

IMPACT OF LASERS ON PRIMARY FREQUENCY STANDARDS AND PRECISION SPECTROSCOPY

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Abstract. - Lasers available at new wavelengths, powers, linewidths, and stabilities have made possible advances in precision atomic frequency standards and spectroscopy. Laser spectrometers with resolving power exceeding 10^{11} have been constructed and used to measure the photon recoil structure of spectral lines and in new tests of relativity. Recent progress in frequency stabilization methods and in laser-cooled ions indicates the possibility of an optical frequency standard with an accuracy and stability of less than 10^{-15} . Diode lasers may enable the construction of an optically-pumped cesium standard with an order-of-magnitude improvement in accuracy over existing primary standards.

I. CESIUM PRIMARY STANDARDS RESEARCH

Introduction. - The U.S. primary frequency standard, NBS-6, is a cesium atomic-beam apparatus with an interaction length of 3.74 meters. The experimental uncertainty that limits its accuracy is the microwave cavity phase shift [1]. This shift is cancelled to some degree by averaging the clock rates obtained from the two directions of atomic beam propagation in the standard. However, the degree of cancellation is limited by the faithfulness of the atomic beam retrace, which is affected by factors of beam axial symmetry.

Since the frequency shift produced by a given cavity phase shift is proportional to the resonance linewidth, one way to improve this standard would be to increase the microwave Q. This factor can be increased in a cesium clock only by lengthening the interaction time with the atoms. It is unlikely that a new primary standard could be made physically longer than NBS-6, because magnetic shielding becomes difficult to achieve for such large dimensions. In addition, a suitable means of cooling neutral atoms in an atomic beam has not yet been developed. Therefore, the microwave linewidth in future cesium clocks will probably not decrease, and any improvement in accuracy must come from a direct assault on the sources of error.

In recent years, the development of reliable single-mode diode lasers has made possible new methods that may significantly reduce the errors found in a cesium clock of given length. While much admirable work has been performed in the past using laboratory-quality diode lasers [2], the advent of low-cost, long-life, commercial lasers with excellent quality now makes long-term operation of an optically-pumped laboratory standard feasible.

Cesium Optical Pumping. - The atomic-state-preparation technique of most interest at the National Bureau of Standards (NBS) is one which promises to change the state of nearly every cesium atom within the atomic beam to a single magnetic sublevel [3,4]. Two lasers can be used for this purpose, one tuned to the ($^2S_{1/2}$, $F = 3 \rightarrow ^2P_{3/2}$, $F = 4$) transition, and the other adjusted to the ($^2S_{1/2}$, $F = 4 \rightarrow ^2P_{3/2}$, $F = 4$) line, both at 852 nm wavelength. The latter laser is plane polarized, with the vector of the electric field parallel to a small magnetic field, which establishes a quantization axis. With both lasers crossing the atomic beam, atoms move from each energy level into every other level, with the exception of the ($^2S_{1/2}$, $F = 4$, $M = 0$) ground-state sublevel. In this case, selection rules prohibit excitation of the atom, and eventually nearly all atoms should fall into this substate. A similar arrangement of lasers will allow the pumping of atoms into the $F = 3$, $M = 0$ state.

A number of advantages result from this process. The most obvious is that the signal should increase considerably, since, instead of being rejected by state selection magnets, atoms are converted to the desired state by optical pumping. In the case of NBS-6, which uses both the ($F = 3$, $M = 0$) and ($F = 4$, $M = 0$) substates, the increase in signal is a factor of eight, and the expected increase in signal-to-noise ratio is consequently $\sqrt{8}$.

A more significant benefit of optical pumping is the generation of a highly symmetrical cesium beam, since optical pumping is largely independent of velocity and position. This feature, combined with a new Cs oven design, should permit a much better retrace of the atomic beam upon reversal. Improved microwave cavity design with smaller cavity windows may help reduce the actual size of phase shifts which vary across cavity windows [5] and lower the total uncertainty in frequency due to cavity phase shift by an order of magnitude.

