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Quantum-mechanical theory of optical coherence

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Laser-based precision spectroscopy and optical frequency comb techniques

General introduction

Light provides the most pertinent example of the dual nature of quantum objects; its oscillatory properties served to verify the electromagnetic theory of Maxwell, and its lumpiness, the photons, signalled the dawn of modern quantum theory.

The electromagnetic phenomena form an integral part of modern technology. They are at work in all electrical motors, and our communication devices utilize their oscillatory behaviour in essential ways. Our radio receivers and mobile phones are all based on the ability of the radiation to sustain well-defined frequency and phase properties.

On the other hand, each device detecting the radiation must be based on the absorption of radiation energy into the material medium. This energy is known to occur in packets, which are now called photons. Absorption of a photon will cause the creation of an excitation which may be amplified and detected. Since the work of Einstein in 1905, we know that the absorption of a quantum of radiation gives rise to one and only one photoemission electron from a solid (Nobel Prize, 1921). Thus the detector counts photoelectrons and not photons, and our information about the behaviour of photons is always indirect. In the process of observation, the photon must be absorbed, and thus it is no longer available afterwards.

The dualism between the two pictures may appear contradictory. They do, however, form the prime example of what is termed *complementarity* in quantum theory, namely the possibility to display either wave or particle properties; albeit they emerge in mutually exclusive limits. From a fundamental point of view, we need to reconcile the two descriptions. We must know how the seemingly smooth oscillatory behaviour of the radiation can manifest itself through the lumpy quantum nature of the field. We thus need both a macroscopic theory to account for the phase properties and a microscopic theory to account for the interaction between the photons and the material absorbing them. The former is given by Maxwell's theory and the latter by quantum electrodynamics.

This year's Nobel Prize in Physics falls in the realm of these aspects of light:

The first part goes to **Roy J. Glauber**, who showed how the quantum theory has to be formulated in order to describe the detection process. This also served to bring out the distinction between the behaviour of thermal light sources and presently common coherent sources such as lasers and quantum amplifiers. This theory uses the formalism of quantum electrodynamics to describe the absorption of a photon in a detector. By correlating

several such detectors, one may obtain higher order correlations, which can display clearly the characteristic features of quantum radiation.

The second part goes jointly to **John L. Hall** and **Theodor W. Hänsch** for their contributions to the development of laser-based precision spectroscopy, including the so-called optical frequency comb technique. These methods make it possible to determine the quantum structure of matter with ever-increasing accuracy and to test fundamental theories, and they also have important applications. Also in precision measurements, quantum effects manifested as the quantum noise, set an absolute limit to the performance of the set-up. In this way the latter field of recognition connects to the former one.

Quantum-mechanical theory of optical coherence

Historical background

The dawn of quantum theory

Towards the end of the nineteenth century, spectral observations became accurate enough to conclude that the emission of a so-called black body was not in agreement with prevailing theories. An almost closed resonant cavity was the theoretical model of such a system, and under the name "Hohlraumstrahlung" its spectral properties were well known. The problem was that applying this knowledge led to expressions that failed to agree with the experimental data.

At this point Max Planck made his seminal contribution (Nobel Prize, 1918); he applied his profound knowledge of thermal entropy to make an *ad hoc* modification of the standard phenomenological approach. As a result, he was able to fit the experiments amazingly well. His result [1], obtained in October 1900, did not explain the physics behind the successful result. Planck, however, proceeded to consider the exchange of energy between the light and the material radiators constituting the thermal environment for the field. He assumed this to take place in finite units, energy quanta. By applying Boltzmann's probabilistic approach to entropy, and "after a few weeks of the most strenuous work of my life", Planck managed to derive his formula [2].

Planck, however, assumed that the energy quantization related to the oscillating radiators surrounding the cavity field. Einstein turned the tables; he realized that the algebraic form of Planck's theory allowed an interpretation in terms of radiation lumpiness, i.e. the radiation was to be considered as consisting of particles, which were termed photons by G.N. Lewis as late as 1926. Einstein applied this hypothesis to a variety of physical phenomena [3]. One of these was the photoelectric effect, which rendered him the Nobel Prize of 1921. In contrast to a common misconception, there were no accurate data on photoemission of electrons at the time of Einstein's publication. Such results were provided after his work by several investigators, culminating in the convincing demonstrations by R.A. Millikan, which were quoted in the citation for his Nobel Prize in 1923.

