NBS TECHNICAL NOTE 680

U. S. DEPARTMENT OF COMMERCE / National Bureau of Standards

Application of Infrared Frequency Synthesis Techniques With Metal-Insulator-Metal Diodes to the Spin Flip Raman Laser

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

Key words: Frequency measurements on tunable lasers; IFS with a tunable laser; infrared frequency synthesis; SFRL frequency measurement; spin flip Raman laser.

I. Introduction

The metal-insulator-metal (MIM) diode[1] 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[3,4] in the stabilized CO_2 laser at 9.4 and 10.6 μm[5,6] and in the methane stabilized helium neon laser at 3.39 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] should help the SFRL achieve the potential for spectroscopy which has long been expected of it. Spectroscopy in this region can be put on a frequency metrology rather than a wavelength metrology basis.[12] 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]

II. CO Laser Frequency Measurement

The frequency of the SFRL is measured in two steps. In the first step, the frequency of the CO pump laser is measured; the second step consists of a laser - SFRL difference frequency measurement. Before proceeding, the essentials of infrared frequency synthesis (IFS) will be briefly reviewed.[14] To measure accurately the frequency of a laser, ν_M, a

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*National Research Council Post Doctoral Associate with National Oceanic and Atmospheric Administration frequency, ν_s, which is close to the frequency to be measured, must be synthesized. The difference between these two frequencies is an intermediate frequency, ν_{IF}, typically less than one GHz. The unknown laser frequency is:

$$\nu_M = \nu_S \pm \nu_{IF}$$

$$\nu_S = \ell\nu_1 1 m\nu_2 + n\nu_{\mu W}.$$

The quantities ν_1 and ν_2 are basis laser frequencies which have been determined by prior synthesis measurement, and $\nu_{\mu W}$ is a microwave frequency. The harmonic numbers ℓ, m, and n are allowed both positive and negative values. The quantity $(1 + |\ell| + |m| + |n|)$ is called the mixing order.[15] The harmonic generation, as well as the mixing which produces the intermediate frequency to be measured, occurs in a suitable MIM diode, typically a sharpened tungsten wire antenna on a nickel base. One particular consideration has been crucial to these experiments. In prior IFS experiments the laser beams have been polarized with the electric field vector in the horizontal plane which contained the antenna. The angles between the laser beams and antenna were then dictated by long-wire antenna theory.[16] According to this theory, the angle between the antenna direction and the direction of the first maximum in the radiation pattern is given by

$$\Theta_m = \cos^{-1}\left(1 - \frac{0.371\lambda}{L}\right)$$

where λ is the wavelength of the radiation being coupled to the antenna and L is the distance between the tip of the tungsten wire and some discontinuity in the wire, either in shape or direction. Although some contention exists as to the type of coupling that prevails at 3.39 μm (perhaps a conical antenna) our experiments indicate that the long wire antenna theory is applicable at 5.3 μm. Our measured value of $\theta_m = 11^\circ \pm 1^\circ$ is commensurate with the antenna length at 100 μm. The unetched diameter of the antennas used in this work is 25 μm.

The infrared frequency synthesis techniques described above are used in the system shown in Fig. 1. The simpler CO pump laser frequency measurement[17] is described first. The CO laser power from the grating is focused onto a MIM diode thru an adjustable iris (for power control) and 12.5 cm focal length lens. The output from the CO_2 laser is also focused onto the diode through an adjustable iris and a 25.4 cm lens. The microwave power is coupled to the diode by aiming a sawed-off section of waveguide (which terminates the microwave circuit) in the general direction of the diode. 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 X 28.359 774 THz) plus .046 607 THz from the klystron plus an 0.000 030 THz IF beatnote, or 56.766 185 ± 0.000 001 THz. The klystron is phased locked to a signal synthesized from the 4th harmonic of an X-band klystron plus the IF locking frequency. The 46.63794 GHz frequency is thus determined by a frequency counter operating at X-band frequencies. The resulting beat note ($\nu_{CO} - 2\nu_{CO_2} - \nu_{\mu wave}$) which is shown in Fig. 2a not only completes the frequency measurement of the CO pump laser but is used to stabilize the CO pump laser by the scheme in Figure 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.[18] 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.

