

Features of stimulated Raman scattering of a laser dye mixture in media with multiple elastic scattering of light

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Introduction

In dye-activated random media, multiple elastic scattering of dye radiation causes its random lasing (RL) under sufficient laser pumping. When the Raman spectrum of the dye overlaps with the continuous RL spectrum, its radiation (i.e. RL) at Stokes frequencies together with laser pumping forms a set of bichromatic pumps (driving forces), the difference in frequencies of which resonates with the corresponding molecular vibrations and induces them (fig.1). The pumping radiation is scattered on this stimulated vibrations and produces stimulated Raman scattering (SRS) at the same Stokes frequencies.

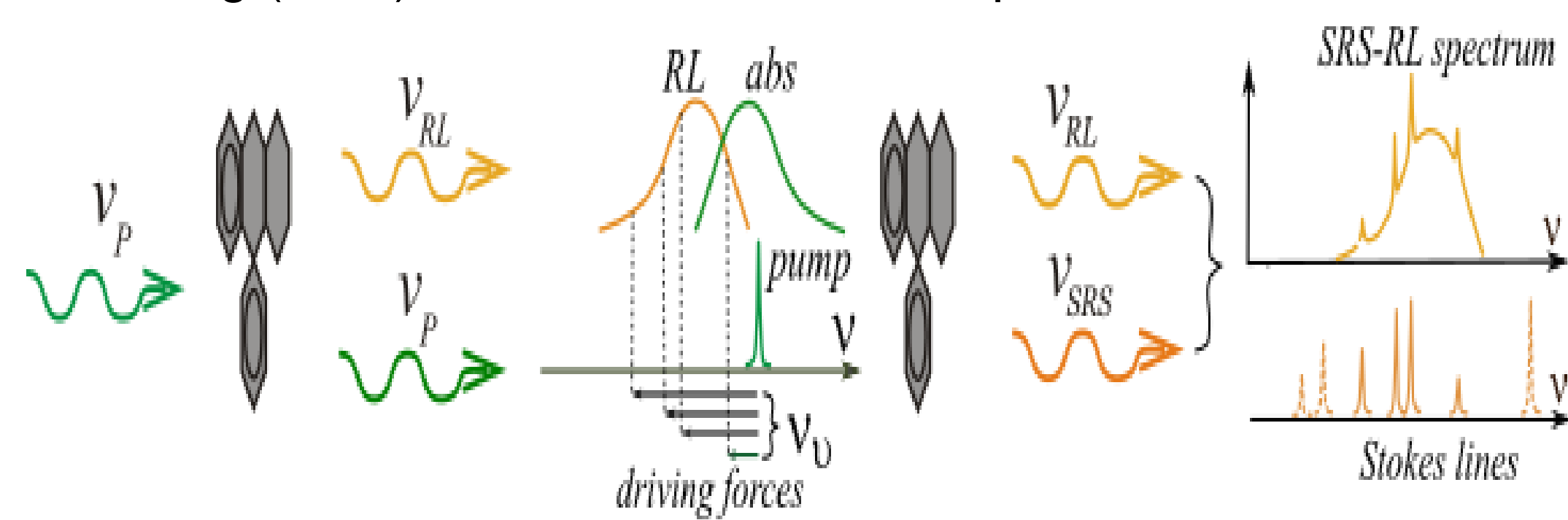


Fig1. Scheme and main steps of SRS-RL arising in a random medium.

Raman amplification operating at these frequencies, enhances the corresponding spectral lines, and all of them become visible in the RL spectrum. It resulting in quasi-line **SRS-RL** spectrum with continuous RL and linear SRS components [1,2].

This situation is similar to active Raman spectroscopy in Stokes region but with internal arising one component (RL) of the bichromatic pumping. The observation of scattered radiation by this method in the Stokes region is impossible due to the interfering luminescence, what forces such measurements to be carried out in the anti-Stokes region (CARS-technique). In the SRS-RL phenomenon, on the contrary, luminescence (through RL) contributes to the appearance and enhancement of Stokes lines what is major advantage of this method.