It thus seemed to be verified that light appeared in energy lumps, which could only be absorbed and emitted in integer units. This picture was extended and elaborated in theoretical works, and it became the basis for the future developments. N. Bohr utilized it in his theory of the hydrogen spectrum in 1913 (Nobel Prize, 1922). Quantization of light was now well established. However, classical Maxwell theory had become the successful basis for electrical engineering. This theory describes the radiation in terms of continuous distributions of energy and well-established phases. The two pictures appeared to be contradictory, which was clearly recognized by Einstein [4]:

"These properties of the elementary processes required by equation (12) [in Ref. [4]] make it seem practically unavoidable that one must construct an essentially quantum theoretical theory of radiation. The weakness of the theory lies, on the one hand, in the fact that it

does not bring any nearer the connection with the wave theory and, on the other hand, in the fact that it leaves moment and direction of the elementary process to “chance”; all the same, I have complete confidence in the reliability of the method used here.”

The emergence of field quantization

Once quantum theory had been developed by Heisenberg, Schrödinger and others, it was obvious that electromagnetic fields should be quantized. It was known how to map the electromagnetic theory onto a set of harmonic oscillators, so the procedure seemed obvious. Dirac [5] was the first to consider this approach in detail. He concludes:

“There is thus a complete harmony between the wave and light-quantum description of the interaction. We shall actually build up the theory from the light-quantum point of view, and show that the Hamiltonian transforms naturally into a form which resembles that for waves.”

With his usual theoretical dexterity Dirac succeeded in obtaining the expression for the rate of spontaneous emission, a characteristic quantum effect. The theory of quantized electromagnetic fields was then developed further by P. Jordan and collaborators, W. Pauli, L.D. Landau and R. Peierls. There were, however, fundamental difficulties that prevented successful applications of the theory.

One problem was inherited from classical physics; the electromagnetic field dragged along with the charge of a moving electron turns out to give an infinite mass to the electron. This is just the first of a series of infinities affecting the straightforward application of quantum theory to fields. This problem was solved only after the Second World War by S. Tomonaga, J. Schwinger and R.P. Feynman (Nobel Prize, 1965). Their *renormalization* program has evolved successfully since, and it now forms the basis for all modern approaches to quantized fields.

Quantum considerations enter optics

When the tools to handle quantum electrodynamics were known, they were applied mainly to high-energy processes. This derives partly from the rapid development of collision experiments and partly from the fact that the requirement of relativistic invariance played a central role in the creation of the theory. It was still naively assumed that the conflict between Maxwell's and Planck's treatments would be of no significance in optical observations. But this state of blissful indifference was not to last.

In 1954-56, R. Hanbury Brown and R.Q. Twiss investigated an interferometric method to determine the angular extension of distant stellar objects, and also made laboratory measurements [6]. They found that the intensity-intensity correlation between photocurrents recorded in two separated detectors displayed a bump when the difference in optical path lengths between the signals was zero. In fact, the correlation function $\langle I(x)I(y) \rangle$, at $x=y$ was found to take twice its value compared with that for widely separated arguments x and y . The authors took this to be a consequence of quantum theory: “The experiment shows beyond question that the photons in the two coherent beams of light are correlated, and that this correlation is preserved in the process of photoelectric emission.” The individual photon had entered the realm of observational optics.

In a paper from 1956, E.M. Purcell [7] indicates that the effect may have a classical interpretation, but he still assumes that it is basically a vindication of the quantum features of light. These arguments constituted the starting point for an intense interest in the relation of quantum considerations to optical observations. This became manifestly obvious when the invention of the laser in 1960 promised the possibility to provide light sources widely different from the conventional thermal ones. Two points of view emerged:

On the one hand, the quantum transition was known to proceed proportional to $(n+1)$, where n is the photon number in the field. In the Hanbury Brown & Twiss phenomenon, the induced photons are few, and this was assumed to account for the factor of two; one photon may induce another one. In a laser, many photons contribute and one may predict a giant effect. On the other hand, there lingered an impression that quantum noise only supplied ripples on the field amplitudes of the classical fields. Thus random function theory would account for the observed effects. As an example we quote [8]: "In the conditions under which light fluctuations are usually measured by photoelectric detectors, the semi-classical treatment applies as readily to light of non-thermal origin as to thermal light, and to non-stationary as well as to stationary fields." The subsequent experimental progress rapidly proved the short-comings of the semi-classical approach. The correct theory was published by Roy Glauber in 1963 [9], and this has been the basis for all subsequent theoretical considerations.

Quantum theory of optical interference experiments Glauber's 1963 contribution

In Ref. [9], Glauber presents the basic features of his quantum theory of optical coherence. The formal features were expanded on in two long articles [10] in the same year. This material was to form the basis for the development of Quantum Optics up to the present time.

