

ly would not be expected to result in preferential production of low-energy bremsstrahlung photons. The appearance of the low-energy bremsstrahlung structure in the region of alignment of the beam with the periodic structure of the crystal suggests the possibility of coherent bremsstrahlung effects. Such an enhancement of the low-energy part of the photon spectrum was predicted by Dyson and Überall,⁵ and discussed in more detail by Überall.⁶ A relativistic electron (or positron) of energy E interacts coherently with a low-energy bremsstrahlung photon of wavelength λ over a path length $L \approx \lambda(E/m_0c^2)^2$, and for an aligned particle the photon intensity is expected to be enhanced by a factor N , equal to the number of crystal atoms contained in the coherence length L . If d is the lattice spacing, then $N = L/d$. Since matrix elements for bremsstrahlung production are small for impact parameters larger than the Thomas-Fermi screening radius a , these enhancement effects should occur for $\theta < a/L$. Accordingly, for incident particles in the energy range of our experiment, N becomes greater than unity for photon energies less than a few MeV, and enhancement should occur for $\theta < 30$ mrad. The effect should be about twice as strong for 28-MeV particles as for the 20-MeV case, and should occur in roughly half the angular range. Our low-energy photon observations are quite consistent with these predictions. These coherence effects occur in addition to any channeling effects, and it should

be noted that the above predictions are for particle trajectories not influenced by the channeling mechanism.

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¹For a general review see various authors, *Can. J. Phys.* **46**, 503-612 (1968).

²J. Lindhard, *Kgl. Dan. Vidensk. Selsk., Mat.-Fys. Medd.* **34**, No. 14 (1966).

³J. U. Anderson, private communication, and p. 547 of Ref. 1.

⁴T. F. Godlove and M. E. Toms, private communication. We thank Dr. Godlove for communicating his results to us prior to publication.

⁵F. J. Dyson and H. Überall, *Phys. Rev.* **99**, 604 (1955).

⁶H. Überall, *Phys. Rev.* **103**, 1055 (1956).

TUNABLE SPIN-FLIP LASER AND INFRARED SPECTROSCOPY

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We investigate the use of a tunable spin-flip Raman laser in infrared absorption spectroscopy. The Raman laser is tunable from 10.9 to 13.0 μm with linearity and resettability of tuning better than $\sim 4 \text{ \AA}$ at $\sim 12.0 \mu\text{m}$. This limit is set by the present measurement capability. A study of the absorption spectrum of NH_3 in the 12.0- μm region shows that the spin-flip Raman laser is superior to a conventional grating spectrometer in attainable resolution.

The recent report of a tunable spin-flip Raman laser¹ in InSb is of considerable significance for infrared spectroscopy. The spin-flip Raman laser in InSb is continuously tunable from 10.9 to 13.0 μm ² (when pumped at 10.6 μm) by varying the dc magnetic field. If sufficiently narrow and stable, such a source might be quite important in infrared spectroscopy. In this paper we re-

port measurements of linearity and resettability of the spin-flip Raman-laser frequency which were made in order to evaluate its suitability for this purpose. At present, linearity and resettability are better than $1:3 \times 10^4$ corresponding to $\sim 4 \text{ \AA}$ at $\sim 12.0 \mu\text{m}$, i.e., $\sim 800 \text{ MHz}$ at 830 cm^{-1} , and are limited by our measurement capability. The emission linewidth is estimated to be ≤ 0.03

cm^{-1} , which is also limited by the measurement techniques. The linewidth, tuning characteristics, and high power output indicate that the spin-flip Raman laser is an excellent source of tunable coherent radiation and should be ideal for infrared-absorption spectroscopy within its spectral range. An absorption spectrum of NH_3 obtained with the spin-flip Raman laser shows that the spin-flip Raman laser is superior to the conventional grating spectrometers in infrared spectroscopy in terms of ultimate resolution and speed.