Another uncertainty of unknown magnitude in present Cs standards is that introduced by Majorana transitions as atoms pass from the large (1 T) fields of the state selection magnets to the small (0.1 T) field of the C-field region [6]. Since the C-field can now be extended to include the optical pumping region, such Majorana transitions should not occur. In addition, pumping of the atoms into a single magnetic sublevel will permit study of Majorana transitions, which should appear as changes in Rabi resonances adjacent to the central clock transition.

The present strength of the C-field is mandated by the presence of asymmetry in the size of microwave resonances near the clock transition. As atoms are

pumped into the ($F = 4, M = 0$) level, these auxiliary features will fade, permitting the reduction of the C-field to a smaller, more stable value. Also, the sign of the C-field is irrelevant in this scheme; therefore, the field can be reversed with no anticipated shifts in clock frequency. This check on systematics is not possible with the current system, since improper sign of the C-field would result in Majorana transitions of unknown effect.

Fluorescence Detection. - Complete removal of state-selection magnets is possible if laser methods are employed to detect atoms that have made the microwave transition. Perhaps the simplest way to accomplish this is with fluorescence detection. In the case where atoms are pumped to the $F = 3, M = 0$ level, a "flop-in" interrogation is possible with a laser tuned to the $F = 4 \rightarrow F = 5$ transition. The advantage of using of this particular transition is that atoms excited to the $^2S_{3/2}, F = 5$ level must decay to the $F = 4$ level, where they are repeatedly excited, contributing many photons per atom to the fluorescence signal. It is possible in this manner to obtain 100% total detection efficiency even when the collection efficiency is considerably less than one. By appropriate adjustment of laser frequency and angle of crossing of the atomic beam, different atomic velocity distributions also may be selected using such fluorescence detection.

A device constructed with laser-induced detection and optical-pumping regions could, perhaps, be operated simultaneously with counter-propagating atomic beams. With appropriate arrangement of pumping and detection regions, only atoms which have made a microwave transition will contribute to the fluorescence signal; this allows distinction between the two beam directions. One direction could be used for locking the microwave frequency, and the other direction for locking the cavity phase shift value. This may even permit the use of separate cavities for the two ends of the Ramsey resonance region. Even if difficulties with background levels and collisions prevent implementation of this technique, switching of the two directions should be much faster with the laser methods, since the cesium ovens can be operated continuously.

There are certain disadvantages of the new methods. One, the light shift of the cesium atoms as they pass through the microwave region, due to the fluorescence light coming from the two laser regions, may be as large as $\Delta\nu \sim 10^{-14}$. The magnitude of this effect was first estimated by A. Brillet (Université Paris-Sud). E. Smith (NBS) has also calculated the shift using a tensorial analysis, which accounts for differences in fluorescence polarization. Care must be taken to reduce the intensity of this fluorescence radiation through the use of apertures and by properly spacing the laser regions from the microwave regions.

A second problem is photon-recoil beam deflection [7]. Since slower atoms are deflected more than faster ones, a velocity-dependent spatial distribution may be formed. The value of frequency shifts due to this effect can be reduced below $\sim 10^{-15}$ by symmetric laser illumination of the beam and proper cavity design [4].

Laser Requirements. - The characteristics of diode lasers appropriate for this task include properties essential to attainment of good signal-to-noise ratio and complete optical pumping, as well as properties important for operating considerations. In the latter category, low cost and ready availability are desirable from the standpoint of construction and repair, and lifetime is important when considering maintenance of a continuously operating standard. Diode lasers are now easily available with projected lifetimes in excess of ten years and costs of about \$100 each. Since manufacturers specify lifetime in terms of power output, however, and not spectral properties, independent measurements are in progress at NBS to measure the length of time diode lasers may be locked to an atomic beam resonance line. To be useful, this lifetime will have to be longer than one year.

In order to completely optically pump each atom, the laser spectral density should be greater than about 10^{-9} mW/cm²-Hz for an atomic beam path length of about a millimeter. With output powers of 3 to 10 mW and laser linewidths of 10 to 100 MHz, this requirement is easily met with a properly collimated beam.

