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Editorial

Raman spectroscopy: contributions to the SPQEO journal

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Abstract. This editorial highlights the focus of *Semiconductor Physics, Quantum Electronics & Optoelectronics (SPQEO)* on the achievements of Raman spectroscopy (RS) in addressing complex challenges in modern semiconductor physics. Built upon the foundational principles of the classical Raman effect, the RS has evolved into a powerful diagnostic technique within materials science, quantum electronics, and optoelectronics. Its inherent advantages – including non-destructive probing, remote sensing capabilities, high spatial resolution, and rapid phase and composition identification – have driven its widespread adoption. Furthermore, breakthroughs such as surface-enhanced Raman scattering have pushed the boundaries of sensitivity, enabling single-molecule detection through enhancement factors reaching 10^{14}–10^{15}. This journal has served as a platform for pivotal RS research across a diverse range of materials systems. Key contributions include the analysis of III-IV-VI group glassy alloys, complex mixed crystals such as $(Cu_6PS_5I)_{1-x}(Cu_7PS_6)_x$ and strain-compensated $Ge_{1-x}Sn_xC_y$ semiconductors. The RS has also proven important in monitoring compositional integrity of copper indium gallium selenide sulfide) thin films for flexible solar cell applications. Moreover, the advances in surface passivation control within Sm-doped SiO_x films were covered. The increasingly important role of RS in the area of nanoscale studies is also elucidated in journal publications. These include enhancement of Raman scattering signals using engineered gold nanoislands, and monitoring of plasmon-induced structural transformations in nanoparticles. All the above achievements highlight that RS is a powerful diagnostic tool that bridges fundamental science and practical application, setting a stage for future breakthroughs.

Keywords: Raman spectroscopy, Raman effect, optical diagnostics, SPQEO journal.

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1. Raman spectroscopy as a tool to study the structure and composition of materials

Raman spectroscopy (RS) is an optical technique used to investigate various types of materials: solids, liquids, and gases [1]. RS is based on Raman scattering effect: inelastic light scattering that provides insights into molecular architecture and the microscopic composition of both solids and soft matter [2, 3]. The effect was discovered by G. Landsberg, L. Mandelstam and C.V. Raman; the latter was awarded the Nobel prize in 1930 [4–6]. Since that time RS has evolved from a fundamental laboratory technique into an indispensable non-destructive characterization tool thanks to its advantages, such as non-destructive and contactless probing, high spatial resolution, fast chemical and phase identification, *etc*. [7–9]. The RS has become even more significant field with emergence of advanced techniques such as surface-enhanced Raman scattering (SERS) and tip-enhanced Raman scattering (TERS). One of the most cited papers in this area (Crossref: 9,286) [10] explored application of the surface-enhanced Raman scattering for optical detection and spectral analysis of single molecules and single nanoparticles. The study demonstrated that the Raman enhancement factor reached 10^{14} to 10^{15}. This enormous enhancement produced a significantly stronger Raman signal with superior stability compared to single-molecule fluorescence signals. Raman spectroscopy has also shown its informativeness and efficiency in the study of graphene [11 (Crossref: 6 866), 12 (Crossref: 6 467)]. The authors note that Raman spectroscopy is an integral part of graphene research. It is used to determine the number and orientation of layers, the quality and types of edges, and the effects of perturbations such as electric and magnetic fields, strain, doping, disorder, and functional groups. This, in turn, provides insight into all sp2-bonded carbon allotropes, since graphene is their fundamental building block.

Fig. 1 illustrates the main areas of Raman technologies. Often it is very difficult to distinguish between these areas, which are deeply and intimately connected and influence each other. For example, fundamental science directly links material characterization and applied Raman spectroscopy.