III. SFRL - CO Pump Difference Frequency Measurement

The problem of measuring the difference frequency between the pump laser and the SFRL output is 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 the angle Θ_m with respect to the beam direction. An arrangement which tips the diode antenna down 11° in the vertical plane while maintaining an 11° 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 research group at Heriot-Watt University has found maxima in the output power at 2 tesla for the $\ell = 0$ Landau level and at 0.2 tesla for the $\ell = 1$ Landau level electrons involved in the spin flip process when using crystals with a 4.5 X 10^{15} cm^{-3} concentration.[19] Following this lead, the 8 X 10^{14} cm^{-3} concentration InSb crystal in our original system was replaced with a crystal with a nominal concentration of 2.5 X 10^{15} cm^{-1}. With this 4 X 4 X 8 mm resonator, a broad maxima with an estimated 30 mw output power near 0.2 tesla was then 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 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 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 between the laser and the 75 GHz klystron. This beat note (shown in Figure 2b) has a frequency of 350 ± 5 MHz. Our spin flip laser frequency (at a magnetic field of 0.2142 tesla) was the CO pump frequency (56.766 185 THz) minus two harmonics of the 0.074 057 THz microwave frequency plus a 350 ± 10 MHz beat as determined by the spectrum analyzer calibration or 56.618 421 ± 0.000 010 THz. Again the 74.057 286 GHz is related to the 7th harmonic of an X band signal which is measured with a frequency counter.

IV. Further Diode Coupling Considerations and Observations

The first step in coupling the desired signals to the diode has been to monitor the rectified signals on the diode when chopping the laser beams. On past occasions this rectified signal has indicated that either the SFRL power or the transmitted pump power was coupled to the diode, although the more general case is to have both coupled simultaneously to some degree. Since the transmitted CO pump is somewhat larger than the SFRL signal, it is often difficult to ascertain that the latter is coupled to the diode.

In order to affect the most favorable situation, we carefully align the InSb resonator normal to a red laser beam which is collinear with the CO pump. After manipulating the SFRL output and transmitted beams with a three dimensional translator to obtain a maximum rectified signal from the diode, a 1.25 GHz spectrum analyzer is then used to monitor the beat notes. The essential features of the spectrum analyzer display are sketched in

Figure 3a. Three classes of beat notes are indicated. Beat notes labelled Class I are intermode beats (possibly off axis modes) associated with a single longitudinal mode. Class II beat notes occur between adjacent axial modes 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 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, beat notes designated as Class II appear, tune rapidly, and disappear as a function of magnetic field. These Class II beats were initially mistaken for those beats denoted by Class III, however they require only the spin flip laser for interpretation. The notes we have designated Class II result from the non linear tuning over an axial mode[20,21,22] and two axial modes oscillating simultaneously near the mode hopping point.[23] These considerations are sketched in Fig. 3b. Because of the varying structure of the aggregate of Class II beat notes (due to intermittent modes as the SFRL frequency is swept) and the smallness of the interval, ΔH, it is difficult to ascertain the value $\Delta\nu/\Delta H$. Our estimate of 0.1 to 0.2 GHz per millitesla is compatible with results obtained elsewhere.[20]

The beat note of prime interest is Class III which completed the SFRL frequency measurement. The amplitude of this depends quite strongly on the microwave power level as expected. In order to obtain the 30 db beat note shown in Figure 2b, 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 impedence ranged from about 100 to 700 Ω.

The 300 MHz beat array of intermode beats at the origin might be some cause for dismay without some elucidation. The intermode beating is a 2nd order process and the modes indicated could thus be relatively weak. Another possibility is that some of the structure is due to harmonics of two modes beating together. The observation of one beat note of the Class 111 variety (4th order mixing) does not necessarily mean that the SFRL is going single mode, but it does indicate that if other modes are present, they are relatively weak compared to the principle mode and probably would not limit resolution of molecular spectra.

