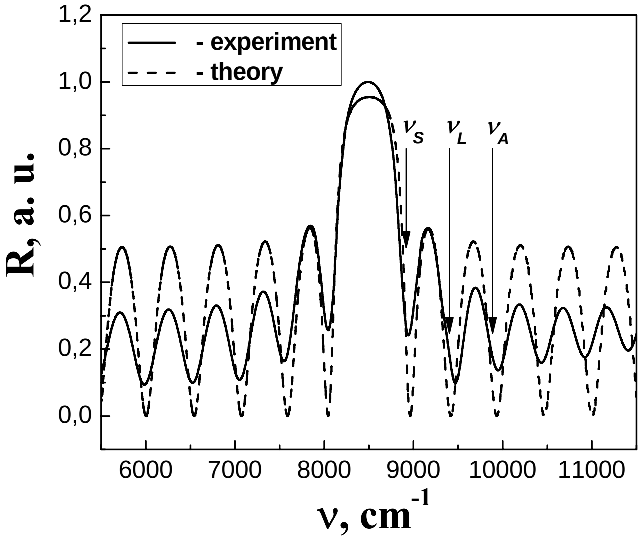

Fig. 1. Reflectance spectrum of multilayer structure PC-1 measured at the normal light incidence and theoretical spectrum calculated by the method of characteristic matrixes. Arrows shows position of pump radiation frequency (ν_L) and frequency of Stokes (ν_S) and anti-Stokes (ν_A) components of Raman scattering

The experimental reflection spectra of these structures are well fitted by the theoretical spectra calculated by the method of characteristic matrixes [8].

Moreover Raman scattering experiments have shown strong increase of Raman scattering intensity in these structures. The enhancement of Raman scattering signal intensity can occur either due to drastic change of the linear optical parameters of explored structure in comparison with a c-Si wafer, or radical modification of a tensor of the Raman susceptibility. It is known that in the case of quasistatic approach when absorption is small the intensity of Stokes (anti-Stokes) component of the Raman scattering will be determined by dispersion of linear optical parameters of structure[9]:

$$I_{S,A} \sim \frac{T_{S,A} T_L \nu_{S,A}^3}{(\alpha_{S,A} + \alpha_L) n_{S,A} n_L} \cdot \left| \vec{e}_L \vec{e}_{S,A} \chi_{S,A} \right|^2 \tag{1}$$

where T_L is the transmission coefficient, α_L is the absorption coefficient, n_L is the refractive index for incident light; $T_{S,A}$ are the transmission coefficients, $\alpha_{S,A}$ are the absorption coefficients, $n_{S,A}$ are the refractive indices, $\chi_{S,A}$ are the Raman susceptibility for Stokes and anti-Stokes components, respectively, $\vec{e}_L, \vec{e}_{S,A}$ are the unit vectors.

When the absorption is small the Raman susceptibility is proportional to the first derivative as follows [10]:

$$\chi_{S,A}(\nu,\theta) \sim \frac{\partial \varepsilon_{eff}(\nu,\theta)}{\partial \nu} \tag{2}$$

where v is the frequency, θ is the angle of incidence.

The effective dielectric function can be expressed from the linear optical characteristics [5,6]:

$$\varepsilon_{eff}(\nu,\theta) = \left[\frac{1+\sqrt{R(\nu,\theta)}}{1-\sqrt{R(\nu,\theta)}} \right]^2, \tag{3}$$

where R is the reflective coefficient

The reflection spectra of analyzed structures show that the reflective coefficient changes dramatically on the boundaries of PBG. Hereupon the effective dielectric function of multilayer structure based on por-Si which generally is a complex quantity, will also change essentially on the boundary of PB. Therefore dependence $\frac{\partial \varepsilon_{eff}}{\partial \nu}(\nu)$ will have strongly pronounced maxima that correspond to boundaries of PB. Thus, according to the Eq. (1) on boundaries of PB a strong increase of Raman scattering signal intensity should be observed that is related to drastic enhancement of the Raman susceptibility.

Raman scattering in samples of PC based on por-Si was studied at excitation by radiation with a wavelength of 1.064 mkm ($\nu = 9398.49$ cm^{-1}). The selection of radiation with the wavelength is related to the fact that on this wavelength absorption both in c-Si, and in the por-Si is small, and also that infrared radiation is widely used in fiber-optic lines. Raman scattering lines on frequency of 520.5 cm^{-1} corresponding to scattering on TO-phonons in c-Si are well distinguishable. Very small intensity of anti-Stokes component is caused by its falls into the interband absorption region for c-Si. Along with Raman scattering lines at spectra there is wide band related to a photoluminescence excited by the pump laser radiation. As photon energy of pump laser ($h\nu_L$ = 1.165 eV) exceeds bandgap of c-Si (E_g = 1.12 eV) and can cause photogeneration of the electron-hole pairs, interband recombination of which leads to appearance of PL. Figure 2 shows that essential modification of the PL spectrum shape for multilayer structures is observed which is related to the presence of PB. Spectral position of PB for analyzed samples of PC explains well the observable changing of PL spectrum. Intensive interband PL shows that in such structures effective photogeneration of free carriers that can be used for control of PB position by excitation light.