However this mythology allows recording only that part of the Raman spectrum which overlaps with RL spectrum. Extension of this region requires expanding the overlapping Raman and RL spectra. In the presented report, this problem is solved by using an additional dye with an RL spectrum that complements the spectrum of the dye under study.

Methology & Samples

The main requirement for an additional dye is the overlap of its RL spectrum with the region of Stokes lines of the dye under study. To clearly confirm the above idea, a pair of well-known dyes rhodamine 6G (R6G) and pyromethene 597 (Pm597) were chosen: the first one corresponds to the investigated one, and the second one is complementary. These dyes have very close absorption and luminescence spectra (inset in Fig. 2) and may be well pumped by the SH of YAG:Nd³⁺ laser (532 nm). At a low dye concentrations (below 3 mmol/l) their emission spectra are close to the luminescence maximum and slightly broaden in the long-wave direction (Figs. 2 and 3, bottom) under high pumping.

But under higher concentration (5 mmol/l) and pumping intensity their RL spectra much expand in long-wave direction (fig 3, top and fig 4, top). Thus, choosing a low concentration of the dye under study (R6G) and a high concentration of the additional dye (Pm597), it is possible to significantly expand the RL spectrum of the additional dye at high pumping, without changing the RL spectrum of the dye under study. In this way, it is possible to obtain a significant part of the RL spectrum, which is formed only by the radiation of an additional dye (Pm597).

