High resolution spectroscopy on Te$_2$: new lines for reference

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Ro-vibrational spectra of different electronic states of molecules are often used as absolute wavelength or frequency standards. These standards are also used to mitigate any slow drift of laser frequency during an experiment. In precision experiment, the two most commonly used molecular standards are iodine and tellurium, both are homo-nuclear diatomic molecules. The former is mostly used as standard for the long wavelength (600 − 900 nm) region, while the tellurium spectrum is widely used in short wavelength (400 − 550 nm) including near ultra violet. A comprehensive data on tellurium spectra can be obtained from the tellurium atlas \cite{1}. However near the 455 nm range where a number of important atomic resonance line, the atlas provides no significant data. We have performed high resolution modulation transfer spectroscopy (MTS) on tellurium molecule in a hot cell in the region close to 455 nm wavelength thereby obtained more than 100 new spectral lines which were not observed before. The resolution of each of these peaks is about few MHz, making them suitable for laser frequency locking.

I. INTRODUCTION

Precision experiments invariably require calibration of device parameters using standards. The most precisely measured physical parameter is frequency. To standardize frequency one uses the energy gap between two electronic levels of an atom that has a narrow natural linewidth. Even though the ground state hyperfine energy splitting of a $^{133}$Cs atom is currently set as the atomic standard, it is likely to be replaced by optical transitions in atoms or ions \cite{2}. In either case, the clock transition is a weakly coupled electronic transition at a short wavelength. These clocks are based on laser cooled atomic samples. Similarly, ion trap or cold atom based quantum information processors are also based on laser cooled atomic samples. Thus laser cooling has now become a standard tool for any precision spectroscopy experiment. The lasers related to cooling or detection are often required to be stable over long hours of experiment and in some cases like stable clock operation, the laser needs to be stable for days. Here stability refers to long time drift of the laser frequency due to thermal or mechanical stresses. These drifts even for the best laser designs can be as high as 10's MHz/hr which is comparable to the cooling transition linewidth in commonly used atoms and ions. There are two ways to mitigate this problem, namely, frequency locking the laser to an ultra-low expansion (ULE) optical cavity mode or to an atomic /molecular standard. The former demands good thermal and mechanical isolation of the cavity such that the cavity length does not drift with time. In the best designs these drifts are about 10 − 100 kHz/hr. The less demanding option is to frequency lock the laser to an atomic or molecular reference spectral line. However an optical cavity can be designed for any wavelength but it is not always possible to find an atomic or molecular spectral line at a desired wavelength. One can in principle use the same atomic species as that of the experiment in order to frequency lock the lasers. In case of multiple species of atoms or ions used in an experiment, the setup becomes complicated due to the requirement of gas cells or hollow cathode lamps for each of the species.

Molecular electronic spectra usually covers a broad range of wavelengths, making them an ideal choice for optical laser frequency standards which does not necessarily be accurate but has to be stable. The most widely used molecules are iodine and tellurium, both are homo-nuclear and hence not IR active. The electronic spectra of iodine shows strong absorption band starting at around 500 nm for the transition between its $X$ and $B$ electronic states which are the lowest two electronic states of the system \cite{3}. Therefore, iodine lines are suitable to frequency lock lasers in the longer wavelength range. The tellurium spectra has been observed starting from ultra violet (UV) wavelength of about 350 nm onward. The main advantage of this molecule is its large spin-orbit coupling and the presence of a large number of isotopes. This combination allows a wide spectral range to be accessible in a single molecular specie \cite{2}.

