

**Femtosecond comb
measurements of the
467 nm electric octupole
transition in $^{171}\text{Yb}^+$**

**Quantum Programme 2001-2004
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Abstract

The frequency of the $^2\text{S}_{1/2}, F=0 \rightarrow ^2\text{F}_{7/2}, F=3, m_F=0$ transition at 467 nm in a single ion of $^{171}\text{Yb}^+$ has been measured with a mode-locked femtosecond Ti:Sapphire laser over the period August 2001 – July 2003. The current best estimate of the transition frequency is $642\,121\,496\,772.3 \pm 0.6$ kHz.

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Authorised for publication on behalf of the Managing Director of NPL by
Prof. Patrick Gill

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Chapter 1

Femtosecond comb measurements of the 467 nm electric octupole transition in $^{171}\text{Yb}^+$

1.1 Introduction

Mode-locked femtosecond lasers have revolutionized the measurement of absolute optical frequency, enabling reference to a locally-realized S.I. second in a single step. A program to develop femtosecond comb capability has been underway at NPL since 2001, and several optical frequency standards have been measured [1, 2, 3, 4]. This paper reports on femtosecond comb measurements made of the 467 nm electric octupole transition in $^{171}\text{Yb}^+$. This extremely forbidden transition makes for an excellent frequency reference and has the advantage that in practice its Q will not be intrinsically limited by the lifetime of the excited state, which is ~ 6 years.

Firstly, the most recent measurements made during the period May – July 2003 are presented. These have the smallest uncertainty of the measurements presented here due to the narrow frequency width of the probe laser. Secondly, these are compared with earlier measurements of 2001 and 2002 which were made with a broader probe laser. Finally, we come to a conclusion on the best estimate and uncertainty that can be put on the transition frequency at present.

1.2 Narrow Linewidth Measurements, May–July 2003

The experimental arrangement and procedure is described in [4], so here we will only mention the changes that have since been made. The repetition rate of the comb is locked to the 10 MHz output of a maser which forms part of the clock ensemble used to generate UTC(NPL). Previously, the carrier envelope offset frequency was simply counted with a frequency counter referenced to the same 10 MHz, whereas now it is also locked to that reference. Improvements in the locking electronics now mean that a counter gate time of 10 s is used rather than 1 s which reduces the time needed to achieve a given statistical uncertainty in the frequency measurement. Figure 1.1 is a schematic of the probe laser system. The ULE cavity is now supported on a

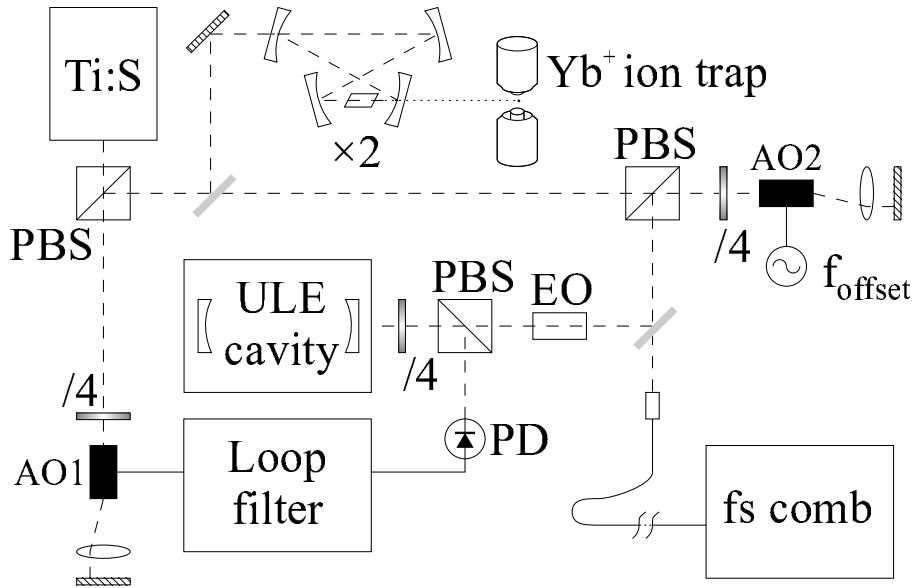


Figure 1.1: Schematic of experimental setup

passive vibration isolation platform [5] and is situated within an acoustic isolation box. This has improved the stability of the cavity with the result that the linewidth of the probe laser has been reduced to < 200 Hz, compared to 4.5 kHz without vibration or acoustic isolation [6]. Figure 1.2 shows a quantum jump spectrum of the $^2S_{1/2}$ – $^2F_{7/2}$ transition.

