A SINGLE ^{199}HG^{+} ION OPTICAL CLOCK*

J.C. Bergquist, S.A. Diddams, C.W. Oates, E.A. Curtis,
L. Hollberg, R.E. Drullinger, W.M. Itano, and D.J. Wineland
National Institute of Standards and Technology, 325 Broadway,
Boulder, Colorado 80305

Th. Udem
Max Planck Institute für Quantenoptik, Hans-Kopfermann Strasse 1,
85748 Garching, Germany

Telephone: 1 303 497 5459
Fax: 1 303 497 7375
e-mail: berky@boulder.nist.gov

1. Introduction

Optical clocks based on laser-cooled atoms and ions potentially have superior stability and accuracy over present-day time standards, but until recently, no practical device was fast enough to count optical cycles in order to generate time. Now that obstacle has been circumvented using pulsed lasers to span an octave from the infrared to the ultraviolet with a grid of equally spaced, phase-coherent frequencies [1-8]. Here we report on work at NIST toward the realization of a highly stable and accurate optical timepiece [9] that uses a femtosecond laser to phase-coherently divide the frequency of a well stabilized laser that is locked to a narrow transition of a single, laser-cooled ^{199}Hg^{+} ion [10,11].

2. Atomic clock basics

All clocks consist of two major components: a device that generates periodic events, and a means for counting, accumulating, and displaying these events. For example, the swing of a pendulum can provide the periodic events that are counted, accumulated, and displayed by a set of gears driving a pair of clock hands. Atomic clocks add a third component: the resonance of a well-isolated atomic transition, which is used to control the oscillator frequency. If the frequency of the oscillator is made to match the transition frequency between two nondegenerate (and largely unperturbed) atomic states, then the clock will have improved long-term stability and accuracy. For an atomic clock based on a microwave transition, high-speed electronics count and accumulate a defined number of cycles of the reference oscillator to mark a unit of time. The same basic concepts apply for an atomic clock based on an optical transition at a much higher frequency. However, if all other factors are equal, the stability of the optical standard will be proportionally higher. This is the principal advantage of an optical clock over a microwave clock. The stability can be quantitatively expressed by the Allan deviation $\sigma_y(\tau)$ which provides a convenient measure of the fractional frequency instability of a clock as a function of averaging time τ [12]. For an oscillator locked to an atomic transition of frequency f_0 and linewidth Δf,

$$\sigma_y(\tau) \approx \left\langle \frac{\Delta f_{rms}}{f_0} \right\rangle_\tau \approx \frac{\Delta f}{\pi f_0} \sqrt{\frac{T}{N\tau}} \tag{1}$$

where Δf_{rms} is the measured frequency fluctuation, N is the number of atoms, and T is the cycle time (the time required to make a single determination of the line center) with $\tau > T$. This expression assumes that other noise sources are reduced to a sufficiently small level such that quantum-mechanical atomic projection noise is the dominant stability limit [13,14]. In this case, $\sigma_y(\tau)$ decreases as the square root of the averaging time for all clocks, so a ten-fold decrease in the short-term Allan deviation leads to a hundred-fold reduction in averaging time τ to reach a given instability. This point is particularly important if we ultimately hope to reach a fractional frequency uncertainty of 10^{-18}, which is anticipated as the limit for many optical clocks [15]. In this case, $\sigma_y(\tau) < 1\times10^{-15}\tau^{-1/2}$ is clearly desirable to avoid inordinately long averaging times.

3. The optical frequency standard

Our optical frequency standard [10] is based on a single, nearly motionless and unperturbed $^{199}Hg^+$ ion that is confined in a cryogenic, spherical Paul trap. The ion is cooled, state-prepared and detected by light that is scattered on the strongly allowed $^2S_{1/2}$ - $^2P_{1/2}$ transition at 194 nm. The $^2S_{1/2}$ (F = 0, m_F = 0) - $^2D_{5/2}$ (F = 2, m_F = 0) electric-quadrupole allowed transition at 282 nm (τ ($D_{5/2}$) $\approx$ 0.09 s) provides the reference for the optical standard (Fig. 1). The natural linewidth of the S-D resonance is about 2 Hz at 1.064 PHz, and recently a Fourier-transform-limited linewidth of only 6.7 Hz (Q = 1.5 × 10^{14}) was observed [10]. We lock the frequency-doubled output of a narrowband and stable 563 nm dye laser [16] to the narrow S-D resonance by the method of electron shelving, whereby each transition to the metastable D-state is detected by the suppression of the scattering of many 194 nm photons on the strongly allowed S-P transition [17,18]. The short-term (1-10 s) fractional frequency instability of the probe laser is $\leq 5 \times 10^{-16}$, which matches well to the best stability predicted by the shot-noise-limited detection of the S-D transition of a single mercury ion. The fractional frequency instability of the single ion mercury standard is expected to be $\leq 1 \times 10^{-15}\,\tau^{-1/2}$ with a fractional frequency uncertainty that approaches 10^{-18} [10]. However, in our present realization of the $^{199}Hg^+$ optical standard, the stability obtained from locking to the ion was degraded