We do not imply by the exhibition of the single beat note in Figure 2b that our system did not exhibit the undesirable situation of more than one mode at some magnetic fields in addition to the mode hopping region. The non-linear mode tuning is another feature which is undesirable from a spectroscopic viewpoint. Clearly the SFRL requires some improvement and recent work elsewhere indicates that these solutions are very close.[10,11]

In order to utilize fully the infrared frequency synthesis techniques described here, it is desirable to improve the SFRL performance in three areas. These objectives, minimization of mode hopping, reduction of cavity pulling, and operation at higher power on a single mode, can possibly be attained by an external cavity operation[10,11] of the spin flip laser. We plan to attempt some of these improvements in the near future. The technique of scanning an external mirror with the magnetic field drive signal seems particularly appealing at present.[11]

We would like to acknowledge helpful discussions with Dr. D. A. Jennings, NBS, Boulder and thank Dr. W. Schade and Dr. S. Miller of the Naval Electronics Lab in San Diego for pointing out their "poor man's" polishing technique which we used to polish our InSb samples. The pleasant association with Dr. J. J. Jimenez (a guest worker from LPTF, Observatoire de Paris) also proved of worth to this experiment.

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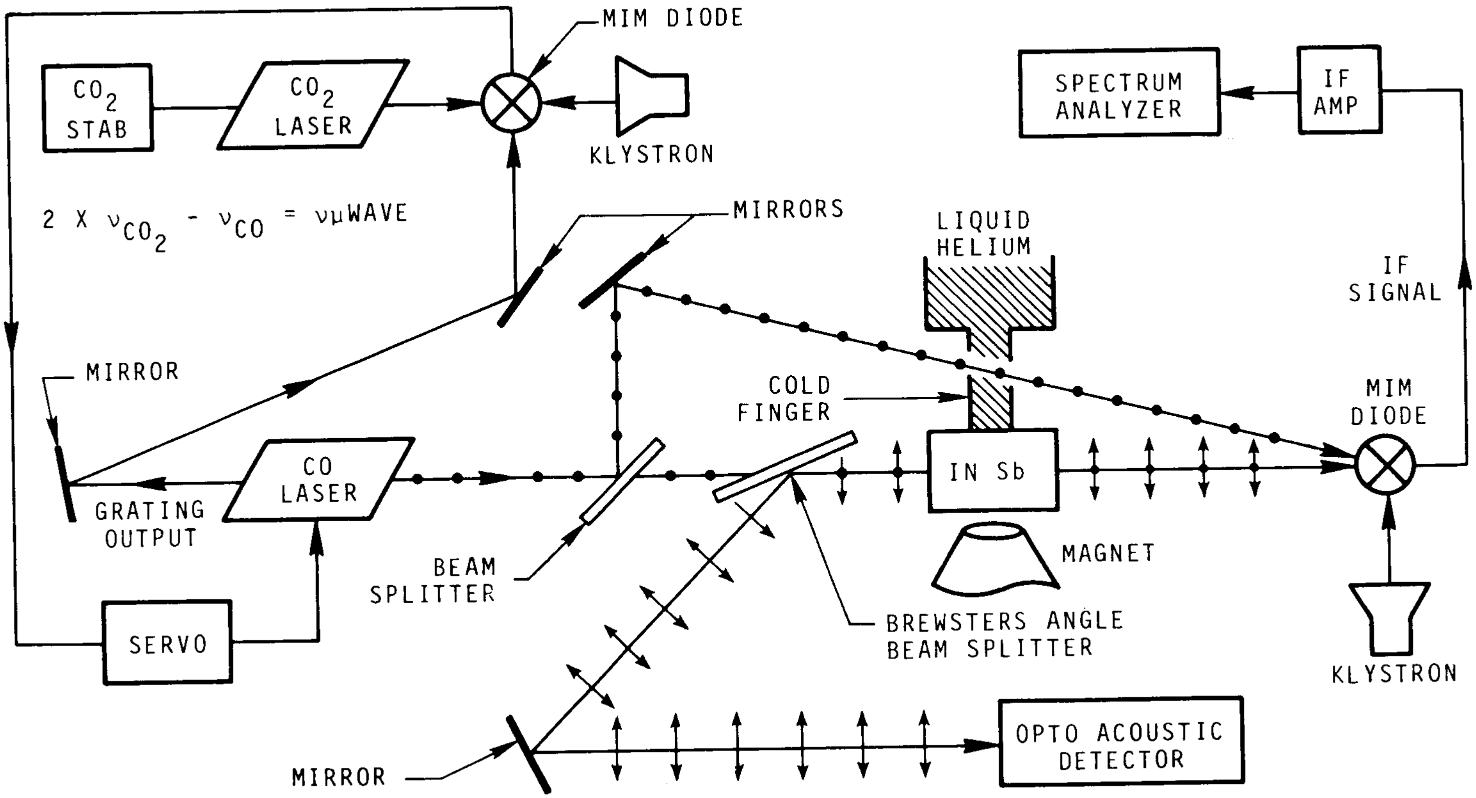

Figure 1. Scheme for making measurements of the spin flip Raman laser frequency. A reference frequency is synthesized from a CO_2 laser standard and a known microwave frequency. This frequency is adjusted to be 30 MHz away from the CO laser frequency, and a servo system consisting of a discriminator, 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 additional pump power provided by the beam splitter output was not required at the frequency used in this experiment. 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.

