sentation become

$$\begin{aligned} d\gamma_a/dt &= [2(1+t^2)]^{-1}\gamma_b \\ &\quad \times \exp\{-i(\beta/2)\int_{-\epsilon}^{t}[(1+t^2)/(\epsilon+t)]^{1/2}dt\}, \\ d\gamma_b/dt &= -[2(1+t^2)]^{-1}\gamma_a \\ &\quad \times \exp\{+i(\beta/2)\int_{-\epsilon}^{t}[(1+t^2)/(\epsilon+t)]^{1/2}dt\}. \end{aligned} \tag{6}$$

This representation is less convenient for analytical work because of the complicated integral in the exponent, but it is quite convenient for numerical work for strong β and small ϵ.

We wish to emphasize the necessity of abandoning the linear approximation for $t(s)$ if a valid model for close crossings is to result. This is particularly important in view of the preoccupation with Weber's equation and its asymptotic properties which has dominated past studies of this problem.[2,6-8] In Ref. 1 we show that the reduction to Weber's equation depends upon assuming the effective linearity of $t(s)$. Equations (6) also show quite clearly why this is not a good idea. If $t(s)$ is taken to be linear, Eqs. (6) are modified to a form which is the same except that the integrand in the exponent is replaced by $T_0(1+t^2)^{1/2}$, with $T_0=\beta/2\sqrt{\epsilon}$. It is evident that if significant coupling occurs near the turning point, the linear approximation is inadequate.

We have computed solutions for a complete grid of parameters (β, ϵ). In Ref. 1 these results are presented in detail and compared with various analytical formulas.

We thank the National Research Council of Canada for support of this work.

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Linewidth of Tunable Stimulated Raman Scattering

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(Received 17 November 1971)

We report direct measurements of the tunable spin-flip Raman (SFR) laser linewidth, obtained by heterodyning the SFR laser output with the output from a cw gas laser. The measured linewidth for the cw SFR laser operating near 5.3 μm is < 1 kHz and is limited by the spectrum analyzer resolution. The measured linewidth of < 1 kHz is the narrowest known for any tunable source of coherent radiation in the infrared.

A recent report of tunable stimulated Raman scattering[1] from the spin flip of conduction electrons in InSb (i.e., the spin-flip Raman laser) has evoked considerable interest in its use in the investigation of numerous physical phenomena. The tunability of the spin-flip Raman (SFR) laser now covers[1-6] a wavelength range from about 9 to 14.6 μm and from 5.2 to 6.2 μm and has been used in high-resolution infrared spectroscopy,[7] transient infrared spectroscopy,[8] pollution detection,[9] and nonlinear optics.[10] A very important parameter of a tunable laser source is the linewidth of its radiation output. Heretofore, only indirect measurement of the linewidth of the SFR laser was available with a quoted number[7] of ~900 MHz. In this paper we report direct measurements of the SFR laser linewidth by heterodyning the SFR laser output with power output from a cw gas laser. We *measure* a linewidth of $\lesssim$ *1 kHz* for the spin-flip Raman laser pumped at 5.3 μm. The linewidth is limited by spectrum analyzer resolution. The SFR laser linewidth calculated from consideration of the amplitude and phase fluctuations in the laser is ~1 Hz. The contribution of the SFR laser linewidth arising from the fluctuations in the magnetic field which determines the SFR laser frequency is judged to be small compared to 1 kHz. The SFR laser frequency ω_s is given by

$$\omega_s = \omega_0 - g\mu_B B, \qquad (1)$$

where ω_0 is the pump frequency, g is the effective g value of the electrons in InSb, μ_B is the Bohr magneton, and B is the magnetic field. Even with the present measured linewidth, the SFR laser is the narrowest-linewidth tunable source of coherent radiation in the infrared.

