High-Sensitivity Spectroscopy with Diode Lasers

R. W. Fox, H. G. Robinson[†], A. S. Zibrov[†], N. Mackie, J. Marquardt,
J. Magyar, L. W. Hollberg

National Institute of Standards and Technology
325 Broadway, Boulder, CO 80303

ABSTRACT

Linewidth reduction of an extended cavity diode laser at 657 nm was accomplished by negative feedback to an intra-cavity ADP crystal. High resolution (170 kHz wide) saturated absorption signals were recorded of the calcium intercombination line which is of interest for a frequency standard. The spectrum of the red $6^2p_{3/2}$-$9^2s_{1/2}$ cesium line in a magneto-optical cell trap was also investigated.

1. INTRODUCTION

The advance of diode lasers into the visible offers opportunities in the area of frequency standards and frequency metrology. Diode lasers stabilized to known spectral features have the potential to be low-cost, high precision frequency markers and length standards. The calcium intercombination line at 657 nm is one such feature that has generated much interest [1,2].

An optical frequency reference may in general be realized by first reducing the short term laser frequency fluctuations by locking to a Fabry-Perot cavity, then locking the cavity to an atomic or molecular line. This approach allows enough signal-to-noise in the short term (< 1 s) for the performance to be limited by systematics associated with measuring the spectral feature, such as field shifts or time of flight broadening. A caveat to this is that the lock to the optical cavity must be relatively immune from offset drifts. Offsets may occur, for instance, when the injection current of a diode system is modulated to produce sidebands for a rf heterodyne lock. Some fraction of the inherent amplitude modulation (am) is detected along with the desired signal, leading to an intensity dependent dc term at the output of the phase sensitive detector. Strategies to deal with this include demodulating at 3f and/or sampling the laser intensity prior to the reference cavity in order to subtract any fluctuations out of the cavity transmission or reflection. However there are both fundamental limits (with regard to the power that must be expended in the sampling channel), and practical limits such as the inhomogeneity of the am across the beam that will induce drift at some level. Thus it is of interest to start with as low residual am from the modulation as possible.

Employing a phase modulator external to the laser cavity to apply sidebands and to reduce the frequency/phase fluctuations is a method that has met with much success.[3] We have positioned the phase modulator inside the extended cavity, where it serves as a frequency modulator. In comparison an external modulator has much lower gain as a frequency transducer. Although external multiple pass configurations have been used, the poor quality of the diode laser beam makes this approach increasingly difficult. A disadvantage of placing the crystal in the laser cavity is the coupling of the output power to the frequency modulation (fm), albeit at a much lower level than direct injection current modulation. However the injection current channel is now available to be part of a closed loop for intensity control, with a servo bandwidth independent of the frequency control loop. If the intensity control loop bandwidth is extended to cover the fm sideband frequency, the residual am and subsequent dc lock drift will be further decreased. Similar approaches using negative feedback to externally placed modulating crystals to eliminate residual am have worked well.[4]

2. LASER FREQUENCY NOISE AND LINEWIDTH REDUCTION

The extended cavity laser consisted of an anti-reflection coated[5] red diode laser operating near 660 nm, a f/1 collimating lens, a 29 mm long Brewster-cut ADP modulator crystal, and a high diffraction efficiency holographic grating.[6] The laser was built on an invar baseplate and enclosed in an aluminum box to reduce acoustics. The grating was mounted on an invar

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support with a simple hinge and a piezo-electric crystal, allowing about 12 Ghz of tuning without mode hops. The zero-order beam from the grating was reflected from a mirror mounted perpendicular to the grating, and served as the output. This configuration reduces the output beam's angular sweep with tuning. The output power was 1 mW to 1.5 mW, depending on the lasing wavelength.

The spectral linewidth of the unlocked (and locked) laser may be examined by a heterodyne measurement using a similar or more stable laser as the local oscillator. In lieu of building two complete systems, the linewidth may also be studied by observing the laser's frequency noise spectrum. This provides information on the spectral distribution and magnitude of the frequency fluctuations that generate the linewidth. The noise spectrum can be measured by using an independent optical cavity as a discriminator. Locking the laser and discriminator cavity together with a low bandwidth (~1 Hz) side-lock conveniently cancels drift and permits faster differential frequency fluctuations between the laser and cavity to be accurately converted to intensity fluctuations.

