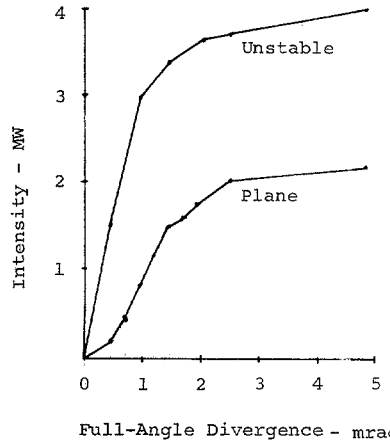

Fig. 1. Integrated angular intensity distribution.

Fig. 1 illustrates the measured angular intensity distribution of the Q-switched laser using either the unstable or parallel-plane mirror resonators and pertinent parameters are compared in Table I.

A significant practical advantage of the negative-branch unstable resonator was found in its relative insensitivity to angular misalignment. Initial alignment could be easily achieved and maintained over long periods without resort to special components.

In conclusion, we have found that efficient energy extraction and near diffraction-limited beam quality together with stable, non-critical alignment could be achieved with a negative-branch unstable resonator applied to a Q-switched Nd:YAG laser. Comparison of experimental results clearly shows the superiority of the unstable resonator form over the conventional parallel-plane mirror system.

4.12 Laser Transition Measurements for Nd-Glass, E. M. Dianow, A. Ya Karasik, L. S. Kornienko, A. M. Prokhorov, I. A. Shcherbakov, *USSR*

(15 min)

(Received late; see page 83)

Session 5

Wednesday, May 28, 1975 1:30-5:00 P.M.

Tunable Lasers and Isotope Separation

Chairman: O. Judd, *Los Alamos Scientific Laboratory, Los Alamos, N. M. 87544*

5.1 Applications of Tunable Infrared Lasers *(Invited)*, P. L. Kelley, *MIT Lincoln Lab., Lexington, Mass.*

(30 min)

No summary.

5.2 Tunable $Hg_{1-x}Cd_xTe$ Infrared Lasers, J. P. Sattler, B. A. Weber, and J. Nemarich, *Harry Diamond Laboratories, Adelphi, Md. 20783*

(15 min)

We report on the development of new magnetically-tunable infared lasers using the semiconductor alloy $Hg_{1-x}Cd_xTe$. Tunable spin-flip Raman lasing and tunable recombination lasing have been obtained from this material using chopped-CW and TEA CO_2 lasers for optical pumping.

Semiconductor band theory and experimental results have indicated that crystals of $Hg_{1-x}Cd_xTe$ with small band gaps should have highly tunable Raman and recombination radiation, owing to the small conduction band effective mass. In addition, by selecting the appropriate composition of this material with respect to a pump-laser line, one should be able to reduce the spin-flip lasing threshold because of resonant enhancement effects. Our experiments indicate that these expectations are indeed warranted for this material, which offers great promise as a tunable infrared laser. Initial measurements using a TEA CO_2 pump laser have yielded a large first-Stokes tuning rate of $3.4\ cm^{-1}/kG$[1].

The experiments with the chopped-CW source utilized a conventional grating-tunable CO_2 laser that produced 2 W in the TEM_{00} mode. The output beam was externally chopped to a 2% duty cycle at 73 Hz to reduce crystal heating. This radiation with $E_p \| H$ was focused using a 26-cm focal length Ge lens onto the $Hg_{1-x}Cd_xTe$ crystal held at 12 K on a cold finger in a superconductive magnet. The crystal was a $0.5 \times 3 \times 6$-mm parallelopiped, and was pumped in collinear geometry parallel to the 3-mm length and perpendicular to H. The crystal had an n-type carrier concentration of $5.4 \times 10^{14}\ cm^{-3}$, a mobility of $1.1 \times 10^5\ cm^2/V\text{-sec}$ and a cadmium fraction, x, of 0.234. The output radiation was analyzed using a 0.5 m spectrometer, a Ge:Cu detector, and a lock-in amplifier. The spin-flip radiation could be produced using all pump lines of the CW laser from the $P(16)$ to the $P(30)$ of the (00°1-02°0) band. Using the 9,52-μm line, the emission tuned from 9.67 to 9.96 μm by varying the magnetic field from 4 to 13 kG. A greater tuning range was obtained with the TEA laser pump.[1] The threshold for stimulated emission was measured to be 150 mW, corresponding to an intensity of $200\ W/cm^2$. The output polarization was perpendicular to the pump polarization in accordance with spin-flip selection rules.

The recombination radiation was observed for all CW-CO_2 laser pump lines between 9.22 and 9.55 μm. When using the 9.22-μm pump line, the emission tuned from 9.59 to 9.36 μm by varying the magnetic field from 0 to 20 kG. Various cavity modes could be selectively excited by adjusting the focusing and alignment of the pump laser beam with respect to the crystal faces. The wavelength, polarization, and output power of the recombination radiation depended on the magnetic field and pump wavelength.

