CO_2 : $^{192}OsO_4$ laser: absolute frequency of optical oscillations and new possibilities

Yu. S. Domnin, N. B. Roshelyaevskiĭ, V.M. Tatrenkov, P. S. Shumyatskiĭ, O. N. Kompanets, A. R. Kukudzhanov, V. S. Letokhov, and E. L. Mikhaĭlov
All-Union Scientific-Research Institute of Physico-Technical Radiotechnical Measurements and Institute of Spectroscopy, USSR Academy of Sciences

(Submitted 12 July 1979)
Pis'ma Zh. Eksp. Teor. Fiz. **30**, No. 5, 269–272 (5 September 1979)

The absolute frequency of a CO_2:OsO_4 laser is measured. The possibility of measuring with the standard accuracy the frequencies of lasers operating at transitions of shorter wavelengths (> 28 THz) and setting up an experiment for verification of the hypothesis of variation of the universal constants is discussed.

PACS numbers: 42.55.Dk, 06.30.Ft

1. The most accurate measurements of the absolute frequencies of lasers in the submillimeter and IR ranges have heretofore been made with the error of $\gtrsim 6\times 10^{-10}$.[1,2] The traditional scheme of measurements is a circuit consisting of

 0021-3640/80/170249-04$00.60

several stages, in which the unknown laser frequency at each stage is measured by comparing it with the sum or difference frequency of a given harmonic of the laser emission of the preceding stage and a harmonic of a klystron with a known frequency measured with respect to a standard source. A possible way of increasing the accuracy of the absolute measurements of laser frequencies is modification of the traditional system of synthesizing their frequencies by using an independent secondary frequency standard based on a CO_2 : $^{192}OsO_4$ laser operating at the $P(14)$ line at the wavelength of 10.53 μm.

In this paper we report the first absolute frequency measurement of a CO_2 : $^{192}OsO_4$ quantum standard, which was obtained by stabilizing the frequency of a CO_2 laser at the $P(14)$ line with respect to a narrow nonlinear resonance in the vibrational-rotational spectrum of a $^{192}OsO_4$ molecule. The CO_2 : $^{192}OsO_4$ standard is the first optical standard whose frequency was measured with an accuracy of 3×10^{-11}.

2. The selection of the $^{192}OsO_4$ molecule to obtain narrow nonlinear resonances suitable for stabilizing the frequency of a CO_2 laser with high accuracy is determined by the absence of nuclear spin of the ^{192}Os and ^{16}O isotopes and its large molecular weight. The spectrum of this molecule is free from the hyperfine structure and the resonance splitting due to the recoil effect as well as the displacement of its frequency due to quadratic Doppler effect are negligibly small (5×10^{-13} and 0.36×10^{-15} deg^{-1}, respectively). In addition, as shown in Refs. 4 and 5, good coincidence of the frequency of the nonlinear resonances of $^{192}OsO_4$ with the center of the amplification line is characteristic, as are the small values of the broadening and displacement of resonance frequencies due to pressure. Because of this the CO_2 : $^{192}OsO_4$ quantum standard is a promising candidate for the secondary frequency standard in the intermediate IR range with the frequency reproduction accuracy of $\sim10^{-13}$–10^{-14}.

3. An experiment to synthesize the frequency of the CO_2 : $^{192}OsO_4$ laser was carried out by using a method proposed in Ref. 3, whereby the laser frequency was compared to the sum frequency of the eighth harmonic of a D_2O laser frequency and the 7.5-GHz klystron frequency. Both the klystron and D_2O laser frequencies were synchronized with respect to a cesium frequency standard through an intermediate HCN laser and a set of SHF sources by using a phase frequency self-tuning system.

The CO_2:$^{192}OsO_4$ laser had the following characteristics: the power output (without beam broadening) was directed to the nonlinear external absorption cell $\sim$100 mW, the cell length was 2.5 m, and the OsO_4 vapor pressure in the cell was $\sim$2.5 Pa. The width at half height of the $^{192}OsO_4$ nonlinar resonance at the $P(14)$ line frequency was approximately 200 kHz and the relative contrast was 2%. The CO_2 laser frequency stabilization network was the static, first-order regulation loop with the attenuation frequency of approximately 100 Hz. The long-term frequency stability of the CO_2 : $^{192}OsO_4$ laser over a period of several days was $\leqslant3\times10^{-11}$ and was basically determined by imperfections of the design model, by the presence in the beam of parasitic amplitude modulation, and by the insufficiently effective optical decoupling between the laser and the external cell.

The power output of the auxiliary CO_2 heterodyne-laser which was phase-coupled (at a frequency difference of $\sim$4 MHz) to the CO_2:$^{192}OsO_4$ laser was 0.5 W, the power output of the D_2O laser was $\sim$40 mW, and that of the klystron was $\sim$10 mW.

The outputs of all three sources were mixed in a point tungsten-nickel diode. Under these conditions, the rf beat signal in the 7-MHz band exceeded the noise by 20 dB (in the 50-kHz band).