As mentioned above, for the enhancement of Raman scattering efficiency in multilayer silicon structures it is necessary, that component of Raman scattering gets on the edge of PB. Therefore PC were fabricated, having basic PB, lying near to a line of excitation radiation (see Fig. 1). The smooth change of PB position was produced by variation of the angle of incidence of excitation radiation on explored sample that led to shift of PB to high-frequency region. Therefore there was a smooth change of $\varepsilon_{eff}(\nu)$ that led to change of Raman scattering efficiency on TO-phonons in c-Si (ν_S = 520.5 cm^{-1}). It is visible from angular dependence of Stokes component intensity for sample PC-1 that at a certain incidence angle of excitation light on the sample (~10º) the characteristic maximum is observed. The observable maximum of Raman scattering intensity at an incidence angle of pump radiation witch equals to 10º corresponds to hitting of the Stokes component in the edge of PB. At the same time, anti-Stokes component frequency for the given sample will be out of PB. The highest value of quantity that leads to growth of quantity of the Raman susceptibility and according to the equation (1) growth of Stokes component intensity is thus reached. Note that almost fivefold growth of Raman scattering intensity signal observed in experiment gives only inferior estimate of quantity of the Raman susceptibility growth as in the conditions of spent experiment with growth of an incidence angle scattered light collecting also got worse which has been proved in check experiments with samples c-Si.

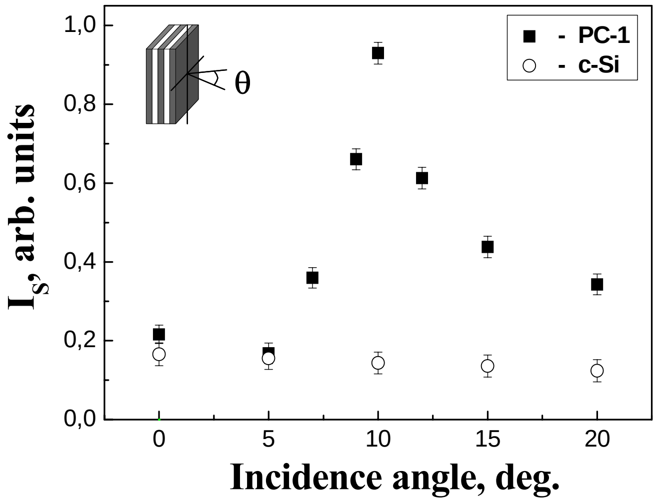

Fig. 2. Dependence of the Stokes scattering intensities for the sample PC-1 and c-Si substrate from incidence angle of excitation light (λ = 1.064 μm). The inset shows geometry of the experiment.

Conclusion

Thus, the obtained results demonstrate the possibility of significant enhancement of the Raman scattering intensity of infrared light on the photon band gap edge in periodic multilayer structures produced from porous silicon. The increase of the Stokes component intensity of the Raman scattering on silicon phonons can be explained by the essential increment of the Raman scattering tensor components on the edge of the photon band gap. This effect can be used for the reduction of the threshold for the stimulated Raman scattering as well as for an improvement of the parameters of the Raman laser on silicon. Furthermore effective photogeneration of free carriers in these structures gives possibility of control of photon band gap position and as consequence the value of Raman scattering intensity amplification by intensity of pumping radiation.

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КЕУЕКТІ КРЕМНИЙ НЕГІЗІНДЕГІ КВАЗИПЕРИОДТЫ НАНОҚҰРЫЛЫМДАРДЫҢ ЖАРЫҚТЫҚ КОМБИНАЦИЯЛЫҚ ШАШЫРАУ ЕРЕКШЕЛІКТЕРІ

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Теориялық және тәжірибелік тұрғыдан жақын инфрақызыл жарығының комбинациялық шашырау интенсивтілігін фотондардың жұтылу шекарасында кеуекті кремнийдің сыну көрсеткіштері әртүрлі, алма-кезек орналасқан қабаттарынан тұратын көпқабатты периодты құрылымы үшін ұлғайту мүмкіндігі көрсетілген. Алынған нәтижелер бұл құрылымдардың комбинациялық шашырау эффективтілігін арттыру үшін матрица ретінде қолдану перспективтілігін көрсетеді.

ОСОБЕННОСТИ КОМБИНАЦИОННОГО РАССЕЯНИЯ СВЕТА В КВАЗИПЕРИОДИЧЕСКИХ НАНОСТРУКТУРАХ НА ОСНОВЕ КРЕМНИЯ

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Теоретически и экспериментально показана возможность усиления интенсивности комбинационного рассеяния ближнего инфракрасного света на краю поглощения фотонов в периодической многослойной структуре, состоящей из чередующихся слоев пористого кремния с высоким и низким показателями преломления. Полученные результаты показывают перспективность использования этих структур в качестве матрицы для усиления эффективности комбинационного рассеяния.