The linewidth of a laser source is determined by the amplitude and the phase fluctuations in the radiation phenomenon which leads to the stimulated emission. The laser linewidth is given by the Schawlow-Townes expression[11]

$$\Delta\nu_L = 8\pi h\nu_L[\Delta\nu_c \Delta\nu_{sp}/(\Delta\nu_c + \Delta\nu_{sp})]^2 P^{-1}, \qquad (2)$$

where $\Delta\nu_L$ is the laser linewidth far above laser threshold, h is Planck's constant, ν_L is the laser frequency, P is the laser power output, $\Delta\nu_c$ is laser cavity linewidth,[12] and $\Delta\nu_{sp}$ is the spontaneous emission linewidth for the radiation process. For typical operating conditions of the SFR laser in the 5–15 μm range, Eq. (2) predicts a laser linewidth of ~1–100 Hz. (The exact numbers applicable to the present experimental conditions will be given later.) To measure such narrow linewidths, conventional spectrometers are clearly unsuitable. Earlier attempts to estimate the SFR laser linewidth by monitoring a molecular absorption[7] line led to an upper limit of ~900 MHz. It was not appreciated at that time that to measure the extremely narrow linewidth a very precise control on the magnetic field is necessary if the magnetic field is being swept in monitoring the molecular absorption. This follows from the enormously large tuning rate for the SFR lasers arising from the effective g value of -50 for electrons in InSb.[13] Such a large g value gives an experimentally observed tuning rate[1-6] of ~70 MHz/G. For molecular absorption lines in the infrared, typical Doppler widths are of the order of ~70–100 MHz, which implies that a proper estimate of the SFR laser linewidth by measuring molecular absorption will require a precision control of the magnetic field of the order of a small fraction of a gauss. SFR lasers heretofore were limited to a minimum magnetic field of ~14 kG for their operation because of the quantum-limit considerations.[2] Such fields require the use of superconducting solenoids where control of magnetic field to a fraction of a gauss at $B \gtrsim 10$ kG is difficult if not impossible. The ideal way of measuring a laser linewidth is by heterodyning its power output with another laser whose linewidth is either known or is expected to be much narrower than the linewidth of the laser to be measured. Here again, to bring the beat frequency within measurement range of present infrared detectors requires the same precise control of the magnetic field mentioned above. In addition, the field is also required to remain constant. Because of these reasons, no really good measurements of SFR laser linewidth were available to date.

The situation has changed considerably with the operation of the SFR laser at magnetic fields as low as 400 G,[14] where a conventional electromagnet with its inherent stability and precision control of the field can now be utilized to measure the linewidth of the SFR laser. We have directly measured the SFR laser linewidth by heterodyning the low-field SFR laser operating at $B \approx 1713$ G with a cw CO laser line. The experimental setup used is shown in Fig. 1. The cw CO laser with a grating inside the cavity is made to simultaneously oscillate on two transitions of CO: $P_{9-8}(12)$ at 1888.32 cm^{-1} and $P_{9-8}(13)$ at 1884.35 cm^{-1}. The grating is adjusted such that the power output on the $P_{9-8}(12)$ transition is about 10 times that on the $P_{9-8}(13)$ transition. The entire power output of ~6.0 W was focused with a 10-cm BaF$_2$ lens onto an InSb sample having an electron concentration of 1×10^{15} cm^{-3}. The sample, 2 mm×2 mm×9.24 mm long was immersed in liquid He at 1.4–1.6 K. The CO laser polarization was parallel to the magnetic field. The 2 mm×2 mm faces, which are parallel to the magnetic field, serve to form the SFR laser cavity. The CO pump radiation enters the InSb sample through one of the 2 mm×2 mm faces as shown in Fig. 1. The $P_{9-8}(12)$ transition of the CO laser acts as the pump for the SFR laser. The magnetic field is supplied by either a small permanent

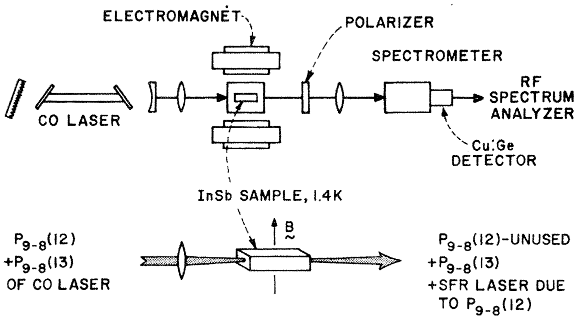

FIG. 1. Experimental setup for measuring the SFR laser linewidth by a heterodyne technique.