The frequency measurements of the CO_2 : $^{192}OsO_4$ laser were carried out on different days during a two-week period. The results of processing of four series of measurements, each lasting nearly three hours and consisting of 20 measurements, showed that the frequency of the CO_2 : $^{192}OsO_4$ standard [the $P(14)$ line at $\lambda = 10.53\ \mu$m] in units of the atomic time scale is

$$\nu = 28\ 464\ 676\ 938{,}5 \pm 1{,}0\ \text{kHz}.$$

4. Thus, the absolute frequency of the CO_2:$^{192}OsO_4$ quantum standard was measured for the first time with the relative error of 3×10^{-11}. The accuracy of measurement was limited principally by the frequency instability of the CO_2:$^{192}OsO_4$ laser; this accuracy can be improved by two orders of magnitude by using the known methods.[4,5] This will permit measurement of the absolute frequencies of the lasers operating at shorter wave lengths with practically standard accuracy. Determination of the absolute frequencies of CO_2:$^{192}OsO_4$ also permits one to use it subsequently as a frequency standard which can be used to measure laser frequencies in the IR and optical ranges without the use of the submillimeter lasers for continuous synchronous control of its frequency according to the standard.

The precision measurements of the absolute frequency of the CO_2:$^{192}OsO_4$ standard make it possible to compare the frequencies of the quantum transitions of atoms and molecules, which are determined by different fundamental constants,[6,7] in order to verify the hypothesis[8] about the time variation of the physical constants (the expected relative annual variation of these constants is approximately 7×10^{-11}). As is well known,[9] the transition frequency of the atomic sublevels of the hyperfine structure, which depends on the magnetic moment, of the nucleus is expressed in terms of such a set of constants

$$\omega_{\text{hyperfine structure}} \sim c\, g_I\, \alpha^2 R \frac{m_e}{m_p},$$

where g_I is the gyromagnetic proton ratio, α is the hyperfine-structure constant, R is the Rydberg constant, m_e and m_p are the masses of the electron and the proton, respectively, and c is the speed of light. On the other hand, the frequency of the vibrational transition of the polyatomic molecule is[10]

$$\omega_{\text{vib.}} \sim \sqrt{k/M},$$

where M is the reduced mass of the atoms, which are responsible for the given coupling and k is the quasi-elastic constant. The latter can be expressed in terms of the well-known constants: $kx^2 \sim E_e\sqrt{(m_e/m_p)}$, where x is the amplitude of a normal vibration ($x \sim a_0$ is the Bohr radius) and E_e is the electron energy ($E_e \sim R$). As a result, the frequency ratio of the hyperfine and vibrational transitions is proportional to

$$\frac{\omega_{\substack{\text{hyperfine}\\\text{structure}}}}{\omega_{\text{vib}}} \sim \frac{g_I e^4}{(c\hbar)^{3/2}}\left(\frac{m_e}{m_p}\right)^{1/4} = g_I\, e\, \alpha^{3/2}\left(\frac{m_e}{m_p}\right)^{1/4},$$

where e is the electron charge and $\hbar$ is Planck's constant. This shows that by using the atomic-time standard and matching generator frequencies, we can obtain information on the time variation of the obtained set of constants from periodic measurements of the absolute frequency of the standard operating at the vibrational-rotational transition of the molecule.

Although this hypothesis about the instability of the physical constants contradicts the general and special theories of relativity[8] and it encounters a number of other serious objections,[11] the final solution of the problem can be obtained experimentally. The absolute measurements of the frequency of the CO_2:$^{192}OsO_4$ laser constitute the beginning of such an experiment.

[1]K.M. Evenson and F.R. Peterson, Laser Spectroscopy of Atoms and Molecules, G. Walter Ed., "Problems of Applied Physics" series, Russ. translation M., Mir, 1979.
[2]K.M. Evenson, D.A. Jennings, F.R. Peterson, and J.S. Wells, Laser Spectroscopy III (Proc. Third Intern. Conf., Jackson Hole, Wyoming, July 4–8, 1977), J.L. Hall and J.L. Carlsten, Eds., Springer-Verlag Berlin, Heidelberg, New York, 1977, p. 65.
[3]Yu.S. Domnin, V.M. Tatarenkov, and P.S. Shumyatskiĭ, Kvant. Elektron. **2**, 2612 (1975) [Sov. J. Quantum Electron. **5**, 1426 (1975)].
[4]O.N. Kompanets, A.R. Kukudzhanov, V.S. Letokhov, V.G. Minogin, and E.L. Mikhaĭlov, Zh. Eksp. Teor. Fiz. **69**, 32 (1975) [Sov. Phys. JETP **42**, 15 (1975)].
[5]O.N. Kompanets, A.R. Kukudzhanov, and E.L. Mikhaĭlov, Kvant. Elektron. **4**, 2016 (1977) [Sov. J. Quantum Electron. **7**, 1150 (1977).
[6]R.H. Dicke, Science **129**, 621 (1959).
[7]N.G. Basov and V.S. Letokhov, Usp. Fiz. Nauk **96**, 585 (1968) [Sov. Phys. Usp. **11**, 855 (1970)].
[8]P.A.M. Dirac, Proc. Roy. Soc. London **A165**, 199 (1958).
[9]I.I. Sobel'man, Vvedenie v teoriyu atomnykh spektrov [Introduction to the Theory of Atomic Spectra], FML, 1963, p. 261.
[10]M.A. Ek'yashevich, Atomnaya i molekularnaya spektroskopiya [Atomic and Molecular Spectroscopy], FML. 1962, p. 611.
[11]Ya.B. Zel'dovich, Usp. Fiz. Nauk **78**, 549 (1962) [Sov. Phys. Usp. **5**, 931 (1963)].