A Mechanism of Rubidium Atomic Clock Degradation: Ring-Mode to Red-Mode Transition in rf-Discharge Lamps

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***Abstract*— In the vapor-cell atomic clock, long-term stability can be influenced by slow variations in the discharge lamp's output via the light-shift effect. Additionally, over a multi-year mission lifetime the lamp's aging can degrade its optical pumping efficiency. Understanding the mechanism(s) that drives these changes is particularly important for spacecraft devices, where the atomic clocks are called upon to function continuously and reliably for many years. Here, we consider the two well-known, but little studied, modes associated with alkali rf-discharge lamp operation: the ring mode and the red mode. Consistent with previous research, we find that the ring mode is best for optical pumping, and that the clock-signal amplitude degrades significantly when the lamp operates in the red mode. Examining the emission spectrum as the lamp transitions between these two modes, we show that the ring-mode to red-mode transition is driven by radiation trapping within the lamp.**

I. Introduction

In the mid-sixties, Robert Shaw [1] initiated the first detailed investigations into the plasma characteristics of alkali rf-discharge lamps used for optical pumping in atomic clocks. Shaw discovered that the discharge lamp operated in two distinct spectral "modes," which he referred to as the ring mode and the red mode; these are shown in Fig. 1. In the ring mode, which corresponded to relatively low lamp temperatures, the lamp's emission "appeared primarily white… with a red ring near the bulb walls." In contrast, at higher lamp temperatures Shaw found that the red mode "was less intense and appeared red throughout the discharge." Shaw noted that under nominal conditions the transition between these two modes was "abrupt" near a lamp temperature of 123 ^{o}C and that the transition temperature depended on rf power. Additionally, Shaw found that in the ring mode both alkali and noble gas lines were present in the lamp's emission; while in the red mode only alkali lines appeared. To date, the mechanism driving the ring-mode to red-mode transition has never been uncovered. Here, we consider two questions: 1) what is the ring-mode to red-mode transition mechanism and 2) what effect does the ring-mode to red-mode transition have on a rubidium clock's operation. As will be discussed subsequently, we find that the ring-mode to red-mode transition is driven by radiation trapping at elevated lamp temperatures, and that when this transition occurs the atomic clock signal decays because of a loss in optical pumping efficiency. The transition therefore represents a potential mechanism for atomic clock failure, and we suggest that the noble-gas lines from the lamp be used to herald too large a lamp temperature.

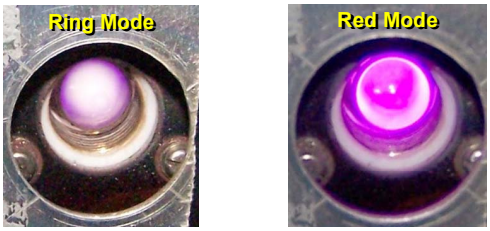

Figure 1. Pictures of a Rb rf-discharge lamp operating in the ring mode (left) and red mode (right). In the figure, the mode change derives from a change in the lamp's temperature.

II. Discharge Lamp Spectroscopy

In our first series of studies, we examined the spectrum emitted by a Rb/Xe rf-discharge lamp. Briefly, we imaged the lamp's emission in the plane of an optical fiber, which we could scan vertically across the image's face. The fiber went to a spectrometer, which allowed us to study the various spectral components emitted by the lamp separately. Examining the radial variation of xenon light and rubidium second resonance light,[1] we found that the xenon emission appeared to come uniformly from the lamp volume, while the rubidium light was principally located near the lamp walls. This is consistent with previous work [2,3], indicating that Rb

[1] We measured second resonance light because it is much less sensitive to radiation trapping effects.

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atoms are ionized in the discharge and that atomic resonance lines are generated near the lamp walls, where ion/electron recombination is most probable. Visually, we found that the ring-mode to red-mode transition occurred around 165 °C in these experiments.[2] Further, we found that the radial distribution of xenon light and rubidium light did not change as we transitioned from ring mode to red mode, suggesting that at no time was xenon appreciably ionized. If xenon had been ionized, its radial distribution would have mimiced that of rubidium.