At present, the ac Stark shift is the largest systematic shift, due to the high light intensity required to drive the octupole transition. Previously, the probe beam was focused to a spot size of $5\ \mu\text{m}$ and typically a power of 1 mW for a pulse duration of 300 ms was required to excite the transition with 50 % probability. Now that the linewidth has been reduced and the spectral intensity increased, it has been possible to de-focus the probe beam by 1–2 Rayleigh ranges to reduce the total intensity whilst maintaining the same transition rate. Having a larger spot size at the ion reduces errors due to drifts in the alignment. Rather than making an absolute measurement of the intensity experienced by the ion, the frequency is measured as a function of the probe power and then extrapolated to zero power to obtain

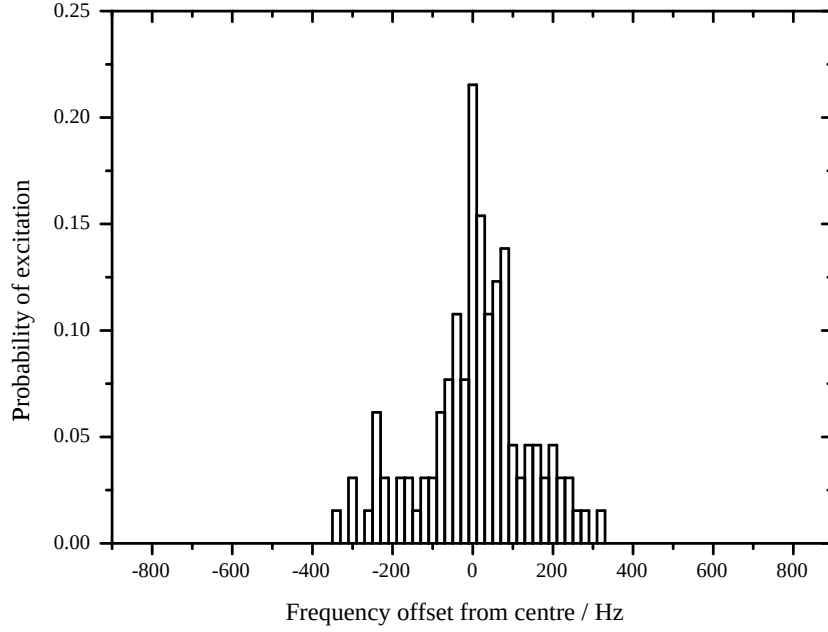


Figure 1.2: Spectrum of the $^2S_{1/2}$, $F=0 \rightarrow ^2F_{7/2}$, $F=3$, $m_F=0$ transition in $^{171}\text{Yb}^+$. A Gaussian fit has a FWHM of 177 ± 10 Hz.

the absolute frequency. The spot size and focusing arrangement remain fixed and alignment on the ion is checked periodically and re-optimized if necessary.

Figure 1.3 shows the result of measurement over three separate days in May 2003. Each data point is typically the mean of 15-20 quantum jumps taken in a single scan over a period of 5 mins. The frequency of light locked to the ULE cavity is continually measured by the femtosecond comb which in combination with the frequency offset applied to AO2 (see Fig. 1.1) determines the absolute frequency of the probe laser at each quantum jump. The vertical error bars represent the standard error on the mean of the data obtained during a single scan. The horizontal error bars represent the uncertainty due to power drift, evaluated by measuring the power before and after a scan, plus an estimate of the alignment uncertainty. Extrapolation to zero power with a weighted least-squares fit, gives a transition frequency 642 121 496 772.66 kHz. From here on, for the purpose of brevity, we will simply use the frequency difference from 642 121 496 770 kHz, so for example, the number just given is written 2.66 kHz. The uncertainty on the intercept, calculated from the uncertainties ascribed to each point is 0.02 kHz. Judging from the spread of data, this appears to be an underestimate and indeed a χ^2 test reveals that the distribution is far from being statistical and that type-B [7] errors dominate. To perform this test, the horizontal error bars are converted into effective vertical error bars, by multiplying them by the slope of the weighted fit and adding them in quadrature to the existing vertical error. The reduced χ^2 is 3.4, which for 27 degrees of freedom is highly improbable and therefore is clearly non-statistical. It would appear then that either the known systematic errors (power, alignment) have been under-estimated, or fluctuation in another unaccounted-for systematic shift is