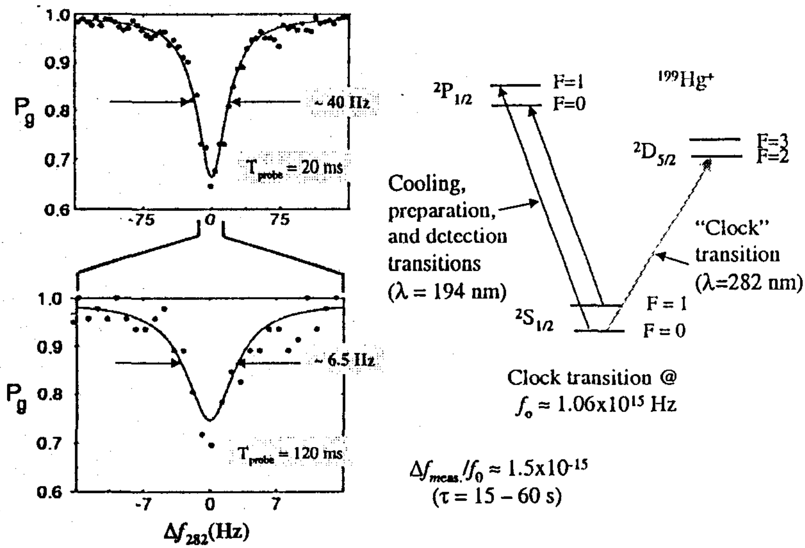

Figure 1. *Quantum-jump absorption spectra of the* $^2S_{1/2}(F=0) \bullet {}^2D_{5/2}(F=2)$ $\bullet m_F = 0$ electric-quadrupole transition and a partial energy level diagram of $^{199}Hg^+$ with the relevant transitions indicated. $\bullet f_{282}$ is the frequency detuning of the 282 nm probe laser, and P_g is the probability of finding the atom in the ground state. The upper spectrum is obtained with a Rabi excitation pulse 20 ms long (averaged over 292 sweeps) and the lower spectrum corresponds to an excitation pulse 120 ms long (averaged over 46 sweeps). The linewidths are consistent with the Fourier-transform limit of the respective pulse times [10].

primarily because the signal contrast was less than 100 % (see the spectra in Fig. 1) and there was substantial dead time between probes of the S-D resonance. Hence, $\sigma_y(\tau) \leq 2 \times 10^{-15}$ for averaging times up to ~ 30 s (where the frequency stability of the laser is dominantly controlled by the cavity), at which point $\sigma_y(\tau)$ began to average down as $\tau^{-1/2}$ (as the control of long term frequency fluctuations is transferred to the ion) [9,10].

4. The frequency divider

Figure 2 shows the optical clock in more detail, consisting of the optical frequency standard and the femtosecond-laser-based optical clockwork [9]. The Hg^+ standard provides high accuracy and stability, but to realize a countable clock output, we must phase-coherently convert the optical signal to a lower frequency. The