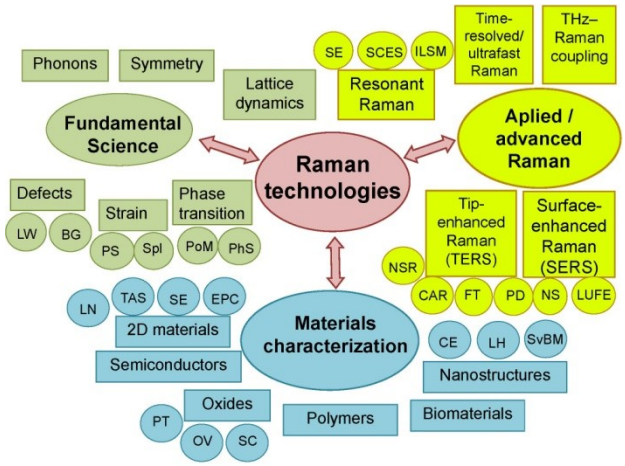

Fig.1. Conventional classification of Raman spectroscopy areas. Here, the following abbreviations are used. *Fundamental science:* LW – linewidths; BG – background; PS – peak shifts; Spl – splitting; PoM – polymorphs; PhS – phase state; *Materials characterization:* LN – layer number; TAS – twist angle & stacking; SE – strain engineering; EPC – electron–phonon coupling; PT – phase transitions; OV – oxygen vacancies; SC – strong correlations; CE – confinement effects; LH – local heating; SvBM – surface *vs* bulk modes; *Applied/advanced Raman:* NSR – nanoscale spatial resolution; CAR – combined AFM + Raman; FT – frontier technique (still TRL 3–4); SE – strong enhancement; SCES – selective coupling to electronic states; ILSM – ideal for low-signal materials; PD – phonon dynamics; NS – nonequilibrium states; LUFE – links to ultrafast electronics.

2. Applications of Raman spectroscopy in materials research

Raman scattering, together with other spectroscopic methods, has been used to study the structure and vibrational properties of glassy III-IV-VI group alloys [13, 14]. In [13], amorphous $Ga_{7.9}Ge_{11.5}Te_{80.6}$ alloys were investigated, in particular, using Raman spectroscopy. The Raman spectra exhibited the main bands at 88, 118 and 138 cm^{-1}. For Te-Te, Ge-Te, and Ga-Te bonds, the stretching vibrational frequencies were similar**.** It was Established that: (i) the band at 88 cm^{-1} in the spectrum is attributed to the oscillation modes of GeTe or trigonal Te; (ii) the main vibrational mode (symmetric breathing or stretching mode) of GaTe4/2 tetrahedra strongly overlaps with the corresponding band of GeTe4/2 tetrahedra due to the similar bond strengths of Ga-Te and Ge-Te bonds and similar atomic masses; (iii) the bending modes of GaTe4/2 and GeTe4/2 tetrahedra are close to 92 cm^{-1}; (iv) the stretching modes of tetrahedral units GaTe4/2 and GeTe4/2 lie in the region of 115 to 125 cm^{-1}; (v) the oscillatory modes of GaTe4/2 tetrahedra are observed at 124…135 cm^{-1}; (vi) the peak at 140 cm^{-1} corresponds to the oscillatory modes of Te-Te. The observed bands in the Raman scattering spectra of the Ga-Ge-Te alloys show that these glasses contain different nanophases. Research on $Ge_{16}Sb_{24}Se_{60}$ glass [14] reveals that the structure of this glass complies with the chemically ordered network model, where Ge-Se and Sb-Se bonds dominate the bonding configuration. When halogen elements are deficient, M-M bonds (where M represents Ge or Sb) emerge within the network structure.

The paper [15] deals with mixed crystals $(Cu_6PS_5I)_{1-x}(Cu_7PS_6)_x$ grown using direct crystallization technique. The following distinctive features of the Raman spectra were identified: (i) for the end-point compounds, relatively weak bands typical only for Cu_6PS_5I (539 cm^{-1}) and Cu_7PS_6 (143 cm^{-1} and 226 cm^{-1}) were observed; (ii) the maxima at 143 cm^{-1} and 226 cm^{-1} were also observed for the mixed $(Cu_6PS_5I)_{1-x}(Cu_7PS_6)_x$ crystals of the Cu_7PS_6-rich compositional interval ($0.84 < x < 1$), which, together with the absence of the high-frequency band at 539 cm^{-1}, clearly correlates with the $P2_13$ structure of the Cu_7PS_6-rich phase; (iii) the band 539 cm^{-1} was observed for the intermediate $(Cu_6PS_5I)_{1-x}(Cu_7PS_6)_x$ compositional range with coexisting $P2_13$ and $F43m$ phases that correlates with the Cu_6PS_5I-type face-centered structure.

