The resonant third order non-linear susceptibility in InSb has been employed to generate a magnetically tunable cw laser sideband near 5 \( \mu \)m.

The third order non-linear conduction-electron susceptibility \( \chi^{(3)}(-\omega_{\text{gen}}, \omega_1, \omega_2, -\omega_3) \) has been shown \([1,2]\) to be strong in semi-conductors having non-parabolic conduction bands. The degenerate form of this non-linearity (\( \omega_1 = \omega_2 \)) has been used \([1]\) to generate the difference frequencies \( \omega_{\text{gen}} \) given by \( 2\omega_1 - \omega_3 \) in InSb and several other semiconductors, with \( \omega_1 \) and \( \omega_3 \) being provided by a pair of Q-switched CO\(_2\) lasers operating near 9.6 and 10.6 \( \mu \)m. Input powers of the order of \( 10^2 \) and \( 10^3 \) W produced difference frequency power levels of around \( 10^{-3} \) W. More recently a strong resonance in the degenerate nonlinearity has been demonstrated in InSb \([3,4]\) when the magnetic Landau energy level intervals \( \omega_c, 2\omega_c \) and the spin-splitting interval \( \omega_s \) are magnetically tuned into coincidence with the laser difference frequency \( \omega_1 - \omega_3 \). The non-degenerate 4-wave mixing process \( \omega_{\text{gen}} = \omega_1 + \omega_2 - \omega_3 \) has also been observed \([4]\) in InSb using frequencies \( \omega_1, \omega_2, \omega_3 \) obtained from fixed frequency CO lasers; the difference frequency \( \omega_{\text{gen}} \) so produced was therefore not tunable, and the mixing process was only strong at the particular magnetic fields satisfying the resonant condition \( \omega_1(\omega_2) - \omega_3 = \omega_s \). This letter now reports the generation of a magnetically tunable sideband on a fixed frequency CO laser beam near 5 \( \mu \)m via a resonant 4-wave mixing process, the frequencies \( \omega_1 \) and \( \omega_3 \) being provided by fixed frequency CO laser beams, and the frequency \( \omega_2 \) being generated within the InSb crystal by spin-flip laser action \([5]\) induced by one of the incident CO laser beams (\( \omega_3 \)). This non-degenerate 4-wave process is therefore automatically resonant (\( \omega_4 = \omega_3 - \omega_2 \)) at all magnetic fields, enabling the sideband frequency \( \omega_{\text{gen}} \) to be continuously magnetically tuned whilst retaining the strong enhancement of \( \chi^{(3)} \) under resonant conditions.

The experimental apparatus employs a highly stable cw CO laser constructed by Edinburgh Instruments Ltd., equipped with an intra-cavity diffraction grating to obtain laser line selection. Adjustment of this grating enables any chosen pair of CO laser transitions, differing in frequency by typically 3 to 4.5 cm\(^{-1}\), to oscillate simultaneously. Fine adjustment of the grating further allowed the power ratio of the two oscillating lines to be varied from 1 to about 20, whilst adjustment of the discharge current enabled the total laser power to be set at will up to the maximum level of 300 mW used in these measurements. The cw beam from the CO laser was focussed with a 15 cm BaF\(_2\) lens onto a 2 x 2 x 8.5 mm polished crystal of 8 x 10\(^{14}\) cm\(^{-3}\) InSb mounted on a helium cooled cold finger between the poles of a high homogeneity (<1 in 10\(^6\) variation over 1 cm) high stability (field variation <1 part in 10\(^5\)) electromagnet, with the magnetic field stabilised by a feedback loop operating from a Hall probe. The laser beams propagated along the long axis of the crystal in the Voigt configuration with electric vector parallel to the magnetic field, and emergent radiation was analysed with a 15 cm monochromator and Cu:Ge detector.

The input face of the InSb crystal was uncoated (natural reflectivity \(\sim 36\%\)) whilst the output face of the crystal was antireflection coated to give a reflectivity of \(< 3\%\). The principal purpose of this coating was to increase the threshold of spin-flip laser action \([5]\). With this coating and the experimental parameters described above, a laser beam power of less than 100 mW was not sufficient to produce stimulated spin-flip laser action in
Fig. 1. Monochromator scan of radiation emerging from InSb crystal with gain × 1 (A), gain × 8 (B), gain × 210 (C). The expected position of the sideband is marked by the arrows. Linewidths are limited by the resolution of the monochromator.