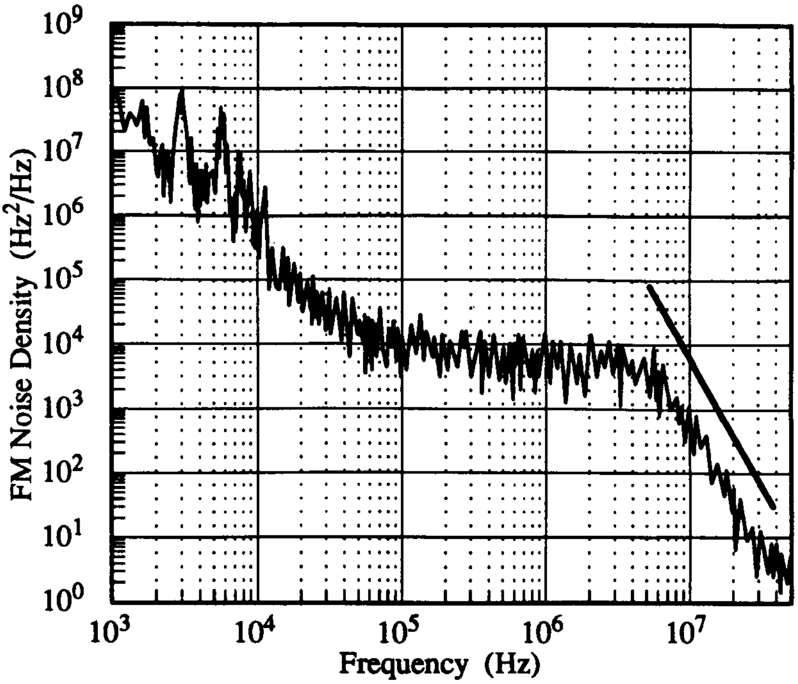

Fig. 1 The unlocked extended cavity laser frequency noise spectrum. The line indicates the discriminator cavity roll-off slope.

The 12 cm extended cavity length resulted in a measured frequency noise level of approximately 8000 Hz^2/Hz (see fig. 1). Below about 50 kHz the fluctuations increase in a typical 1/f fashion, obliging the noise density to rise as $1/f^2$. A number of distinct acoustic peaks are evident in figure 1; these result from structural resonances of the grating mount. The frequency noise data was taken with a $\Delta\nu$=8 MHz linewidth discriminator cavity in transmission. The data shows the cavity roll-off (-40 dB/decade) at 8 MHz, indicating that the laser's frequency noise spectrum is flat out to at least 30 MHz. At about 30 MHz the measurement noise floor causes a deviation from -40 dB/decade. The fast frequency noise level apparent between 50 kHz and 30 MHz indicates an inherent Lorentzian laser linewidth of about 25 kHz ($\Delta\nu$= $\pi \cdot 8000$ Hz^2/Hz). This is additionally broadened by the low frequency noise.