Tellurium spectrum has been studied extensively in many contexts. The most extensive work has been done by J. Cariou, P. Luc, Atlas du spectre d'Absorption de la Molecule de Tellure, CNRS, Paris, 1980 \cite{1}. Subsequent work has been done to extend this atlas on both sides of the spectrum available in the atlas \cite{5,8}. While longer wavelength region can be made accessible by thermally exciting the molecule (having higher temperature of the gas cell), short wavelength spectra are generated by the strong spin-orbit coupling present in the molecule. The progress made in identifying a plethora of spectral lines starting in UV and continuing all the way above 600 nm makes a good choice as reference for quantum optics, quantum information and precision measurement experiments \cite{9,12}. However,
FIG. 1: (Color online) Schematics of our setup for the MT spectroscopy where a saturated pump beam is sent through a tellurium vapour cell and a weak probe beam in the opposite direction. The beat signal, generated by this pump-probe beam after going through the Te cell, is picked up on the photodiode (PD) with a responsivity of 0.64 W/A and then demodulated the signal by mixing the ac part of it with the modulation frequency \( f_1 \) of electro-optic modulator (EOM) which has been employed to create the sidebands to the laser carrier frequency \( f_0 \). The lineshape of the Te lines have been observed in the oscilloscope after doing the low-pass filtering (LPF) of the demodulated signal. The spectras thus, having a good signal to noise ratio that are free from any background as shown in the schematic, shows a potential for use in obtaining optical frequency reference. PBS- polarizing beam splitter; BS- beam splitter; L- lens.

the reference wavelength requirement of these experiments are generally very specific and related to the atomic species being probed. Therefore the reference spectral line should not be farther than 1 GHz away from the required frequency to be able to frequency lock and shift by the use of standard acusto-optic modulators (AOM). So far, the reported spectral lines in tellurium are restricted in the 410 – 502 nm region \[13-15\] while the atlas also includes some of the UV spectra \[16\]. However, the region of interest for specific atoms are either the 470 – 490 nm or closer to the 450 nm related to emission lines from barium and ytterbium ions. Here we report experimentally measured more than 100 new lines close to 455 nm wavelength and spreading over a spectral range of 200 GHz. The spectral resolution of these lines are about few MHz mainly limited by the natural linewidth.

**II. EXPERIMENTAL DETAILS**

The experiment has been performed on a gas cell filled with solid \(^{130}\)Te\(_2\) in low pressure neon buffer gas. To obtain sufficient vapour pressure of tellurium the cell is heated up to 530°C. Uniformly heated cell is a necessity to avoid solid deposit on the windows thus reducing the optical signal. A clamp heater has been used to heat the cell uniformly while the setup is mounted inside a heat shield which is then insulated using glass wool. A detailed thermal, mechanical and optical design of the cell is given in \[17\]. The experimental setup for the modulation transfer spectroscopy (MTS) is shown in figure \[1\]. Modulation Transfer Spectroscopy has been chosen as opposed to saturation spectroscopy in order to avoid background noise in the spectral signal. In MTS two counter- propagating laser beams are used, one
of which is modulated at a frequency of $\sim 5.8$ MHz. Due to the applied modulation on the pump beam, there are three frequencies of the pump and one frequency of the probe beam interacting simultaneously with the molecules. Thus a four-wave mixing mediated by the Tellurium molecules takes place inside the cell leading to a probe spectra which is a derivative of the saturation absorption resonance spectrum as shown in figure 2. The line shape of each individual spectral line can be fitted using a known theoretical model to obtain the center frequency as well as the width of the spectral line. In order to scan over a wide range of frequencies yet having a good resolving power we have used external cavity diode laser (ECDL) which is described in ref. [12]. The laser line width is estimated to be below 1 MHz with an output power of about 40 mW. As shown in the figure 1, a part of the light is sampled out to simultaneously observe the wavelength in a Fizeau type wavemeter from Toptica while a second part probes a single Barium ion which is used as an absolute wavelength reference. The laser frequency can be scanned over 100 GHz without any mode hop by scanning the voltage applied to the grating of the ECDL laser. However for the measurements reported here, the scan was performed piece-wise in order to maintain linearity of the piezo expansion as a function of the applied voltage.