In Ref. 1 we reported the tunability of the spin-flip Raman laser in a $n_e = 3 \times 10^{16} \text{-cm}^{-3}$ InSb sample to be from ~ 11.7 to $13.0 \mu\text{m}$ when pumped at $10.6 \mu\text{m}$. With the use of the low-concentration samples,² the overall tuning range of the spin-flip Raman laser has been extended from ~ 10.9 to $\sim 13.0 \mu\text{m}$, as seen in Fig. 1. The reason for the extension of the tuning range with lower electron concentration has its origin in the factor which limits the low-magnetic-field operation of the spin-flip Raman laser and will be discussed elsewhere.² The peak spin-flip Raman-laser power was ~ 30 - 100 W for an input power of 1.5 kW obtained from a Q-switched CO_2 laser having a pulse repetition rate of 120 Hz . For spectroscopic applications, it is of great interest to investigate how the fine tuning of the spin-flip Raman laser behaves, especially over frequency shifts of the order of a few Raman cavity modes. These experiments were carried out on a $1.3 \times 10^{16} \text{-cm}^{-3}$ InSb sample.² The sample length was $\sim 5 \text{ mm}$ and the Raman-laser cavity dimen-

sion l (see Fig. 3 of Ref. 1) was 2.01 mm . This corresponds to a cavity-mode separation of $\sim 0.624 \text{ cm}^{-1}$. The InSb surfaces forming the Raman-laser cavity had no high-reflectivity coatings and thus had a reflectivity of $\sim 36\%$ determined by the specular reflection. When we include a calculated free-carrier absorption of $\sim 0.1 \text{ Np}$ per pass in the cavity (see Ref. 2), corresponding to the present carrier concentration and magnetic fields, we obtain a Raman-cavity finesse³ of ~ 2.5 . When the magnetic field was tuned from ~ 30 to $\sim 40 \text{ kG}$, the spin-flip Raman-laser output power showed a 10% modulation as a function of the magnetic field. The separation between the peaks was $\sim 0.631 \text{ cm}^{-1}$, confirming that the output-power fluctuations are caused by the spin-flip Raman emission line sweeping past the cavity resonances. In order to measure the fine tuning of the spin-flip Raman laser and to see if the output frequency was changing gradually or if it was jumping from one cavity mode to the next, the wavelength of the spin-flip Raman output was accurately measured with a $\frac{3}{4}$ -m grating spectrometer equipped with a 75 -groove/mm, $102\text{-mm} \times 102\text{-mm}$ grating. The spectrometer slits were set to give a resolution of $\sim 0.15 \text{ cm}^{-1}$, which is about the limit of the present grating. The measured spin-flip Raman-laser linewidth was seen to be $\sim 0.16 \text{ cm}^{-1}$ and no evidence of simultaneous oscillation on two cavity modes was found. (The emission linewidth of the spin-flip Raman laser can be estimated to be $< 0.03 \text{ cm}^{-1}$.) In addition, the output wavelength was found to tune linearly with the magnetic field and showed no signs of hopping from one cavity mode to the next. The maximum periodic nonlinearity of the output wavelength was measured to be $\sim 4 \text{ \AA}$ (at ~ 12.0), corresponding to $\sim 0.026 \text{ cm}^{-1}$ at 830 cm^{-1} . Even this nonlinearity was not due to the spin-flip Raman-laser frequency being pulled and pushed⁴ by the cavity modes but could be shown to arise from the periodic error in the spectrometer drive screw with a periodicity of $\sim 1.5 \text{ cm}^{-1}$. Thus the frequency pulling and pushing, if present, is much smaller than the 0.026 cm^{-1} quoted above. This is a surprisingly small number, but it may not be entirely unreasonable since in absence of any high-reflectivity coatings on the InSb Raman cavity, as mentioned earlier, the cavity Q is rather low and the cavity finesse³ is only ~ 2.5 . Thus, with a linewidth of less than $\sim 0.03 \text{ cm}^{-1}$ and a linearity of tunability and resettability far exceeding $1:3 \times 10^4$ (i.e., $\leq 0.026 \text{ cm}^{-1}$ at 860 cm^{-1}) the spin-flip Raman laser