In [16], the effect of flash annealing (FLA) in the submillisecond range on the microstructural and chemical composition of copper indium gallium selenide sulfide (CIGSS) films deposited by magnetron sputtering at low temperature (300 °C) on a flexible polyimide substrate covered with a thin Mo film was investigated. Two vibrational bands were observed near 190 and 280…290 cm^{-1}. Addition of sulfur to the crystal lattice of chalcopyrite (CH) leads to a shift of the A1 mode to 195 cm^{-1}. The weaker peak at 280…290 cm^{-1} may be attributed to the Cu_xSe phase or the A1 mode of $Cu(InGa)S_2$. The shoulder near 150 cm^{-1} may be attributed to the Cu-poor ordered vacancy domains of chalcopyrite. One can see that the FLA leads to insignificant increase in the intensity of the A1 mode of Cu(InGa)SSe and a slight shift of its peak position from 195.3 to 194.9 cm^{-1} due to the increase in the Cu/(Ga + In) ratio. The slight increase of the intensity can be associated with a small increase of the CH crystallites size. The contribution of the Cu_xSe phase band becomes more pronounced after FLA treatment. It is shown therefore that annealing with a pulsed lamp leads to a more homogeneous polycrystalline structure with a grain size close to 1300 nm and increased Cu concentration, which results in a decrease in the concentration of defects in the material and ideal tetragonality of the CH lattice.

The paper [17] focuses on study and fabrication of stable, cost-effective $Ge_{1-x}Sn_x$ films doped with carbon (C), while evaluating the effectiveness of non-equilibrium laser annealing technology for their crystallization. Amorphous $Ge_{1-x}Sn_x$ and $Ge_{1-x}Sn_xC_y$ films were deposited by magnetron sputtering of corresponding target materials onto Ge-buffered silicon substrates. The samples underwent rapid annealing using a scanning continuous-wave laser (wavelength λ = 455 nm), with variable energy density (fluence) and were subjected to regular thermal annealing for comparison. Analysis of the Raman spectroscopy results leads to the following conclusions: (i) laser annealing is significantly more efficient than thermal annealing; (ii) $Ge_{1-x}Sn_xC_y$ films with the Sn content up to 4.2 at.% substituted in the Ge

lattice demonstrate grain sizes reaching 30 nm, which is three times larger than the sizes of the grains formed by thermal annealing; (iii) co-introduction of C significantly improves the surface morphology; (iv) minor carbon incorporation into the Ge lattice substantially reduces the amount of tin required for transition from indirect band gap to direct band gap electronic structure; (v) laser annealing allows obtaining $Ge_{1-x}Sn_xC_y$ films with a slightly higher (~0.4 at.%) content of tin atoms in the Ge lattice. Key conclusions of the research: combining magnetron sputtering with scanning laser annealing holds promise for fabricating stress-compensated polycrystalline $Ge_{1-x-y}Sn_xC_y$ films. This technique stabilizes metastable solid solutions and opens new possibilities for obtaining direct-bandgap GeSn materials.

3. Raman-related phenomena

K. Michailovska *et al.* [18] studied the effect of Sm and Ni impurities in the ncs-Si–SiO_x:Sm structures formed by high-temperature air annealing of SiO_x films doped with Sm during thermal co-evaporation in vacuum of silicon monoxide and metallic Sm. The presence of crystalline silicon nanoparticles in SiO_x films doped with Sm and annealed at 970 °C was confirmed by Raman spectroscopy. It was assumed that the mechanism of SiO_x decomposition stimulation during annealing is due to the reaction of Sm atoms with oxygen of the oxide matrix, which leads to a local increase in the concentration of Si–Si bonds and nucleation of Si particles near the impurity atoms. Passivation of the ncs-Si surface due to Si–O–Sm bonds significantly slows down their additional oxidation with atmospheric oxygen during annealing in air.