It is also necessary, both for pumping and detection, that the laser frequency fluctuations be small. Measured standard deviations per unit frequency interval for typical commercially available devices are less than 10 kHz/ $\sqrt{\text{Hz}}$ for frequencies below 10 kHz [8]. At higher frequencies, the frequency noise decreases, and should not introduce appreciable noise through the pumping process.

Long-term ($10^2 \lesssim \tau \lesssim 10^4$ s) diode laser frequency stability of about 10 kHz has been demonstrated at NBS [3], where one laser was simply locked to a rubidium vapor absorption cell, and light shift of the rubidium ground-state hyperfine splitting was interpreted in terms of laser frequency drift. Although it is probably not needed, greater stability is expected if the lock is made to an atomic beam or a saturated absorption cell.

Finally, laser intensity fluctuations also appear as undesirable noise, especially when a cycling transition is used for detection. If we assume a cesium beam magnitude of 10^{10} atoms/s, fractional laser intensity noise, $\Delta I/I$, per unit frequency interval should be less than $10^{-5}/\sqrt{\text{Hz}}$. Measurements at NBS indicate that some of the diode lasers tested have intensity noise less than $10^{-6}/\sqrt{\text{Hz}}$ [3]. Other authors have measured noise as small as $\sim 10^{-7}/\sqrt{\text{Hz}}$ [9].

Newly developed diode lasers appear to have the characteristics needed for use in laboratory cesium standards. Implementation of these new light sources, with various other modifications of the traditional cesium beam clock, may enable the construction of a cesium primary standard with an order of magnitude improvement in accuracy.

II. PRECISION LASER SPECTROSCOPY AND METROLOGY

Introduction. - Lasers have contributed importantly to atomic and molecular spectroscopic investigations of high precision and resolution, and they promise to play a vital role in the development of future precise frequency standards. The special features of a single-mode laser as a radiation source are due to the spatial and temporal coherence of the electromagnetic waves emitted in macroscopically different parts of the laser. Compared with the light of conventional sources, laser light is characteristically more intense, directional, spectrally pure, and coherent. As a result, the use of lasers has become widespread throughout the many fields of science and dominates modern metrology and spectroscopy. Their spectral purity makes possible resolution of fine details, and the lasers themselves provide several ways to eliminate unwanted broadening of spectral lines. Lasers can be used to detect, measure, cool, and study small numbers of atoms and molecules (even one!). Complicated spectra can be simplified by selective laser excitation and induced emission. Transient and interference methods can reveal relaxation rates, collision rates, and splitting of energy levels. For many years, these methods were limited by the natural coincidence of emission and absorption frequencies of the laser and the atomic systems under study. But the development of dye lasers, color-center lasers, diode lasers, etc., has given spectroscopists and metrologists widely tunable sources of intense, narrowband radiation throughout the optical spectrum. Thus, lasers complement and greatly enhance the sensitivity and application range of classical spectroscopic methods, such as absorption, fluorescence, level crossing and Raman spectroscopy to name a few.

In an article of this size and scope, we will concentrate on but a small part of precision laser physics. We will briefly summarize the development of ultra-stable lasers and some of the methods used to eliminate spectral broadening. Much of the work presented will be a quick review or preview of some of the excellent work to be presented at this conference.

Stable Lasers. - Metrology and spectroscopy were two of the evident applications of the laser from the very beginning. Lasers were recognized as the optical analogs of radio frequency or microwave oscillators, but they did not present an immediate, easy solution to high-resolution optical spectroscopy. An important problem is that the spectral linewidth of lasers frequently limit resolution. The width of the spectral interval within which the laser emission is concentrated is determined by the temporal instabilities of the system as a whole (resonator plus active medium). Until recently, the monochromaticity of the best optical sources was far poorer than the best microwave and γ -ray sources. In recent years, the situation has changed due to remarkable progress in the frequency stabilization of lasers. The spectral width, $\Delta\nu$, of free-running optical

gas lasers is generally many kHz ($\Delta\nu/\nu \gtrsim 10^{-11}$) and for widely tunable dye lasers significantly worse ($\Delta\nu/\nu \gtrsim 10^{-8}$). However, good physical designs and active servo control methods can dramatically minimize the spectral width of these lasers [10-14]. Both gas and dye lasers with an emission linewidth of a few tens of hertz have been constructed [15-17] and have provided the basis for the development of spectrometers with resolving powers exceeding 10^{13} .

