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Polarized reflectance spectroscopy of aluminum nanoantennas on the surface of emitting GeSiSn/Si heterostructures

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Abstract. The study presents an investigation of the optical properties of rectangular-shaped aluminum nanoantenna arrays formed on the surface of an emitting GeSiSn/Si heterostructure with multiple quantum wells. The positions of the localized surface plasmon resonance modes excited along the long (*L*) and short (*S*) sides of the examined nanoantennas are determined utilizing the technique of Fourier-transform infrared reflectance anisotropy spectroscopy. Experimental results demonstrate that both *L*- and *S*-modes are located in the near-infrared range, and as the lateral dimensions of the nanoantennas increase, the modes' positions shift towards lower energies with an increase in the intensity of the resonance. The *S*-mode appears in the spectra as an overlay on the more pronounced *L*-mode with an intensity an order of magnitude lower. The geometry of the nanoantennas arrays with the resonance position near the photoluminescence peak of $Ge_{0.84}Si_{0.076}Sn_{0.084}$/Si heterostructures ($E \approx 0.65$ eV) is characterized.

Keywords: polarized reflectance, reflectance anisotropy spectroscopy, RAS, aluminum nanoantennas, localized surface plasmon resonance, LSPR

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Поляризационная спектроскопия отражения алюминиевых наноантенн на поверхности излучающих гетероструктур GeSiSn/Si

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Аннотация. В работе представлены результаты исследования оптических свойств массивов алюминиевых наноантенн прямоугольной формы, сформированных на поверхности гетероструктуры GeSiSn/Si с множественными квантовыми ямами. По спектрам ИК фурье-спектроскопии анизотропного отражения определены положения мод локализованного поверхностного плазмонного резонанса, возникающего вдоль длинной (*L*) и короткой (*S*) сторон рассматриваемых наноантенн. Экспериментально показано, что обе моды расположены в ближнем ИК-диапазоне и при увеличении латеральных размеров наноантенн смещаются в сторону меньших энергий с увеличением интенсивности резонанса. *S*-мода проявляется в спектрах в виде наложения на более интенсивную *L*-моду и имеет интенсивность на порядок меньше. Среди исследованных образцов определена геометрия массивов наноантенн с резонансом вблизи энергии пика фотолюминесценции гетероструктур $Ge_{0.84}Si_{0.076}Sn_{0.084}$/Si $E \approx 0.65$ эВ.

Ключевые слова: поляризационная спектроскопия отражения, спектроскопия анизотропного отражения, алюминиевые наноантенны, локализованный поверхностный плазмонный резонанс

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Introduction

The integration of photonic devices into silicon electronics is possible through the development of light detectors and emitters based on GeSiSn alloy nanoheterostructures. Such semiconductor structures achieve operating range in the near and mid-infrared (IR) spectrum down to 8 microns. This characteristic renders them highly promising for use in fiber-optic systems, photonic integrated circuits and sensors made on a single silicon chip [1]. The sensitivity and emission properties of these heterostructures can be controlled and enhanced through the creation of hybrid systems involving plasmonic metal nanoparticles on the surface of GeSiSn/Si heterostructures.

The interaction between electromagnetic radiation and metal nanoparticles induces the oscillation of free electrons within the metal, resulting in localized surface plasmon resonance (LSPR) [2]. The outcome of this resonance is a pronounced amplification of the electric field in the vicinity of the surface by several hundred times. The confinement of electromagnetic radiation energy within a nanovolume, owing to the LSPR phenomenon, finds broad applications predominantly associated with the enhancement of luminescence in various emissive structures [3].

The frequency and intensity of LSPR depend on the size, shape and dielectric environment of metal nanoparticles [2]. In practice, this renders them a universally applicable physical system for manipulating emissive structures across a broad optical spectrum. Typically, silver or gold is employed as the material for plasmonic nanoparticle production, owing to their strong interaction with light and lower losses [4]. As an alternative, aluminum can be used in a wider optical range with plasmon resonance in nanoparticles varied from ultraviolet to mid-infrared with limitations in a narrow part of the optical spectrum around 800 nm caused by interband transitions [5].

To investigate the formed metal nanoparticles, one can employ polarized reflectance spectroscopy, specifically utilizing its variant, the Fourier-transform infrared (FTIR) reflectance anisotropy (RA) spectroscopy method [6]. This optical technique has demonstrated its efficiency as a robust tool for examining plasmon resonances in metal nanoparticles [7].

Thus, the purpose of this work is to study the optical properties of aluminum nanoantenna arrays formed on the surface of an emitting GeSiSn/Si nanoheterostructure using methods of polarized reflectance spectroscopy.