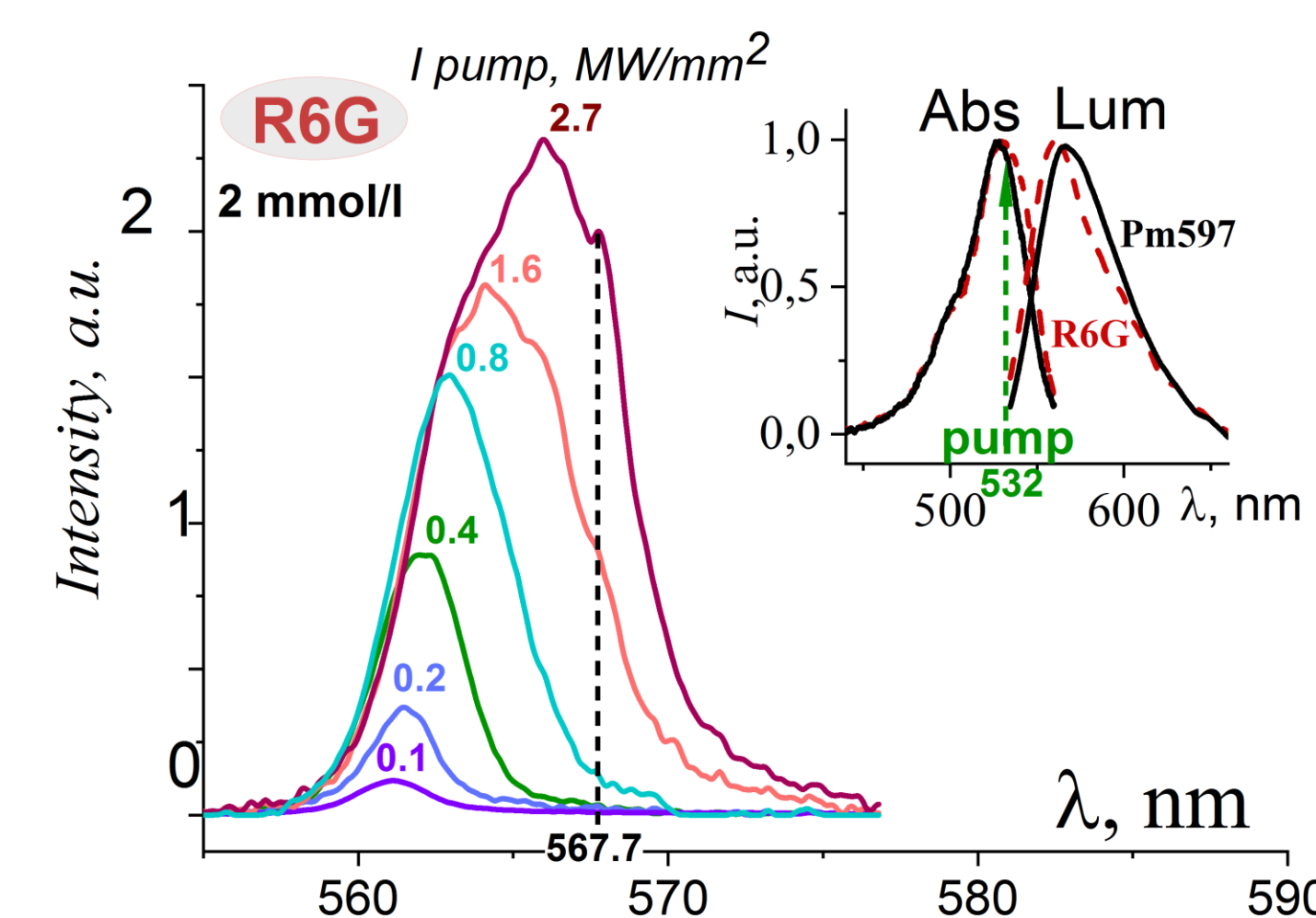


Fig2. Dependence of SRS-RL spectrum of R6G at 2 mmol/l concentration on pumping intensity. On insertion: mutual disposition of luminescence and absorption spectra of R6G and Pm597.

The study of the SRS-RL phenomenon was carried out on vesicular polymer films about 15 μm thick, dyed with a mixture of R6G and Pm597 dyes. The concentration of the studied dye (R6G) was kept constant at 2 mmol/l, and the concentration of the additional dye (Pm597) was changed: 1.25, 2.5, and 5 mmol/l.