In the 1963 publication, Glauber made the following points:

Detection in photon correlation experiments must be based on a consistent application of quantum electrodynamics. Thus all multi-photon experiments must be based on the fact that, once a photon has been absorbed, the state of the field has been changed so that the next absorption event occurs against a different initial state than the previous one. In particular, a state with only n photons, can only have correlations up to n :th order. This implies the use of normally ordered expectation values for the optical detection processes. As the consecutive absorption processes are based on different states of the field, its state ought to be characterized by correlations to all possible orders, and the description in terms of classical noise is not sufficient. In particular, experiments like those by Hanbury Brown & Twiss are described by a consistent calculation of two-photon interference effects. Their factor of two derives simply from the property of Gaussian fluctuations to give

$$\begin{aligned} \langle I(x)I(y) \rangle &\propto \\ \langle a^+(y) a^+(x)a(x)a(y) \rangle &= \langle a^+(y) a(y) \rangle \langle a^+(x)a(x) \rangle + \langle a^+(y) a(x) \rangle \langle a^+(x)a(y) \rangle, \end{aligned}$$

which explains the factor of two when $x=y$.

In interference experiments, the phase of the light is important, and then the state is best represented in terms of *coherent states*, and defining a distribution function on these, Glauber introduced the concept of a quasidistribution into Quantum Optics. These are quantum descriptions of the state, which have straightforward relations to classical phase space distributions. Glauber shows that in certain cases they can be given by a diagonal representation in the coherent states. They do, however, display clearly non-classical features; thus, for example, for some simple quantum states they do not satisfy the positivity of a probability distribution. If the distribution is positive, we can give the state a classical interpretation. Glauber shows that the thermal light sources correspond to a Gaussian distribution, thus justifying, in this case, the use of fluctuation theory. The case of an ideal laser source shows no correlations of the Hanbury Brown & Twiss type.

The papers carry out an analysis of the formalism for photon detection based on normally ordered correlation functions, which are conveniently described by the diagonal representation now called the P-function or the Glauber-Sudarshan representation. Glauber points out that the photon absorption statistics from a laser cannot be described by any simple stochastic behaviour, Gaussian or Poissonian, but require a detailed knowledge of the quantum state of the device. This observation formed the basis for much subsequent work on formulating a quantum theory of lasers, parametric amplifiers and photon correlation experiments.

The coherent states were known from harmonic oscillator physics, but Glauber introduced them as basic entities to describe optical fields. They are eminently suitable for this, because like classical signals they possess both amplitude and phase. However, being an exact quantum description, they may be applied down to the intensity level where the quantum granularity of light influences the observations. At the same time, they provide a convenient tool to extract the classical limit so useful in the applications of optical signals to communication and high precision measurements. The classical description emerges, but the fundamental quantum fluctuations are still present, setting the ultimate limit to what accuracy is attainable in principle.

The mathematical formalism of quantized fields was developed in parallel with Glauber's work on their applications. E.C.G. Sudarshan [11] drew attention to the use of coherent state representations for the approach to classical physics; at this point he refers to Glauber's work. Together with J.R. Klauder he proceeded to develop the mathematical formalism of Quantum Optics; their approach is presented in their textbook [12]. After the initial contributions, many authors applied Glauber's results to the rapidly evolving experimental situation in optical physics, thus creating the field today called "Quantum Optics".

The present status of Quantum Optics

Quantum Optics developed into a multifarious and challenging field of research. The experiments brought measurements down to the level of single photons in the field and a few atoms, also allowing devices of ultimate stability. Thus the special quantum features of the theory are of the utmost importance.

Glauber himself summarizes his theory and its applications in the lectures [13]. A generation of theoreticians have utilized and developed these results; among them are D.F. Walls [14] and M.O. Scully [15], who have laid a solid foundation for the experimental activities. L. Mandel has used the theory to design many ingenious experiments illuminating the quantum nature of light signals [16]; his student H.J. Kimble has continued to push the field in new directions.

Technical developments in the field of Quantum Optics have made it necessary to consider the quantum character of the light signals:

It has become possible to create squeezed states. These have quantum fluctuations anisotropic in the phase, and one of the quadratures is less uncertain than the coherent states. In principle, such states allow the minimization of the quantum noise effects on ultra high precision measurements.

One can also observe the effect opposite to that of Hanbury Brown & Twiss, namely antibunching. In this case, the photons occur less bunched than in the totally random fashion of a Poisson distribution. For photon statistics, this is a pure quantum phenomenon.

In the limit of low intensity, only a few photons are involved, and this can be applied in secure quantum communications, the topical field of quantum computing and the recording of ultra-weak signals in high-precision experiments. In all these situations, a good understanding of the basic theory is required, as quantum effects set the fundamental limit to what can be achieved; technical noise can be eliminated, quantum noise cannot.

Another field of applications of the quantum approach to optics is offered by the possibilities to test fundamental aspects of quantum theory. In spite of the success quantum theory enjoys in applications, the interpretation of the theory has not reached any consensus. Thus we still need to push the experiments further and further into the quantum regime in the hope of gaining new insights into the workings of the formalism.