III. MEASURED SPECTRAL LINES

The spectral lines obtained in this experiment are tabulated in Tab. 1. The relative frequencies are given with respect to the line no. 82 which was previously known spectral line (line no. 1677) from Atlas [1]. The intensity observed for each line is normalized to the line no. 82, thus it is referred to as relative intensity. The laser power varies as a function of the frequency which has been normalized while calculating the relative strength.

| Line no. | Wavenumber(cm$^{-1}$) | Rel. frequency(GHz) | Rel. strength |
|----------|------------------------|---------------------|--------------|
| 1        | 21948.5268             | 112.82              | 0.01         |
| 2        | 21948.64822            | 109.18              | 0.14         |
| 3        | 21948.68792            | 107.99              | 0.01         |
| 4        | 21948.76464            | 105.69              | 0.01         |
| 5        | 21948.85837            | 102.88              | 0.24         |
| 6        | 21948.87204            | 102.47              | 0.91         |
| 7        | 21948.97778            | 99.3                | 0.08         |
| 8        | 21948.98812            | 98.99               | 0.02         |
| 9        | 21949.00647            | 98.44               | 0.07         |
| 10       | 21949.06317            | 96.74               | 0.29         |
| 11       | 21949.09086            | 95.91               | 0.14         |
| 12       | 21949.14123            | 94.4                | 0.03         |
| 13       | 21949.15457            | 94                  | 0.08         |
| 14       | 21949.22462            | 91.9                | 0.04         |
| 15       | 21949.25397            | 91.02               | 0.02         |
| 16       | 21949.3301             | 85.74               | 0.11         |
| 17       | 21949.44077            | 85.4                | 0.04         |
| 18       | 21949.50114            | 83.61               | 0.04         |
| 19       | 21949.50715            | 83.43               | 0.27         |
| 20       | 21949.52049            | 83.03               | 1.56         |
| 21       | 21949.62523            | 79.89               | 0.01         |
| 22       | 21949.6836             | 78.14               | 0.01         |
| 23       | 21949.91543            | 71.19               | 0.11         |
| 24       | 21949.9201             | 71.05               | 0.03         |
| 25       | 21949.93811            | 70.51               | 0.13         |
| 26       | 21949.98615            | 69.07               | 0.33         |
| 27       | 21950.1119             | 65.3                | 0.04         |
| 28       | 21950.17895            | 63.29               | 0.27         |
| 29       | 21950.22131            | 62.02               | 0.08         |
| 30       | 21950.29403            | 59.84               | 0.18         |