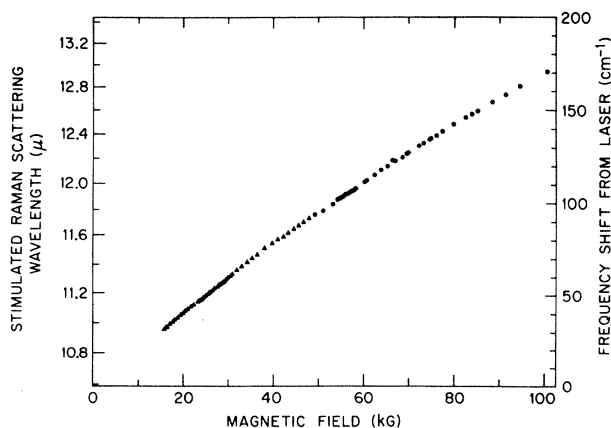


FIG. 1. Tuning characteristics of the spin-flip Raman laser pumped with a CO_2 laser at $10.6 \mu\text{m}$. (The frequency ω_s of the spin-flip Raman laser is given by $\omega_s = \omega_0 - g\mu_B B$, where ω_0 is the pump-laser frequency, g is the effective g value of the electrons, μ_B is the Bohr magneton, and B is the magnetic field.)

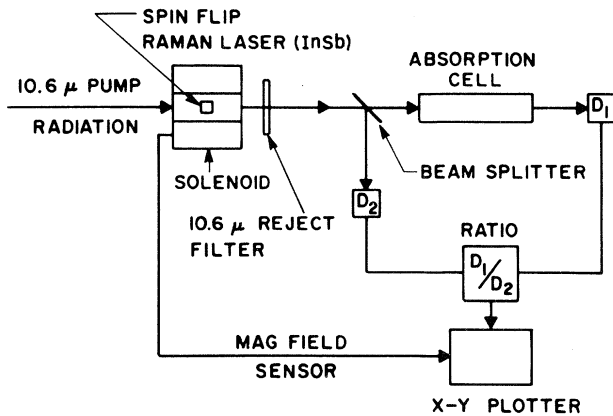


FIG. 2. Experimental setup for using the tunable spin-flip Raman laser as an infrared spectrometer source.

should be far superior to conventional grating spectrometers in use in infrared spectroscopy.

The measurements of spin-flip Raman-laser linewidth described above are clearly limited by the spectrometer resolution. In order to confirm and/or improve our estimate of $\leq 0.03 \text{ cm}^{-1}$ for the linewidth, and to evaluate the capability of the tunable spin-flip Raman laser as an infrared spectrometer, we have measured the absorption of NH_3 in the range⁵ 800-900 cm^{-1} , using the spin-flip Raman laser as the source. Figure 2 shows an outline of the experimental setup used. The spin-flip Raman laser employed an $n_e \approx 1.3 \times 10^{16} \text{ cm}^{-3}$ InSb sample. The rest of the setup is self-explanatory. The output gave the NH_3 absorption versus B . The magnetic field values were converted to frequency using the tunability curve in Fig. 1. Figure 3(a) shows a portion of the absorption spectrum of NH_3 (ammonia pressure ~ 10 Torr and absorption length ≈ 15 cm) as a function of frequency in the range 846-855 cm^{-1} . It should be pointed out that absolute calibration of frequency from the magnetic field is limited to $\sim 0.1 \text{ cm}^{-1}$, but the relative calibration is better than 0.03 cm^{-1} . The numbers on the peaks identify the transitions. The interesting point is that the $sP(6, 1)$ and $sP(6, 2)$ are resolved in our trace. The spacing is seen to be $\sim 0.05 \text{ cm}^{-1}$. This confirms our earlier estimate of a spin-flip Raman-laser linewidth of $\leq 0.03 \text{ cm}^{-1}$. Unfortunately, now we are limited to a certain extent by the Doppler broadening and pressure broadening of the NH_3 absorption line. To improve on the absorption spectrum further, we need to use longer absorption paths together with lower NH_3 pressures and lower temperatures. The trace in Fig. 3(a) should be compared with