Materials and Methods

Arrays of aluminum nanoantennas were formed through electron nanolithography on the surface of a heterostructure with multiple $Ge_{0.84}Si_{0.076}Sn_{0.084}$/Si quantum wells, where the well and barrier widths are 1 nm and 7 nm, respectively. The growth of the heterostructure was carried out using molecular beam epitaxy [1]. The nanoantennas were deposited on an area of 500×500 μm^2.

Figure 1 illustrates the geometry of the arrays structure, exemplified by an image of array #3 captured through scanning electron microscopy (SEM). The designations of the lateral dimensions of the nanoantennas, which distinguish each array structure from one another, are provided in the figure. These dimensions are detailed in Table.

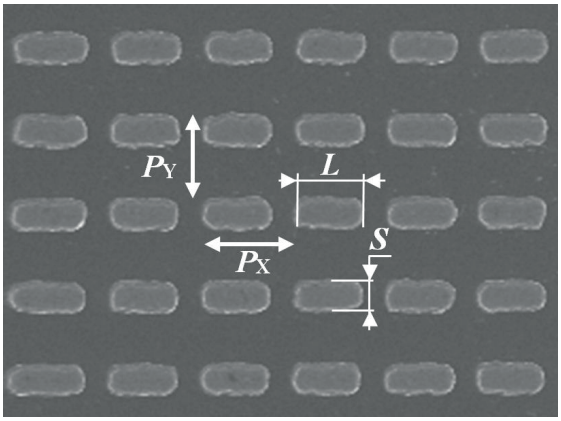

Fig. 1. Image of the nanoantenna structure array #3 obtained by scanning electron microscopy. The nominal height H of all nanoantennas is 50 nm

Table

Parameters of the studied aluminum nanoantennas arrays formed on the surface of the $Ge_{0.84}Si_{0.076}Sn_{0.084}$/Si radiating heterostructure with multiple quantum wells

sample	L, nm	S, nm	H, nm	P_x, nm	P_y, nm
#1	200	140	50	485	435
#2	265	147		478	435
#3	360	157		485	435

Polarized reflectance spectra were measured using a Vertex 80 Fourier-transform infrared (FTIR) spectrometer with radiation linearly polarized along the long L and short S sides of the nanoantennas. The modulation spectra of reflectance anisotropy were measured using an original setup, implemented on the basis of the same FTIR spectrometer and described in detail in [6]. The essence of the latter method is to modulate the direction of linear polarization of the probe radiation along two perpendicular directions x and y lying in the plane of the samples surface. The value being measured synchronously corresponds to the normalized difference in reflectance coefficients:

$$\frac{\Delta R}{R} = 2\frac{R_x - R_y}{R_x + R_y},$$

where R_x and R_y represent the reflectance coefficients when the plane of the incident radiation linear polarization is aligned along the corresponding direction in the surface plane. In this study, the x and y directions coincide with the long (L) and short (S) sides of the aluminum nanoantennas, respectively (see Fig. 1).

Results and Discussion

From each of the presented nanoantenna arrays, an ordinary reflectance spectrum R was obtained with probe radiation linearly polarized along the L and S sides. The reference reflectance

spectrum was measured from a gold-coated mirror, exhibiting a high reflectance range from 2.3 eV to mid-IR energies. A typical reflectance spectrum is shown in Fig. 2 (top) utilizing array #3 as an example. Analyzing ordinary reflectance spectra proves challenging due to the difficulty in identification of areas where the differences in reflectance spectra with linear polarization direction **E** along *L* (**E**//*L*) and *S* (**E**//*S*) are marginal against the general background of the total reflected radiation. In such cases, the modulation method of reflectance anisotropy spectroscopy is advantageous, wherein only the polarization-dependent component of the reflectance spectra is detected. Fig. 2 (bottom) illustrates the resulting reflectance anisotropy spectrum from array #3.

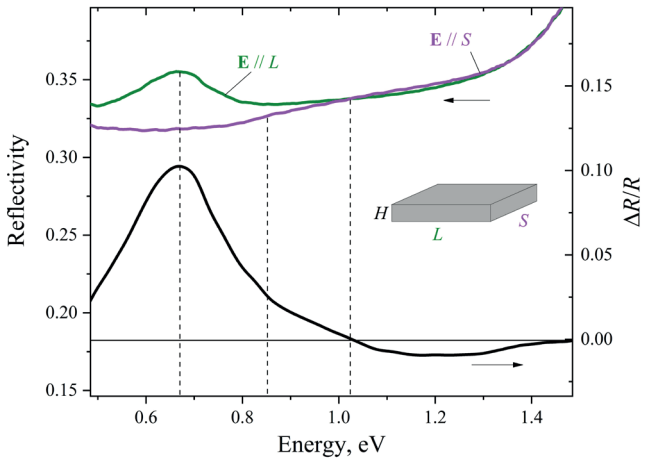

Fig. 2. Polarized reflectance spectrum (top) of nanoantenna array #3 with linear polarization of the probe radiation parallel to the *L* side (green curve) and *S* side (purple curve). Bottom – modulation reflectance anisotropy spectrum of the same structure

