Earlier investigations [3] indicated that the formation rate of the B* proton exciplex is probably slower than that of the A* proton exciplex. This fact would explain the sensitivity of B* laser emission to various processes which compete with the population of the B* state.

On the basis of the experimental facts any one of these processes which have been enumerated below can cause a decrease in the B* population density below the laser threshold value:

1) The formation of A* exciplexes quenching the B* laser emission at low HCl-concentration.
2) A quenching process due to Cl^--ions in strongly acidified solutions leading to radiationless deactivation of both exciplex states A* and B* [4].
3) The laser emission from the state N* of the neutral molecule.

We are now carrying out further investigations in order

1) to identify the structure of the proton exciplex states,
2) to explain the mechanism of the photochemical protonation processes,
3) to explain the mechanism of quenching the B* laser emission if the laser threshold of the neutral 4-MU is exceeded.

We are investigating oxygen-alcylated 4-MU derivates in this context and compare the results of measurements of the 4-MU N*-, A*-, B*-laser intensities, as a function of the excitation intensity, with theoretical dependencies obtainable from the numerical solution of the laser rate-equations associated with different proton exciplex formation mechanisms.

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Tunable Stimulated Raman Emission Generated by a Dye Laser

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One well-known method of generating tunable stimulated Raman emission (SRE) makes use of a Raman medium, with a Raman shift varied by an external magnetic field and pumped by a fixed-frequency laser. This method is realized with the spin-flip Raman laser [1].

In this paper we want to discuss a different approach to tunable SRE. The basic idea is to pump an ordinary Raman medium having a fixed Raman shift by a tunable laser. Dye lasers are promising pumping sources because they are capable of generating high powers over a broad tunability range. Powerful flashlamp-pumped dye lasers are made, for instance, from brilliant sulphaflavine (508 – 574 nm) [2], rhodamine 6G (570 – 630 nm) and a mixture of rhodamine 6G and cresyl violet (630 – 710 nm) [3]. They cover a spectral range with a width larger than the Raman shift corresponding to the Q_1 component of the fundamental vibrational transition of hydrogen (4155 cm^{-1}) [4]. As shown by Table 1, the Stokes and anti-Stokes SRE lines up to the 4-th step, generated by these lasers in hydrogen, may together with the dye laser emission cover the entire spectrum from the UV to the far IR. Hydrogen is a favorable medium because of its large Raman shift and because it is to a large extent free from absorption and dispersion in the region of interest.

The feasibility of SRE generation in H_2 by dye laser pumping was investigated in a preliminary experiment. The experimental setup is shown in Figure 1. A 20 mm long dye cell was longitudinally pumped by a passively Q-switched ruby laser. The 400 mm long dye laser cavity consisted of a roof prism and a resonant reflector (LAK 10-flat). From $5 \cdot 10^{-5}$ molar solutions of 3.3′-diethylthiatricarbocyanine bromide (DTTC) in ethanol or in DMSO, output powers up to 85 MW were obtained when pumping pulses of 330 MW peak power and 10 nsec duration were applied. The dye laser emission band was 13 nm wide with its central wavelength at 795 nm (ethanol) or 830 nm (DMSO). For spectral narrowing, interference filters and/or Fabry-Perot-etalons (FPE) were inserted into the cavity. The dye laser beam was focussed by a 200-mm focal length lens into a 300-mm long vessel containing room-temperature hydrogen at a pressure of 200 atmospheres. For power measurements by planar vacuum photocells (ITL), the different SRE beams emerging through an exit window at the oposite end of the vessel were spatially separated by a 3-prism-set. Spectra were recorded on Polaroid film type 47, using a grating spectroscope.

Table 1. Potential tunability ranges.