The tuning data of these lasers for $Hg_{0.766}Cd_{0.234}Te$ have been analyzed using band theory, and the effective-mass ratio has been determined to be 9.77×10^{-3}. This value compares favorably with those obtained by other experimenters using other techniques for similar alloy ratios. The results obtained are used to project the tuning behavior of $Hg_{1-x}Cd_xTe$ crystals with different x-factors and electrical parameters.

[1] J. P. Sattler, B. A. Weber, and J. Nemarich, *Appl. Phys. Lett.*, vol. 25, p. 491, 1974.

5.3 A Highly Stabilized, Tunable CW Dye Laser and an Accurate Determination of the Rydberg Constant, J. B. West, R. L. Barger, and T. C. English, *National Bureau of Standards, Boulder, Col. 80302*

(15 min)

Recently it has been demonstrated[1] that remarkable frequency stability may be obtained from a CW dye laser. By using a wide-band (0-100 kHz) servo system to lock the dye laser frequency to a transmission fringe of a high finesse optical cavity, residual frequency fluctuations of less than 50 kHz for short times (20 μ sec) and 100 Hz for long times (10 sec) were obtained. However, due to thermal fluctuations in the reference optical cavity, long term drifts on the order of 1.5 MHz/min were observed.

Modifications to the laser system have resulted in a significant improvement in performance.[2] A high optical quality dye jet has replaced the dye cell and wavelength tuning is now accomplished using a tilted birefringent filter. Lasing in a single longitudinal cavity mode is achieved with two intracavity etalons. Error signals for the frequency control servo are derived from the signal difference between the transmission fringe of an external high finesse cavity and a reference beam bypassing the cavity. Low frequency corrections (0-1 kHz) are applied directly to the laser output mirror using a piezioelectric driver. High frequency corrections (1 kHz-5 MHz) are applied by varying the voltage across a deuterated KDP crystal in the laser cavity. Present frequency fluctuations are 1 kHz for short times and 300 Hz for 300 seconds. Long term frequency drifts are virtually eliminated by locking the length of the servo cavity to a fringe of a 3.39 μm He-Ne local oscillator laser which is frequency-offset-locked to a methane stabilized laser.[3] By using this technique, the long term frequency drifts of the dye laser should approach that of the methane-stabilized laser (about a part in 10^{13}), and the variable frequency-offset-locking technique allows the dye laser frequency to be tuned over 2.5 GHz while stabilized to this degree.

The amplitude of the dye laser output is stabilized to one part in 10^4 with a high speed

[1] R. L. Barger, M. S. Sorem, and J. L. Hall, *Appl. Phys. Lett.*, vol. 22, p. 573, 1973.

[2] R. L. Barger, J. B. West, and T. C. English, in preparation.

[3] R. L. Barger and J. L. Hall, *Phys. Rev. Lett.*, vol. 22, p 4, 1969.

servo system which uses an ADP crystal and linear polarizer combination. Present plans call for the entire dye laser and servo cavity to be enclosed in a pressure box. This will allow continuous pressure tuning of the laser over a 0.2 nm wavelength range without disturbing the frequency servo lock.

Presently, experiments are in progress which use this stabilized laser to saturate the Balmer α transition of the hydrogen atom. The result of these experiments should be a significant improvement in the accuracy of the Rydberg constant. Saturation of the transition in a beam offers specific advantages over experiments done with a discharge. In particular, shifts and broadening due to collisions and the Stark effect should be nearly eliminated. For these reasons, our experiments use a beam of metastable hydrogen atoms.

An RF discharge is used to produce ground state hydrogen atoms which are then collimated using a multichannel array of 5 μm diameter capillary tubes. Metastable hydrogen atoms (2^2S) are produced by bombarding ground state atoms with electrons near threshold energy ($\approx$11 eV). Metastable atoms in the beam are detected downstream by applying a dc electric quenching field. A photomultiplier tube monitors the resulting Lyman-α emission which is proportional to the number of metastable atoms in the beam. Laser saturation of the hydrogen 2^2S-3^2P transition occurs just upstream from the detector. Thus, the saturation is observed as a decrease in the Lyman-α emission.

The laser wavelength is measured with a frequency-controlled Fabry-Perot interferometer[4] which is accurate to about 10^{-5} order.

Saturation of the 2^2S-3^2P transition should provide a linewidth on the order of 30 MHz. If, however, the 3^2P state is used as an intermediate state and an RF field is simultaneously applied to induce transitions from the 3^2P to 3^2S state, a linewidth of 1 MHz should be realized.[5] After careful corrections for systematic errors, this method should provide an accuracy of at least one part in 10^9 for the Rydberg constant.

[4] R. L. Barger and J. L. Hall, *Appl. Phys. Lett.*, vol. 22, p. 196, 1973.

[5] D. E. Roberts and E. N. Fortson, *Phys. Rev. Lett.*, vol. 31, p. 1539, 1973.

5.4 Laser Stimulation of Chemical Reactions (*Invited*), R. Deslattes, *National Bureau of Standards, Gaithersburg, Md.*

(30 min)

No summary.

5.5 Laser Isotope Separation (*Invited*), B. B. Snavely, *Lawrence Livermore Laboratory, Livermore, Calif. 94550*

(30 min)

No summary.