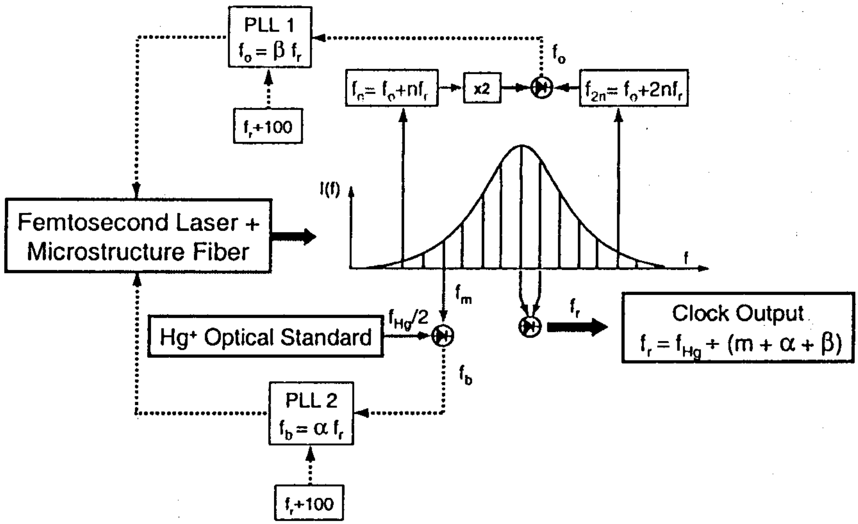

Figure 2. Schematic of the self-referenced all-optical atomic clock. Solid lines represent optical beams, and dotted lines represent electrical paths. The femtosecond laser, having repetition rate f_r, combined with the spectral broadening microstructure fiber produces an octave-spanning comb of frequencies in the visible/near infrared spectrum, represented by the array of vertical lines in the center of the figure. As depicted above this comb, the low frequency portion of the comb is frequency-doubled and heterodyned against the high frequency portion, yielding the offset frequency f_o that is common to all modes of the comb. Additionally, an individual element of the comb is heterodyned with the optical standard laser oscillator ($f_{Hg}/2$ = 532 THz) whose harmonic is locked to the clock transition frequency of the single $^{199}Hg^+$ ion. This yields the beat frequency f_b. Two phase-locked loops (PLL) control f_o and f_b with the result that the spacing (f_r) of the frequency comb is phase-locked to the Hg^+ optical standard. Thus f_r is the countable microwave output of the clock, which is readily detected by illuminating a diode with the broadband spectrum from the frequency comb. See the text for further details.

clockwork that divides the 1.064 PHz optical frequency to a countable microwave frequency f_r is based on a femtosecond laser and a novel microstructure optical fiber. The Ti:sapphire femtosecond ring laser emits a train of pulses (~25 fs duration) at the nominal repetition rate of f_r = 1 GHz [19]. The frequency-domain spectrum of the pulse train is a uniform comb of phase-coherent continuous waves separated by f_r. The frequency of the n^{th} mode of this comb is $f_n = nf_r + f_o$ [20,21], where f_o is a frequency offset common to all modes that results from the difference between the group- and the phase-velocity inside the laser cavity [22]. If the frequency comb of the laser covers an entire octave, then f_o can be measured by frequency doubling an infrared mode (n) and heterodyning it with an existing mode ($2n$) in the visible portion of the comb [3,7]. The heterodyne signal yields the frequency difference $2(nf_r + f_o) - (2nf_r + f_o) = f_o$. Only recently with the development of microstructure silica fibers [23,24] has the required octave-spanning spectrum been attained with high repetition rate, low-power, femtosecond lasers. The unique dispersion properties of the microstructure fiber provide guidance in a single spatial mode (~1.7 μm diameter) with zero group velocity dispersion near 800 nm [23]. Because temporal spreading of the pulse is minimized, peak intensities in the range of hundreds of GW/cm^2 are maintained over a significant propagation length, thus providing enhanced spectral broadening due to self-phase modulation. With approximately 200 mW (average power) coupled into a 15 cm piece of microstructure fiber, the total spectral width is broadened from ~15 THz to ~300 THz (spanning from ~520 nm to ~1170 nm).

In addition to f_o, a second heterodyne beat f_b is measured between an individual comb element $f_m = mf_r + f_o$ (m is an integer) and the 532 THz local oscillator of the Hg^+ standard. As shown in Fig. 2, two phase-lock loops (PLL) are employed to control f_o and f_b, thereby fixing the clock output f_r. PLL-1 forces $f_o = \beta f_r$ by controlling the pump power of the femtosecond laser [7]. Similarly, PLL-2 changes the cavity length of the femtosecond laser with a piezo-mounted mirror, such that $f_b = \alpha f_r$. The constants α and β are integer ratios implemented with frequency synthesizers that utilize $f_r/100$ as a reference. In this manner, the frequencies of both PLL's are phase-coherently linked to f_r such that all oscillators employed in the clock are referenced to the 532 THz laser oscillator itself. When f_o and f_b are phase-locked, every element of the femtosecond comb, as well as their frequency separation f_r, is phase-coherent with the laser locked to the Hg^+ standard [25]. With no other frequency references as an input, we realize all aspects of a high accuracy, high-stability, optical atomic clock: a stable laser that is locked to a narrow atomic reference, and whose frequency is phase-coherently divided to give a pulsed microwave output that can be recorded with a counter [9].