Wavelength/nm from	to	pumping lasers	
530	679		
679	947	1st	step of Stokes SRE
947	1560	2nd	
1560	4440	3rd	
4440	(∞)	4th	
433	530	1st	step of anti-Stokes SRE
368	433	2nd	
319	368	3rd	
282	319	4th	

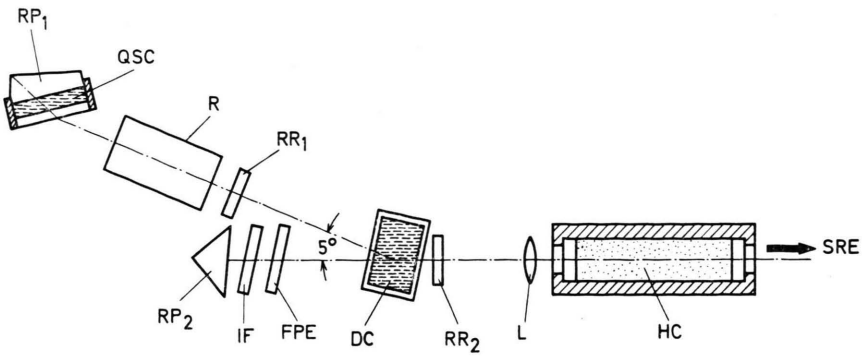

Fig. 1. The experimental setup.
R: ruby-laser head; RP_1: roof prism;
OSC: Q-switch dye cell;
RR: resonant reflector.
DC: dye-laser cell, RP_2: roof prism;
IF: interference filter;
FPE: Fabry-Perot-etalon;
RR: resonant reflector; L: lens;
HC: hydrogen cell.

85 MW of broadband, nonpolarized dye laser power, i. e. a spectral power density of 6.5 MW/nm, were not sufficient to excite SRE. When, however, the bandwidth was reduced to 3 nm using an intracavity interference filter (Fig. 2 b), which reduced the power to 58 MW but increased the spectral power density to about 19 MW/nm, bright 1-st and 2-nd anti-Stokes SRE was observed. The laser was tuned by tilting the interference filter. The tunability range within which the dye laser power exceeded the SRE threshold was found to be 7 nm, centered at 826 nm. Figure 2 c shows a SRE spectrum with the first anti-Stokes line at 612 nm and the second anti-Stokes line at 487 nm.

In further experiments the spectral density of the pumping radiation was increased by employing 1-mm, 0.25-mm, 0.15-mm and 0.05-mm thick FPE's, coated with 60% reflecting dielectric mirrors. This resulted in multiple-line spectra with up to 20 lines, and line widths between 0.2 and 0.01 nm. Combining the interference filter used to obtain Figs. 1 b and 1 d with a 0.05-mm thick FPE resulted in two lines each about 0.2 nm wide. In all cases SRE was observed. Figures 1 e and 1 f show the dye laser spectra obtained with a 0.05-mm FPE and the corresponding anti-Stokes lines. The individual lines of each spectrum are separated by $\Delta\lambda = \lambda^2/2\,n\,t$, where λ is the wavelength of the dye laser or Raman emission and $n\,t = 0.075$ mm is the optical thickness of the FPE. In this case tuning was achieved by changing the solvent.

A certain instability of SRE output was probably caused by variations in the transverse mode structure [5] of the dye laser from shot to shot and by increasing damage in the ruby. Therefore, reliable power measurements were difficult. With dye laser powers between 45 and 60 MW, and with a 0.05-mm FPE inside the dye laser cavity, the 1-st anti-Stokes powers ranged from 0 to 125 kW, the 2-nd anti-Stokes powers from 0 to 2 kW. The first Stokes line was registered as a burn mark on exposed polaroid at a wavelength of 1.2 microns. The 2-nd Stokes line (2.6 microns) was not expected to be observed because we used glass optics.

With the present setup the pump powers to obtain SRE were inconveniently high. Our attempts to drastically reduce the SRE threshold and to increase the tunability range will include the construction of high-power, narrow-band, mode-controlled dye lasers, which are pumped by flashlamps. We will also provide an appropriate resonant cavity for the Raman laser and test some other promising Raman media.

