ABSOLUTE SPIN-FLIP RAMAN LASER FREQUENCY MEASUREMENTS WITH METAL–INSULATOR–METAL DIODES

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Infrared frequency synthesis techniques with metal–insulator–metal (MIM) diodes have been extended to include absolute frequency measurement of a spin-flip Raman laser (SFRL). As a result of this extension, spectroscopy in the 5.3 μm region more readily can be put on a frequency rather than a wavelength metrology basis. Additional observations with the diode are in qualitative agreement with recent work relating to non-linear tuning over axial SFRL modes.

The point contact, tungsten-nickel diode [1], also referred to as the metal–insulator–metal (MIM) diode, has been used with success to measure the frequency of several important gas lasers over the past few years. These measurements have led to a new value for the velocity of light [2], and to new secondary frequency standards [3] in the methane stabilized [4] helium neon laser at 3.39 μm and in the stabilized [5] CO_2 laser [6] at 9.4 and 10.6 μm. We report here an extension of frequency synthesis techniques with the MIM diode to include a tunable laser: namely, the spin-flip Raman laser [7] (SFRL) at 5.3 μm [8,9]. This new development, when coupled to recent developments of external cavity operation of the SFRL [10,11], provides a method using an inexpensive device to help the SFRL to achieve the potential for spectroscopy which has been expected of it. Spectroscopy in this region can be put on a frequency metrology [12] rather than a wavelength metrology basis. The capabilities of making frequency measurements at 5.3 μm have acquired added importance by virtue of a recent class of two photon experiments at 10.6 μm with the CO_2 laser [13].

The frequency of the SFRL is measured in two steps. In the first step, the frequency of the CO pump laser is measured; in the second step the pump laser – SFRL difference frequency is measured. The infrared frequency synthesis techniques described elsewhere [1,3,14] are used in the system shown in fig. 1. The simpler CO pump laser frequency measurement is described first [15,16]. In the particular experiment here, the frequency of $P_7(17)$ of the CO laser is two harmonics of the P(18) line of the CO_2 laser frequency standard (2 × 28.359 774 THz) plus 0.046 607 THz from the klystron plus a 0.000 030 THz IF beat note, or 56.766 185 ± 0.000 001 THz.

The resulting beat note ($\nu_{CO} - 2\nu_{CO_2} - \nu_{\mu\mathrm{wave}}$), with a typical 30 dB signal-to-noise ratio, not only completes the frequency measurement of the CO pump laser but is also used to stabilize the CO pump laser by the scheme in fig. 1. Over 100 CO lines lie within 40 GHz of the second harmonic of some CO_2 laser line and these CO frequencies may be measured and stabilized in this manner. The CO laser would then have nearly the same long term stability as the CO_2 laser, and its absolute frequency would be known to within a part in 10^9. This procedure increases the utility of the CO laser as a pump for a tunable Raman spin-flip laser to be used for high resolution spectroscopy [17]. Also, as a monitor of the CO laser operation, one can observe the output from the diode and make appropriate adjustments to insure that the CO laser is operating in a single mode.

The problem of measuring the difference frequency between the pump laser and the SFRL output is