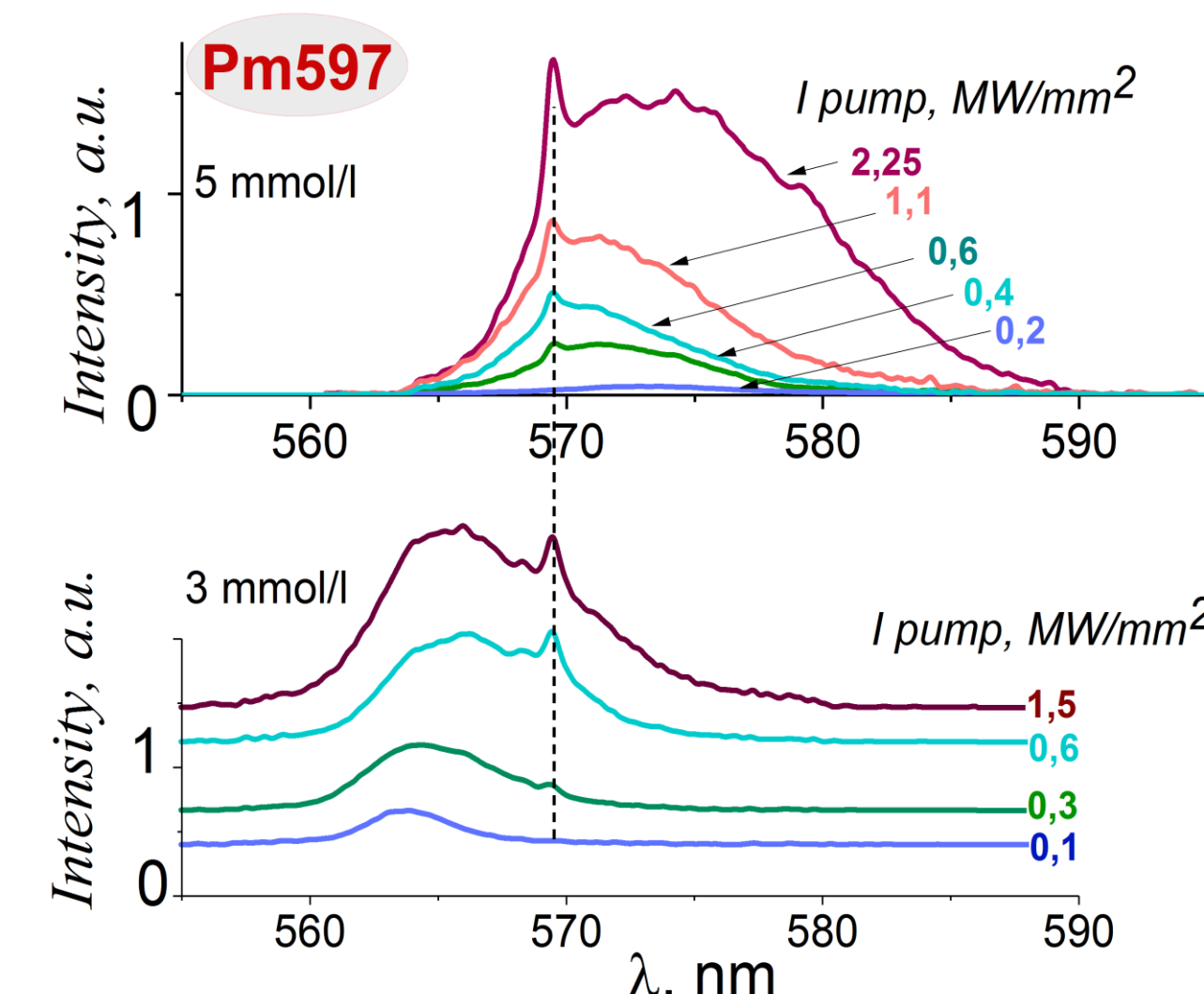


Fig3. Dependence of SRS-RL spectrum of Pm597 at low (2 mmol/l, bottom) and high (5 mmol/l, top) concentrations on pumping intensity.

Multiple light scattering in the films is caused by close packing of micron gaseous vesicles. The radiation spectra were single-shot registered with diffraction spectrograph of 0.2 nm resolution in backward geometry at helium temperature. The samples were pumped by the SH of Q-switched YAG:Nd³⁺ laser (15 ns) with intensity 0.05 - 2.7 MW/mm².

Results and discussion

The main results are presented on the fig.5, where the SRS lines of Pm597/R6G mixture are displayed depending on their concentrations in units of mmol/l (a - c) in comparison with the Stokes lines of SRS spectra of individual dyes (d).

