

Site-selective spectroscopy of electronic states of fullerene derivatives

Razbirin B.S.¹, Sheka E.F.², Nelson D.K.*¹, Starukhin A.N.¹,
Degunov M.Yu.¹, Shukov I.V.¹

¹*Ioffe Institute, 194021, St. Petersburg, Russia*

²*Peoples' Friendship University, 117923, Moscow, Russia*

*e-mail: d.nelson@mail.ioffe.ru

Knowledge of the detailed electronic structure of fullerene derivatives is very essential for understanding their molecular structure, photophysical and chemical properties. Optical spectroscopy is a powerful method of obtaining information about electronic and vibronic states of molecules. However, absorption and luminescence spectra of fullerenes and their derivatives in condensed state are usually inhomogeneously broadened even at low temperatures, which prevents from using all the variety of spectroscopic techniques for their characterization.

Traditionally, one attempts to overcome the inhomogeneous broadening by using crystalline Shpolskii's matrices which provide fine-structured optical spectra. However, in the case of fullerenes and their derivatives, this measure is successful for a very limited number of species [1–3] while a predominant majority of the spectra remain broad. This forces to look for another way to solve the problem. Quite suitable has turned out the site-selective laser spectroscopy, first application of which to fullerene C₆₀ derivatives is presented in the paper.

A structuring of purely electronic transition (0-0) bands was observed under monochromatic excitation by laser beam within the region of vibronic bands. The substitution of the broad 0-0 emission band by a multiplet of narrow lines is explained by a combine action of several reasons among which there are 1) a weak electron-phonon interaction that provides the intense narrow zero-phonon lines accompanied with relatively weak phonon sidebands for all electronic and vibronic transitions; 2) the absence of the electron excitation transfer over different impurity centers; and 3) the electron-vibrational resonance in the excited state. The vibrational analysis of the fine-structured luminescence spectra revealed under the site selective excitation made possible to determine the frequencies of a number of intramolecular vibrations in the excited state of species under study.

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