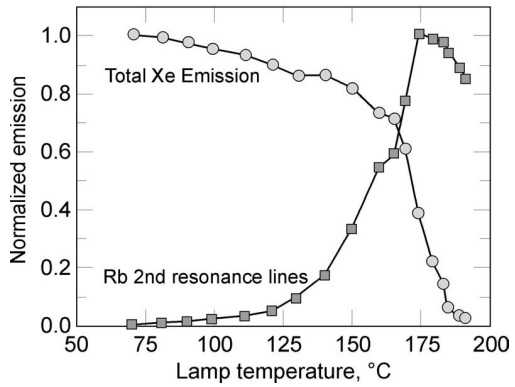

Figure 2. Circles correspond to the normalized intensity of a sum of prominent xenon spectral lines, while squares correspond to the intensity of rubidium second resonance emission (i.e., $6^2P_{1/2,3/2} \rightarrow 5^2S_{1/2}$) at 422 and 420 nm.

Figure 2 shows the change in the lamp's spectral emission with temperature. As the lamp temperature crossed the threshold for the ring-mode to red-mode transition, the xenon light fell precipitously. Near the same temperature, the rubidium emission rose dramatically, and then fell at a somewhat higher temperature. Further, using several lines of xenon we were able to deduce an electronic temperature for the xenon atoms (i.e., T_{Xe}), which indicated the extent to which upper levels of xenon were populated in the discharge. For lamp temperatures below ~ 150 °C, T_{Xe} was constant at roughly 5200 °C. However, between 150 and 175 °C T_{Xe} also fell precipitously.

As discussed elsewhere [4], we attribute these observations to the role of radiation trapping [5] in the dynamics of the rf-discharge. At low lamp temperatures, and consequently low rubidium density, radiation trapping is not significant. In such cases, multi-step ionization of rubidium takes place from the ground state [6], and the electron temperature of the discharge is associated with the rate limiting step in the ionization process (i.e., the rubidium 5S to 5P excitation step at 1.6 eV). As the lamp temperature increases, the rubidium density increases, and consequently the photons emitted by the 5P to 5S decay become trapped inside the vapor; this gives rise to a metastable 5P population. Multi-step ionization under such conditions need not originate in the ground state, but can occur starting from atoms trapped in the 5P state. Consequently, under radiation trapping conditions the rate-limiting step in the ionization process is associated with excitation out of the 5P state, which requires less electron kinetic energy (i.e., ~ 1.0 eV). Therefore, under radiation trapping conditions we expect the discharge's electron temperature to cool, giving rise to a reddening of the plasma emission and a loss of xenon spectral lines.

III. Clock Signal Amplitude

If radiation trapping is the source of the ring-mode to red-mode transition, then we should expect to see some evidence of this effect in the clock's 0-0 hyperfine transition. Specifically, it is well known that radiation trapping gives rise to self-reversal in alkali discharge lamps, as trapped photons spectrally diffuse from the center of the resonance line to the lineshape wings [7,8]. Therefore, in the lamp temperature range of 150 to 175 °C, we should expect to see some consequence of radiation trapping in the optical pumping rate that generates the atomic clock signal.

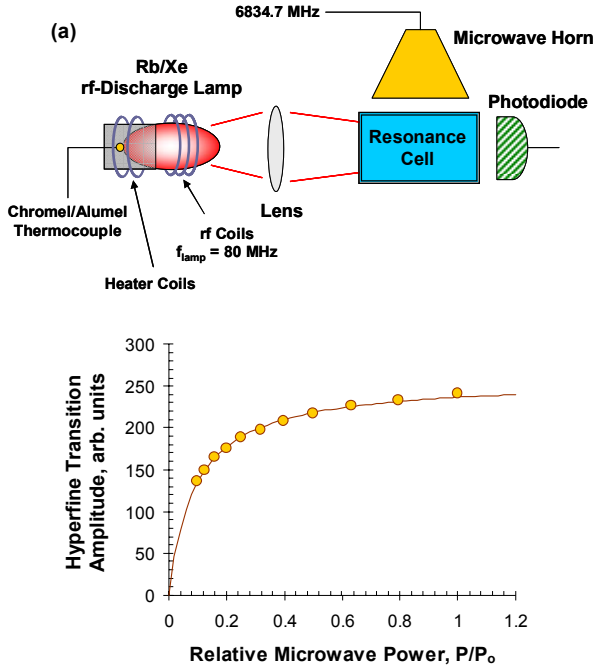

Figure 3. (a) Block diagram of the experiment examining the 0-0 hyperfine transition amplitude as a function of rf-discharge lamp temperature. (b) Illustrative example of the data, where we measure the hyperfine transition amplitude as a function of relative microwave power for a fixed lamp temperature.