On the other hand, the coherent state representation offers a tool to carry the quantum theory over into the classical regime. Here the amplitude and phase of the field become well-determined variables, and they can be used for communication and ultra-high precision measurements. Choosing the parameters of the experiment propitiously, one may neglect the underlying quantum fluctuations and regard the signals as well-defined classical field amplitudes.

Laser-based precision spectroscopy and optical frequency comb techniques

The need for precision measurements of atomic structures and optical frequencies

The history of science tells us that many advances have been spurred by measurements of unprecedented precision revealing new structures and phenomena. This is particularly true in atomic spectroscopy, where increasing spectral resolution led to the observation of atomic fine structure (due to the electronic spin), hyperfine structure (due to the nuclear spin), and volume isotopic shifts (due to the different charge distributions of the nuclei of isotopic species of an element). External magnetic and electric fields give rise to Zeeman and Stark energy level structures. More subtle, quantum electrodynamic effects give rise to the Lamb shift. A number of Nobel Prizes in physics have been awarded for the study of atomic structures and their interpretations. By pushing to ever higher precision and resolution, we are likely to detect new phenomena. Ultimately, a precision approaching 1 part in 10^{18} might be achievable. At very high precision, questions about the constancy of optical transition frequencies over time can be asked, an aspect related to the constancy of the fundamental constants themselves. Possible asymmetries between matter and anti-matter may also be revealed. The possibility to determine optical transition frequencies very accurately is also closely related to obtaining better atomic clocks. This in turn will allow better GPS systems, better space navigation and improved control of astronomical telescope arrays. The Nobel Prize to J.L. Hall and T.W. Hänsch this year is based on these developments.

Historical background

There are many technical features in the spectrometers limiting the resolution achievable. However, there are also more basic limits to the resolution; for example, the movements of the atoms give rise to a Doppler broadening, which is proportional to the transition frequency. In the visible region, corresponding to a transition frequency of about 10^{15} Hz, this broadening is about 1 GHz. However it becomes negligible for transitions in the radiofrequency or microwave regions. More fundamentally, the finite lifetime of an excited state gives rise to a natural linewidth for the transition connecting it to the ground state. This latter limitation to spectral resolution is related to the limited time available for

the measurement. The natural linewidth is in the MHz range for an excited lifetime in the 100 nanosecond range, and exceedingly small when the level is metastable.

In the quest to achieve a high spectral resolution, various methods have been developed. By making measurements of the internal level splittings in a long-lived atomic state it is possible to benefit from the natural linewidth being small. This is used in the atomic beam magnetic resonance technique (Nobel Prize to I.I. Rabi, 1944), where the transition frequency between the two hyperfine structure levels in ^{133}Cs at 9.2 GHz can now be determined with almost 15 digits precision, forming the fundament for the current definition of the second. This precision can be attained only by increasing the interaction time of the moving particles, using so-called Ramsey fringe techniques (Nobel Prize to N.F. Ramsey, 1989). Optical resonance techniques (Nobel Prize to A. Kastler, 1966) also provide a resolution down to the Heisenberg limit in optical pumping (ground state) or optical double resonance experiments (excited states). The fact that the Doppler broadening is normally very detrimental in the visible regime can be eliminated by using special techniques such as saturation or two-photon spectroscopy. The Nobel Prize in 1981 was awarded to N. Bloembergen and A.L. Schawlow for their contributions to the development of laser spectroscopy. A drastic approach towards the solution of the Doppler problem is to reduce the velocities, which is achievable with laser cooling techniques. The forces exerted by light can also be used to bring about spatial confinement of the atoms (Nobel Prize to S. Chu, C. Cohen-Tannoudji and W.D. Phillips, 1997). Ions can be trapped in electromagnetic field arrangements (Nobel Prize to H.G. Dehmelt and W. Paul, 1989). A combination of cooling and trapping with evaporative cooling ultimately made Bose-Einstein condensation possible, creating coherent matter with the particles brought almost to a complete stand still (Nobel Prize to E.A. Cornell, W. Ketterle and C.E. Wieman, 2001).

Laser-based high-precision spectroscopy

The development of masers and lasers, rewarded with the Nobel Prize in 1964 to Ch.H. Townes, N.G. Basov and A.M. Prokhorov, has provided high-frequency oscillators with a very narrow bandwidth when continuous-wave, single-mode operation is achieved. Owing to the action of the laser resonator, a line-width much narrower than the width of the transition can be achieved. The line-width and stability of the output will largely be limited by mechanical vibrations, acoustic noise etc. Such effects can be strongly reduced by locking the laser frequency to sharp interference fringes of specially designed, high-finesse passive interferometers using electronic feed-back. A stability below the one Hz level can be achieved. Such lasers can then also be locked to sharp atomic and molecular transitions. J.L. Hall and collaborators (at JILA, Boulder; operated by NIST, National Institute of Standards and Technology, and University of Colorado) have developed the powerful frequency stabilization schemes allowing fundamental measurements to be made.