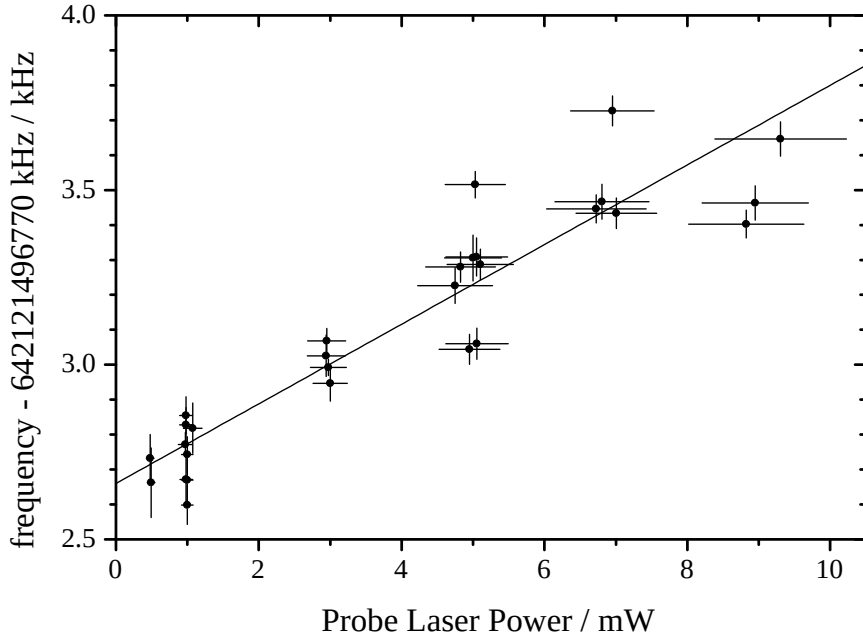


Figure 1.3: Frequency of the octupole transition as function of probe power, May 2003. The solid line is a weighted fit to the data with slope $0.11(1) \text{ kHz (mW)}^{-1}$ and intercept $2.66(2) \text{ kHz}$.

influencing the transition frequency. Either way, using the weighted fit and simply taking the error on the intercept is unjustified. An alternative presentation corrects for the Stark shift by subtracting from each point the value of the weighted fit to the data, as shown in Fig. 1.4 for the May data. One can see from this that the scatter of the data is large relative to the size of the error bars. The most conservative estimate treats the distribution as being rectangular, with a standard error, $(\text{semi-range})/\sqrt{3}$, and uses the intercept of an unweighted fit. This gives a revised value, $2.67 \pm 0.16 \text{ kHz}$.

Figure 1.5 shows the result of a measurement made on a single day in July 2003. Extrapolation to zero power with a weighted fit, gives a transition frequency 2.63 kHz . The calculated uncertainty on the intercept is 0.03 kHz , but again the distribution is non-statistical. The reduced χ^2 is 4.2 which, for 21 degrees of freedom is highly improbable. Treating the distribution as rectangular and using the intercept of an unweighted fit to the data gives a revised value, $2.52 \pm 0.21 \text{ kHz}$.

The second largest systematic shift acting on the transition frequency is the second-order Zeeman shift and this needs to be taken into account in order to obtain the unperturbed transition frequency. Figure 1.6 shows frequency as a function of the square of the magnetic field. A fit to this yields a coefficient of $-1.72 \pm 0.03 \text{ mHz } (\mu\text{T})^{-2}$, in good agreement with a previous determination [6]. The data in figures 1.3 and 1.5 were taken at a field of $188(4) \mu\text{T}$, therefore a correction of 0.06 kHz must be added. The only other shift that needs to be taken into account is an offset in the maser frequency to which the comb is referenced, leading to a correction of -0.03 kHz . Other systematic shifts have been evaluated in detail [8]

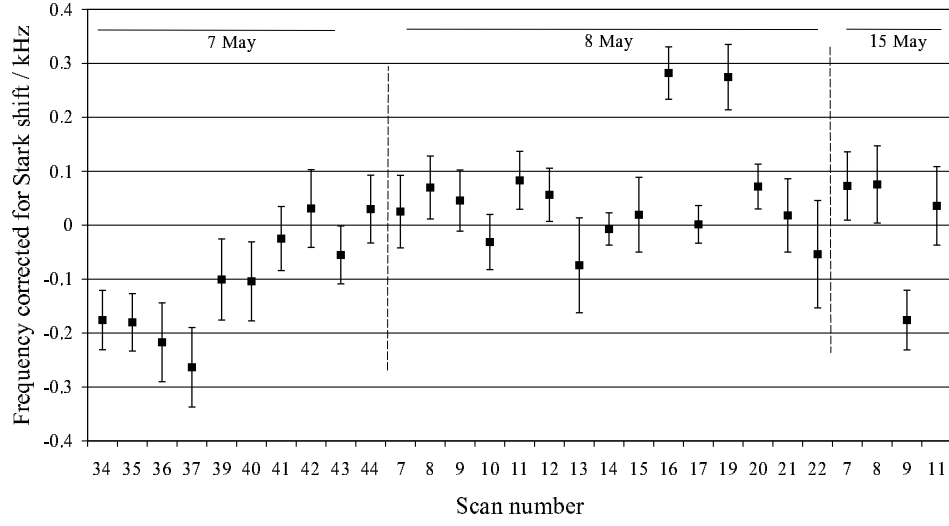


Figure 1.4: Frequency of the octupole transition corrected for Stark shift and with data points plotted sequentially, May 2003.