Long-term frequency stabilization of lasers has improved to $\lesssim 10^{-13}$ [18,19] due, in large part, to the development of nonlinear sub-Doppler techniques which produce narrow optical resonances in atomic and molecular systems. These resonances give stable and accurate frequency markers with linewidths often limited either by natural lifetime or transit time broadening. Important theoretical and experimental strides toward the reduction of transit (or interaction) time broadening recently have been made [20-24]. This progress presents the realistic possibility of an optical frequency source with stability and accuracy exceeding 10^{-15} [25]. We will return to the subject of narrow lines presently.

Another problem that confronted the pioneers of laser physics was that the early lasers could be tuned only over a very small percentage of their center frequency. While this was not fatal for all precision measurements, nor to optical frequency standards, spectroscopy was limited to those molecular lines that happened to coincide with available laser wavelengths. Now, however, this roadblock essentially has been removed with the development of a number of different kinds of lasers with varying degrees of tunability. Most notably, fluorescent organic dyes [26-28] have become widely used as a laser medium whose emission bands provide a substantial degree of tunability. Other kinds of optical oscillators have also been developed, such as parametric oscillators, semiconductor diode lasers, high-pressure optically-pumped gases, spin-flip Raman lasers, and color-center lasers. Many of these last examples are especially well-suited for tunable sources of coherent infrared radiation.

A final annoyance to those who wish to do highly precise optical frequency metrology/spectroscopy, is the rather poor absolute accuracy limitation ($\sim 10^{-10}$ - 10^{-11}) of optical frequency measurements, while relative precision has now exceeded 10^{-14} . Recent work at the National Bureau of Standards (NBS), Boulder, Colorado and the National Research Council of Canada (NRC) indicates that $\lesssim 10^{-13}$ absolute accuracy is possible [29]. An accurate absolute frequency measurement in the visible (especially a measurement of a frequency-stabilized laser) will likely add impetus to a new definition for either the meter or the speed of light, c [30]. Another consideration is that the optical laser may well provide a frequency standard with improved performance relative to the present Cs frequency standard [18,19]. Practicability of that laser as frequency/time standard is ruled out until a frequency multiplication [31] and/or division [32] scheme connecting optical and microwave domains is successful. Even without absolute accuracy, tunable optical laser spectrometers with high stability and precision provide the capability to do numerous fundamental physical measurements [33].

Line Narrowing Methods. - The resolution of even an ideal spectrometer with infinite resolving power is limited by the width of spectral lines. Frequently, this width is due to the Doppler broadening caused by the random thermal motion of free atoms at low pressure. Whereas atoms in gases can be otherwise relatively unperturbed, their spectral lines are broadened by the Doppler effect because they are moving in all directions with a wide range of thermal velocities. The resulting Doppler broadening blurs important detail of spectral lineshape or structure. Hyperfine and fine structure are often hidden beneath the Doppler-broadened profile, even though each atom still retains its typically much narrower linewidth.

Although some methods to reduce Doppler broadening (e.g., collimated beams, gas cooling, Dicke narrowing) have been known and used in spectroscopy for years, they are not without their problems. A well-collimated beam contains few atoms or molecules. Moreover, to realize the elimination of first-order Doppler broadening provided by the collimated beam, the light collection angle must be equally small, so that only light perpendicular to the atomic beam is collected. Needless to say, signal-to-noise ratio is not large. But, in principle, a laser can overcome all of these difficulties, since it can be bright enough to excite nearly all of the atoms in the beam, or even to excite them many times over. With optical cycling, an easily detectable amount of light can be emitted by a small number of molecules in the beam. In addition, the laser light can be collimated to ensure that its angular spectrum is at least as small as that of the atomic beam. Consequently, a 4π collection angle is possible, permitting high resolution of even weak spectral lines without analyzing the wavelength of the fluorescence. Double-resonance and optical-pumping schemes are also possible. These methods were used at MIT [34] to measure the hyperfine structure of a molecular iodine line with resolution sufficient to measure not only the nuclear quadrupole, but also the magnetic octupole interaction. Recently, Lichten and his collaborators used laser-atomic beam methods to improve the measurement uncertainty in the Rydberg constant to one part in 10^9 [35].