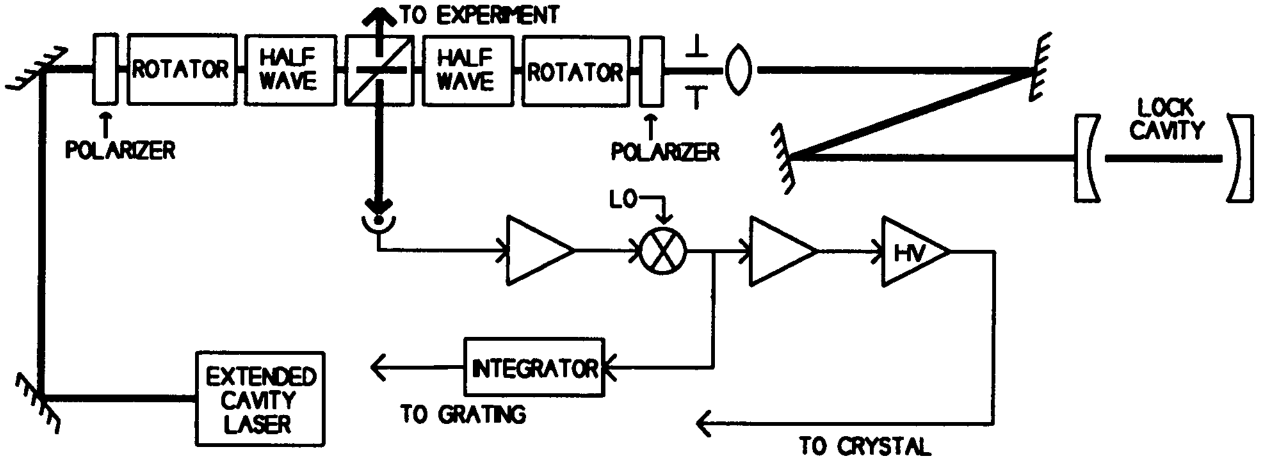

Figure 2. The extended cavity laser was locked to a 2.5 MHz linewidth optical cavity by the rf heterodyne technique. The rotator-polarizer pairs provide 35 dB of isolation each.

The frequency fluctuations at rates above 100 kHz comprise relatively little of the laser linewidth. This is a consequence of the fast frequency noise level, H_0, and the corresponding integrated phase noise. For a white fm noise spectrum, the phase noise integrated[7] beyond a given arbitrary frequency f, i.e. integrated from f to infinity, is H_0/f. Note that significant contributions to the laser linewidth appear only when the integrated phase noise density approaches 1 rad^2.(8) The measured noise level indicates the integrated phase fluctuations above 100 kHz are on the order of $H_0/f \sim 0.1$. Therefore we expect a sharp reduction of the linewidth when the noise below 100 kHz is reduced, in contrast to the situation of a solitary diode laser where the frequency noise in the MHz range is important.

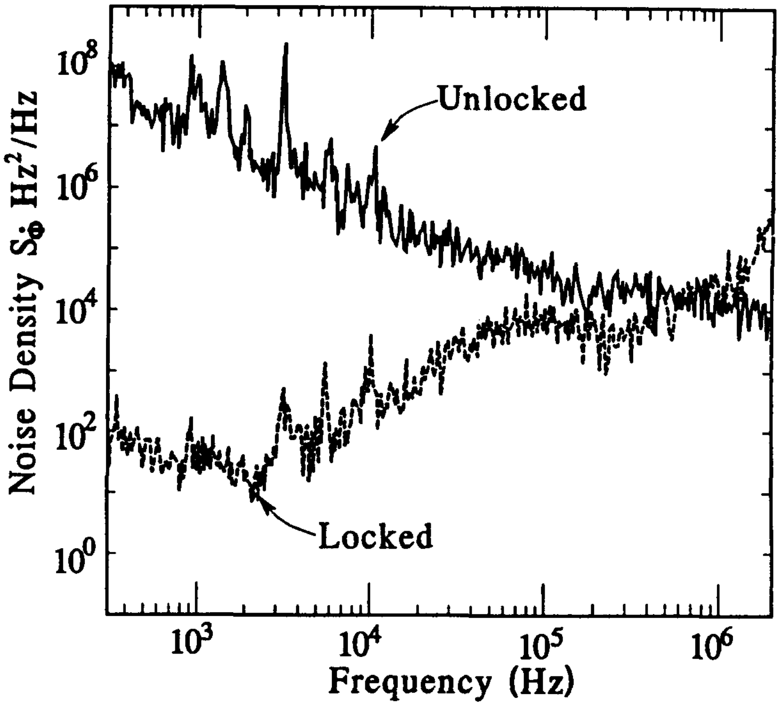

Fig. 3 The unlocked and locked error signal out of the mixer. The noise density values agree well with an independent measurement, fig. 4

The non-confocal cavity used for the lock had a linewidth of 2.5 MHz, a free spectral range of 1 GHz and a piezo-electric transducer (pzt) for tuning. A conventional rf heterodyne set-up [9] provided the error signal as shown in figure 2. Two negative feedback loops were implemented to reduce the frequency noise. An integrating loop ($f_{0\,dB}$ ~300 Hz) to the grating pzt primarily served to limit the necessary dynamic range of the faster loop. This rather low bandwidth was dictated by mechanical resonances of the grating structure, the fundamental resonance being at 3 kHz. The faster (ADP) loop had approximately 80 dB of gain below 100 Hz, which was then rolled-off with double-pole and single-pole sections to a unity gain frequency of ~1 MHz. Compensation was made for the lock-cavity phase shift and much of the crystal high-voltage driver's phase shift. The bandwidth and consequently the attainable low-frequency gain were limited by the residual uncompensated phase shift of the high-voltage driver. Higher frequency fluctuations could be corrected by using a low-voltage higher-bandwidth feedback path to the other side of the crystal.