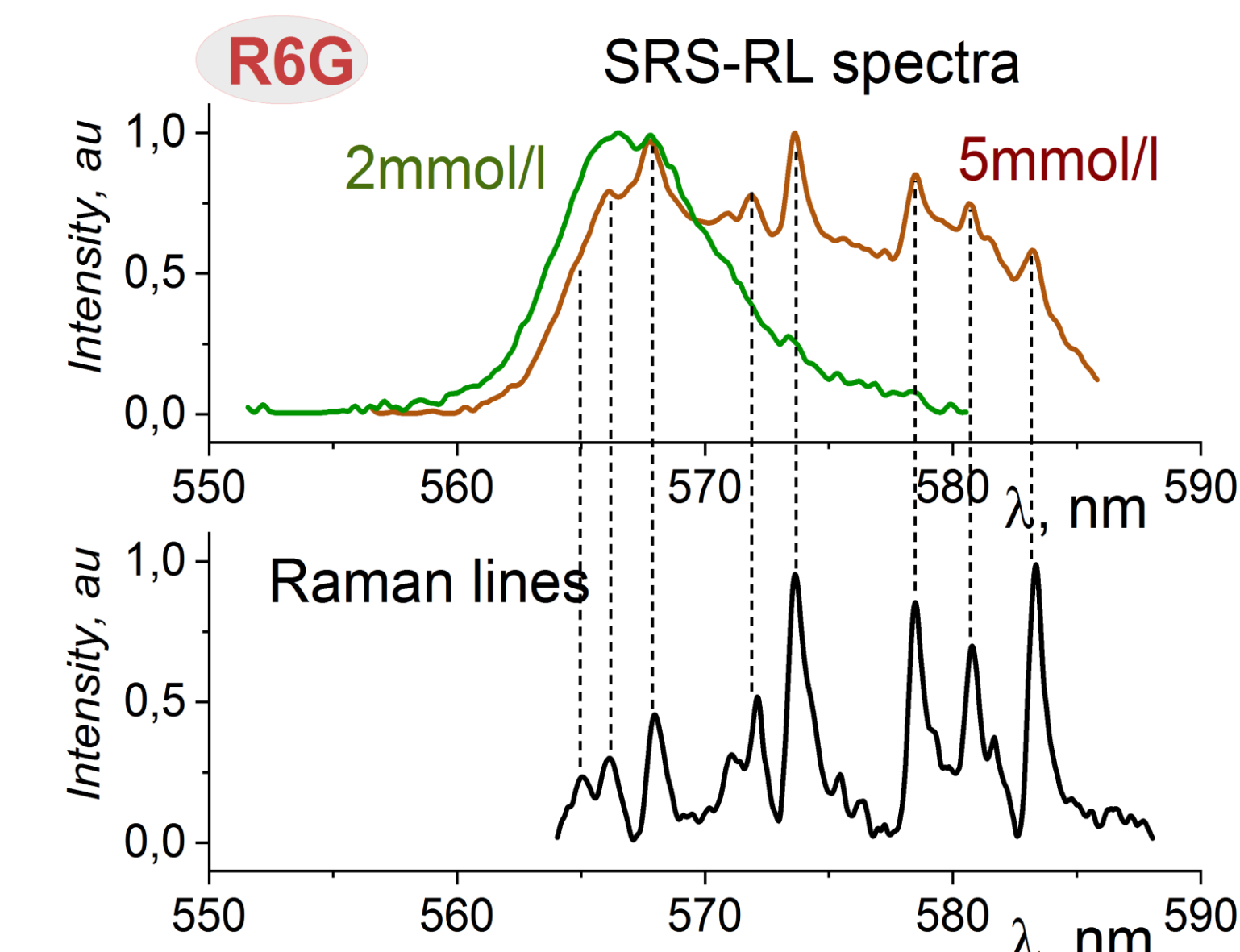


Fig4. SRS-RL spectra of R6G in vesicular film (top) and Stokes Raman lines of R6G appearing under stimulated Raman scattering in a random medium. (bottom).

To interpret these data, one should pay attention to the fact that R6G at the concentration of 2 mmol/l cannot show its Raman lines in SRS-RL of the dye mixture under the influence of its own RL, because its most intense Raman lines are located in the longer-wavelength region (Fig. 4), where its RL radiation is absent. They appear only under highest concentration.

