Review of the proposal to launch a new research theme
"Modern Trends and Developments in Raman Microspectroscopy and Photoluminescence for Condensed Matter Studies"
and the project
"Ultrasensitive SECARS microspectroscopy and luminescence biolabeling with core-shell nanostructures".

The proposed new research theme is a harmonic and natural continuation of theme No. 04-4-1111-2013/2017 "Multimodal platform for Raman and nonlinear microscopy and microspectroscopy for condensed matter studies", which is successfully coming to an end.

I would like to begin my assessment with a very important point that the Frank Laboratory of Neutron Physics, JINR, has Russia's only and world's unique functioning setup, namely, the picosecond microspectrometer of coherent anti-Stokes Raman scattering, or CARS microscope. Though Raman spectroscopy is a rather new field at JINR, where this research started in 2013, the indisputable advantage of the highly skilled team engaged in this field is their continuous aspiration to keep up with the dominant world trends of research in Raman spectroscopy and microscopy. Since the Raman spectrum is very informative while its intensity is extremely low, various schemes and methods are under development in the world for considerably increasing scattering efficiency of the light that carries information on the structure of the medium and physical and chemical processes occurring in it. Among these methods, SERS (surface enhanced Raman scattering), CARS (coherent anti-Stokes Raman scattering), and TERS (tip enhanced Raman scattering) are of particular interest. These trends are reflected in the titles of the new theme and project on research and development in highly-contrast ultrasensitive Raman microspectroscopy.

The results of three years' activities on mastering quite a complicated CARS method that underlie the design of the setup and developing the SERS method with application to this setup on the initiative of the theme leader have allowed the scientists of the Raman Spectroscopy Sector to come to grips with the problem of combining these two mutually enhancing methods. Microscopy of surface enhanced coherent anti-Stokes Raman scattering (SECARS) by molecules localized on SERS-active surfaces is attractive because the theoretically achievable scattered light intensity gain for SECARS is orders of magnitude higher than for SERS. In addition, being higher than the pump light frequencies, the scattered anti-Stokes light frequency allows, unlike the case of ordinary SERS, radically tuning from overshadowing Stokes luminescence typical of most interesting samples for biological, chemical, and material science research. This holds out hope of considerably extending, due to nonlinear optics, the range of objects detected by optical methods and achieving sensitivity at the level of single-few molecules in the region under investigation. **It is also worth noting that there are only few single works on this topic in the world literature.**

The undoubted merit of the project is also the proposal to create and develop a single complementary optical platform for the biovisualization of objects using Raman microscopy and luminescence methods based on core-shell nanostructures.

Summing up, I note that the project "Ultrasensitive SECARS microspectroscopy and luminescence biolabeling with core-shell nanostructures" proposed within the new theme is of high current importance and scientific significance and deserves support for its implementation at FLNP, JINR, in the coming three years.

V. V. Smirnov
Professor, Head of the Optical Spectroscopy Department
Prokhorov General Physics Institute, Russian Academy of Sciences

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