Most of the laser power was reserved for the saturation spectroscopy, leaving 30 μW incident on the detector used for the lock channel. This incident power was just sufficient to allow intensity noise limited operation. The 1 MHz bandwidth allowed enough low frequency gain to impart this limiting noise floor onto the laser at frequencies below 10 kHz.

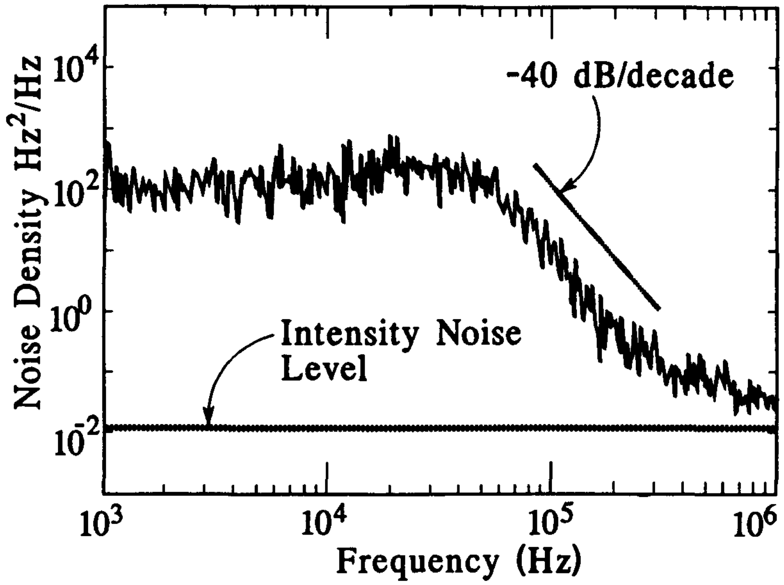

Fig. 4 The locked extended cavity laser frequency noise spectrum. The data was obtained with a 50 KHz linewidth discriminator cavity.

The ADP crystal transfer function was ~0.78 MHz/volt. The crystal was bonded on one side to a ceramic mount and acoustically damped to reduce piezo-electric resonances in the 90 kHz range. The modulation frequency was chosen to be well above the cavity bandwidth to produce a broad lock acquisition range. Modulation frequencies of 12 MHz and 26 MHz were used. However the modulation index was limited by the available synthesizer power, so the crystal was driven as a series resonant LC circuit at the modulation frequency. Q's of about 10 yielded adequate sideband amplitudes.

Reduction of the frequency noise and attendant linewidth were observed with an independent high quality reference cavity used as a discriminator. The reference cavity is constructed from a ULE glass cylinder with optically contacted mirrors. It has a 50 kHz linewidth, a 3 GHz free spectral range, and is mounted inside a vacuum housing (~10^{-6} Pa) to reduce index drifts and acoustic noise. Frequency noise measurements were obtained by side-locking to this high finesse reference cavity with slow feedback ($f_{0\,dB}$ ~1 Hz) to the $\Delta\nu$=2.5 MHz "lock" cavity pzt. The resultant fm to am conversion is indicative of the laser's frequency fluctuations only to the extent that the two cavities do not exhibit common mode vibrations. The hermetically sealed lock cavity was hard-mounted to the optical table and the reference cavity was supported in a "cradle" fashion by 2 fine wires that allowed pendulum modes but strongly damped high frequency oscillations. We believe that the frequency region of "common mode" vibrations was << 100 Hz.

The locked laser exhibits frequency fluctuations (see fig. 4) at the 200 Hz^2/Hz level out to several hundred kHz. At approximately 400 kHz the increasing laser noise (due to the falling servo gain) and measurement noise floor (due to detector/intensity noise) combine to deviate the slope from -40 dB/decade. The laser noise increases to the meet the open-loop level of 8000 Hz^2/Hz as the servo gain drops to unity at about 1 MHz. Inadequate servo-loop phase margin causes an increase at this point over the open-loop noise, but in terms of fluctuations that contribute to the linewidth this is not significant since the integral of the phase noise contribution from this frequency region is negligible compared that of the lower frequency region. This noise spectrum indicates[10] a linewidth reduced to less than 1 kHz, sufficient for high resolution saturated spectroscopy of Calcium in an oven.