Figure 3a shows a block diagram of the experimental arrangement. Light from our Rb/Xe rf-discharge lamp passed through a cylindrical glass resonance cell (L = 3.9 cm and 2R = 2.2 cm) containing isotopically enriched Rb^{87} along with 39 torr of N_2 and 61 torr of Ar. The resonance cell temperature was maintained at approximately 64 °C; the transmitted light intensity was detected with a Si photodiode, and the resonance cell was placed in the near field of a microwave horn (15 dB gain).

For the experiment, we fixed the lamp temperature and then measured the 0-0 hyperfine transition amplitude as a

[2] Lamp temperature should only be taken as "indicative", since it was difficult to accurately and consistently place the thermocouple in the commercial lamp-bulb housing.

function of relative microwave power, P/P_o. This is illustrated in Fig. 3b. To analyze the data, we recognized that the 0-0 transition amplitude, S, can be written as [9]

$$S = \frac{A_o R\gamma\beta P}{(2R+\gamma)^2\left[(\gamma+R)^2+\beta P\right]}, \qquad (1)$$

where, R is the optical pumping rate, βP corresponds to the square of the microwave Rabi frequency driving the 0-0 transition, and γ is the intrinsic linewidth of the 0-0 resonance. Rearranging Eq. (1), we get

$$\frac{1}{S} = \frac{(2R+\gamma)^2}{A_o R\gamma}\left[1+\frac{(\gamma+R)^2}{\beta P}\right]. \qquad (2)$$

We know define the saturation power, P_{sat}, as $(\gamma+R)^2/\beta$ and the microwave-power-independent signal amplitude, α, as $A_o R\gamma/(2R+\gamma)^2$, and rewrite Eq. (2) as

$$\frac{1}{S} = \frac{1}{\alpha}+\frac{\left(P_{sat}/P_o\right)}{\left(P/P_o\right)}.^{3} \qquad (3)$$

Note that while α and P_{sat} both depend on the optical pumping rate, they are related to different aspects of the atomic clock signal: α is a measure of the relative change in light intensity (i.e., $\Delta I/I_o$), while P_{sat} is a measure of the linewidth of the 0-0 transition.

Fitting our data to Eq. (3), we obtained α and the microwave saturation power as functions of lamp temperature. Normalizing to the dc transmitted light intensity, I_o, Fig. 4 shows $\Delta I/I_o$ as a function of lamp temperature. For the circled data, we always let the lamp equilibrate at T_{Lamp} = 108 °C before raising the temperature to any particular value. For the data shown as squares, we let the lamp equilibrate at T_{Lamp} = 208 °C before lowering its temperature to a particular value. Thus, the two sets of data correspond to starting the measurements from either the ring mode or the red mode, respectively. Clearly, with regard to the amplitude of the 0-0 hyperfine transition, there is no hysteresis.

[3] In the experiment, the lamp was very close to the resonance cell and as the lamp temperature increased so too did the resonance cell temperature. By measuring the cell temperature separately, and correlating cell temperature to lamp temperature, we were able to correct our data for this effect. Briefly, given weak optical pumping conditions we have $|\Delta I|/I_o \equiv |N_{on}-N_{off}|\sigma L << 1$, where N_{on} and N_{off} are the number densities of atoms in the absorbing state with the microwaves on and off-resonance, respectively, and σ is the absorption cross-section. Thus, we can write $|\Delta I/I_o| = N(T_c)\Delta\rho\sigma L$, where $\Delta\rho$ is the microwave induced change in the density matrix elements and T_c is the cell temperature. Expanding $N(T_c)$ in a Taylor series about $T_c = T_o$, we have

$$N(T_c) = N_o + (T_c - T_o)\frac{dN}{dT_c} = N_o\left(1+\frac{(T_c-T_o)}{N_o}\frac{dN}{dT_c}\right).$$

We corrected our data by dividing the raw $|\Delta I|/I_o$ values by the term in brackets on the right-hand side of the above equation.