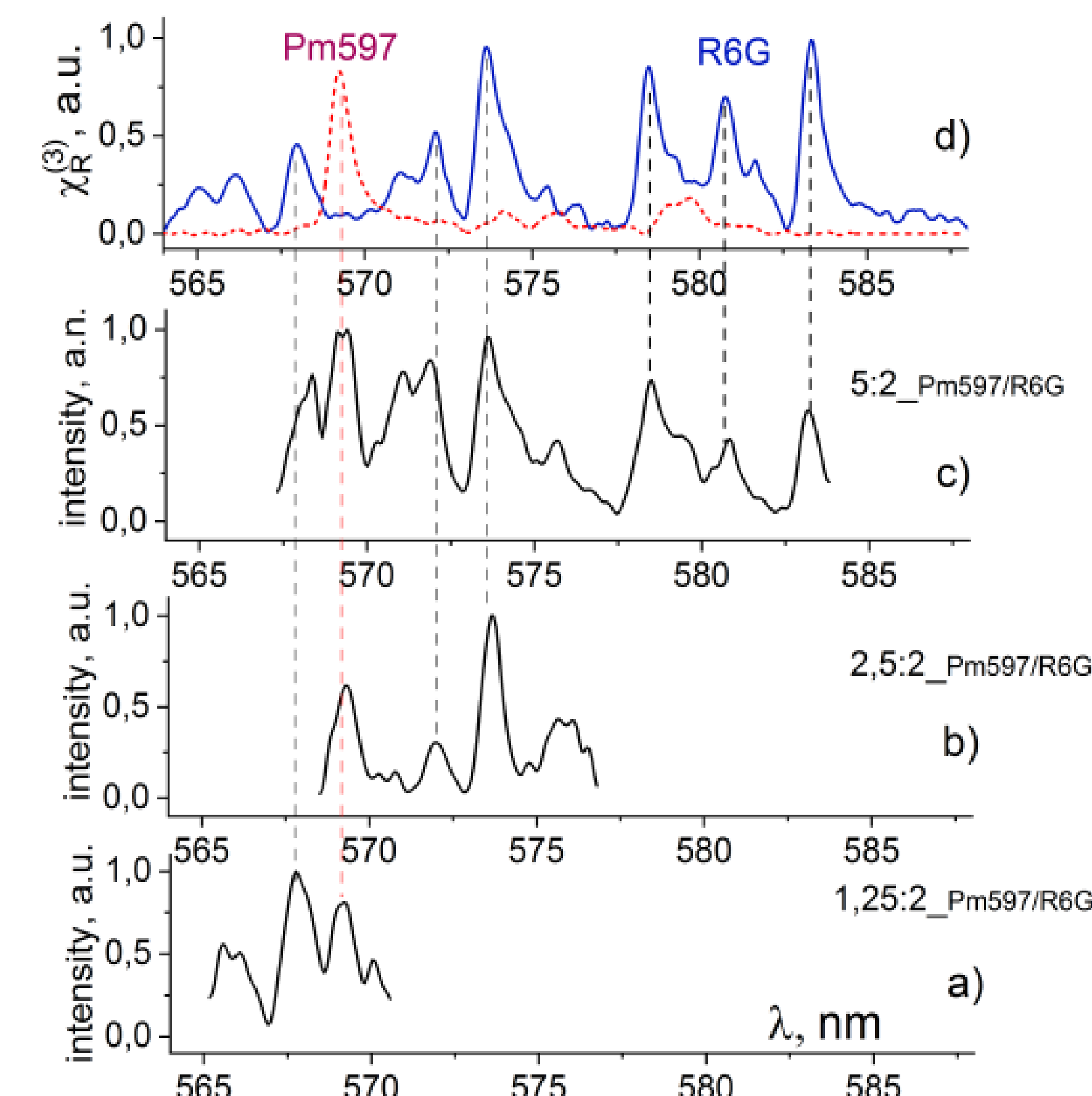


Fig5. Dependence of SRS-RL spectrum of Pm597/R6G mixture on the Pm597 concentrations: 1.25/2 mmol/l (a); 2.5/2 mmol/l (b); 5/2 mmol/l (c). Stokes lines of SRS spectra of individual dyes (d).

But at the same time, Pm597 at 5 mmol/l extends its own RL spectrum (Fig. 3, top) to the location of the R6G Raman scattering lines. This extension increases with concentration growth of Pm597 (Fig. 4) and at 5 mmol/L its RL spectrum covers the entire range of Raman scattering R6G (see fig. 5).

As follows from fig5 there is a correlation between the gradual appearance of Raman lines in the SRS-RL spectrum of the mixtures (fig.5) and the extension of the RL spectrum of Pm597 under its concentration increasing (fig.3). Taking into account the absence of R6G's (at 2 mmol/l) own RL radiation in the long-wave region, this indicates the stimulation of the SRS of rhodamine R6G by the RL radiation of Pm597.

