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PRECISION WAVELENGTH METROLOGY WITH A FOURIER TRANSFORM SPECTROMETER

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Abstract – We have investigated the intrinsic accuracy of the optical frequency scale in spectra acquired by a Fourier Transform Spectrometer (FTS). The uncalibrated accuracy of the FTS optical frequency axis is about 1 part in 10^5. This uncertainty can be reduced by at least two orders of magnitude using a multiplicative calibration correction derived from a single wavelength standard line. The work reported here describes a new approach to accurate calibration of the wavenumber scale for a UV-visible FTS, which we have used to measure accurate wavenumbers and Ar pressure shifts for the prominent lines of ^{198}Hg.

Keywords: Fourier Transform Spectrometer, wavenumber calibraton, Hg, pressure shift

1. INTRODUCTION

The wavenumber scale in spectra acquired by a Fourier Transform Spectrometer (FTS) is affected by two main phenomena -the finite size of the aperture and imperfect alignment of the sample and the reference beams ("cosine error"). A "cosine error" of about 1 part in 10^5 may be good enough for many spectroscopic applications, but the absolute accuracy can be significantly improved by applying a multiplicative correction to the frequency scale based on a spectral line of well-known wavelength. Using this approach the uncertainties can be reduced by at least two orders of magnitude. By using a single line as a wavelength standard the effects of the finite aperture size and the "cosine error" can be simultaneously corrected with an appropriate calibration factor k_{eff}:

$$\sigma_{\mathrm{TRUE}} = (1+ k_{eff}) \, \sigma_{\mathrm{APPARENT}},$$

where σ_{TRUE} is the correct wavenumber, and $\sigma_{\mathrm{APPARENT}}$ is the measured, uncorrected wavenumber.

If the wavenumber scale for an unknown spectrum is to be calibrated using a known standard line, it is essential that the FTS be illuminated in the same way by the unknown and calibration sources. The most convenient approach is to employ as reference line a wavelength standard in the investigated source (intrinsic calibration source). If no suitable line is emitted by the source, as is often the case for sealed electrodeless discharge lamps, it is often necessary to use an extrinsic calibration source [1]. In this work we describe the use of a laser line and its harmonic as an extrinsic calibration source and as a diagnostic tool for testing the validity of the single point calibration approach. By using this approach we have obtained accurate wavenumbers and Ar pressure shift parameters for the most prominent lines of ^{198}Hg.

2. EXPERIMENT

The essential components of the experiment are the FTS, the laser system, the electrodeless discharge lamp, and a small integrating sphere. The FTS used in this work was designed primarily for use in the ultraviolet and is comprehensively described in several papers [2, 3]. Although the limiting resolution of the instrument is 0.025 cm^{-1}, the typical resolution used in our experiments was 0.03 cm^{-1}.

The electrodeless discharge lamps (EDL) employed here were constructed of fused quartz with an internal envelope containing a few mg of ^{198}Hg and Ar buffer gas surrounded by a water jacket through which cooling water was flowed to stabilize the lamp temperature at 8 °C. The lamps were filled with Ar at pressures of 33, 400, 930 and 1330 Pa.

The laser system was a single-frequency cw ring dye laser pumped by an Ar ion laser. It was equipped with an intracavity frequency doubler and operated with the dye DCM. The wavenumber of the UV beam generated by frequency doubling in a cw system has a ratio of 2:1 to the wavenumber of the red fundamental beam to a very high degree of accuracy. The fundamental frequency of the dye laser was stabilized by locking it to the "t"-component of the hyperfine transition R41 (6-5) of molecular iodine (σ = 15379.221566 cm^{-1}). By stabilizing the dye laser to an iodine line we obtained red and UV reference lines with wavenumbers known to better than 2 parts in 10^9 [4].

The integrating sphere of 10 cm diameter played a central role in our experiment. The sphere was used to couple light from the mercury EDL, the fundamental laser beam, and the frequency doubled laser beam into the FTS. The exit opening of the sphere simulates an extended incoherent source, thus ensuring that there will be no shifts due to non-uniform illumination of the FTS entrance aperture.

3. RESULTS AND DISCUSSION

The first goal of our experiment was to investigate the calibration accuracy of the FTS using the single point calibration approach. Approximately 140 spectra were recorded over a period of several months. From each of these spectra we determined the ratio of the apparent wavenumbers of the frequency-doubled and fundamental laser beams. This ratio should be exactly two. Since either of the laser lines could be used with equal confidence as a reference line for a single point correction of the wavenumber scale, the deviation of the ratio from two establishes a lower limit for the uncertainty of the calibration.