Continued...
| Line no. | Wavenumber(cm⁻¹) | Rel. frequency(GHz) | Rel. strength |
|---------|------------------|---------------------|--------------|
| 31      | 21950.33606      | 58.58               | 0.89         |
| 32      | 21950.35507      | 58.01               | 0.01         |
| 33      | 21950.37608      | 57.38               | 0.40         |
| 34      | 21950.39743      | 56.74               | 1.11         |
| 35      | 21950.41144      | 56.32               | 0.06         |
| 36      | 21950.44046      | 55.45               | 0.04         |
| 37      | 21950.50217      | 53.6                | 0.04         |
| 38      | 21950.53986      | 52.47               | 0.01         |
| 39      | 21950.59657      | 50.77               | 0.04         |
| 40      | 21950.75868      | 45.91               | 0.01         |
| 41      | 21950.76769      | 45.64               | 0.02         |
| 42      | 21950.82673      | 43.87               | 0.08         |
| 43      | 21950.87209      | 42.51               | 1.07         |
| 44      | 21950.88744      | 42.05               | 0.01         |
| 45      | 21950.89778      | 41.74               | 0.02         |
| 46      | 21950.91212      | 41.31               | 0.10         |
| 47      | 21950.9248       | 40.93               | 0.04         |
| 48      | 21950.94815      | 40.23               | 0.01         |
| 49      | 21950.97583      | 39.4                | 0.06         |
| 50      | 21951.06356      | 36.77               | 0.02         |
| 51      | 21951.19832      | 32.73               | 0.03         |
| 52      | 21951.23601      | 31.6                | 0.04         |
| 53      | 21951.26336      | 30.78               | 0.03         |
| 54      | 21951.34609      | 28.3                | 0.18         |
| 55      | 21951.37077      | 27.56               | 0.16         |
| 56      | 21951.38111      | 27.25               | 0.02         |
| 57      | 21951.38945      | 27                  | 0.20         |
| 58      | 21951.44182      | 25.43               | 0.91         |
| 59      | 21951.46784      | 24.65               | 0.02         |
| 60      | 21951.49319      | 23.89               | 0.01         |
| 61      | 21951.54589      | 22.31               | 0.02         |
| 62      | 21951.58859      | 21.03               | 0.07         |
| 63      | 21951.62828      | 19.84               | 0.27         |
| 64      | 21951.64663      | 19.29               | 0.03         |
| 65      | 21951.67098      | 18.56               | 0.03         |
| 66      | 21951.68933      | 18.01               | 0.02         |
| 67      | 21951.7072       | 17.63               | 0.67         |
| 68      | 21951.73769      | 16.56               | 0.89         |
| 69      | 21951.74103      | 16.46               | 0.02         |
| 70      | 21951.82375      | 13.98               | 0.13         |
| 71      | 21951.8411       | 13.46               | 0.01         |
| 72      | 21951.93483      | 10.65               | 0.27         |
| 73      | 21951.99053      | 8.98                | 0.06         |
| 74      | 21952.04057      | 7.48                | 0.02         |
| 75      | 21952.06192      | 6.84                | 0.02         |
| 76      | 21952.12696      | 4.89                | 0.56         |
| 77      | 21952.16799      | 3.66                | 0.44         |
| 78      | 21952.17933      | 3.32                | 0.02         |
| 79      | 21952.18467      | 3.16                | 0.01         |
| 80      | 21952.2327       | 1.72                | 0.01         |
| 81      | 21952.27373      | 0.49                | 0.02         |
| 82      | 21952.29007      | 0                   | 1.00         |
| 83      | 21952.35212      | -1.86               | 0.02         |
| 84      | 21952.3788       | -2.66               | 0.10         |
| 85      | 21952.40115      | -3.33               | 0.16         |
| 86      | 21952.40916      | -3.38               | 0.06         |
| 87      | 21952.45519      | -4.91               | 0.01         |
| 88      | 21952.51223      | -6.66               | 0.01         |
| 89      | 21952.57827      | -8.64               | 0.01         |
| 90      | 21952.63164      | -10.24              | 0.04         |
| 91      | 21952.69965      | -10.48              | 0.27         |

