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This report summarizes research progress on long-term laser frequency stabilization for the period January 1, 1976 - December 31, 1976. It includes a list of publications published under AFOSR support.

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LONG-TERM LASER FREQUENCY STABILIZATION

U. S. Air Force Office of Scientific Research

Contract F44620-76-C-0079

PROGRESS REPORT

covering the period

January 1, 1976 – December 31, 1976

Principal Investigator: Shaoul Ezekiel

March 14, 1977

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SUMMARY OF RESEARCH

The aim of the research program is the long-term stabilization of the frequency of the 5145 Å argon ion laser by using an I_2 resonance, observed in a molecular beam, as a reference. The major accomplishments during the past year and the status of the program are outlined below.

a. Long-Term Stabilization

The 5145 Å argon ion laser has been locked to a hyperfine transition in a molecular beam of I_2. Two methods were used to measure the long-term frequency drift. The first method is the simpler one, and only requires one additional molecular beam apparatus. Here, the laser frequency drift is monitored by measuring the shift of the laser frequency with respect to the center of the same I_2 transition observed in an independent molecular beam. In the second and more conventional method, two argon lasers are independently stabilized to adjacent I_2 transitions in independent molecular beams. The two laser outputs are mixed on a fast photodiode, and the resulting beat frequency gives an accurate measure of frequency stability. Both methods yielded similar results; i. e., better than one-part-in 10^{13} in an integration time of about 100 seconds. This is the highest stability yet achieved in the visible region of the spectrum.

b. Intensity Shifts

The shift in the stabilized laser frequency as a function of laser intensity was found to be less than two-parts-in 10^{14} for a 1% change in laser intensity at 1 mW. This value is still an upper limit at present, pending a more detailed study.

c. Shifts Due to External Magnetic Fields

The effect of external magnetic fields was investigated by applying a 50-gauss field at various angles with respect to the I_2 interaction region. A shift of two-parts-in 10^{13} per gauss was measured. This type of shift can, of course, be reduced significantly by using suitable magnetic shielding. At present, there is no magnetic shielding.

d. Frequency Reproducibility

The reproducibility of the frequency of a molecular beam stabilized laser depends on several factors, the most important of which is the orthogonality between the laser and molecular beam. Any deviation from orthogonal excitation of the I_2 beam results in a shift of the center frequency of the I_2 transition, as well as a nonsymmetrical broadening of the line due to the velocity distribution of molecules in the beam. Frequency shifts caused by nonorthogonality can be reduced considerably by reflecting the laser beam back through the I_2 beam by means of a precision corner reflector. In this way, two iodine transitions are observed which are symmetrically displaced with respect to the true line center. For a small misalignment angle, the two I_2 transitions overlap such that the overall effect of retroreflection is to convert a shifted line to an unshifted but broadened line as long as the intensities of the two laser beams are equal. Clearly, the smaller the misalignment angle, the less the contribution due to unequal intensities.

In our reproducibility experiments, we determined the orthogonal alignment between laser and molecular beam by deflecting the laser beam and observing the width of the I_2 transition as a function of angle. In this way, we were able to achieve orthogonal alignment within 5×10^{-5} radians, which corresponds with an error in the line center of 24 kHz. This misalignment error was reduced further by reflecting the laser beam back through the I_2 beam with a one-second of arc corner cube reflector. The reflected beam, which was 10% less intense than the incident beam, was prevented from reentering the laser by making the laser beam strike the corner cube slightly off center, thus inducing a slight translation in the reflected beam. The procedure for reproducing the laser frequency was to unlock one of the stabilized lasers and remove the alignment mirror and corner reflector; then to realign the laser beam, as described above, and measure the new beat frequency with respect to the fixed laser. The results of 5 attempts gave a standard deviation in the reproducibility of 1.5×10^{-12}, which is the best that has yet been obtained for a laser in the visible region.

e. Line Shape of I_2 Hyperfine Transitions

The line shape of I_2 hyperfine structure observed in a molecular beam was examined with an instrumental resolution of one-part-in 10^{10}. This is the highest resolution yet achieved in the visible region of the spectrum. The experimental setup employs a 5145 Å argon ion laser which is long-term stabilized (one part in 10^{13}) to one hyperfine component in a molecular beam of I_2; the stabilized laser frequency is precisely tuned, by an external acousto-optic scheme, to excite a neighboring hyperfine component in a second I_2 molecular beam. The measured linewidth is 75 kHz (HWHM). The natural width for the transition is inferred to be 35 kHz from lifetime data. In the present measurement the contributions of Doppler and transit time-broadening is approximately 10 kHz; Zeeman broadening in the Earth's field is estimated to be less than 5 kHz and collisional broadening is expected to be negligible in the beam. Line broadening caused by laser jitter is estimated to be 20 kHz. The observed line shape has been compared with theoretically predicted line shapes that take into consideration the various broadening mechanisms we have mentioned. It was found to be predominantly Lorentzian.