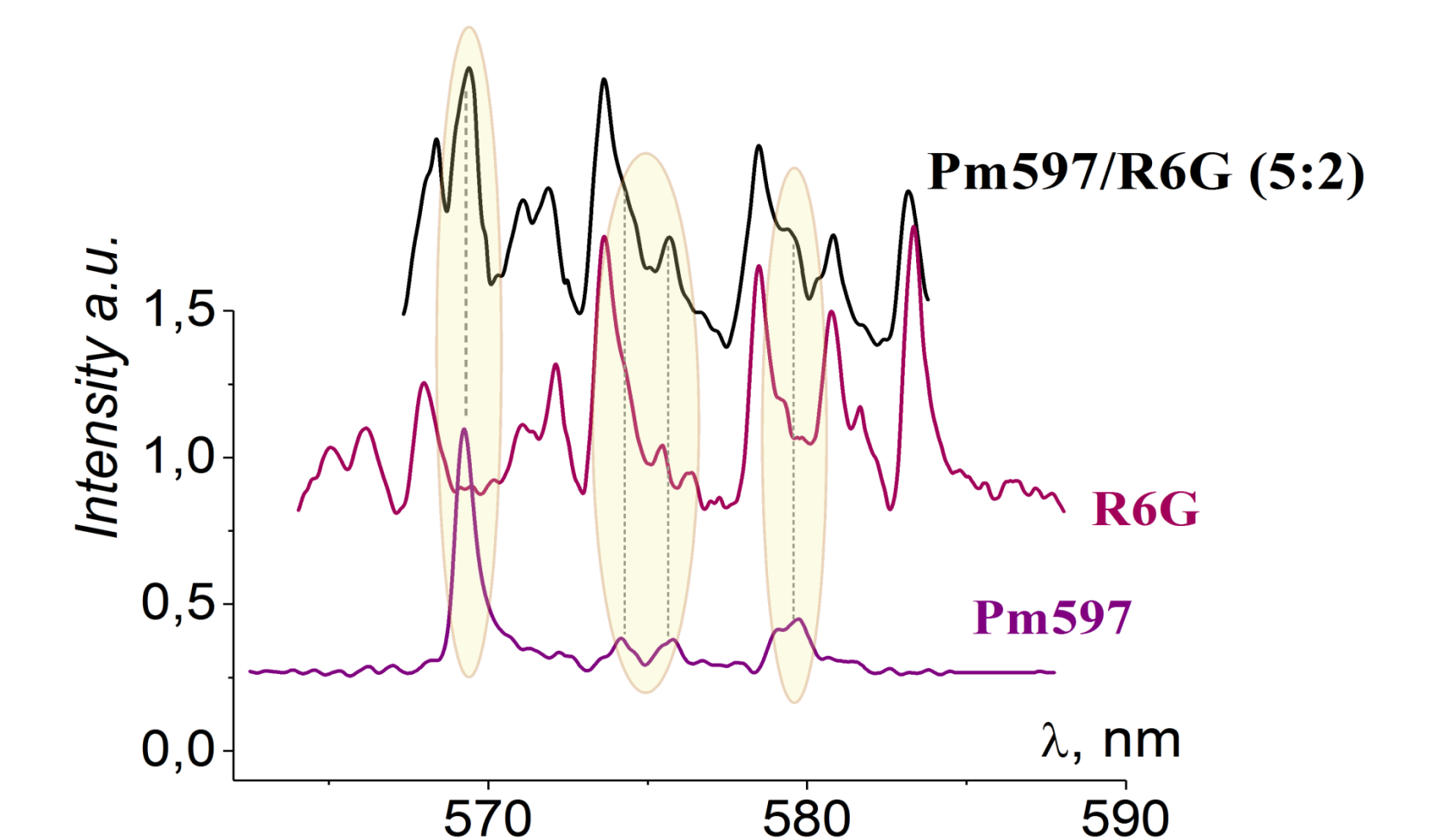


Fig6. SRS lines exhibition of Pm597 of highest concentration in dyes mixture Pm597/R6G.

The RL emission of Pm597 stimulates SRS in the mixture on its own Raman lines also (compare Fig. 3, top, and Fig. 5). The number of these lines increases with the expansion of the RL spectrum under its concentration increases as in the film with sole Pm597, as well as in the film with only Pm597. This is shown more clearly in Fig. 6, where the SRS lines of Pm 597 are highlighted in the SRS-RL spectrum of a more concentrated mixture (5/2 mmol/l). Because these lines are much weaker compared to the R6g lines, the latter show up very clearly, making their identification and separation very easy.

Conclusions

It is shown that the use of an additional dye in a mixture with the studied one allows to significantly expand the registration range of the Raman lines of the latter dye with the help of SRS-RL phenomena.

For this, it is necessary to select such an additional dye and experimental conditions (dye concentration, pumping etc.) that ensure the RL radiation of the additional dye in the spectral region of the Stokes Raman lines of the dye under study.

Bibliography

1. Yashchuk V. P., Tikhonov E. A. and Prigodyuk O. A. JETP Lett., V.91 (2010), No4, p.174.
2. Yashchuk Vasil P. Laser Phys. V. 25 (2015), p. 075702.