*Continued...*
| Line no. | Wavenumber(cm⁻¹) | Rel. frequency(GHz) | Rel. strength |
|---------|-------------------|---------------------|--------------|
| 92      | 21952.70703       | -12.5               | 0.02         |
| 93      | 21952.74372       | -13.6               | 0.01         |
| 94      | 21952.8131        | -15.6               | 1.00         |
| 95      | 21952.82011       | -15.89              | 0.16         |
| 96      | 21952.93252       | -19.26              | 0.16         |
| 97      | 21952.98022       | -20.69              | 0.02         |
| 98      | 21953.01991       | -21.88              | 0.01         |
| 99      | 21953.13032       | -25.19              | 0.01         |
| 100     | 21953.16968       | -26.37              | 0.13         |
| 101     | 21953.21838       | -27.83              | 0.01         |
| 102     | 21953.23306       | -28.27              | 0.01         |
| 103     | 21953.24907       | -28.75              | 0.01         |
| 104     | 21953.36148       | -32.12              | 0.01         |
| 105     | 21953.37282       | -32.46              | 0.02         |
| 106     | 21953.39417       | -33.1               | 0.01         |
| 107     | 21953.41852       | -33.83              | 0.04         |
| 108     | 21953.44087       | -34.5               | 0.09         |
| 109     | 21953.48257       | -35.75              | 0.03         |
| 110     | 21953.54361       | -37.58              | 0.16         |
| 111     | 21953.60131       | -39.31              | 0.01         |
| 112     | 21953.61165       | -39.62              | 0.01         |
| 113     | 21953.64868       | -40.73              | 1.33         |
| 114     | 21953.69071       | -41.99              | 0.01         |
| 115     | 21953.69705       | -42.18              | 0.01         |
| 116     | 21953.70939       | -42.55              | 0.03         |
| 117     | 21953.74041       | -43.48              | 0.01         |
| 118     | 21953.74808       | -43.71              | 0.03         |
| 119     | 21953.7791        | -44.64              | 0.19         |
| 120     | 21953.83681       | -46.37              | 0.03         |
| 121     | 21953.89819       | -48.21              | 0.01         |
| 122     | 21953.93555       | -49.33              | 0.04         |
| 123     | 21953.95089       | -49.79              | 0.20         |
| 124     | 21953.95623       | -49.95              | 0.01         |
| 125     | 21954.03161       | -52.21              | 0.02         |
| 126     | 21954.04929       | -52.74              | 0.01         |
| 127     | 21954.05496       | -52.91              | 0.04         |
| 128     | 21954.08565       | -53.83              | 0.02         |
| 129     | 21954.09232       | -54.03              | 0.01         |
| 130     | 21954.12868       | -55.12              | 0.91         |
| 131     | 21954.15136       | -55.8               | 0.03         |
| 132     | 21954.16737       | -56.28              | 0.02         |
| 133     | 21954.17471       | -56.5               | 0.02         |
| 134     | 21954.18472       | -56.8               | 0.89         |
| 135     | 21954.19906       | -57.23              | 0.01         |
| 136     | 21954.20907       | -57.53              | 0.01         |
| 137     | 21954.22875       | -58.12              | 0.07         |
| 138     | 21954.28445       | -59.79              | 0.08         |
| 139     | 21954.56598       | -68.23              | 0.11         |
| 140     | 21954.57365       | -68.46              | 0.02         |
| 141     | 21954.594         | -69.07              | 0.03         |
| 142     | 21954.60434       | -69.38              | 0.03         |
| 143     | 21954.61368       | -69.66              | 0.01         |
| 144     | 21954.65438       | -70.88              | 0.36         |
| 145     | 21954.69407       | -72.07              | 0.89         |
| 146     | 21954.72042       | -72.86              | 0.01         |
| 147     | 21954.75578       | -73.92              | 0.01         |
| 148     | 21954.79481       | -75.09              | 0.04         |
| 149     | 21954.80181       | -75.3               | 0.04         |
| 150     | 21954.80515       | -75.4               | 0.01         |
| 151     | 21954.8305        | -76.16              | 0.01         |
| 152     | 21954.85752       | -76.97              | 0.19         |
**TABLE I – continued from previous page**

| Line no. | Wavenumber(cm\(^{-1}\)) | Rel. frequency(GHz) | Rel. strength |
|----------|-------------------------|--------------------|--------------|
| 153      | 21954.89488             | -78.09             | 0.17         |
| 154      | 21954.97326             | -80.44             | 0.02         |
| 155      | 21955.02096             | -81.87             | 0.17         |
| 156      | 21955.16506             | -86.19             | 0.78         |
| 157      | 21955.19608             | -87.12             | 0.01         |
| 158      | 21955.26713             | -89.25             | 0.01         |
| 159      | 21955.27347             | -89.44             | 0.02         |
| 160      | 21955.28415             | -89.76             | 0.03         |
| 161      | 21955.37721             | -92.55             | 0.01         |
| 162      | 21955.38588             | -92.81             | 0.00         |
| 163      | 21955.39055             | -92.95             | 0.01         |
| 164      | 21955.47328             | -95.43             | 0.01         |
| 165      | 21955.53532             | -97.29             | 0.02         |
| 166      | 21955.557               | -97.94             | 0.06         |
| 167      | 21955.61537             | -99.69             | 0.02         |
| 168      | 21955.68342             | -101.73            | 0.06         |
| 169      | 21955.78583             | -104.8             | 0.56         |

**IV. CALIBRATION OF THE FREQUENCIES**

All the Te\(_2\) lines tabulated in Tab. I have been observed by scanning the laser frequency over 100 GHz on both side of the S\(_{1/2}\)-P\(_{3/2}\) transition (455.4 nm) of barium ion. As mentioned before the whole range is scanned in piece-wise manner. Each piece of measurement is achieved by applying triangular voltage to the ECDL piezo at an amplitude \(\sim 10 - 20\) V about a offset voltage at 20 Hz frequency. The off-set voltage has been changed step-wise to cover the whole range of the frequency scan. The scale of the 100 GHz frequency spectra has been obtained by stitching each neighborhood frequency range having more than one overlapping spectral lines. Within each scan range, about seven Te\(_2\) lines have been observed [12]. Even though the spectral lines are within 7 GHz range, the peizo has been scanned by more than 10 GHz such that non-linearity near the edge of the scan can be avoided. The piezo scanned voltage and the Te\(_2\) lines, both has common time axis and this time axis is converted to frequency using the conversion relation which is given by