Note that $\Delta I/I_o$ peaks at around 185 °C and then falls at higher lamp temperatures. This is consistent with Fig. 2, which shows a peak in the rubidium 2nd resonance emission at roughly the same temperature with a subsequent fall off in intensity. We interpret this data in the following way. As the lamp temperature increases there is first an increase in rubidium vapor density, which translates into brighter rubidium lamp lines and consequently higher optical pumping rates. However, as radiation trapping becomes ever more severe, photons diffuse to the lineshape wings. These spectrally-diffused photons are off-resonance with respect to the rubidium absorption lines in the resonance cell, and so the optical pumping rate falls. This decrease in optical pumping rate due to spectrally-diffused photons will likely be compounded by radiation-trapping effects that alter the lamp's discharge dynamics. Specifically, as the electron temperature cools due to radiation trapping, the rate of rubidium ionization in the lamp should also be suppressed.

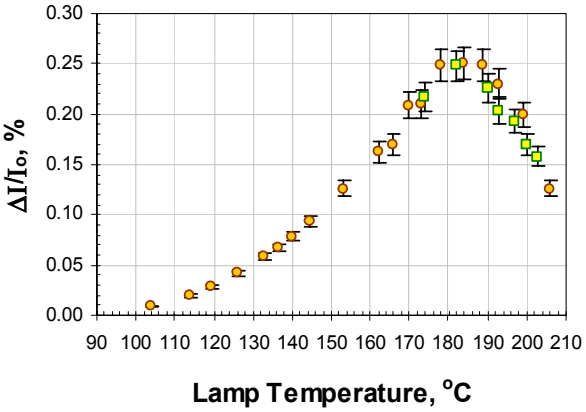

Figure 4. From the saturation curve fits, this figure shows the (microwave-power-independent) relative change in the photodiode light intensity as a function of lamp temperature. As described in the text, circles correspond to raising the lamp temperature from 108 °C for each data point, while squares correspond to lowering the lamp temperature from 208 °C for each data point.

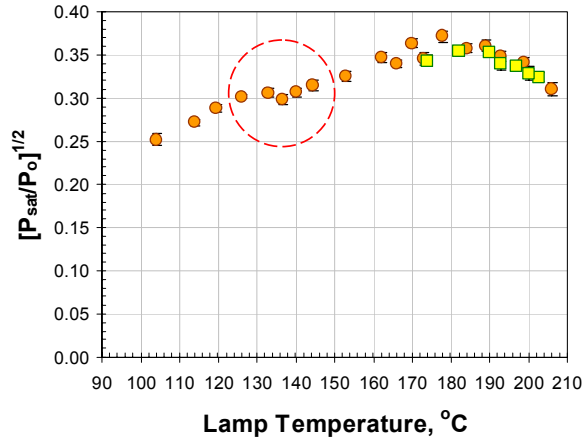

Figure 5. Square-root of the relative saturation power for the 0-0 hyperfine transition as a function of lamp temperature. Note that $[P_{sat}/P_o]^{1/2}$ is a measure of the linewidth of the 0-0 transition. Again, circles correspond to raising the lamp temperature from 108 °C for each data point, while squares correspond to lowering the lamp temperature from 208 °C for each data point. Hysteresis is not observed.

Figure 5 shows the square-root of the relative saturation power as a function of lamp temperature. This data conveys complimentary information to that shown in Fig. 4, since $[P_{sat}/P_o]^{1/2}$ is a measure of the 0-0 transition's linewidth. As the figure shows, for lamp temperatures below ~ 170 ^oC, P_{sat} increases with lamp temperature. This is due to the increase in optical pumping rate with lamp temperature discussed with regard to Fig. 4. Additionally, at high temperatures we observed a decrease in P_{sat}, consistent with the fall-off in signal amplitude shown in Fig. 4. Together, Figs. 4 and 5 provide compelling evidence that there is a decrease in optical pumping rate at high lamp temperatures, which is the result of radiation trapping within the plasma of the rf-discharge lamp. Finally, we note that there is a dip in the saturation power at $T_{Lamp} = 135\ ^oC$. At present, we do not understand the origin of this effect, though it was repeatable in our measurements. It is worth noting that there was a corresponding dip in the xenon emission at roughly the same lamp temperature.

IV. Conclusions

We have investigated the ring-mode to red-mode transition in alkali rf-discharge lamps. Contrary to Shaw's original explanation, which required a change in the ionized species in the two modes (i.e., ring mode $\Rightarrow$ noble-gas ionization, red mode $\Rightarrow$ alkali ionization), our work suggests that the two modes are differentiated by the role of radiation trapping in the discharge's dynamics. Briefly, in the presence of radiation trapping, the first excited state of the alkali atom becomes (effectively) metastable. Ionization of alkali atoms can therefore be achieved at lower electron energies, and as a consequence there is a cooling of the discharge. However, in both modes it is the alkali that is the dominantly ionized species, and noble-gas light is generated via inelastic collisions with electrons. As the discharge cools, the electrons have less energy to excite the noble-gas atoms, and therefore noble-gas emission is reduced in the red mode.