Modulating the laser frequency with the ADP crystal was accompanied by measurable residual amplitude modulation. The level of am depended slightly upon the particular mode and wavelength. This can be understood by noting that the residual am will be a function of the slope of the composite gain curve $G(\nu)$ of the extended cavity laser at the lasing wavelength. The grating reflection and the residual etalon of the AR coated chip are the two primary contributors to $dG(\nu)/d\nu$. A modulation index of β=1.0 at 12 MHz was accompanied by 0.015% amplitude modulation. Modulation via the injection current at the same modulation index (as determined by the average of the induced sideband amplitudes) resulted in 1.7% amplitude modulation.

The above mentioned residual am could be reduced by a simple feedback loop to the injection current. However, this may in general cause a change in β and the phase of the modulation, which is a cause for concern if it effects the lock discriminator. Although to first order the lock discriminator is insensitive to small changes in the signal phase,[11] for high-quality locks this may be a noise term.

3. CALCIUM SATURATION SPECTROSCOPY

The calcium intercombination line at 657 nm exhibits a number of desirable properties of a spectral reference line. Among other attributes it has a 400 Hz natural linewidth and a simple ground state configuration ($^1S_0$) that should allow nearly all of the atoms present to take part in the absorption process. A laser cooled calcium beam or neutral atom trap approach may well be required to take advantage of it's full potential. However calcium saturated absorption in an oven provides excellent frequency stability in a simple system.

A high density calcium beam about 6 cm long was created in an oven with uncoated bk-7 glass windows. The background gas pressure was kept at less than $1.3\cdot10^{-3}$ Pa (10^{-5} Torr) to avoid pressure broadening. The windows were partially

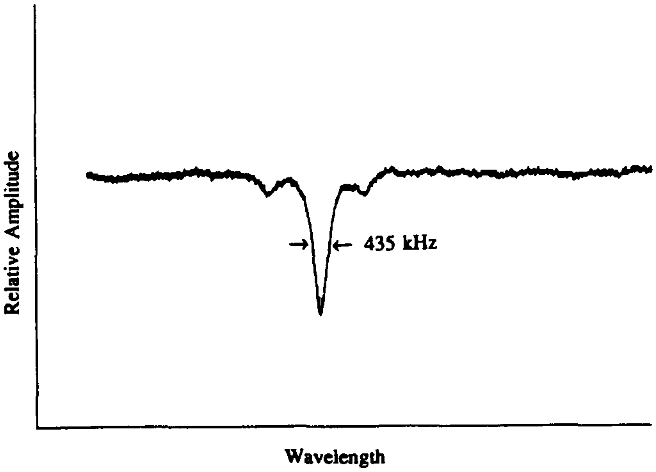

Fig. 5 435 kHz wide absorption of the calcium intercombination line at 657 nm. The linewidth is limited by time-of-flight broadening. The sidebands are spaced by 2.3 MHz.

protected from calcium deposition by the oven design.

A pair of coils provided a transverse magnetic field that was aligned with the laser polarization. This allows only $\Delta M_F=0$ transitions, which have only a very small quadratic Zeeman sensitivity (~10^8 Hz/T^2). The saturating beam power was approximately 400 µW, and was reflected by a flat mirror to serve as the probe beam. The Doppler absorption at the operating temperature was 10% and 1.4 GHz wide. The Doppler curve was narrowed somewhat due to a directional calcium flux in the oven. The saturation feature (see fig. 5) was about 5% of the Doppler absorption. The hot calcium and a 2 mm diameter laser beam size resulted in a short interaction time (~1 µs), limiting the observed linewidth to 435 kHz. The laser's spatial profile was shaped by a pair of anamorphic prisms and a telescope to provide a 7 mm beam, with which 170 kHz linewidths were obtained. Further reductions will require a better spatial mode. Lock-in detection at 40 kHz resulted in signal-to-noise ratios of ~10^3 (1 Hz bandwidth) at modulation levels that produced no modulation broadening.