Atomic hydrogen is arguably the most fundamental atomic system one can study, allowing accurate theoretical calculations to confront precise experimental data. As early as 1972 T.W. Hänsch together with A.L. Schawlow described a first laser spectroscopy measurement of this kind on hydrogen, in which a narrow-band tuneable dye laser, developed by him, could be used to resolve the Lamb shift in an excited atomic state. Hänsch has then, together with his students, pushed hydrogen spectroscopy towards its limits in a sequence of papers measuring the 1s-2s optical frequency, the Rydberg constant and Lamb shifts, first at Stanford University and after 1986 at MPQ in Garching and the Ludwig-Maximilians Universität, München. The 1s-2s transition is of particular interest because the long lifetime of the upper state allows extremely narrow lines.

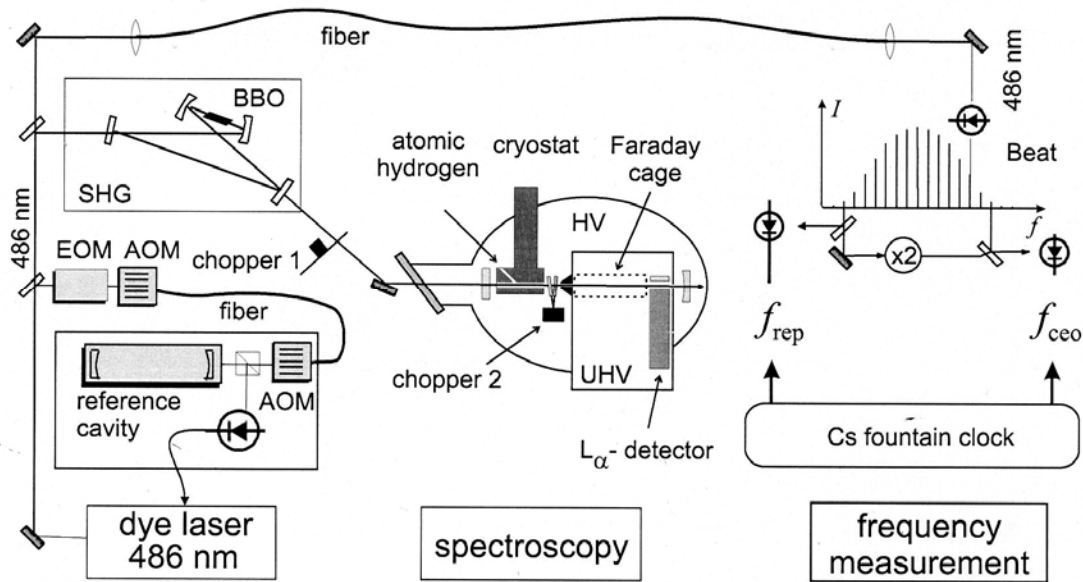


Fig. 1. Set-up for precision determination of the 1s-2s transition in atomic hydrogen. A stabilized narrow-band laser at 486 nm is frequency doubled to 243 nm to induce the Doppler-free two-photon transition in cryogenically cooled hydrogen. Longer interaction times are achieved by pulsing the hydrogen beam and restricting the detection to the slow tail using delayed irradiation. The frequency of the blue laser is measured in terms of the cesium atomic clock using an optical frequency comb generator (From Ref. [17]).

Doppler-free two-photon absorption spectroscopy, a method suggested by V. P. Chebotayev and co-workers in 1970, using two photons at about 243 nm, is employed in a set-up of the kind shown in Fig. 1.

This intrinsically precise transition can be taken advantage of only when the laser output frequency is stabilized according to the principles developed by J.L. Hall [18,19]. Ultrastable resonance cavities, sometimes suspended in vacuum and at highly temperature-stabilized conditions, are utilized. The techniques also include active devices using acousto-optic frequency shifters and electro-optic phase modulators as indicated in Fig. 1. A crucial step was actually taken in 1984 in a joint paper by Hall and Hänsch [20].

The 1s-2s interval is now determined to be 2,466,061,413,187,103 (46) Hz, while the Rydberg constant has the value $109,737.31568525$ (73) cm^{-1} . Further groups, for instance in Paris and Oxford, have contributed to the precision spectroscopy of hydrogen, but Hänsch's group has remained the leading one.

Hall has used highly stabilized lasers for fundamental measurements including the Michelson-Morley [21,22] and the Kennedy-Thorndyke [23] experiments with confidence levels highly improved over earlier work.

Through the work of Hall and Hänsch, the precision of optical laser spectroscopy now is similar to that of microwave atomic clocks at the 10^{-15} level. While further improvements of the latter ones might be limited, optical clocks are likely soon to widely outperform the microwave techniques. These trends are shown in Fig. 2.