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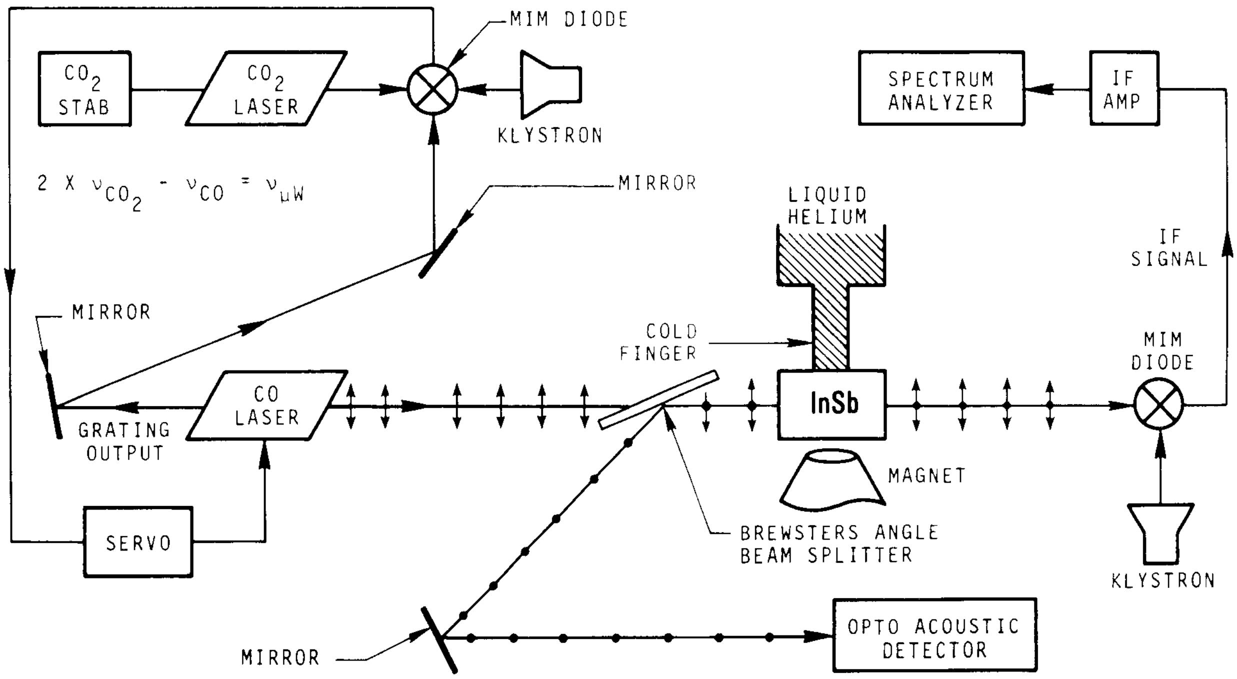


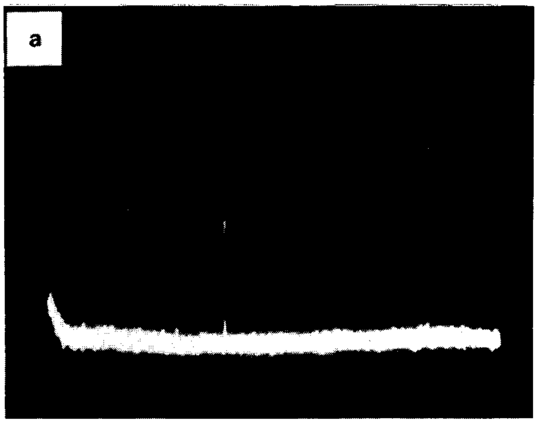
Fig. 1. Scheme for making measurements of the spin-flip Raman laser frequency. A reference frequency is synthesized from a CO_2 laser standard and a known microwave frequency. This frequency is adjusted to be 30 MHz away from the CO laser frequency, and a servo system, consisting of a discriminator, DC amplifier, and piezoelectric driver unit, maintains the beat note at 30 MHz. The 0–1 tesla magnetic field is controlled by a Hall probe and the InSb crystal is cooled by a liquid helium cold finger. The forward going SFRL signal and transmitted pump signals are processed in the MIM diode on the right for a difference frequency measurement. The reverse propagating SFRL signal is reflected off a Brewsters angle beam splitter and will be used with an opto acoustic detector to map the spectral features on which frequency measurements will be made.

complicated by three factors. First, the spin-flip and pump signals have mutually orthogonal polarizations, which originally presented difficulties in coupling to the diode antenna. Second, the power output from the SFRL is low compared to levels generally used for synthesis in MIM diodes. Third, the large collinearly-transmitted pump signal makes it difficult to establish that the weaker SFRL output has been coupled to the diode.