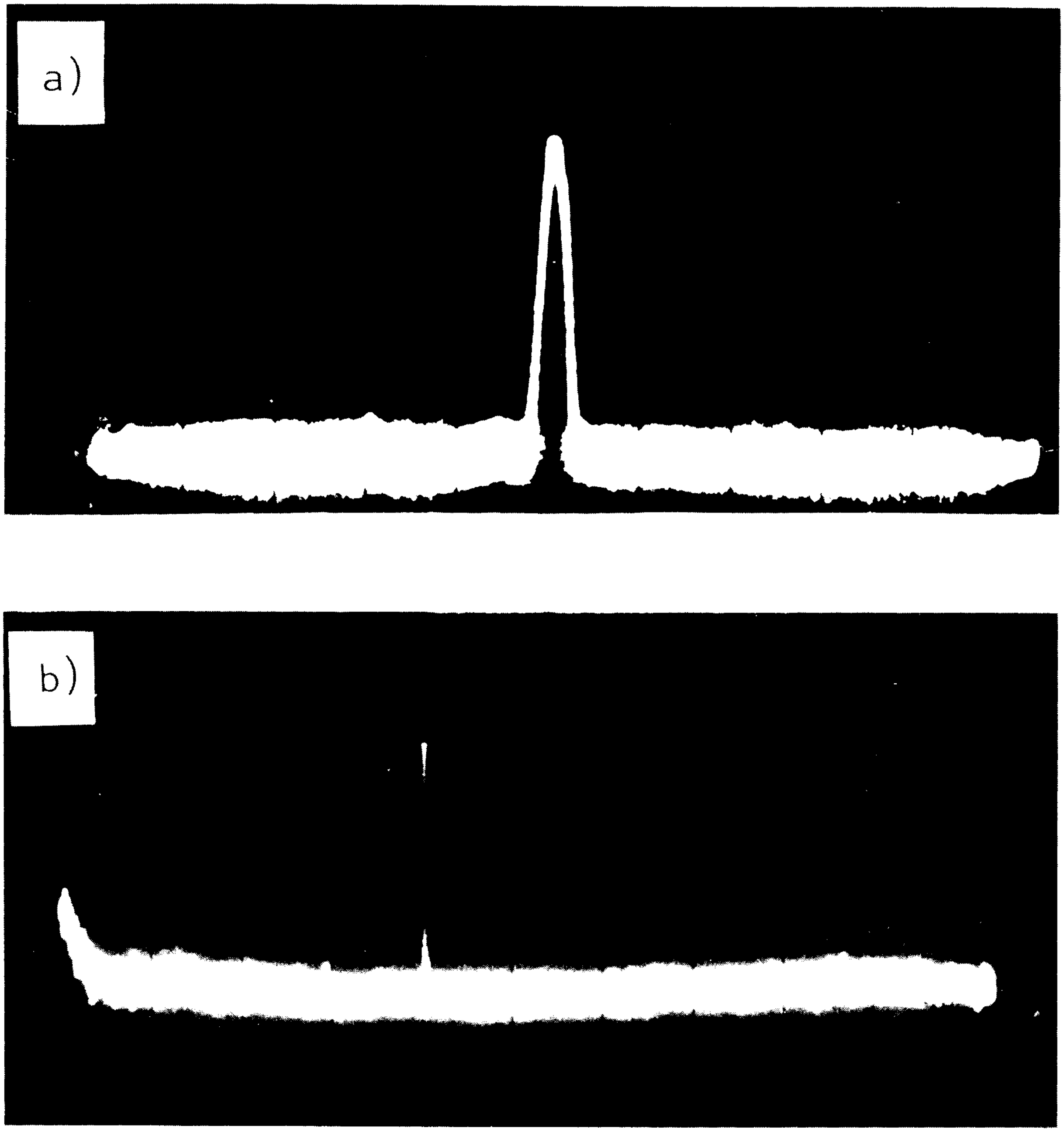

Figure 2. a) Beat note from metal-insulator-metal (MIM) diode when irradiated with outputs from CO$_2$ laser, a CO laser and a 46.6 GHz microwave source. The center frequency is 30 MHz with 1 MHz per division dispersion, 1 ms per division sweep rate and 30 khz bandwidth. Display is logarithmic with 10 db per division. The signal to noise ratio is slightly over 30 db.

b) 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, 5 msec per division sweep rate, single sweep, and 300 khz bandwidth. Signal to noise ratio is slightly less than 30 db on this 10 db per divison scale. This beat note jittered around over a 20 MHz range at this particular point of operation for the spin flip Raman laser.