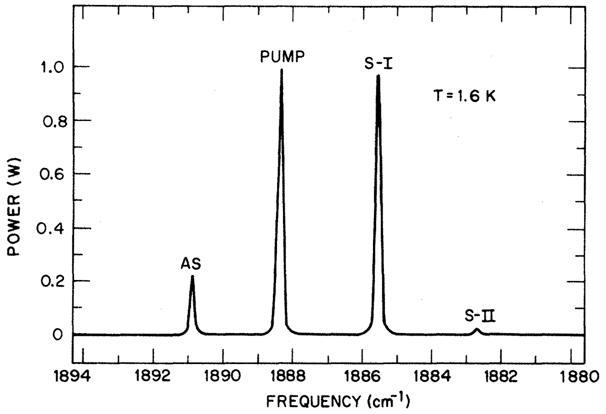

FIG. 2. Low-field output spectrum of the SFR laser at $B\approx 1141$ G. The magnetic field is provided by a permanent magnet. The input pump power on the $P_{9-8}(12)$ transition is ~ 2.5 W. Notice that for the purposes of this analysis the CO laser was oscillating only on the $P_{9-8}(12)$ transition.

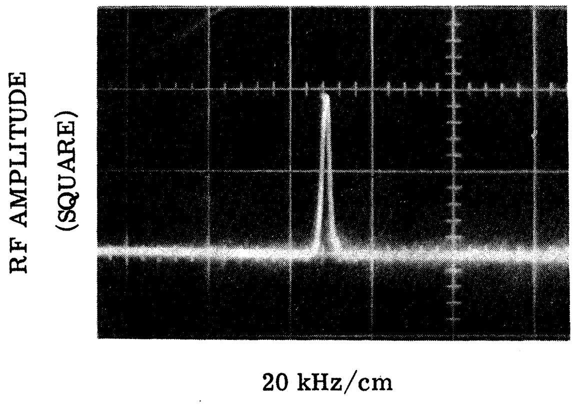

FIG. 3. Spectrum analyzer display of the rf beat between the $P_{9-8}(13)$ transition of the CO laser and the SFR laser pumped with the $P_{9-8}(12)$ transition of the CO laser at $B\approx 1713$ G. Beat frequency is 82 MHz. Sweep width is 20 kHz/cm with a resolution of 1.0 kHz. Sweep rate is 0.005 sec/cm. Exposure time is 0.1 sec.

magnet or a 4-in. Varian electromagnet. Sizable Raman conversion efficiencies are obtained even at these low magnetic fields as seen in Fig. 2. Here a fixed magnetic field of ~ 1141 G was provided by a permanent magnet, and the CO laser was made to oscillate only on the $P_{9-8}(12)$ transition.

For the heterodyne experiment, the magnetic field, now provided by the electromagnet, is adjusted such that the I Stokes SFR laser frequency nearly coincides with the $P_{9-8}(13)$ transition of the CO laser emerging simultaneously from the InSb sample. The SFR laser polarization is normal to the magnetic field[14,15] and normal to the polarization of the $P_{9-8}(13)$ transition. To obtain heterodyne signals, a polarizer at 45° is inserted in the beam to give parallel components of electric field polarizations for the SFR laser output and $P_{9-8}(13)$ transition. The low-resolution spectrometer precedes a fast Cu:Ge photoconductive detector (4.2 K) in order to remove all power output but that at wavelengths near the $P_{9-8}(13)$ transition. The SFR laser power output is ~ 1 W and the $P_{9-8}(13)$ CO transition power is ~ 200 mW. The output from the photodetector is analyzed for its rf content by using a Tektronix 1L20 spectrum analyzer.

Figure 3 shows the best frequency near ~ 82 MHz obtained between the $P_{9-8}(13)$ transition of the CO laser and the SFR laser at $B\approx 1713$ G. The rf sweep rate of 20 kHz/cm with a 1.0-kHz resolution gives us an effective beat frequency width of *~1 kHz*. From this we estimate a *SFR laser linewidth of* <1 *kHz* which is limited by the spectrum analyzer resolution. It was seen that there was a considerable jitter in the position of the beat frequency from one spectrum analyzer sweep to the next. The jitter was ~ 40 kHz over a time range of few seconds. This jitter is ascribed to the magnetic field fluctuations. However, from sweep to sweep, the width of the beat note remained at $\lesssim 1$ kHz.