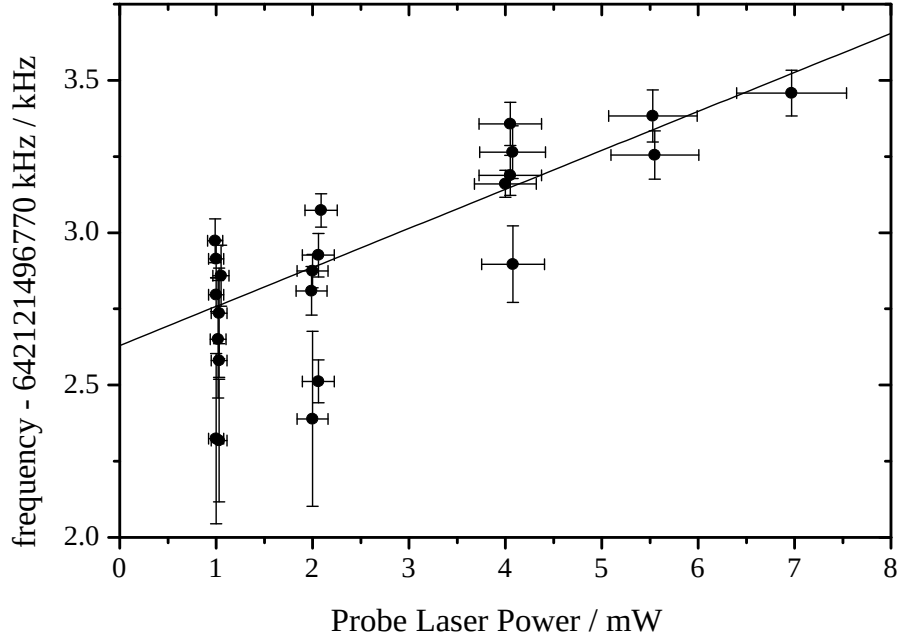


Figure 1.5: Frequency of the octupole transition as function of probe power, July 2003. The solid line is a weighted fit to the data with slope $0.13(1) \text{ kHz (mW)}^{-1}$ and intercept $2.63(3) \text{ kHz}$.

but are negligible at this level. The corrected frequencies for the measurements in May 2003 and July 2003 are

$$2.69 \pm 0.16 \text{ kHz}$$

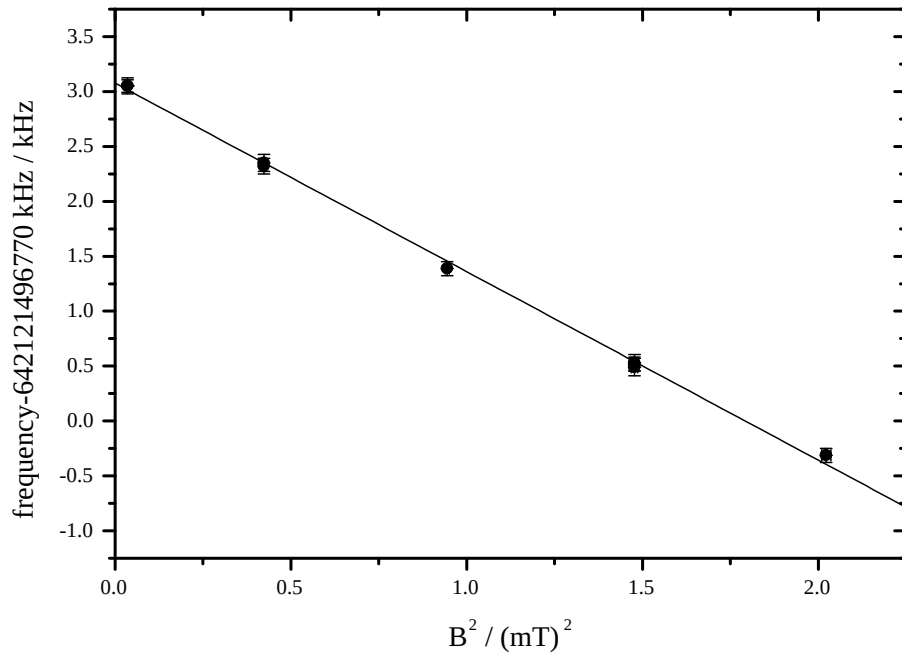


Figure 1.6: Frequency of the octupole transition as function of the square of the magnetic field. The solid line is a weighted fit to the data with slope $-1.72(3) \text{ mHz } (\mu\text{T})^{-2}$. $\chi^2 = 4.1$, which for 7 degrees of freedom has a probability, $P(\chi^2 > 4.1) = 0.14$.

and

$$2.55 \pm 0.21 \text{ kHz}$$

respectively and are in good agreement with each other.