\[
f_1 = f_0 + \frac{\Delta f}{\Delta t} \Delta t,
\]

where \(f_1\) is the unknown frequency and \(f_0\) is the calibrated frequency. The slope \(\frac{\Delta f}{\Delta t}\) is the rate of frequency scan as obtained from the linearity fit of scanned voltage-time measurement and \(\Delta t\) is the time difference between the resonances \(f_0\) and \(f_1\) respectively. This linear transformation holds provided the piezo scanning voltage is linear with time, otherwise non-linearity has to be taken into account. Therefore the range of scanned voltage is selected such that a linear fitting shows a \(\chi^2\) \(\sim 0.99\). The first piece of the scan range contains a well known barium resonance line S\(_{1/2}\)-P\(_{3/2}\) which is used as \(f_0\) as in eq. (1). This frequency then acts as the absolute frequency for the frequency calibration. The frequency of the rest of the Te\(_2\) lines are determined by successive use of eq. (1). Thus the uncertainty with which any of the resonance center frequencies can be measured is thus determined by the uncertainty of the rate of frequency scan, the accuracy in time with which each resonance zero crossing can be determined, and the uncertainty of the frequency measurement \(\delta f_0\). The uncertainty on the rate as obtained from the linearity as well as the uncertainty on the time measurement is negligible as compared with the \(\delta f_0\), which is 30 MHz given by the resolution of the wavemeter.

In order to verify the correctness of this spectral stitching method, the frequency of almost all the lines have been measured individually by actively locking the blue laser to the corresponding Te\(_2\)-line and these are compared with our calibrated data. Even though, these lines are 100 GHz away from the barium line, the frequencies match very well within the uncertainty dominated by the wavemeter uncertainty. The known Te-lines found in the Atlas [1] are compared with our measured data. In particular, the line numbers 3, 6, 15, 20, 31, 34, 43, 58, 67, 78, 82, 94, 105, 113, 130, 134, 145, 154, 156, 162 and 169 in the above table have been identified as the line numbers staring from 1667 to 1687 respectively in the Atlas data. The measured frequencies of the line numbers 31, 43, 67, 82, 113, 130, 162 and 169 are found to be in good agreement with the Atlas data within the wavemeter uncertainty whereas for the rest of the matched lines, the difference in frequency varies from 40 – 100 MHz.
FIG. 2: The modulation transfer spectroscopy signal of a sample resonance line (line no. 83 in the Table I) as a function of the laser frequency. The * denotes the experimental points along with the error bar while the solid line is a fit of the line-shape function with a reduced $\chi^2 \sim 0.99$. 

V. LINE-SHAPE AND ERROR DETERMINATION

The line center of individual resonance lines and their uncertainties has been obtained from the zero-signal crossing of the MTS spectrum. Figure 2 shows one of the resonance spectrum fitted with a line-shape function. The line shape of a MTS resonance is devoid of any background slope unlike frequency modulation spectrum. This shape can be described by a theoretical lineshape function as shown in [18].

The best fit provides the linewidth to be 20.9(4) MHz for the line shown in fig. (2). Thus with an electronic suppression (about 100) it is possible to frequency lock the laser with a bandwidth of a few hundred kHz for almost all lines shown in this work.

VI. SUMMARY AND DISCUSSION

In this article we provide more than 100 measured new resonance lines of the Te$_2$ molecule beyond the known lines from the Atlas with a frequency uncertainty of 30 MHz over a range of 200 GHz. This is particularly important in terms of quantum optics, communication and computing experiments as most of the cooling lasers lies in the shorter wavelength range of the visible spectrum. In terms of laser frequency locking this range covers atomic resonance lines of atoms and ions including barium.

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