However, it is the high intensity per unit bandwidth, characteristic of lasers, which makes it possible virtually to eliminate the first-order Doppler broadening in gas samples. This is done with either of two nonlinear techniques, saturated absorption or two-photon spectroscopy [36]. In each of these methods, two laser beams are oppositely directed through the same region of the gas. At least one beam is intense enough to importantly alter the properties of the atoms with which it interacts, so that these atoms can be identified by the other beam. This permits the selection of atoms which have zero velocity component along the beam direction.

The power of these techniques and their numerous modifications has opened a plethora of physical experiments, spectroscopic measurements, and metrological studies. Spectrometers with resolution exceeding 10^{-11} were constructed [20,21]

and used to resolve the splitting of spectral lines by photon recoil, which is peculiar to saturated-absorption spectroscopy [15,16]. Tests of special relativity were improved [37]. The Lamb shift of the ground state in hydrogen was measured with an experimental precision of 4×10^{-3} and was found to be in good agreement with theory [38]. Saturation labeling and polarization labeling of complex molecular spectra were developed [39]. The experiments and experimental techniques are far too numerous to list in an article of this length, but there are many excellent reviews and conference proceedings [36,40].

With the high resolution afforded by sub-Doppler spectroscopy and frequency-controlled, actively-stabilized lasers, many researchers deemed it prudent to pursue the resolution limitations imposed by interaction time broadening and second-order Doppler broadening. Interaction time broadening is due to and is inversely proportional to the finite transit time of the atoms through the radiation field. A particularly simple but powerful scheme to reduce transit time broadening greatly and attain spectrally narrow lines is the method of optical Ramsey fringes [20,21]. Basically, the method depends on the initial preparation of a coherent superposition of quantum states by an electromagnetic driving field, followed at some later time by a second (and, for saturated absorption, a third) phase-coherent, field-absorber interaction. The field-phase information embedded into the absorber's memory in the first interaction gives rise to the interference fringes in the second, temporarily separated interaction. These interference fringes give line narrowing with spectral width determined by the longer transit times through the whole system, and not just through an individual beam. Utilizing this method, researchers at NBS achieved full resolution of the recoil splitting of the Ca intercombination line at 657 nm [13] and clearly demonstrated, for the first time, a laser resolving power exceeding 5×10^{-12} . Transit time broadening can also be reduced by longitudinal interaction schemes [Hall, J. L., private communication]. Very benign electromagnetic storage vessels for ions have been developed, which entirely eliminate transit time problems [22,25].

Second-order Doppler broadening can be reduced only by decreasing the speed of the atoms under study or by interacting with a single-vector velocity group (e.g., longitudinal interaction with a well-collimated atomic beam or by pulsed-laser method [15,41]). Decreasing the speed is possible by cryogenic cooling, but many materials condense if they are cooled much below room temperature. Cooling to very low temperatures without condensation can be done by radiation pressure from a suitable laser [23]. In this method, atoms lose momentum and slow down by scattering photons from a light beam coming in the opposite direction. In this way, the radiation pressure of the light reduces gas temperature. The exciting experiments, spectroscopy, and numerous developments and advantages of laser-cooled, trapped ions will be fully treated with other papers in this conference. The prognosis is excellent for optical and microwave frequency standards with

accuracy and stability better than 10^{-15} . Thus, it is now possible to do precision spectroscopy and metrology that are limited in resolution only by the natural linewidth of the quantum system(s) under investigation [42] or by the fundamental linewidth of the probe.

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"The metre is the length equal to the distance travelled in a time interval of 1/299 792 458 of a second by plane electromagnetic waves in a vacuum."

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