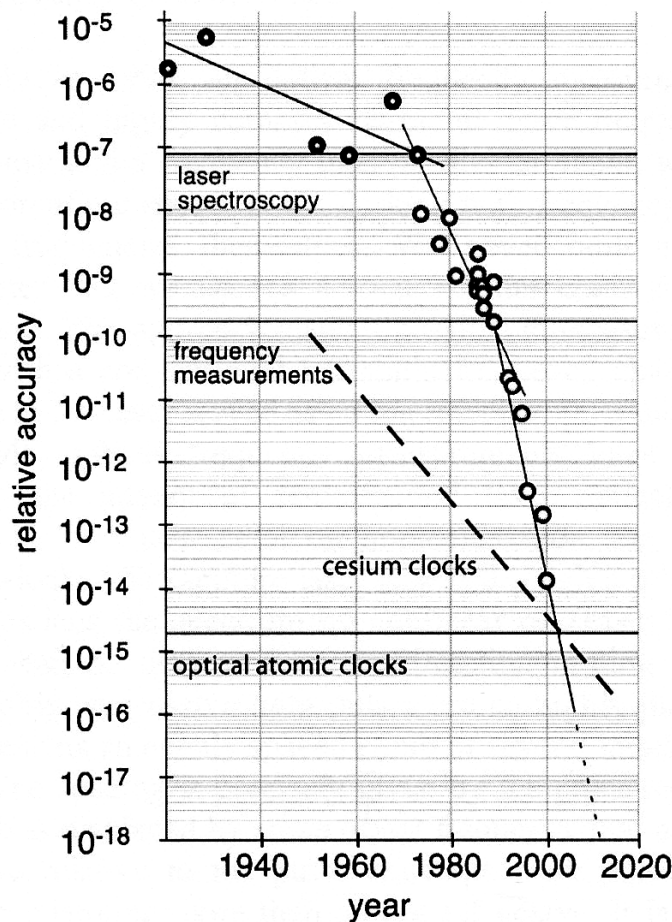


Fig. 2. Developments in relative accuracy in precision spectroscopy. The level 10^{-15} has been achieved in optical as well as microwave based systems, where the former now are taking the lead (From Ref. [24]).

The definitions of the units for length and time have undergone a continuous development, once having been coupled to the size of the earth and its motion. Since 1889 the *General Conference on Weights and Measures* is the authority dealing with physical unit matters. In 1960 the metre was defined as a certain number of wavelengths of a spectral line in ^{86}Kr , and in 1967 the second obtained its present definition as being 9,192,631,770 oscillations of the radiation inducing the hyperfine transition in ^{133}Cs mentioned above. With improved measurement methods, the velocity of light could now be determined with even better precision by multiplication of frequency and wavelength for a stable radiation source. The meter definition, unfortunately being based on a slightly asymmetric spectral line, quickly became the limitation. In a process, in which J.L. Hall [25,26] and many others were greatly involved, the metre was redefined through coupling it to the second when one in 1983 stated, that the velocity of light in vacuum is 299,792,458 m/s, in accordance with the best measurements, but now with the uncertainty zero! This meant that 1 metre is the distance light travels in $1/299,792,458$ s.

In order to find the wavelength of a frequency stabilized laser source expressed in the metre unit, its frequency should thus be measured and the defined value of the velocity of light should then be divided with the measurement result. To determine an optical frequency around 10^{15} Hz by relating it to the hyperfine frequency in Cs around 10^{10} Hz turned out to be very complicated. Long chains of highly stabilized and phase-locked lasers, which were frequency multiplied and combined with stable microwave sources, were developed at few highly specialized laboratories in the world and only a small number of optical transitions were determined. The new definition of the meter became

unusable for most practical purposes. A new way to accurately measure optical frequencies became very urgent.

The development of optical frequency comb techniques

Optical frequency comb techniques, constituting a very important development in high-precision metrology, solved the mounting problems in an ingenious way. The new measurement methods are based on fundamental relations between cavity modes in a continuously operating laser and their interference, which leads to a repetitive train of short pulses. However, a prerequisite is that the cavity modes, spaced by

$$\Delta\nu = c/2L,$$

(c is the velocity of light, L the length of the resonator) have a constant internal phase relation, since otherwise the interference becomes random. The more modes are locked together, the shorter the pulses become. This reflects the properties of the Fourier transform; a 5 fs pulse requires the locking of about 10^6 modes, covering a large fraction of the visible region. Thus, only laser media with a broad gain profile (dyes, titanium-doped sapphire, etc.) can be used for femtosecond pulse generation. At mode-locking, a small "ball" of light bouncing between the mirrors is created by interference inside the cavity, and part of the light is coupled out as an external beam with the pulse separation equalling the cavity round trip time