In [19], substrates with laterally ordered arrays of gold nanoislands fabricated by interference lithography were developed. They demonstrated the efficiency in enhancing the Raman scattering signal of a common analyte dye (crystal violet (CV)) comparable to commercial SERS substrates, with the lowest detected CV concentration of ~10^{-6} to 10^{-7} M. These substrates remained reusable without any performance degradation after thermal treatment at 300 °C. Furthermore, we observed a fundamental difference in the effect of substrate annealing on the Raman signal intensity: annealing performed prior to analyte deposition yields markedly different results compared to subsequent in situ annealing (with deposited molecules).

I.Ya. Yaremchuk *et al.* [20] analyzed optical properties of nanocomposite materials based on plasmon metallic spherical nanoparticles. The authors compared four different effective theories. It was shown that size distribution in nanocomposite materials based on plasmon metallic spherical nanoparticles causes an inhomogeneous broadening and change in the amplitude of the plasmon band. The lowest shift of the resonance characteristics for copper-based nanocomposite was predicted by all the effective theories.

Studies of optical properties of As_2S_3 and Se films deposited on gold nanostructured substrates by thermal spraying showed that the chemical mechanism of SERS is the primary factor contributing to enhancement of the Raman signal from chalcogenide films [21]. Adjusting SERS substrate parameters to tune the position of the substrate plasmonic band for resonance with the excitation laser radiation, the plasmonic amplification effect can be enhanced. Beyond enhancement, localized plasmon resonance within the gold NPs induces localized heating of the surrounding chalcogenide film, triggering local structural transformations.

The effectiveness of SERS substrates based on star-shaped gold nanoparticles was investigated in [22]. The electric field of a radiating dipole is enhanced not only by setting the frequency of the plasmon absorption band at the frequency of the exciting laser radiation, but additionally due to significant contribution to Raman scattering of the so-called “hot spots” that exist at the edges of the stars. Research indicates that these nanostructures effectively enhance Raman scattering signals being excited by lasers at wavelengths of 532 and 632.8 nm, which are close to the peak wavelengths of the plasmon absorption band. By adjusting the inherent geometric parameters of these nanostructures, the peak position of the plasmon absorption band can be tuned to align closely with the laser excitation wavelengths. Analysis of the Raman scattering spectrum of R6G molecules revealed that the Raman signal gain can reach up to four orders of magnitude.

Using micro-Raman spectroscopy, detailed vibrational studies of L-asparagine and L-glutamine amino acids adsorbed onto aluminum foil were conducted under various excitation wavelengths [23]. The vibrational spectrum of the microcrystalline acid exhibits two distinct ranges: (i) the low-frequency region associated with lattice vibrations; and (ii) the mid-to-high-frequency range associated with molecular vibrations. The study demonstrates significant differences in the Raman spectra of the L-asparagine and L-glutamine, despite their similar polarity and charged nature, with their molecular structures differing only at the methyl group of the main chain. Different crystal structures also influence their polarization features, which depends on crystallization of aggregates. It was proved therefore that micro-Raman spectroscopy provides high-quality, reproducible vibrational spectra of solid-state amino acids and can be used for amino acid identification or elucidating the nature of molecular adsorption on metal surfaces.

4. Conclusion

Raman spectroscopy that provides direct access to optical phonons and molecular coupling has become an important method of solid state research. Raman scattering is sensitive to nanoscale changes in phonon symmetry, damping, and coupling strength, allowing correlation of local lattice dynamics. It resolves local strain and symmetry breaking. It is used to determine the number and orientation of layers, the quality and types of edges, and the effects of perturbations such as electric and magnetic fields, strain, doping, disorder, and functional groups. This, in turn, provides insight into all sp2-bonded carbon allotropes, since graphene is their fundamental building block.