5.6 Application of Photodeflection to Spectroscopy,[1] A. F. Bernhardt,[2] D. E. Duerre, J. R. Simpson, and L. L. Wood, *Lawrence Livermore Laboratory, Livermore, Calif. 94550*

(15 min)

The laser deflection of a single isotopic component of an atomic beam has recently been demonstrated. The technique relies on the selective absorption of laser beam photons by one isotopic component of an atomic beam to transfer momentum from the laser beam to that component. In a well-collimated atomic beam, the momentum acquired by many absorption and spontaneous re-emission events per atom will be sufficient to deflect the isotopic component out of the original beam.

This physical separation permits mass analysis of the deflected beam and highly reliable mass assignment of various isotopic hyperfine components. In addition, peaks from different isotopic components which cannot be relatively resolved optically, can be unequivocally resolved from the associated mass spectrometer data. A plot of mass peak height as a function of laser frequency gives a separate absorption curve for each mass number. The isotopic hyperfine structure of the barium 5536 Å resonance was investigated in this manner. Frequency shifts for all peaks of isotopes 134 through 137 were measured relative to 138 with $\leqslant$8 MHz error. The natural linewidth of the transition is 20 MHz. The absorption peaks of ^{134}Ba, ^{136}Ba and ^{135}BaF = 5/2, which could not be resolved optically, were resolved from the mass spectromer data. This is the first experimental resolution of these peaks.

The accuracy of the photodeflection method of spectroscopy is limited by reading error of the reference peak and the peaks plotted from mass spectrometer data. In the limit of a spectrum analyzer of great finesse and negligible laser linewidth, this method has an accuracy of about one tenth the natural linewidth of the transition. Since the natural linewidth of 5535 Å Ba resonance is $\sim$20 MHz, it would be possible to achieve an accuracy of about $\pm$2 MHz in the determination of the hyperfine structure of this resonance line.

The separation efficiency in the barium experiment has been measured. A simple expression for this efficiency has also been derived. The most important parameter in this expression is the branching ratio into the metastable 6s5d $^1D_2$ state from the upper 6s6p $^1P_1$ level of the 5536 Å resonance. The deflection process terminates for a given atom when it decays into the $^1D_2$ level rather than returning to the ground state. The previously accepted value for this ratio is 1/24. This would imply a separation efficiency in our experiment of less than 10^{-4}. Instead, we measured an efficiency of more than 70%. Our result implies that the accepted branching ratio of 1/24 is high by just over a factor of 30.

[1] Work performed under the auspices of the U. S. Atomic Energy Commission.

[2] Also Fannie & John Hertz Foundation Fellow at the Dept. of Applied Science, University of California, Davis-California.

5.7 Experimentally Measured Photoionization Cross Sections and Excited State Lifetimes in Uranium Vapor,[1] R. W. Solarz, *Lawrence Livermore Laboratory, Livermore, Calif. 94550*

(15 min)

The chemistry and spectroscopy of uranium are especially complicated and a complete knowledge of the behavior of this important element is required in order to assess the promise of various schemes for an isotope separation process. Some of the parameters needed for assessing the varied approaches are, for example, hyperfine splittings, lifetimes, bound state absorption cross sections, and relative photoionization cross sections from laser populated excited states. Until now most of the available spectroscopic information has been obtained using classical techniques such as high resolution absorption spectroscopy. Attempts to analyze the resulting spectra have been partially successful; nonetheless much of the data has defied analysis making it clear that alternate experimental approaches are required. While there is currently an excellent line list available for the U I and U II systems, many levels have not been assigned. In addition little information exists concerning hyperfine structure and no data exists on lifetimes, ionization cross sections, and high lying levels of the neutral and ionized species. Furthermore there are several experimental problems unique to the uranium system which require attention in any experimental set-up. This paper will present recent laser spectroscopic experiments aimed at measuring lifetimes and photoionization cross sections in uranium vapor. Experiments to map out high lying odd parity levels in uranium will also be presented.

It is well known that most isotope separation schemes, whether employing physical or chemical extraction processes, require a relatively long-lived laser populated level in order to function at peak efficiency. In a search for such long-lived states we have begun a systematic effort to map the lifetimes of uranium levels in the visible region of the excitation spectrum. A uranium oven operated at about 2000°C provides a cloud of primarily $^5L_6^\circ$ ground state and $^5K_5^\circ$, 620 cm^{-1} populated vapor. A Molectron commercial N_2 pumped dye laser is used to bring either population to a predetermined excited state. The resulting fluorescence decay is monitored through a half meter monochromator-phototube-boxcar arrangement. Advantages of this method over alternate techniques will be discussed and the experimental results presented.

Also well known is that a primary difficulty in a two step photoionization process is that the cross sections for ionization are typically quite small resulting in the requirement of a large laser for the second state of the separation process. In order to minimize this requirement an experiment to determine relative photoionization cross sections in uranium vapor has been carried out. In this experiment a high temperature uranium oven is again utilized. The resulting

[1] This work was performed under the auspices of the U. S. Atomic Energy Commission.