Long wire antenna theory indicated that for best coupling the antenna should be rotated in the plane of polarization by a specific angle, θ_m, with respect to the beam direction. An arrangement which tips the diode antenna down the proper angle in the vertical plane while maintaining equivalent projection in the horizontal plane with respect to the beam direction has permitted 5 μm signals with either polarization to be coupled to the diode. The angle for optimum coupling, θ_m, is related to the antenna length, l, and the wavelength of the radiation λ, by the equation [18]:

$$\theta_m = \cos^{-1}\left(1 - \frac{0.371\lambda}{l}\right) .$$

By using an InSb crystal with a nominal concentration of 2.5×10^{15} cm^{-3}, a broad power maxima [19] near 0.2 tesla with an estimated 30 mW output power was obtained. This point of operation was selected since the frequency difference between the CO pump and the SFRL was predicted to be about 150 GHz corresponding to the 2nd harmonic of an available klystron. Since the power available from the SFRL was low, it was deemed desirable to keep the mixing order [20] as low as possible in the initial experiments. The SFRL power density is increased at the diode junction by focusing it down with a 2.5 cm focal



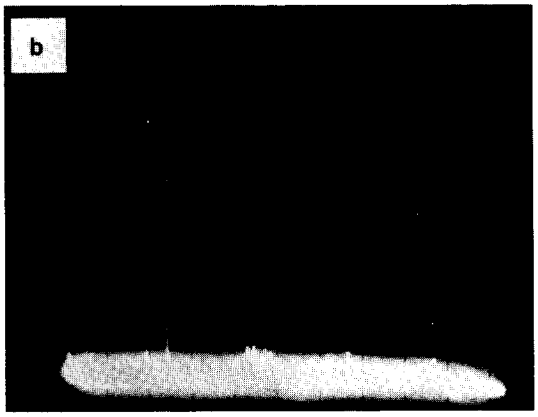


Fig. 2. a) Beat note from MIM diode when irradiated with outputs from a spin-flip Raman laser, a CO laser, and a 74.06 GHz microwave source. Center frequency is 370 MHz with 20 MHz per division dispersion, 300 kHz bandwidth, and single sweep. Signal-to-noise ratio is slightly less than 30 dB on this 10 dB per division scale. This beat note jittered around over a 20 MHz range at this particular point of operation for the spin-flip Raman laser.
b) Same as a) except that magnetic field is now 0.4216 tesla instead of 0.2142 tesla as above. This beat note is associated with 4 harmonics of the 74 GHz microwave frequency or a CO pump – SFRL difference frequency of 295.59 GHz. The center frequency is 1070 MHz, with 100 MHz per division dispersion, and the vertical scale is linear rather than logarithmic on this single sweep.

length lens. Sufficient transmitted pump power (although the radii of curvature of the pump and SFRL wave fronts are not generally equal) was also coupled to the diode through the same lens to produce a beat note among the lasers and the 2nd harmonic of the 74 GHz klystron. The beat note shown in fig. 2a has a frequency of 350 ± 2 MHz. The corresponding spin-flip laser frequency (at a magnetic field of 0.2142 tesla) was the CO pump frequency (56.766185 THz) minus two harmonics of the 0.074057 THz microwave frequency plus a 350 ± 2 MHz beat as determined by the spectrum analyzer calibration or 56.618421 ± 0.000003 THz. We point out, however, that our SFRL frequency is not stable to this degree at present.

In more recent experiments, we have used an InSb resonator with a nominal concentration of 8×10^{14} cm^{-3} and have observed SFRL-CO pump laser frequency differences corresponding to the 2nd, 3rd, and 4th harmonics of the 74 GHz klystron. Shown in fig. 2b is a beat note corresponding to a difference frequency of about 296 GHz, or nearly 10 cm^{-1}.

In the process of making these frequency measurements, several other types of beat notes are observed and some are useful in this experiment. The essential features of the spectrum analyzer display are sketched in fig. 3a, where three classes of beat notes are indicated. Beat notes labelled Class I are intermode beats associated with a single longitudinal mode of the SFRL, presumably from off axis modes and some of their harmonics. Class II beat notes occur between adjacent axial modes of the SFRL and the Class III beats result from beating of signals from the CO pump laser, the SFRL, and the microwave source. The SFRL coupling is indicated by intermode beat notes of the Class I designation. The focusing lens is further manipulated and the diode impedence is varied to maximize the amplitude of these beat notes. An additional periodic variable is the magnetic field since the SFRL gain reflects the resonator modes to some degree. When the amplitude-bandwidth area for Class I beat notes is minimized by varying the magnetic field, beat notes designated as Class II appear, tune rapidly, and disappear periodically as the magnetic field is varied by an amount corresponding to a mode ($c/2Ln$) of the InSb resonator. These Class II beats were initially mistaken for those beats denoted by Class III, however they require only the spin-flip laser for generation. The notes we have designated Class II result from the non-linear tuning over an axial mode [21–23] and two axial modes oscillating simultaneous-