5. High-stability clock output

With both PLL's closed, the ~1 GHz microwave output has the value of $f_r = f_{Hg}/(m + \alpha + \beta)$. If we choose the signs of beats f_o and f_r such that $\alpha = -\beta$, then f_r would be an exact sub-harmonic of $f_{Hg}/2$. The stability of the 532 THz laser should be transferred to each element of the femtosecond comb, in addition to f_r. We obtain f_r from the bandpass-filtered photocurrent generated with ~5 mW of the broadened comb light incident on a p-i-n photodiode. We have measured the instability of f_r by subtracting it from the output of a synthesizer that is referenced to a hydrogen maser for which $\sigma_y(1s) \approx 2.5 \times 10^{-13}$. The stability of this difference frequency is then analyzed with both a high-resolution counter and a dual-mixer time measurement system [26]. Both results are consistent with the resolutions of the respective measurements and the maser stability, demonstrating that the 1 s stability of f_r is at least as good as that of the hydrogen maser [9].

Before we can conclusively state that a microwave signal with stability matching that of the optical standard can be obtained from the optical clock, f_r needs to be compared to an oscillator with stability significantly better than the hydrogen maser. This reference oscillator could be either the microwave output of a second optical clock, or the high stability output of a cryogenic microwave oscillator [27]. Lacking these, we can still verify the stability of the comb in the optical domain, and thereby infer the expected stability of f_r, by comparing one element of the optical comb to a second optical standard with high stability. For example, we can detect, filter, and count the heterodyne beat signal between a single element of the comb at 456 THz and a frequency-stabilized diode laser locked to the $^1S_0$ - $^3P_1$ intercombination transition of a laser-cooled ensemble of calcium atoms [28,29]. The Allan deviation of the heterodyne signal between the Hg^+ stabilized comb and the Ca-stabilized optical standard is shown as the triangles in Fig. 3. For $\tau < 10$ s the Allan deviation averages down roughly as $9 \times 10^{-15}\ \tau^{-1/2}$, which is higher than that of the Hg^+ standard (recall that the Hg^+ standard stability for $\tau < 10$ s is primarily controlled by the cavity (see Sec. 3)), but consistent with that expected of the Ca standard in its present configuration. It is also consistent with the monotonic increase in the 1 s instability of the heterodyne beat frequency as the stability of the Ca standard is degraded by using shorter probe times and lower resolution signals [9]. However, for $\tau > 10$ s, frequency and phase fluctuations introduced by the 180 m long optical fiber that transmits the 532 THz light to the femtosecond system begin to degrade the stability observed between the Ca standard and the Hg-referenced comb. We have measured the fiber-induced noise by double-passing the light through the optical fiber and find that the average fractional frequency fluctuations are between 2 to 4×10^{-15} for $1\ s < \tau < 10$ s. For $\tau > 10$ s, the fiber noise averages down as $1/\tau$. Finally, for $\tau > 30$ s the instability of the Hg^+ standard in its present configuration contributes to the measured instability at approximately the same level as the Ca standard. Nonetheless, the measured fractional frequency instability decreases with averaging to $\sim 1.5 \times 10^{-15}$ at 100 s.