f. Status of the Understanding of I_2 Hyperfine Structure

The hyperfine structure of I_2 within the bandwidth of the 5145 Å argon laser is associated with the P(13) and R(15) lines in the 0-43 band. The spacings between the hyperfine structure transitions were determined with a precision of one part in 10^{11}, using a heterodyne technique employing two argon lasers individually stabilized to various I_2 transitions.

In order to describe the hyperfine structure accurately it was necessary to construct a Hamiltonian that included the following interactions:

$$H_{HFS} = H_{NEQ} + H_{SR} + H_{TSS} + H_{SSS}$$

The first three terms of the Hamiltonian are, respectively, the nuclear electric quadrupole, the magnetic spin rotation and tensor spin-spin interactions. The fourth term in the Hamiltonian is the scalar part of the nuclear spin-spin interaction which results from the indirect electron coupled spin-spin interaction.

The coupling strengths associated with every term in the Hamiltonian were varied in a least-squares computer program to obtain the best fit to the data. The results showed that the inclusion of the tensor and scalar nuclear spin-spin interaction dramatically reduced the standard deviation of the fit 2.6 kHz for the R(15) transition.

The high resolution available in the present experiment has allowed the precise determination of both ground- and excited-state coupling constants for the quadrupole, spin-rotation action, tensor and scalar spin-spin interactions. These are given below for the R(15) transition.

$$eQq' - eQq'' = 1893.9757 \pm 0.0015 \text{ MHz}$$

$$C' - C'' = 187.520 \pm 0.02 \text{ kHz}$$

$$D'_t - D''_t = 101.916 \pm 0.30 \text{ kHz}$$

$$D'_s - D''_s = 2.21 \pm 0.22 \text{ kHz}$$

$$eQq' = -559.8142 \pm 0.200 \text{ MHz}$$

$$eQq'' = -2453.7900 \pm 0.200 \text{ MHz}$$

$$C' = 187.520 \pm 0.02 \text{ kHz}$$

$$C'' = 0.0$$

$$D'_t = -101.660 \pm 0.5 \text{ kHz}$$

$$D''_t = 0.256 \pm 0.5 \text{ kHz}$$

$$D'_s = -2.216 \pm 0.22 \text{ kHz}$$

$$D''_s = 0.00$$

g. Short-Term Stabilization of High-Power Argon Laser

Recently, we have been investigating the short-term frequency stabilization of a high-power (15 W) commercially-made argon laser (Spectra-Physics Model 171). The free-running laser jitter of this laser is about 20 MHz, which is mainly due to cooling water noise, power supply fluctuations and plasma noise. By using an intracavity electro-optic phase modulator as a fast response length transducer, we were able to reduce the 20 MHz jitter to about 26 kHz. This was accomplished by locking the laser frequency, by means of a wideband feedback loop, to the side of the resonance of an external Fabry-Perot interferometer. The bandwidth of the feedback loop was ~1 MHz. The laser jitter was measured by heating two independently stabilized high-power argon lasers.

h. Long-Term Stabilization of High-Power Argon Laser

The multiwatt argon laser described in (g) was also long-term stabilized by locking the reference Fabry-Perot reference to a transition in an I_2 molecular beam. A long-term stability of seven parts in 10^{14} was achieved in an integration time of 1000 seconds.

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T. J. Ryan, Sc.D. Thesis entitled, "Argon Laser Frequency Stabilization Using an Iodine Molecular Beam Reference," February 1973.

L. A. Hackel, S.M. Thesis entitled, "Molecular Beam Techniques for High-Resolution Spectroscopy," February 1973.

D. G. Youmans, S.M. Thesis, entitled, "Molecular Beam Stabilized Argon Laser," February 1973.

L. A. Hackel, Sc.D. Thesis, entitled, "Precision Laser Spectroscopy of I_2 Hyperfine Structure," September 1974.

D. G. Youmans, Ph.D. Thesis, entitled, "Performance of Molecular Beam Stabilized Argon Laser," September 1974.

Personnel Who Have Participated in the Program in the Past Year

Dr. Shaoul Ezekiel	–	Associate Professor
Dr. Lloyd Hackel	–	DSR Staff
Richard Hackel	–	Graduate Student
Steven Goldstein	–	Graduate Student