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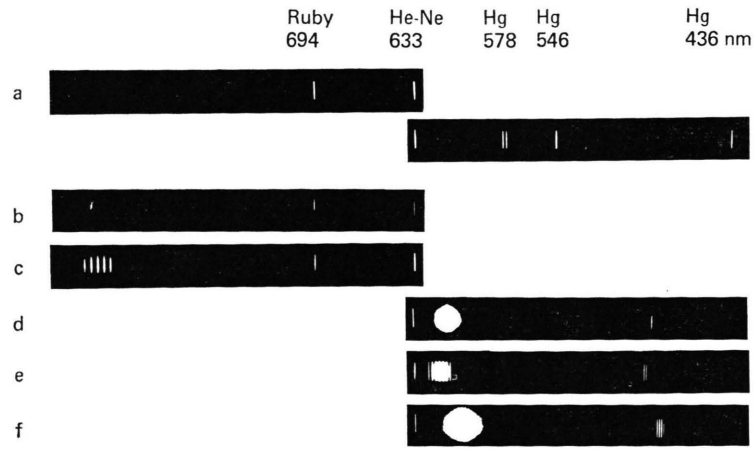

Fig. 2. a) Reference lines; b) dye laser emission at 830 nm, narrowed by an interference filter (FWHM = 11 nm, maximum transmission 95%) inside the cavity; c) multiple-line dye laser spectrum obtained with a 0.05-mm thick quartz FPE with dielectric coatings (60% reflection) inside the cavity; d) 1st and 2nd anti-Stokes lines generated by the dye laser with interference filter inside the cavity; e), f) anti-Stokes lines generated by the multiple-line dye laser. – The DTTC was dissolved in DMSO (a to e) and in ethanol (f). Because of the film sensitivity relative intensities cannot be judged from the spectra. Some of the lines are broadened by strong overexposure.

* *Note added in proof:* After completion of the manuscript, it was brought to our attention that tunable Raman emission was generated, using a neodym laser tuable within a range of 200 cm^{-1} and pyridine as a Raman medium. R. V. AMBARTSUMYAN, V. M. APATIN, and V. S. LETOKHOV, JETP Letters **15**, (6), 237 [1972].

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Inkohärente Lichtstreuung an Ladungsdichteschwankungen schwach ionisierter Plasmen in Magnetfeldern

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Incoherent Scattering of Light from Charge-Density Fluctuations in Weakly Ionized Magnetoplasmas

The spectral density of light scattering from weakly ionized plasmas embedded in a homogeneous magnetic field $\boldsymbol{B}$ is given by the work of WILLIAMS and CHAPPELL [1, 2] and KLINGENBERG [3]. The spectral density has been computed for electron plasmas and is analyzed for the case $\boldsymbol{k} \perp \boldsymbol{B}$ ($\boldsymbol{k}$ is the difference between the wave vectors of the incident wave and the scattered wave) in the regimes $k D \gg 1$ and $k D \ll 1$ (D is the Debye length).

Wird ein Plasma durch eine monochromatische elektrische Welle (Laser) angeregt, so entsteht eine Streustrahlung, deren spektrale Intensität gemessen werden kann.

WILLIAMS und CHAPPELL [1, 2] und KLINGENBERG [3] betrachten ein Dreikomponentenplasma, in dem die Stöße zwischen geladenen und neutralen Teilchen durch einen BGK-Ansatz [4] beschrieben werden. Aus den Zweiteilchen-Korrelationsfunktionen läßt sich die spektrale Dichte $S(\boldsymbol{k}, \omega)$ der Streustrahlung berechnen:

$$S(\boldsymbol{k}, \omega) = -\frac{2(kD)^2}{\omega} \operatorname{Re} \frac{i(1-B_i/F_i)\, B_e/F_e}{1-B_i/F_i-B_e/F_e} \qquad (1)$$

$\omega = \omega_2 - \omega_1$ und $\boldsymbol{k} = \boldsymbol{k}_2 - \boldsymbol{k}_1$ bezeichnen die Differenzen der Frequenzen und Wellenzahlvektoren von einfallender $(\omega_1; \boldsymbol{k}_1)$ und gestreuter $(\omega_2; \boldsymbol{k}_2)$ Welle; D ist die Debye-Länge. Die Größen B_e, F_e bzw. B_i, F_i lassen sich auf einfachere Integrale K_e bzw. K_i zurückführen:

$$B_e = -\frac{1}{(kD)^2}(F_e + i\omega K_e); \qquad (2\,a)$$

$$B_i = -\frac{1}{(kD)^2}(F_i + i\omega K_i) \qquad (2\,b)$$

$$F_e = 1 - \nu_{e0} K_e; \qquad F_i = 1 - \nu_{i0} K_i \qquad (3\,a, b)$$

ν_{e0} bzw ν_{i0} sind die Stoßfrequenzen der Elektronen bzw. Ionen mit den Neutralteilchen.