More recently, we have implemented active cancellation [30,31] of this fiber noise and have further improved the signal-to-noise in the Ca standard. Data taken under these conditions reveal a fractional frequency instability of 7×10^{-15} at $\tau = 1$ s. These results are plotted as the square data points in Fig. 3 [9]. In this case we cannot place great significance in the stability for $\tau > 1$ s for two reasons. First, the Allan deviation for averaging times $\tau > 1$ s is calculated from the juxtaposition of 1 s averages. Such data analysis is known to result in errors for certain noise processes [32]. Second, for this specific data the 532 THz laser oscillator was not locked to the $^{199}Hg^+$ ion, and therefore it was necessary to subtract out the smooth and predictable drift (~1 Hz/s) of the Fabry-Perot cavity to which this laser is stabilized. However, neither of these affect the measured 1 s Allan deviation, for which we find an upper limit of 7×10^{-15} for the optical comb. Again, this 1 s

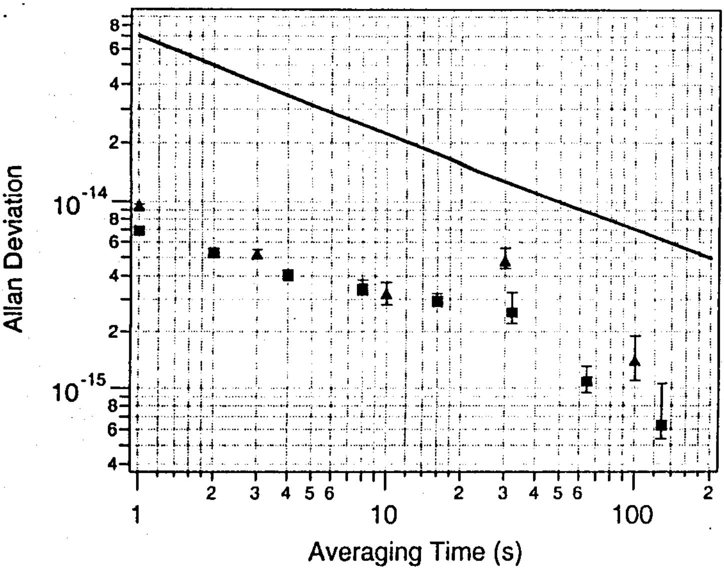

Figure 3. Measured stability of the heterodyne signal between one element of the femtosecond comb and the Ca optical standard at 456 THz (657 nm). The femtosecond comb is phase locked to the 532 THz laser oscillator. The triangles are the stability data without cancellation of the additive fiber noise. The squares are the measured stability with active cancellation of the fiber noise and improved stability in the Ca standard. These results are about an order of magnitude better than the best stability reported with a cesium microwave standard, which is designated by the solid line [34].

instability is consistent with that of the Ca standard in its improved configuration. Similar stability in the ~1 GHz clock output remains to be verified.

Finally, when f_r and f_0 are detected and counted with respect to the frequency of the hydrogen maser (which acts as a transfer standard to the NIST realization of the SI second [33]), an absolute measurement of the $^{199}Hg^+$ clock transition can be made [11], f_{Hg} = 1 064 721 609 899 143 (10) Hz. The statistical uncertainty of our measurements is about +/- 2 Hz, limited in part by the fractional frequency instability of the maser at our measurement times and in part by the accuracy determination of the cesium standard. The systematic uncertainty of +/- 10 Hz assigned to f_{Hg} is based on theoretical arguments in lieu of a full experimental evaluation. A second $^{199}Hg^+$ standard has been constructed toward making a full evaluation.

6. Conclusion

In conclusion, we have constructed an optical clock based on the 1.064 PHz (282 nm) electric-quadrupole transition in a laser-cooled, single $^{199}Hg^{+}$ ion. The optical frequency is phase-coherently divided to provide a microwave output using a mode-locked femtosecond laser and a microstructured optical fiber. The short-term (1 s) instability of the optical output of the clock is measured against an independent optical standard to be $\leq 7 \times 10^{-15}$. This optically-referenced femtosecond comb provides a countable output at 1 GHz, which ultimately could be used as a higher accuracy reference for time scales, synthesis of frequencies from the RF to the UV, comparison to other atomic standards, and tests of fundamental properties of nature.

Acknowledgements

This report is based in part on references 9 and 10. The authors are grateful to A. Bartels (GigaOptics GmbH) for his valuable assistance with the femtosecond laser. We are also indebted to R. Windeler (Lucent Technologies) for providing the microstructure optical fiber. We further acknowledge many illuminating discussions with J. Hall, S. Cundiff, J. Ye, and F. Walls. This work was supported in part by the Office of Naval Research.