1.3 Comparison with earlier frequency measurements

Measurements were also made with the femtosecond comb in August 2001 and March 2002. The probe laser at these times was broader having a FWHM of 4.5 kHz. In August 2001 a value for the transition frequency of 2.6(1.3) kHz was obtained [9]. Here measurements were made on a single day and at just one probe power, therefore the Stark shift had to be corrected for by making an estimate of the absolute intensity incident upon the ion. A large uncertainty in measuring the spot size and hence intensity at the ion led to a $\sim 30\%$ uncertainty in the Stark shift which accounts for the relatively large error bar on this measurement. In March 2002, data were taken over four days and at various powers allowing for extrapolation to zero power and leading to a more precise determination of 1.26 ± 0.23 kHz [4].

There is good agreement between the most recent values and that obtained in 2001. However, there is clearly a discrepancy with the value obtained in 2002. The recent values are more than 6σ away from this result. This leads one to look very carefully over the measurements and corrections made.

1.3.1 Re-analysis of March 2002 data

On re-examination of the March 2002 data an error was identified in the Zeeman shift correction applied, which was traced back to an incorrect field calibration. It was thought that the applied field was $310 \mu\text{T}$, however, a factor of two error in the calibration of the field (originating from an incorrect calculation of the Lande g_F -factor) means that the actual field applied was $620 \mu\text{T}$. Due to the quadratic dependence of the second-order Zeeman shift on magnetic field, this led to a factor of four error in the Zeeman shift correction applied. Another smaller correction should also be applied due to the new evaluation of the Zeeman shift coefficient. This has now been determined as $-1.72 \pm 0.03 \text{ mHz } \mu\text{T}^{-2}$ compared to $-1.9 \pm 0.3 \text{ mHz } \mu\text{T}^{-2}$, which was the best known value at the time of publication. The correction applied was $+185(34) \text{ Hz}$, but in the light of the field calibration error and the new value for the Zeeman shift coefficient, this should have been $+655(35) \text{ Hz}$. Adding the difference between these two corrections to the frequency gives a number $1.73(23) \text{ kHz}$. The error was also present in the analysis of the August 2001 data, and correcting for this gives a number of $3.1(1.3) \text{ kHz}$. This goes some way to explaining the large discrepancy, however, the 2003 and 2002 numbers are still different by 4σ .

In the analysis of the 2003 data it is clear from the χ^2 of the weighted fit that the size of type-B errors has been underestimated, therefore the analysis of the 2002 data was reviewed to see if the uncertainties had been underestimated there. The graph from [4] is reproduced in Fig. 1.7. Here the data were grouped according to day and power. The reduced χ^2 value for the fit came out as 1.37, which for 7 degrees of freedom has a probability, $P(\chi^2 > 1.377) = 0.21$. As such there was insufficient evidence for underestimation of the uncertainty. The major source of systematic uncertainty is the stability of the alignment of the tightly focused probe beam on the ion. This alignment was re-optimized several times during each day. On reflection, a more appropriate grouping of the data is by alignment, because