$$T = 2L/c .$$

For a 1 metre long resonator the pulse separation is about 7 ns. The early development was related to a description of an improved frequency resolution through repeated interaction with an electromagnetic field, as is the case for the Ramsey fringe technique. M.M. Salour in a paper together with C. Cohen-Tannoudji demonstrated the effect for double pulse excitation [27]. Hänsch simultaneously demonstrated the case of multiple pulse interaction and a corresponding increased resolution [28]. Hänsch soon extended the concept to an infinite pulse train from a mode-locked laser in studies of the 3s-4d transition in sodium. He realized that it was more fruitful to consider the phenomena observed as the atomic interaction with the continuous sharp laser modes in the frequency domain rather than using the multiple Ramsey fringe language. Hänsch's paper with Eckstein and Ferguson [29] was thus the starting point in the development of frequency comb techniques. In published conference presentations, Hänsch developed these concepts as early as 1976 and 1977. Similar considerations of high-resolution spectroscopy using mode-locked lasers were made by Baklanov and Chebotayev [30], also related to increasing the intensity of the 1s-2s two-photon transition in hydrogen. Chebotayev and collaborators in 1991 discuss an early frequency comb technique [31]. However, due to the early death of Chebotayev in 1992 the influence of these activities on the practical realization of optical frequency comb techniques became limited.

Around 1990 Hänsch, with H.R. Telle and D. Meschede, realized a frequency chain comprising visible or near-infrared laser oscillators only [32]. This was an important step towards facilitating the task of measuring optical frequencies. At the end of the 1990s Hänsch and collaborators started to use the frequency comb structure of the mode-locked titanium sapphire lasers to bridge large frequency intervals in a simplified frequency chain to relate optical frequencies ultimately to the cesium clock.

The frequency f_n of a particular mode can be expressed as an integer multiple of the mode separation (i.e. the pulse repetition rate $f_{\text{rep}}=1/T$) plus a carrier envelope offset frequency f_{CEO} :

$$f_n = n f_{\text{rep}} + f_{\text{CEO}}, \text{ where}$$

$$f_{\text{CEO}} = (\Delta\phi/2\pi) f_{\text{rep}} \text{ (Carrier envelope offset frequency).}$$

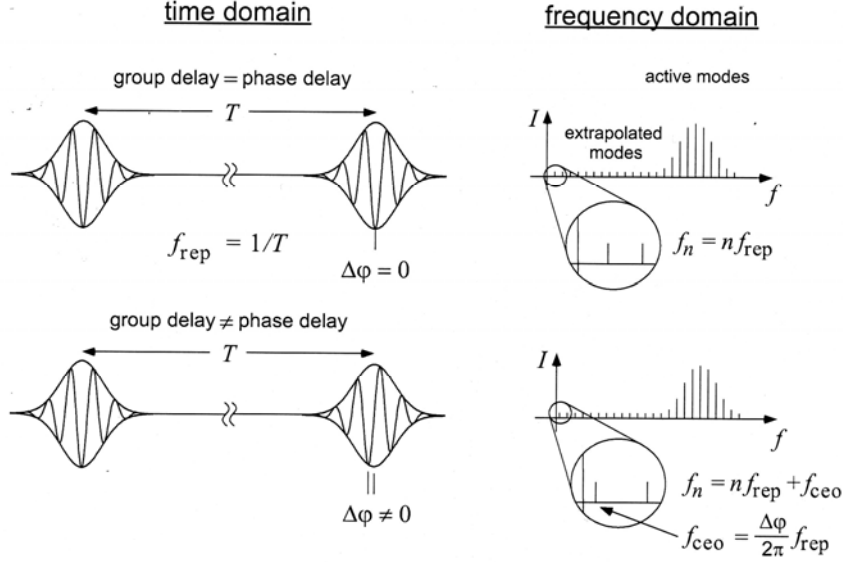


Fig. 3. Time and frequency representations of femtosecond radiation. In the general case the electrical field of the laser light moves under the pulse envelope. The frequency comb can be extrapolated down to frequency 0, and then there is generally an off-set f_{CEO} , which must be determined (From Ref. [17]).