Die Integrale K_e und K_i sind in einer für numerische Auswertungen zweckmäßigen Form darstellbar (KLINGENBERG [3]):

$$K_e = \lim_{s \to -i\omega} \sum_{n=-\infty}^{\infty} X_{ne} \int_{-\infty}^{-\infty} du \frac{f_e(u)}{s + i k u \cos\Theta + \nu_{e0} + i n \omega_c}, \qquad (4\,a)$$

$$K_i = \lim_{s \to -i\omega} \sum_{n=-\infty}^{\infty} X_{ni} \int_{\infty}^{\infty} du \frac{f_i(u)}{s + i k u \cos\Theta + \nu_{i0} + i n \Omega_c}. \qquad (4\,b)$$

In (4) bezeichnet s eine komplexe Variable; ω_c bzw. Ω_c die Gyrofrequenz der Elektronen bzw. Ionen; Θ den Winkel zwischen dem Wellenzahlvektor $\boldsymbol{k}$ und der Richtung des äußeren Magnetfeldes $\boldsymbol{B}$; f_e bzw. f_i die eindimensionale Maxwell-Geschwindigkeitsverteilungsfunktion der Elektronen bzw. Ionen. In den Größen X_{ne} bzw. X_{ni} treten modifizierte Bessel-Funktionen $I_n(\lambda)$ erster Art der Ordnung n auf:

$$X_{ne} = \exp\left\{-\frac{k^2 v_e^2 \sin^2\Theta}{\omega_c^2}\right\} I_n\left(\frac{k^2 v_e^2 \sin^2\Theta}{\omega_c^2}\right); \qquad (5\,a)$$

$$X_{ni} = \exp\left\{-\frac{k^2 v_i^2 \sin^2\Theta}{\Omega_c^2}\right\} I_n\left(\frac{k^2 v_i^2 \sin^2\Theta}{\Omega_c^2}\right). \qquad (5\,b)$$

v_e bzw. v_i ist die thermische Geschwindigkeit eines Elektrons bzw. Ions.

Im Grenzfall verschwindender Stoßfrequenzen ν_{e0}, ν_{i0} ergibt sich mit (2) bis (5) aus (1) das Ergebnis von SALPETER [5] [Gl. (17)].

Im folgenden diskutieren wir nur jenen Teil des Streuspektrums, für den allein die Elektronenbewegung wesentlich ist, d. h. $\omega \gg k v_i$ und $\omega \gg \Omega_c$. Dann gilt $B_i/F_i = 0$ und (1) vereinfacht sich zu

$$S(\boldsymbol{k}, \omega) = \frac{2(kD)^2}{\omega} \frac{\varepsilon_i(\boldsymbol{k}, \omega)}{\varepsilon_r^2(\boldsymbol{k}, \omega) + \varepsilon_i^2(\boldsymbol{k}, \omega)}, \qquad (6)$$

wobei für die Dielektrizitätskonstante ε_L (Index L kennzeichnet, daß nur Raumladungsschwankungen berücksichtigt wurden) gilt

$$\varepsilon_L = \varepsilon_r + i\,\varepsilon_i = 1 - (B_e/F_e). \qquad (7)$$

Die Maxima der Spektrallinien liegen für $|\varepsilon_i| \ll |\varepsilon_r|$ bei Frequenzen $\omega = \omega_M$, die sich als Lösungen der Dispersionsgleichung $\varepsilon_r(\boldsymbol{k}, \omega) = 0$ ergeben.

In der Umgebung von $\omega = \omega_M$ liefert (6) für jeden Wert von $\boldsymbol{k}$ ein Lorentz-Profil

$$S(\boldsymbol{k}, \omega) = \frac{2(kD)^2}{\omega_M \varepsilon_r'|_\omega} \frac{A(\boldsymbol{k}, \omega_M)}{(\omega - \omega_M)^2 + A^2(\boldsymbol{k}, \omega_M)} \qquad (8)$$