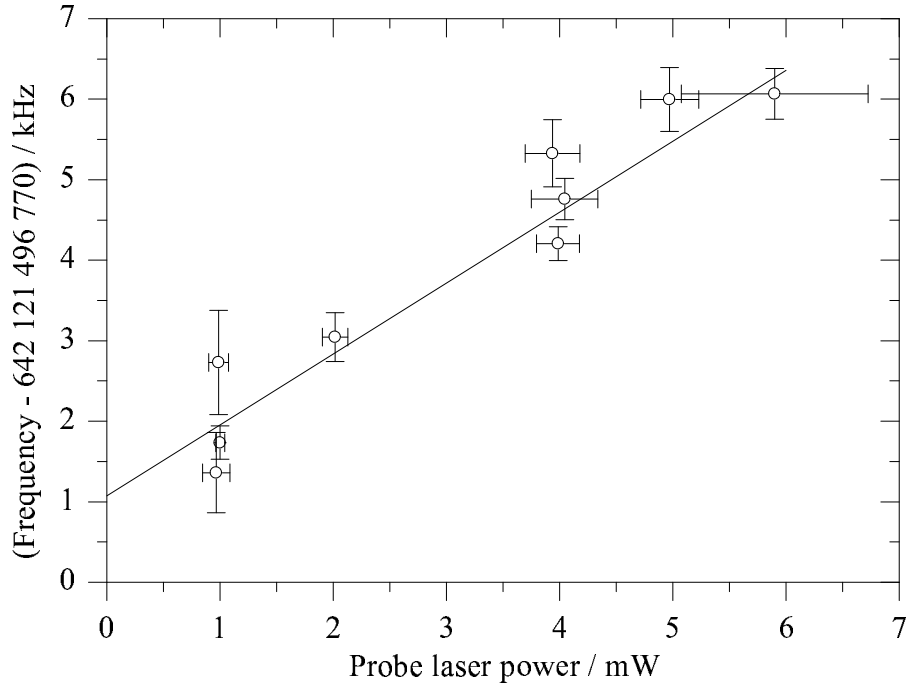


Figure 1.7: March 2002 data reproduced from [4].

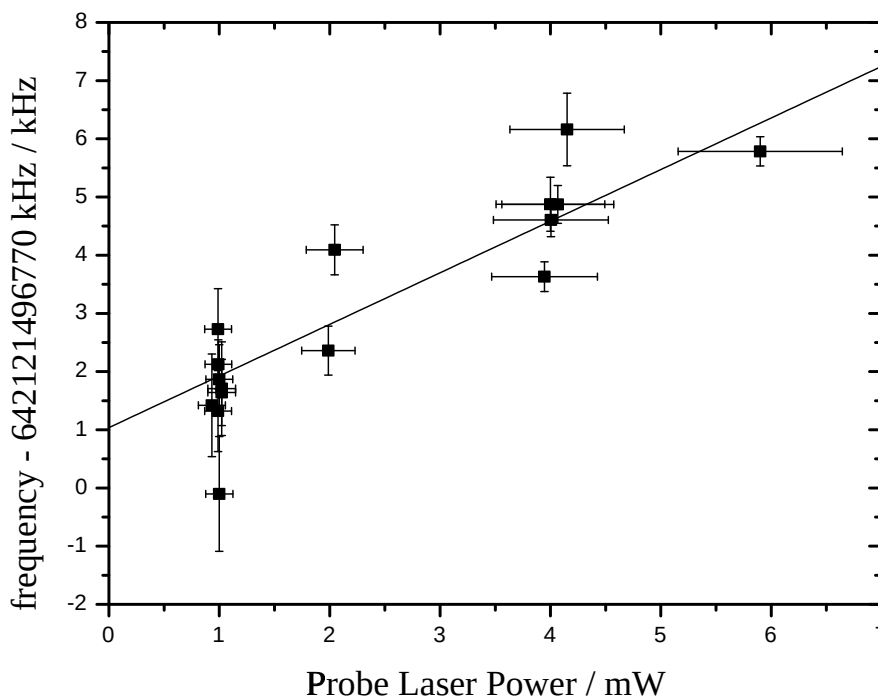
the horizontal error bar assigned to each point is largely due to this alignment uncertainty. Figure 1.8 shows the 2002 data plotted as a function of probe power and grouped according to alignment. Also, the data from the first day of measurement has been omitted because the experimental parameters were not as well controlled as they were on subsequent days. The uncertainty in the intercept is 280 Hz which is a little larger than before. The reduced χ^2 comes out as 1.58, for which the probability, $P(\chi^2 > 1.58) = 0.08$ with 14 degrees of freedom, so there is some evidence of non-statistical behaviour but not enough to suggest further expansion of the error bars. Correcting the frequency for the Zeeman shift and maser offset gives a value

$$1.69(28) \text{ kHz}$$

which is offset from the May 2003 value by just over three times their combined uncertainty.

1.3.2 Further experiments in May 2003

Further measurements were made in May 2003 to try and reproduce the conditions in March 2002. Initially the 2003 measurements were taken with the probe laser de-focused by 1-2 Rayleigh ranges to reduce sensitivity to probe alignment, whereas in the 2002 measurement the focus of the probe laser was positioned at the ion to maximize the excitation rate. The probe beam was therefore, repositioned with its focus at the ion, thus increasing the intensity and therefore the sensitivity of the frequency to alignment. Secondly, the acoustic isolation box was removed and the vibration isolation reduced so that the probe linewidth was broadened back to ~ 2 kHz. The probe laser remained focused. Figure 1.9 shows the measurements