SPQEO journal highlights achievements in this area, particularly in the field of materials research, namely: (i) the structure and vibrational properties of glassy alloys of groups III-IV-VI have been studied; (ii) specific features of the Raman spectra for the mixed $(Cu_6PS_5I)_{1-x}(Cu_7PS_6)_x$ crystals have been found; (iii) Raman scattering spectra have been used to monitor changes in the chemical composition of copper-indium-gallium sele-nide sulfide (CIGSS) during flash annealing; (iv) Raman diagnostics of stress-compensated polycrystalline direct-bandgap $Ge_{1-x-y}Sn_xC_y$ materials has revealed new possibilities for obtaining direct-bandgap GeSn materials.

SPQEO also draws attention to phenomena related to Raman scattering: (i) control of ncs-Si surface passivetion through Si–O–Sm bonds during high-temperature air annealing of Sm-doped SiO_x films; (ii) demonstration of the efficiency of Raman signal amplification on substrates with laterally ordered arrays of gold nanoislands fabricated by interference lithography in comparison with commercial SERS substrates; (iii) monitoring localized structural transformations induced by localized plasmon resonance in gold nanoparticles using Raman spectroscopy; (iv) behavior of star-shaped gold nanoparticles for forming efficient SERS substrates; (v) using vibrational spectra of micro-Raman spectroscopy of solid-state amino acids to identify these acids or to elucidate the nature of molecular adsorption on metal surfaces.

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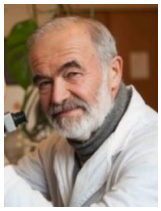

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Rudko G.: verification, formal analysis.
Maksimenko Z.: resources, data curation.
Belyaev A.: supervision,
All the authors: writing – review & editing.

Раманівська спектороскопія: внески до журналу SPQEO

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Анотація. У цій статті проаналізовано висвітлення журналом SPQEO досягнень в сфері раманівської спектроскопії (РС) у вирішенні складних задач сучасної фізики напівпровідників. Основана на класичному ефекті Рамана, РС стала потужним методом діагностики у матеріалознавстві, квантовій електроніці та оптоелектроніці. Притаманні їй переваги, включаючи неруйнівний характер вимірювань, можливості дистанційного зондування, високу просторову роздільну здатність та експресність аналізу хімічного та фазового складу, сприяли її широкому поширенню. Більше того, найновіші досягнення, а саме поверхнево-посилене раманівське розсіювання (SERS), розширили межі чутливості, дозволивши виявляти окремі молекули завдяки коефіцієнтам посилення порядку 10^{14}–10^{15}. В журналі SPQEO опубліковані результати раманівських досліджень широкого спектра напівпровідникових матеріалів і структур. До важливих результатів відносяться аналіз склоподібних сплавів III-IV-VI груп, складних змішаних кристалів, таких як $(Cu_6PS_5I)_{1-x}(Cu_7PS_6)_x$ та напівпровідників $Ge_{1-x}Sn_xC_y$ з компенсацією деформації. РС також виявилася важливим методом моніторингу хімічного складу тонких плівок селеніду сульфіду міді-індію-галію (CIGSS), призначених для створення гнучких сонячних елементів. Подальші досягнення включають контроль пасивації поверхні у плівках SiO_x, легованих Sm. У журнальних публікаціях також відображена зростаюча роль РС в області нанофізичних досліджень. До них належать вивчення посилення сигналів комбінаційного розсіювання за допомогою штучно створених золотих наноострівців та моніторинг структурних перетворень, індукованих плазмонами, у наночастинках. Усі вищезазначені досягнення підкреслюють, що РС є потужним діагностичним інструментом, який є важливим як для фундаментальної науки, так і для практичних застосувань, і закладає базу для подальшого розвитку обох цих напрямків.

Ключові слова: Раманівська спектроскопія, ефект Рамана, оптична діагностика, журнал SPQEO.