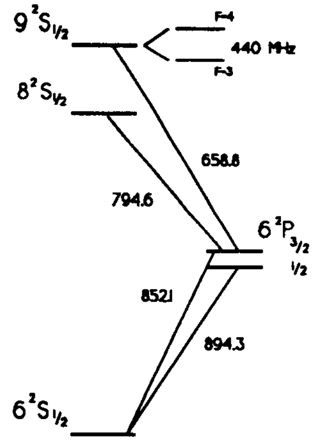

Fig. 6 Cesium energy level diagram. Wavelengths are in nm, and the hyperfine splitting of the 9s state is shown.[12]

4. CESIUM TRAP SPECTROSCOPY

Cesium magneto-optic cell traps may well serve as the basis for the next generation of atomic frequency and time standards. There is a cesium transition (6p$_{3/2}$ to 9s$_{1/2}$) at 658.8 nm which is close to calcium in wavelength and within the tuning range of the laser. We note that the essentially Doppler-free absorption from the 6p$_{3/2}$ levels is a potentially useful indication of trap characteristics. The 852 nm cooling laser in this trap is typically locked several linewidths to the red of the ground state F=4 to F=5 transition (see fig. 6), and a second laser tuned 9 Ghz away re-pumps atoms in the lower ground state level back into the cycling transition.

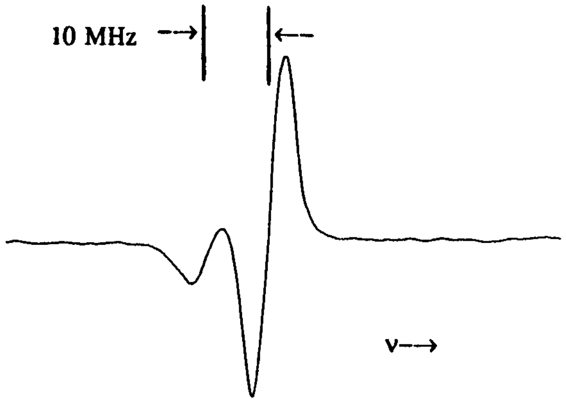

Fig. 7 Cs derivative spectroscopy at 658.8 nm. The larger peak is 1% absorption. The cooling laser is tuned to the red of the 852.1 nm transition.

The red absorption from the upper trapping level was measured with a weak probe beam (~10 µW, 0.05 mW/cm^2) approximately the same diameter as the cluster of trapped atoms. An optical fiber was used as a spatial filter and conveniently transmitted the red probe light from the locked laser to the trap. Etalon fringes with a period of 4 MHz and spurious low frequency amplitude noise resulting from polarization changes also caused by the fiber were reduced by a double beam subtraction configuration. FM modulation at 50 kHz and lock-in detection with a 1 ms time constant were employed, yielding signal-to-noise ratios of ~200. The absorption detection limit extrapolated to 1 s was $3\cdot10^{-6}$, limited by mismatching of the double beam configuration that allowed laser and fiber noise to be detected. The 1% absorption (see fig. 7) of the 6p$_{3/2}$ F=5 → 9s$_{1/2}$ F=4 transition indicates a trap population of ~$2\cdot10^8$ atoms, which agrees well with 852 nm fluorescence measurements.

We observe a double peak absorption spectrum which is preliminarily attributed to Autler-Townes splitting (AC Stark effect) caused by the 852 nm cooling laser fields. A frequency separation that varies as the square root of the intensity and an asymmetry that depends on detuning from line center is expected,[13] and is consistent with our observations.

5. SUMMARY

A 12 cm long extended cavity laser at 657 nm stabilized to a precision reference cavity by electronic feedback to an intra-cavity ADP crystal has been described. The residual am that accompanies fm is approximately 100 times less when the laser is modulated with the crystal rather than the injection current. This laser was used to measure linear doppler-free absorption from the cesium 6 $^2p_{3/2}$ state in a magneto-optical trap. In addition we used the laser for non-linear absorption experiments of the calcium intercombination line. Linewidths of 170 kHz were observed, limited by laser spatial mode size and quality.

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† Present address: Dept. Physics, Duke University, Durham, N. C.

‡ Permanent address: P. N. Lebedev Physics Institute, Leninsky Prospect, 53, 117924, Moscow, Russia

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