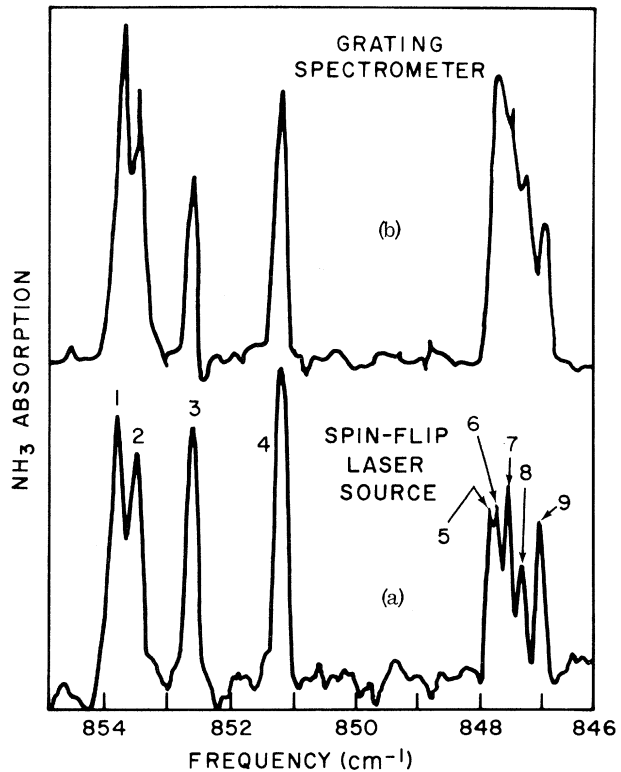


FIG. 3. (a) Absorption spectrum of NH_3 at a pressure of 10 Torr taken with the spin-flip Raman-laser spectrometer shown in Fig. 2. The identification of the transitions is as follows: (1) $aP(4, 0)$, (2) $aP(4, 1)$, (3) $aP(4, 2)$, (4) $aP(4, 3)$, (5) $sP(6, 1)$, (6) $sP(6, 2)$, (7) $sP(6, 3)$, (8) $sP(6, 4)$, and (9) $sP(6, 5)$. (For identification, see Ref. 6.) (b) Absorption spectrum taken with a conventional grating spectrometer, reproduced from Ref. 7.

that in Fig. 3(b) taken with a conventional spectrometer having a 15-cm grating. This is reproduced from Fig. 1(a) of Mould, Price, and Wilkinson,⁷ where the claimed resolution is ~ 0.1 - 0.2 cm^{-1} , and appears to be nearly the best that has been done heretofore using a conventional spectrometer.⁸ The comparison between Figs. 3(a) and 3(b) leaves no doubt about the superior resolution of the spin-flip Raman-laser spectrometer. The $sP(6, 1)$ and $sP(6, 2)$ are not resolved in Fig. 3(b). In addition, the spin-flip Raman-laser spectrometer is extremely fast (because of the relatively high, tunable monochromatic power output); the trace in Fig. 3(a) can be taken in < 1 min. The frequency stability of the spin-flip Raman-laser output was checked by adjusting the magnetic field so that we were at the peak of an absorption line such as $aP(4, 3)$, and by monitoring the absorption at a low NH_3 pressure of ~ 1 Torr for a period of 30 min.