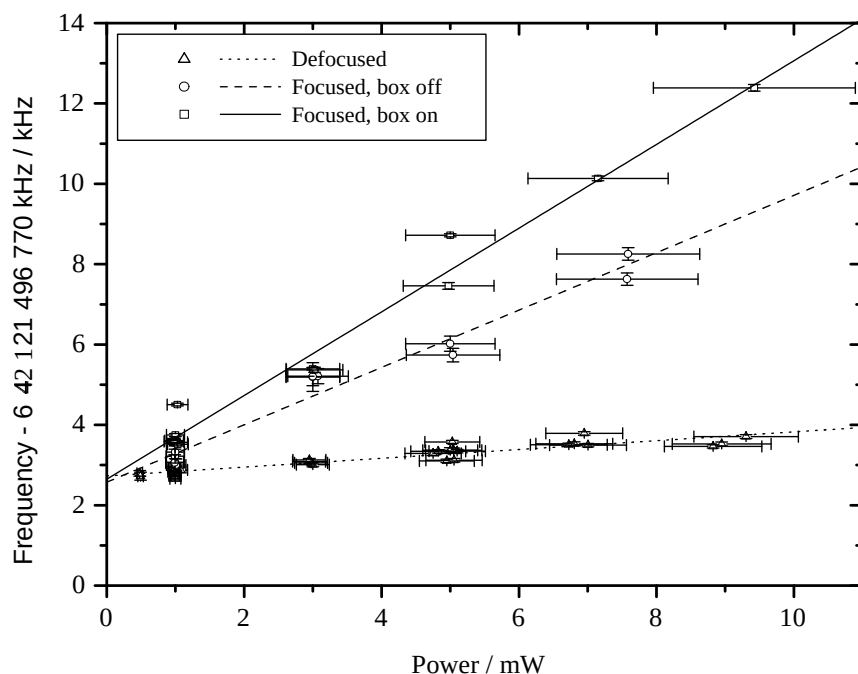


Figure 1.9: Stark shift measurements under different conditions, May 2003. The lines are unweighted fits and the intercepts are 2.72(15), 2.65(37) and 2.57(27) kHz for the de-focused, focused and “box-off” data respectively. The uncertainties for the data were derived by treating the distribution of the residuals as rectangular, due to the large χ^2 values of the fits.

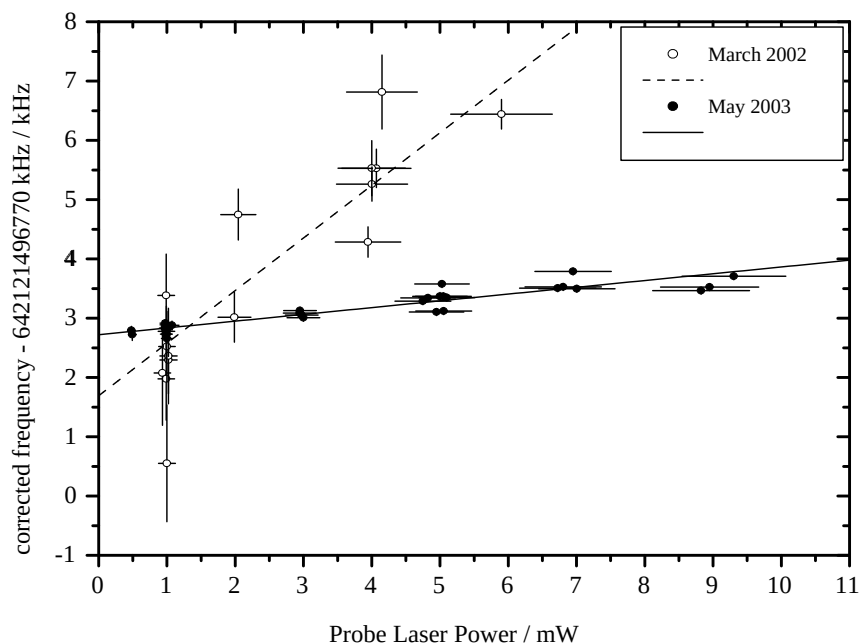


Figure 1.10: Comparison between taken in March 2002 and May 2003.

1.4 Conclusion

The values determined as a result of the four measurement periods have been plotted together in Fig. 1.11. Two aspects of the data are worthy of note. These are, firstly, the uncertainties assigned to the four points and secondly, the spread in the values of the points relative to these uncertainties.