This is illustrated in Fig. 3. While the envelope functions of the pulses are separated by T , the electric field in the pulses does not necessarily have the same periodicity; this results in the carrier envelope offset frequency, which must be determined. In this procedure the phase can also be stabilized. Both f_{rep} and f_{CEO} can be phase-locked to the cesium clock. An arbitrary optical laser frequency f_{opt} can now be determined by measuring the beat frequency f_{beat} between the laser and a close-lying mode in the comb according to

$$f_{\text{opt}} = f_n + f_{\text{beat}} = n f_{\text{rep}} + f_{\text{CEO}} + f_{\text{beat}}$$

In this way the optical frequency of the cesium D_1 line could be measured directly [33]. Hänsch worked together with younger collaborators and students, among them Th. Udem, J. Reichert and R. Holzwarth. They could also show that the comb mode separations were extremely stable, at the 10^{-16} level [34,35]. In Boulder the techniques were developed in parallel [36] and a stability at the 10^{-19} level has later been demonstrated. A difficult part to measure in the expression above is f_{CEO} . However, this difficulty could be overcome by using an extremely wide comb, where a high-frequency mode f_{2n} , can be made to beat against a frequency-doubled low-frequency mode f_n , provided that the comb spans a full octave. The recorded beat frequency is the carrier envelope offset frequency. This relation was also noted by Telle, Keller et al. [37] considering frequency combs. We have

$$2f_n - f_{2n} = 2(n f_{\text{rep}} + f_{\text{CEO}}) - (2n f_{\text{rep}} + f_{\text{CEO}}) = f_{\text{CEO}}$$

Such extreme spectral broadening could be achieved by self-phase modulation of a femtosecond pulse in a piece of photonic crystal fibre, which has air-filled channels and where a high intensity is maintained over a substantial length of the fibre because of group velocity dispersion compensation. Hall and collaborators were the first to demonstrate this particularly simple and useful variety of the optical frequency comb technique (also called self-referencing) [38], closely followed by Hänsch and collaborators [39]. In a joint publication between the Hall and Hänsch groups [40], the frequency of a Nd:YAG laser was measured directly against the cesium clock using the comb technique. Later, the hydrogen 1s-2s frequency could be measured at the 10^{-14} level by Hänsch in a collaboration with the microwave fountain clock groups of C. Clairon and C. Salomon (BNM-SYRTE and Lab. Kastler Brossel, Paris, respectively) [17,41], as already discussed in connection with Fig. 1. In addition, an optical clock based on a single trapped mercury ion could be demonstrated at NIST [42]. The Time and Frequency Division at NIST has for a long time made very significant contributions in precision metrology with researchers including D.J. Wineland, J.C. Bergquist, L. Hollberg and others. In comparisons of the frequency of optical transitions measured with precision at time separations of 1-4 years, possible drifts in the fundamental constants could be investigated by several groups including Hänsch's [17,41,43] and the one at NIST. Within an uncertainty of a few parts in 10^{15} no drifts could so far be established.

Through the development of optical frequency comb techniques, led by Hänsch and with very important contributions from Hall's group, the elaborate previous schemes for optical frequency measurements, which only worked for selected frequencies, have been replaced by a set-up of the size $1 \times 1 \text{ m}^2$, good for precision measurements of any frequency, and even commercially available. A true revolution in optical frequency measurements has occurred, paving the way for the creation of all-optical clocks with a precision that might approach $1: 10^{18}$. Reviews of these developments can be found in Refs [44,45].

The utility of precision laser spectroscopy and optical comb techniques, and further developments

As already discussed, the techniques developed by Hall and Hänsch, allowing extremely precise spectroscopy and optical frequency standards, have many applications of both a fundamental and a practical nature. Tests of fundamental theories can be made at an ever-higher level of precision, regarding relativistic effects, the isotropy of space, possible asymmetries between matter and anti-matter (hydrogen and antihydrogen) and possible drifts in the fundamental constants. Global positioning systems can be refined, very large astronomical telescope arrays can be accurately synchronized and deep space navigation can be facilitated.

Recently the comb technique was extended to the VUV and XUV region by the Hänsch group, and also by the JILA group using harmonic generation in xenon gas, enclosed in an external cavity subject to circulating intense femtosecond pulses. Normally high harmonics generated from femtosecond pulses require such a high intensity that pulses obtained using chirped-pulse amplification (CPA) at a repetition rate of a maximum of a few kHz have been employed. Then no sharp frequency comb is generated, which is, however, the case if the full repetition rate of about 100 MHz can be retained, as is the case in the recent work. High resolution laser spectroscopy, for example, of the interesting 1s-2s transition in He^+ (compared to hydrogen scaled by a factor of 4 towards higher frequencies), seems to be within reach. Ultimately, atomic clocks in the X-ray region may emerge.

Attosecond pulses can be formed if the equidistantly spaced high harmonics are phase-locked together, in a way analogous with the case of a mode-locked laser in the

visible region. In the generation process of single attosecond pulses, as well as for the study of many ultra-high intensity interaction phenomena, it is advantageous if the optical phase is stabilized. Phase stabilization is also of utmost importance in many types of experiments using primary femtosecond pulses. The frequency comb technique thus becomes very important also in time-domain experiments. Actually, Hänsch is closely collaborating with F. Krausz and collaborators (formerly at the Technical University of Vienna, now at the MPQ) within the field of attosecond pulse generation and applications.

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