These conclusions were borne out in our examination of the 0-0 hyperfine transition amplitude as a function of lamp temperature. At low lamp temperatures, where radiation trapping does not play a prominent role, the optical pumping rate rises as a function of lamp temperature. However, in the red mode, the optical pumping rate falls as a function of lamp temperature, consistent with the effect of radiation trapping on resonance photons: radiation trapping causes resonance photons to spectrally diffuse to the wings of the absorption line, where the rubidium atoms in the resonance cell see them as far off-resonance.

With regard to the title of this paper, and the idea of clock degradation, it is clear that certain missions for rubidium clocks (e.g., GPS and Galileo) require the clock's unattended operation for a number of years. The present work shows that if the lamp temperature increases, and a ring-mode to red-mode transition occurs, there could be quite serious implications for clock operation. The transition may not be easy to detect from a simple examination of the clock frequency, since as is apparent from Figs. 4 and 5 the 0-0 hyperfine transition has reasonable amplitude just within the red mode. Thus, on either side of the threshold for the transition the atomic loop will remain locked, and it is only as the lamp temperature increases further that the clock will fail. However, by monitoring the emission of noble gas light in an alkali rf-discharge lamp, as illustrated in Fig. 2, one could effectively monitor the severity of radiation trapping in the lamp's discharge. Specifically, though the amplitudes of the alkali resonance lines change considerably with lamp temperature in the ring mode, the Xe lines only change appreciably when radiation trapping takes on importance. This could be useful as a diagnostic of lamp operation. For example, a reduction of noble gas light intensity could herald an unexpected (and too large) increase in lamp temperature that might eventually lead to the failure of a navigational satellite's atomic clock.

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References

[1] R. W. Shaw, "Spontaneously generated ion acoustic waves in a weakly ionized plasma," Masters thesis, Cornell University, 1964.

[2] H. van Tongeren, "Electron temperature and radial density distribution of Cs ground-state atoms in the positive column of a Cs-Ar D.C. low pressure discharge," Phys. Lett., vol. 37A, pp. 317-318, 1971.

[3] V. S. Zholnerov, I. M. Nekrylova, M. V. Spazheva, V. A. Trukhacheva, and V. I. Khutorshchikov, "Spatial distribution of atoms in electrodeless spectral lamps containing rubidium vapor," Opt. Spectrosc. (USSR), vol. 56, pp. 148-149, 1984.

[4] J. C. Camparo and R. Mackay, "Spectral mode changes in an alkali rf discharge," J. Appl. Phys., vol. 101, 053303, 2007.

[5] T. Colbert and J. Huennekens, "Radiation trapping under conditions of low to moderate line-center optical depth," Phys. Rev. A, vol. 41, pp. 6145-6153, 1990.

[6] L. Vriens, "Multistep ionization in the positive column of low-pressure Na-Ne and Ne discharges," J. Appl. Phys., vol. 49, pp. 3814-3820, 1978.

[7] F. Fermeersch, V. Fiermans, J. Ongena, H. A. Post, and W. Wieme, "Monte Carlo investigation of imprsonment of resonance radiation with partial frequency redistribution," J. Phys. B: At. Mol. Opt. Phys., vol. 21, pp. 1933-1937, 1988.

[8] A. F. Molisch, G. J. Parker, B. P. Oehry, W. Schupita, and G. Magerl, "Radiation trapping with partial frequench redistribution: Comparison of approximations and exact solutions," J. Quant. Spectrosc. Radiat. Transfer, vol. 53, pp. 269-275, 1995.

[9] J. C. Camparo and R. P. Frueholz, "Linewidths of the 0-0 hyperfine transition in optically pumped alkali-metal vapors," Phys. Rev. A, vol. 31, pp. 1440-1448, 1985; J. C. Camparo and R. P. Frueholz, "Saturation of the 0-0 hyperfine transition linewidth enhancement factor in optically pumped alkali-metal vapors," Phys. Rev. A, vol. 32, pp. 1888-1889, 1985.