Fig. 1 shows the measured deviations in the ratio from approximately 140 spectra taken over a period of months during which the FTS was realigned several times. The data show a scatter in the ratio from about -3 parts in 10^8 to about +4 parts in 10^8. The mean of all the ratios reveals a slight bias to the positive side of less than 1 part in 10^8. The entire set of results indicates that the accuracy of the single point calibration approach for this instrument is limited to about 2 parts in 10^8.

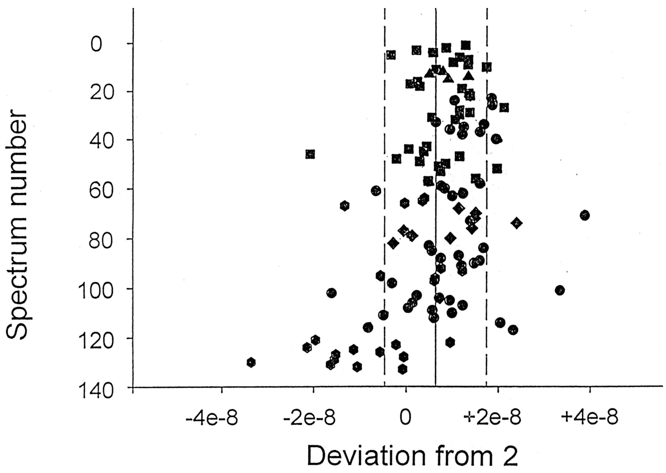

Fig. 1. Deviations in the ratio of the wavenumbers of the frequency-doubled and fundamental laser beams from the expected value of exactly 2 for a series of approximately 140 independent spectra.

A second goal of the experiment was to measure the wavenumbers of the most prominent lines of ^{198}Hg and investigate their pressure-shifts at four different pressures of Ar. For the measurements we used a correction factor k_{eff} calculated as the average of the values determined from the fundamental and frequency-doubled laser lines. As expected [5], we observed that all of the lines shifted to the red with increasing Ar pressure. This indicates that the essential contribution to the shift comes from an attractive long-range interatomic interaction of the van der Waals type. Within the experimental uncertainty these shifts all vary linearly with perturber density.

In Fig. 2 we show the dependence of the wavenumber of the 404.7 nm line of mercury (7 $^3S_0$ - 6 $^3P_0$) on Ar pressure. The behavior of this line is representative of the others we observed. As predicted by the Lindholm-Foley impact theory a linear dependence on the argon pressure was found:

$$\Delta \upsilon_0 = (1/2\pi c)\sigma_1 < v > N_{Ar}$$

where $\Delta \upsilon_0$ is the mercury line shift, N_{Ar} is the argon pressure, σ_I is the shift cross section, and <v> denotes the mean relative velocity.

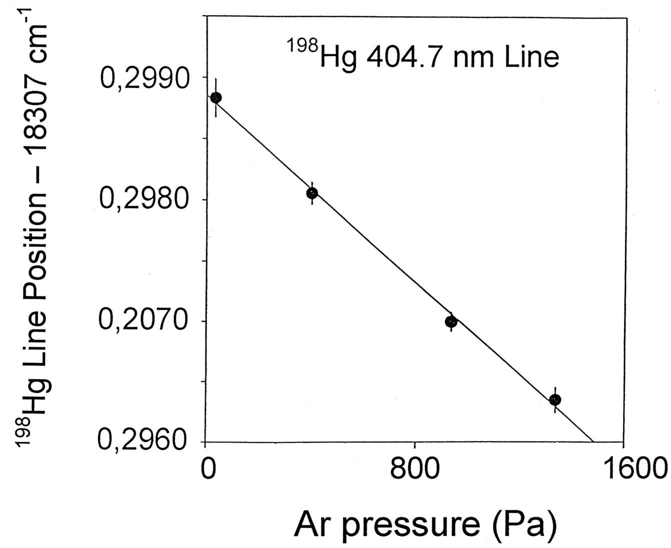

Fig. 2. The dependence of the wavenumber of the ^{198}Hg 404.7 nm line of mercury (7 $^3S_0$ - 6 $^3P_0$) on Ar pressure.

4. CONCLUSION

A dye-laser line stabilized to the Doppler-free absorption line R41 (6-5) of I_2 and that same laser line frequency-doubled were combined in an integrating sphere with emission from a Hg EDL and observed with the FTS. The limiting uncertainty of our FTS was determined from the difference in the correction factor as calculated from the measurements of the optical frequency of the laser fundamental and its harmonic in each spectrum.

Generally, our measurements of the wavenumbers of ^{198}Hg lines are in excellent agreement with the data available in ref. [5], but are determined with a higher accuracy. The red shift observed for all mercury lines with increasing Ar pressure indicates that the essential contribution to the line-shift comes from the attractive long-range interatomic interaction of the van der Waals type. The assumption of this shape of interatomic interaction provides values for the ^{198}Hg line-shift constants.

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