Upon comparison of the RA spectra with ordinary reflectance spectra, it becomes evident that, in addition to the prominent peak associated with plasmon resonance along *L* side, a distinct resonance also emerges in the structure along the *S* side. This resonance spans a broad energy range of 0.82–1.4 eV and differs in intensity from the *L*-mode by almost an order of magnitude. The energy range where the S mode appears partially overlaps with the similar range of the *L*-mode, causing the resonance along the *S* side to manifest as an overlap with the intense peak excited along *L* in the reflectance anisotropy spectra. The inflection in the RA spectrum corresponds to the initiation of *S*-modes in its low-energy region. It is worth noting that in the case of the nanoantenna array #3, the resonance excited along the *S* side is relatively discernible in the polarized reflectance spectra. This distinction arises because structure #3 comprises relatively large nanoparticles among the nanoantenna arrays presented in the study and the resonance intensity is proportionate to the volume of such particles [5].

Hence, to ascertain the spectral position of the resonant modes across all nanoantenna arrays, reflectance anisotropy spectra were measured. The resultant curves are depicted in Fig. 3. As the lateral dimensions of nanoantennas increase, the resonant *L*- and *S*-modes are predictably shifted towards lower energies. This shift is anticipated as the effective cross section of the metal plane, wherein plasmonic oscillations occur, expands [3, 5]. It is important to note that in the case of arrays #2 and #1 the *S*-mode in ordinary polarized reflectance spectra becomes visually indistinguishable against the background of the entire spectrum. However, in the reflectance anisotropy spectra, the *S*-mode manifests as an overlay on the intense *L*-mode, evident from the inflections of the corresponding curves in the high-energy regions of the whole RA spectrum. Additionally, it is appropriate to mention that the RA spectra of the GeSiSn/Si heterostructure surface without plasmonic nanoantennas exhibit a zero signal, indicating optical isotropy in the specified energy range.

The photoluminescence spectrum of a heterostructure with multiple $Ge_{0.84}Si_{0.076}Sn_{0.084}$/Si quantum wells, on the surface of which arrays of nanoantennas are formed, exhibits a signal peak near 0.65 eV. Consequently, among the presented arrays, the most pronounced plasmonic enhancement of the radiation from such a heterostructure is expected through the interaction of

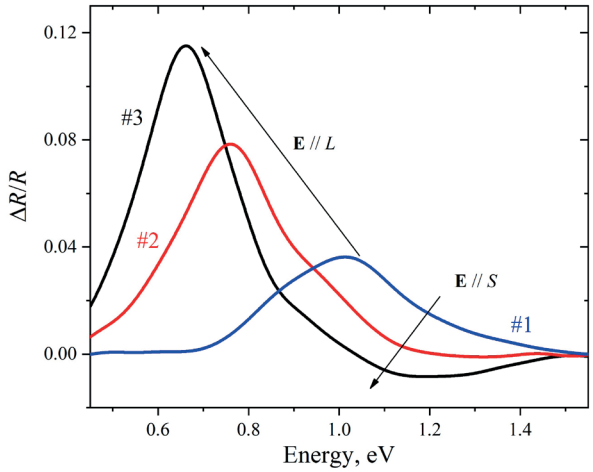

Fig.3. Reflectance anisotropy spectra for each of the nanoantenna arrays. The arrows indicate the dynamics of the resonance energy position with increasing lateral dimensions of the nanoantennas

$Ge_{0.84}Si_{0.076}Sn_{0.084}$/Si with array #3. This expectation arises from the fact that array #3 showcases a maximum of localized surface plasmon resonance at 0.66 eV, while the resonance of structure #1 at 0.65 eV holds a null value.

Conclusion

Thus, we have studied the optical properties of arrays of aluminum nanoantennas formed on a GeSiSn/Si emitting heterostructure using polarized reflectance spectroscopy methods. To control the emission properties of GeSiSn/Si heterostructures, an array of nanoantennas with geometry #3 can be used, since it has an intense resonance at an energy of 0.66 eV, which is located near the photoluminescence peak of the heterostructure ≈ 0.65 eV.

The ability to create nanoparticles with a localized surface plasmon resonance in the near-IR range makes aluminum a suitable material for creating interacting plasmonic and emissive structures in this range.

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