7. References

1. Th. Udem, *et al., Phys. Rev.Lett.* **82**, 3568 (1999).
2. S. A. Diddams *et al., Phys. Rev. Lett.* **84**, 5102 (2000).
3. D. J. Jones *et al., Science* **228**, 635 (2000).
4. M. Niering *et al., Phys. Rev. Lett.* **84**, 5496 (2000).
5. R. Holzwarth *et al., Phys. Rev. Lett.* **85**, 2264 (2000).
6. J. Reichert *et al., Phys. Rev. Lett.* **84**, 3232 (2000).
7. R. Holzwarth *et al., Phys. Rev. Lett.* **85**, 2264 (2000).
8. J. Stenger *et al., Phys. Rev. A* **63**, 021802R (2001).
9. S.A. Diddams, *et al., Science* **293**, 825 (2001).
10. R. Rafac, *et al., Phys. Rev. Lett.* **85**, 2462 (2000).
11. Th. Udem, *et al., Phys. Rev. Lett.* **86**, 4996 (2001).
12. J. A. Barnes *et al, IEEE Trans. Inst. & Meas.* **20**, 204 (1971).
13. W. M. Itano *et al., Phys Rev. A* **47**, 3554 (1993).
14. G. Santarelli *et al., Phys. Rev. Lett.* **82**, 4619 (1999).
15. A. A. Madej and J. E. Bernard, in *Frequency Measurement and Control*, A. N. Luiten, ed. (Springer-Verlag, Berlin, 2001), pp. 153-194.
16. B. C. Young, F. C. Cruz, W. M. Itano, J. C. Bergquist, *Phys. Rev. Lett.* **82**, 3799 (1999).

17. H. Dehmelt, *Bull. Am. Phys. Soc.* **20**, 60 (1975).
18. J.C. Bergquist *et al.*, *Phys. Rev. A* **36**, 428 (1987).
19. A. Bartels, T. Dekorsy, and H. Kurz, *Opt. Lett.* **24**, 996 (1999).
20. A. I. Ferguson, J. N. Eckstein, T. W. Hänsch, *Appl. Phys.* **18**, 257 (1979).
21. J. Reichert, *et al.*, *Opt. Comm.* **172**, 59 (1999).
22. D.J. Wineland *et al.*, in *The Hydrogen Atom*, G.F. Bassani, M Inguscio and T.W. Hänsch, eds. (Springer-Verlag, Heidelberg, 1989) pp. 123-133.
23. J. K. Ranka, R. S. Windeler, A. J. Stentz, *Opt. Lett.* **25**, 25 (2000).
24. W. J. Wadsworth *et al.*, *Electron. Lett.* **36**, 53 (2000).
25. We count the phase-locked beats f_o and f_b with a high resolution counter and verify that they fluctuate about their nominal respective phase-locked values < 100 mHz in 1 s. This implies that the stability of the 532 THz laser oscillator is transferred to the femtosecond comb with a relative uncertainty $< 2 \times 10^{-16}$ in 1 s.
26. S. Stein *et al.*, in *Proceedings of the 1982 IEEE International Frequency Control Symposium*, (IEEE, Piscataway, NJ, 1982), pp. 24-30.
27. S. Chang, A. G. Mann, A. N. Luiten, *Electron. Lett.* **36**, 480 (2000).
28. C. W. Oates, F. Bondu, R. W. Fox, L. Hollberg, *J. Phys. D*, **7**, 449 (1999).
29. C. W. Oates, E. A. Curtis, L. Hollberg, *Opt. Lett.* **25**, 1603 (2000).
30. B. C. Young *et al.* in *Laser Spectroscopy XIV*, R. Blatt, J. Eschner, D. Leibfried, F. Schmidt-Kaler, eds. (World Scientific, Singapore, 1999), pp. 61-70.
31. L.-S. Ma, P. Jungner, J. Ye, J. L. Hall, *Opt. Lett.* **19**, 1777 (1994).
32. P. Lesage, IEEE Trans. Inst. & Meas. **32**, 204 (1983).
33. S. R. Jefferts, *et al.*, in *Proceedings of the 2000 IEEE International Frequency Control Symposium*, Kansas City, 7-9 June, 2000 (IEEE, Piscataway, NJ, 2000), pp. 714-717.
34. P. Laurent *et al.*, in *Laser Spectroscopy XIV*, R. Blatt, J Eschner, D. Liebfried, F. Schmidt-Kaler, Eds. (World Scientific, Singapore, 1999), pp. 41-50.