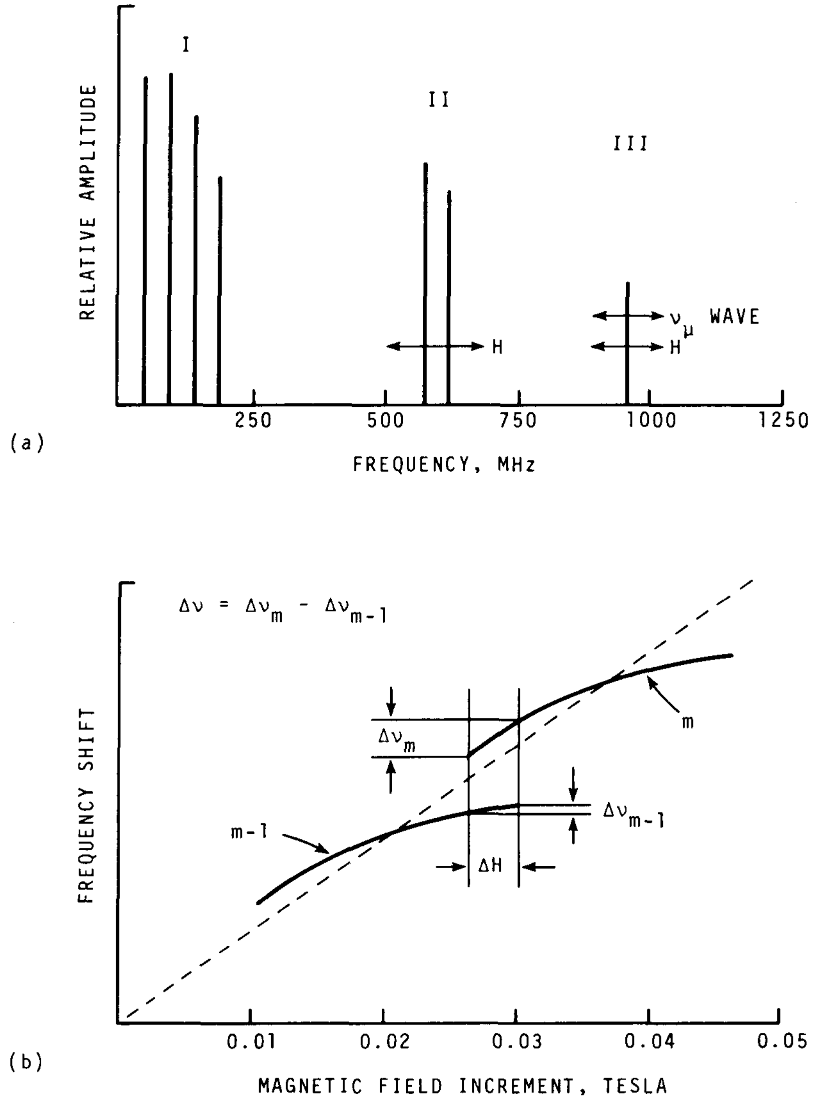

Figure 3 a) Sketch of spectrum analyzer display of diode when irradiated with CO laser, spin flip Raman laser and 74.06 GHz klystron. Three different classes of beat notes are displayed. Class I consists of intermode beats from the spin flip laser and are generally in the region 0-400 MHz. Class II beat 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 Figure 2b) tune with the magnetic field and at twice the 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.)

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