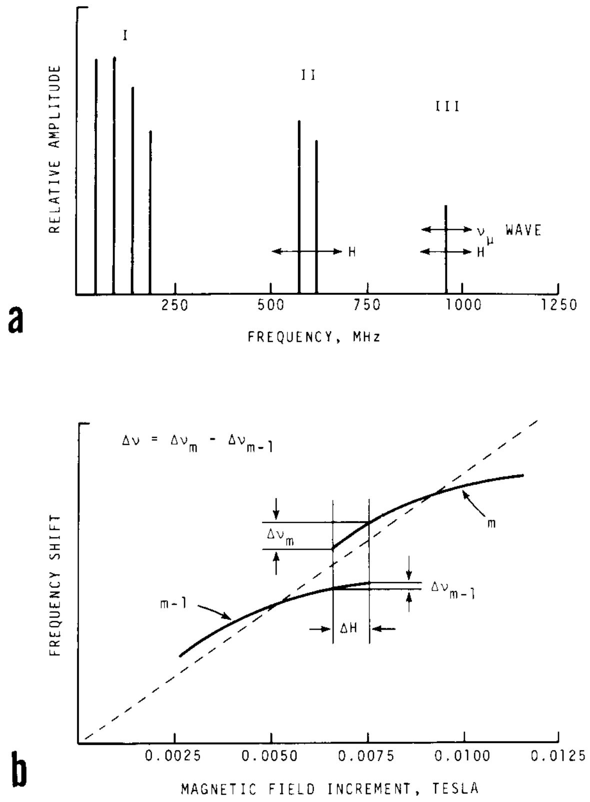


Fig. 3. a) Sketch of spectrum analyzer display of diode when irradiated with CO laser, spin-flip Raman laser and 74.06 GHz klystron. Three different classes of beat notes are displayed. Class I consists of intermode beats from the spin-flip laser and are generally in the region 0–400 MHz. Class II beat notes tune with the magnetic field, but are independent of the microwave frequency. Class III beat notes which are the beat notes of interest for spectroscopic purposes (shown in fig. 2) tune with the magnetic field and at multiples of the tuning rate of the microwave frequency.
b) Non-linear axial modes of the spin-flip Raman laser as a function of magnetic field. ΔH corresponds to the magnetic field interval for mode overlap. $\Delta\nu_m$ and $\Delta\nu_{m-1}$ correspond to the tuning rate of the beginning and end of the axial mode respectively. A measurement of the Class II beat notes tuning rate gives $\Delta\nu/\Delta H$. (The frequency axis is not scaled since the non-linearity has been exaggerated to illustrate the field dependence.)

ly near the mode hopping point [24]. These considerations are sketched in fig. 3b. The Class II beat notes behavior is in qualitative agreement with observations utilizing a different technique [22].

The beat note of prime interest is Class III which completed the SFRL frequency measurement. The amplitude of the beat depends quite strongly on the microwave power level as expected. In order to obtain the 30 dB beat note shown in fig. 2a, we have directed all the available microwave power (which is nominally 500 mW) toward the diode. The transmitted pump power was about 100 mW and the SFRL power estimated to be 30–50 mW. The fraction of any of these levels coupled to the diode of course is not readily determined. Operable diode impedance ranged from about 50 to 700 Ω.

In order to utilize fully the infrared frequency synthesis techniques described here, it is desirable to improve the SFRL performance by minimizing mode hopping, reducing cavity pulling, and operating at higher power on one stable mode. These three objectives can possibly be attained by external cavity operation of the spin-flip laser. Scanning of the external cavity length in synchronism with the magnetic field sweep [11] should reduce the mode hopping and cavity pulling problems.

Finally we would like to point out an additional application of this technique. The SFRL-CO pump difference frequency corresponds to the frequency of a coherent spin motion in the InSb. It has recently been demonstrated that this precessing spin system can be used to generate sidebands on a second CO laser over the range of 5.5 to 6.5 μm [25]. This IFS technique then can be used to determine the variable frequency separation of the sideband in that experiment.

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