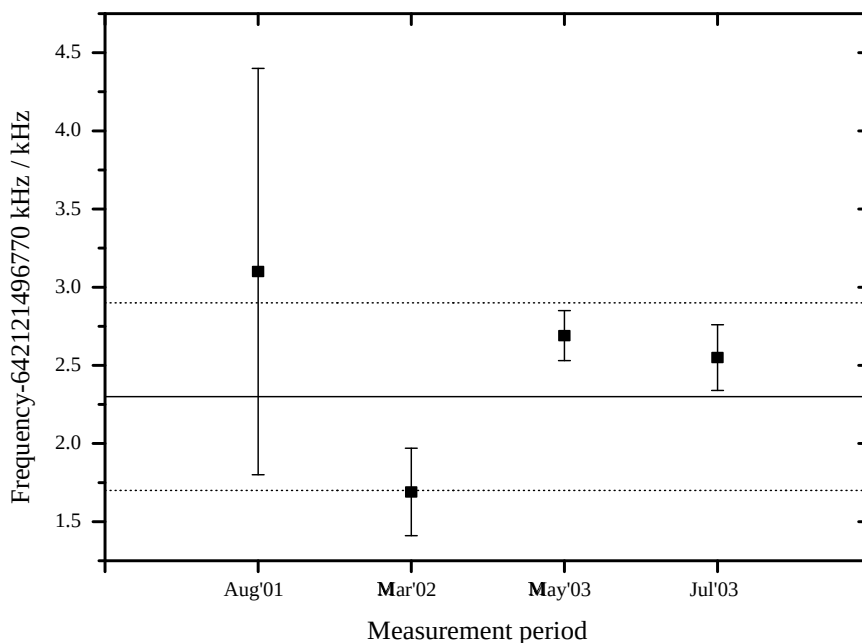


Figure 1.11: Comparison of frequency measurements made in the period August 2001 – July 2003. The solid line is the unweighted mean of the three last points and the dotted lines indicate the frequency plus and minus one standard uncertainty.

The first point has a much larger uncertainty than the other three because the measurement method differed in one vital respect: an absolute measurement of intensity was made to determine the Stark shift rather than a calibration of the Stark shift with power as was done in the latter three measurements. The spot size of the probe beam could only be measured with relative inaccuracy due to its small size ($\sim 25 (\mu\text{m})^2$), therefore a calculation of the intensity based on this area was also relatively uncertain (to within $\sim 30\%$). Calibrating the Stark shift as a function of probe power removes the need to measure the spot size, and as a result the measurements made by this method have smaller uncertainties. The final two points were made with a narrower linewidth probe laser and with a de-focused beam (1-2 Rayleigh ranges). One would therefore expect the uncertainty to decrease in line with the reduction in linewidth (from 4.5 kHz to < 200 Hz), not only because of reduced statistical uncertainty, but also due to reduced systematic uncertainty associated with alignment. The uncertainties on individual scans made over short (~ 5 min.) time-scales did reflect this, however, over a period of a day the scatter in data was much larger than the estimated uncertainty (see section 1.2). This meant

that we could not simply take the weighted mean of the data, but instead the range of data was used to estimate the uncertainty, leading to much larger (~ 200 Hz) error bars. This brings one to consider the scatter of the four measurements plotted above.

There is a significant difference between the second and final two points, indicating variation of a systematic effect which is not taken into account by the estimated uncertainties. It is unlikely that this scatter is due to the Stark shift as this has been carefully controlled and characterized in all the experiments. Indeed, an experiment was performed in which the intensity was increased by a factor ~ 10 , but this did not make a significant difference to the result obtained (see section 1.3.2). Other systematic effects have been evaluated [8], but are at a much smaller level than the extent of the observed variation. Therefore, we cannot account for it in terms of known systematics and the nature of this variation is yet to be understood. Further measurements will be carried out to see if variation continues to occur or if some consistency can be attained. In that it is the largest known systematic, one still suspects that somehow the stability of the Stark shift has been underestimated and that its variation is partly unaccounted for. Therefore, we will make efforts to control the intensity experienced by the ion more rigorously. The beam-pointing stability of the probe-beam will be improved to reduce errors due to alignment drift. As the linewidth is reduced further and the spectral intensity of the probe laser is increased, it will be possible to reduce the intensity even more whilst maintaining the same quantum jump rate. This can be done by further expansion of the spot size at the ion, reducing the alignment sensitivity still more.

To arrive at the best estimate of the transition frequency given the current state of knowledge, we take the unweighted mean of the final three points. The first point is excluded on the grounds that it is an unreliable estimate of the variation in the data, due to its large uncertainty. The uncertainty is estimated from the range of the last three points. For three observations, one standard uncertainty is calculated by multiplying the range by the factor 0.591 [10]. With a range of 1.0 kHz this gives a standard uncertainty of 0.6 kHz. Our best estimate of the frequency of the 467 nm electric octupole transition in $^{171}\text{Yb}^+$ is therefore,

$$642\,121\,496\,772.3 \pm 0.6 \text{ kHz}.$$

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