The region of the InSb crystal illuminated by the CO laser thus contains waves at frequencies $\omega_1$ (1884.4 cm$^{-1}$), $\omega_3$ (1888.9 cm$^{-1}$) and $\omega_2 = \omega_3 - \omega_s$ (1874.7 cm$^{-1}$) at 6.4 kG applied field) and the resonant third order non-linear susceptibility is expected to produce two new frequencies $\omega_1 + \omega_2 - \omega_3 = \omega_1 - \omega_s$ (1870.2 cm$^{-1}$) and $\omega_1 + \omega_3 - \omega_2 = \omega_1 + \omega_s$ (1898.6 cm$^{-1}$). The expected positions of the first of these frequencies is indicated by an arrow in fig. 1, and it is seen that cw emission is observed at this frequency with a power level of $\sim 100 \mu$W. The emission at 1898.6 cm$^{-1}$ lies beyond the optical absorption edge of InSb, and is therefore not expected to be observed. No attempt was made to achieve phase matching of the four wave interaction. Using the dispersion data of Brueck and Mooradian [6] the coherence length for sideband generation at 6.4 kG is estimated to be 0.7 mm. Fig. 2 shows experimental recordings of the shift of the sideband as the magnetic field is changed from 5 kG to 6.4 kG. The strong signal on the high frequency side of the sideband (arrowed) is the stimulated spin-flip emission created by the $\omega_3$ beam, which is incompletely resolved with the monochromator employed in these measurements. The four-wave mixing process thus enables the generation of a tunable sideband frequency $\omega_1 + \omega_2 - \omega_3 = \omega_1 - \omega_s$ displaced by an amount $\omega_s$ from the parent beam $\omega_1$. It is important to note that
the parent beam is not required to be sufficiently strong to produce stimulated spin-flip scattering in its own right (the parent beam was kept well below this threshold in these measurements, as noted earlier). The possibility therefore exists that similar tunable sidebands may be generated upon a CO₂ laser beam, enabling cw magnetically tunable sidebands to be produced near 10 μm; cw spin-flip laser action in InSb at such wavelengths has not so far been observed, due to the high threshold power level required for spin-flip laser action at these wavelengths.

Degenerate 4-wave mixing was also observed with our apparatus by reducing the applied magnetic field until the spin-flip frequency ωₛ was equal to the frequency separation of two CO laser lines, ω₁ (1884.4 cm⁻¹) and ω₁' (1880.0 cm⁻¹). This occurred at a magnetic field of 1810 gauss. At this field resonant energy exchange between the two CO beams and the generated difference frequencies was observed. Fig. 3 shows the magnetic field dependence of the radiation emitted from the end face of the crystal at frequencies ω₁ (1884.4 cm⁻¹), ω₁' (1880.0 cm⁻¹), 2ω₁ - ω₁ (1875.6 cm⁻¹) and 2(2ω₁ - ω₁) - ω₁' (1871.2 cm⁻¹). For this measurement the relative powers in the CO laser beams were \( P_{\omega_1} \sim 25 \text{ mW} \) and \( P_{\omega_1'} \sim 75 \text{ mW} \) (i.e. both beams below the threshold for stimulated spin-flip scattering), and the electric vector was inclined at 45° to the applied magnetic field.

In addition to the generation of the two new frequencies, power gain was observed on the incident beam at 1880.0 cm⁻¹, and depletion of the incident beam at 1884.4 cm⁻¹. The power gain on the incident beam at 1880.0 cm⁻¹ is attributed to Raman gain induced by the beam at 1884.4 cm⁻¹ [4]. The rate of photon conversion from 1884.4 to 1880.0 cm⁻¹ due to stimulated Raman scattering is proportional to \( \text{Im } \chi^{(3)}[^{3}E_0|^2|E_1|^2] \) where \( E_0 \) and \( E_1 \) are the electric vector strengths of the beams at 1884 and 1880 cm⁻¹; it is of interest to note that this power transfer is independent of which of the coupled beams is the stronger, and in fact the configuration employed in these measurements is of a strong “Stokes” beam and a weak “pump” beam. The experimental line shapes show clear asymmetry; this is induced by the interference between the resonant and non-resonant contribution to \( \text{Re } \chi^{(3)} \) previously reported by Yablonovitch et al. [3], and Brueck and Mooradian [4]. (See also ref. [7]). The spin-flip linewidth at this magnetic field is ~5 gauss, about the same as that previously reported by other workers [4] in similarly doped InSb.

In conclusion, the strong spin resonant enhancement in the third order non-linear susceptibility in InSb has been employed to generate a sideband on a CO laser beam which is continuously tunable with magnetic field. The mixing process is automatically spin resonant at all magnetic fields. No attempt has been made to obtain phase matching of the four wave interaction. We suggest that it may be possible to similarly produce tunable sidebands on a CO₂ laser beam, enabling magnetically tunable cw radiation to be generated near 10 μm.

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References