For our laser cavity length of 9.24 mm, $\Delta\nu_c$ can be calculated[12] to be ~ 0.04 cm^{-1}. Inserting the $\Delta\nu_c$, measured ν_L, $\Delta\nu_{\mathrm{sp}}\approx 1$ cm^{-1}, and power output (~ 1 W) into Eq. (2), we obtain $\Delta\nu_L \lesssim 1$ Hz for the SFR laser. Thus the measured linewidth of <1 kHz is considerably larger than the calculated value and improved rf beat-frequency measurements are warranted. From the known current stability of the magnet power supply and the magnet parameters, we believe that the contribution of the fluctuation in the magnetic field to the SFR laser frequency is <1 kHz. However, it is felt that in order to improve upon the present measurements for obtaining a better comparison between calculated and measured SFR laser linewidth, we will have to use a permanent magnet of appropriate field strength. This is being planned.

It should be pointed out that because we are using the $P_{9-8}(12)$ and the $P_{9-8}(13)$ CO transitions from the same laser, the variation in the pump frequency arising from mechanical vibrations etc. of the CO laser make little contribution to the measured width of the beat frequency between the $P_{9-8}(13)$ transition and the SFR laser. Thus

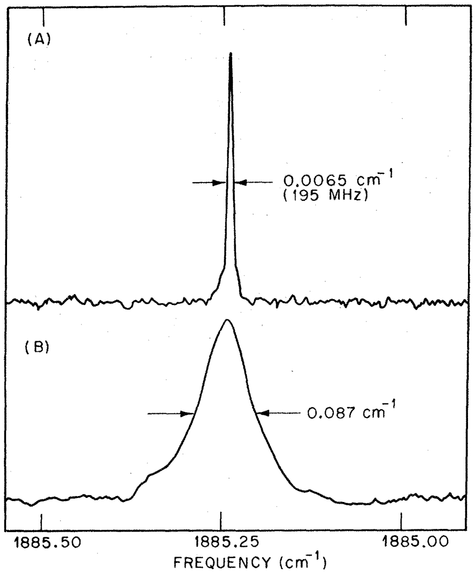

FIG. 4. Absorption spectrum of the 1885.24-cm^{-1} line of H_2O vapor in air at (a) $P_{\mathrm{air}}=30$ Torr, (b) $P_{\mathrm{air}}=760$ Torr.

the measured SFR laser linewidth does not include a contribution from the pump frequency fluctuations. In a practical situation where absolute frequency stability is required, the CO pump laser frequency will need to be well stabilized.

As a further check on the usefulness of the low-field SFR laser in spectroscopy, we show in Fig. 4 the absorption spectrum of the H_2O vapor line[16] at ~1885.24 cm^{-1} measured with the low-field SFR laser spectrometer at $P_{\mathrm{air}}=30$ and 760 Torr. At $P_{\mathrm{air}}=30$ Torr, a linewidth of 0.0065 cm^{-1} (195 MHz) is measured, all of which may be accounted for by the pressure broadening and the Doppler width. At 760 Torr, we see an increase in the linewidth to 0.087 cm^{-1} due to pressure broadening. Incidentally, the measured pressure broadening of this H_2O line is in reasonable agreement with the calculations[16] which yield a linewidth of 0.109 cm^{-1}.

In conclusion, we have directly measured the linewidth of a cw SFR laser by heterodyning its output with a cw gas-laser output. The measurements give an upper limit of 1 kHz for the SFR laser linewidth. This linewidth is determined not by phase fluctuations due to spontaneous emission but by the rf spectrum analyzer resolution. The present measured linewidth is the narrowest linewidth of any source of tunable coherent radiation,[17] and lays to rest any doubts[18] about the true laser nature of the SFR lasers.

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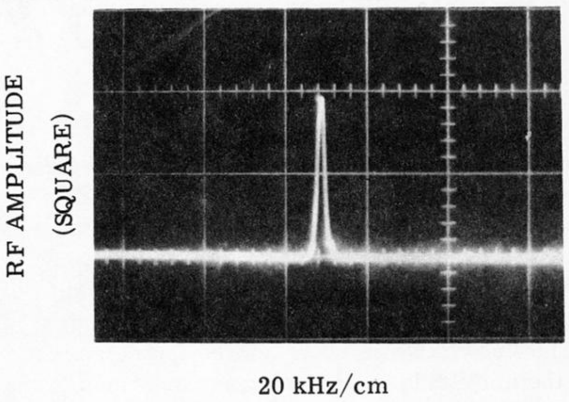

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