There was no measurable change in the absorption, indicating that the frequency stability of the spin-flip Raman laser is at least as good as the estimated linewidth of Raman-laser emission, i.e., 0.03 cm^{-1} or $\sim 1:3 \times 10^4$. The ultimate resolution possible with the spin-flip Raman-laser spectrometer is expected to be much better than what we have shown so far. This is so because the emission linewidth of a quantum oscillator such as the spin-flip Raman laser is expected to be significantly narrower than the present upper limit of 0.03 cm^{-1} arrived at in the present paper.⁴

An additional advantage of the present spin-flip Raman-laser spectrometer lies in the fact that the output occurs in the form of ~ 30 -nsec-wide pulses in normal operation (or ~ 3 -nsec-wide pulses in mode-locked operation).² This should allow time-resolved spectroscopy in the $10\text{-}14\text{-}\mu\text{m}$ range which heretofore has not been possible with high resolution. The spin-flip Raman laser has the drawback of a limited tuning range. However, this disadvantage could be easily overcome by using different pump lasers, higher magnetic fields, and other narrower band-gap semiconductors (in place of InSb), such as $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ or $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ where the g value of the conduction electrons is larger than that in InSb.

In conclusion, the linewidth and fine-tuning measurements both indicate the superiority of the spin-flip Raman laser as the source in infrared spectroscopy as compared with conventional grating spectrometers. Further improvements in the measurement of the linewidth will require heterodyne spectroscopy to pin down the

exact spectral width of the spin-flip Raman-laser output. We have experimentally shown the superiority of the spin-flip Raman laser as a source in absorption spectroscopy as compared with the conventional techniques. It is clear that the immediate applications of the spin-flip Raman laser in infrared spectroscopy and as a local oscillator in heterodyne spectroscopy are eminently reasonable. Further improvements in the characteristics of the spin-flip Raman laser will accrue from its cw operation.⁹

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¹C. K. N. Patel and E. D. Shaw, *Phys. Rev. Lett.* **24**, 451 (1970).

²C. K. N. Patel and E. D. Shaw, to be published.

³M. Born and E. Wolf, *Principles of Optics* (Pergamon, New York, 1959), 2nd ed.

⁴C. H. Townes, in *Advances in Quantum Electronics*, edited by J. Singer (Columbia Univ., New York, 1961), p. 3; W. R. Bennett, Jr., *Appl. Opt. Suppl.* **1**, 24 (1962).

⁵*Tables of Wavenumbers for the Calibration of Infrared Spectrometers* (Butterworths, London, 1961), p. 671.

⁶J. S. Garing, H. H. Nielsen, and K. N. Rao, *J. Mol. Spectrosc.* **3**, 496 (1959).

⁷H. M. Mould, W. C. Price, and G. R. Wilkinson, *Spectrochim. Acta* **13**, 313 (1959).

⁸Recent improvements in detectors may give a factor of ~ 2 improvement in the resolution attainable with a grating spectrometer (K. N. Rao, private communication).

⁹For a discussion of possible cw operation, see Ref. 2.

FORMATION AND INTERACTION OF ION-ACOUSTIC SOLITONS*

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The formation and propagation of ion-acoustic solitons are observed experimentally. The character of a solitary pulse is observed to follow the predictions of the Korteweg-de Vries equation with respect to the shape and velocity of the soliton. The interaction between two solitons is modified significantly by dissipation. However, the nonlinear nature of the interactions is confirmed for solitons moving in the same direction. Solitons moving in the opposite direction and colliding have very little effect on each other.

The formation and propagation of solitons is one of the most interesting results of the nonlinear analysis of dispersive waves in many media. Kennel and Sagdeev¹ showed that the formation of solitons is closely related to the wave structure

of a laminar collisionless shock front. Sagdeev² showed, using the fluid equations, that in a plasma of hot isothermal electrons and cold ions, a single pulse (soliton), traveling slightly faster than the ion sound speed, can propagate without