Precise measurements of atomic energy levels: 

*D lines and fine-structure interval in K*

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We use a diode laser locked to a Rb transition as the frequency reference, along with a scanning Michelson interferometer, to make precise frequency measurements of atomic energy levels. The high precision is possible because the Rb reference frequency is known with submegahertz accuracy. We measured the frequencies of the *D* lines in K and obtained the following values: 12 985.169 60(20) cm$^{-1}$ for the 4$P_{3/2}$ state ($D_1$ line), 13 042.875 95(21) cm$^{-1}$ for the 4$P_{5/2}$ state ($D_2$ line), and 57.706 35(24) cm$^{-1}$ for the 4$P_{3/2}$-4$P_{1/2}$ fine-structure interval. The errors represent approximately an order-of-magnitude improvement over tabulated values.

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Semiconductor diode lasers bring several advantages to the field of laser spectroscopy, the principal one of which is tunability in a relatively small, low-cost package.1 By placing a single-mode diode in an external cavity and using optical feedback from an angle-tuned grating,2 one can make the diode operate at single frequency (single longitudinal mode) with a linewidth of the order of 1 MHz and tunability over several nanometers. In addition, techniques such as saturated-absorption spectroscopy with counterpropagating pump and probe beams can help to eliminate the first-order Doppler effect and permit narrow hyperfine transitions within a given atomic line to be resolved. It is thus possible to get an absolute frequency calibration of a laser by locking to such a transition. Recently, Banerjee et al. showed that a diode laser locked to an atomic transition in Rb can be used along with a scanning Michelson interferometer as a precision wavemeter with which to determine the unknown frequency of a tunable laser.3

In this paper we report using this technique to determine the absolute values of atomic energy levels with an order-of-magnitude greater precision than tabulated values.4 This level of precision is made possible because the absolute frequency of the Rb line used as the reference is known with submegahertz accuracy.5,6 Using this technique, we measured the absolute frequencies of the *D* lines in K with approximately megahertz accuracy. We also determined the fine-structure interval in the 4$P$ state of K. Knowledge of fine-structure intervals is useful in the study of atomic collisions and for relativistic calculations of atomic energy levels. In addition, this technique is applicable to other alkali atoms such as Li and Na, to alkali-like ions, and indeed to any system in which the precision of the tabulated values of energy levels is insufficient.

The interferometer used for the ratio measurement was described extensively in a previous publication.3 For consistency with the terminology there, we call the two lasers “reference” and “unknown.” The basic idea is to obtain the ratio of the two laser wavelengths by using a scanning Michelson interferometer in which both lasers traverse essentially the same path. The reference laser is a frequency-stabilized diode laser tuned to the 780-nm *D*$_2$ line in atomic Rb. A part of its output beam is tapped for Doppler-free saturated-absorption spectroscopy in a Rb vapor cell. The injection current into the diode is modulated slightly to produce an error signal, and the laser is locked to the 5$S_{1/2}$, $F = 2$ → 5$P_{3/2}$, $F' = (2, 3)$ crossover resonance. The frequency of this resonance was measured previously to be 384 227 981.877(6) MHz.5 We have shown that the laser is locked to the peak center with an uncertainty of less than 50 kHz.7 The unknown laser is tuned to the *D* lines of K.

The relevant energy levels of $^{39}$K are shown in Fig. 1. The $D_1$ line is the 4$S_{1/2}$ → 4$P_{1/2}$ transition at 770.1 nm, and the $D_2$ line is the 4$S_{1/2}$ → 4$P_{3/2}$ transition at 767.7 nm. The *D* lines of K are accessed with a single-frequency tunable Ti:sapphire laser (Coherent 899-21). The laser can be tuned from 700 to 800 nm and is frequency stabilized to a temperature-controlled reference cavity that gives it an instantaneous linewidth of 500 kHz. The long-term drift of the laser is only a few megahertz per hour; therefore it is not necessary to stabilize the laser against this drift during the course of the experiment. A part of the laser output is split off for saturated-absorption spectroscopy in a K-vapor cell maintained at a temperature of ~70 °C. After every 5–10 measurements the laser frequency is checked against the saturated-absorption spectrum to ensure that it has not drifted from a given peak.

The saturated-absorption spectra on the $D_1$ and $D_2$ lines for transitions starting from the $F = 2$ ground level are also shown in Fig. 1. The two hyperfine transitions in the $D_1$ line are well resolved. This is so because the hyperfine interval in the 4$P_{1/2}$ state is ~60 MHz, whereas the linewidth in the saturated-absorption spectrum is 20 MHz. The observed linewidth is somewhat larger than the natural linewidth of 6.1 MHz. The primary causes for this increase are misalignment between the counterpropagating beams and power broadening that is due to the pump beam. Collisional broadening in the vapor
The saturated-absorption spectrum on the $D_2$ line presents a slight problem because all the hyperfine levels in the $4^P_{3/2}$ state lie within a range of 30 MHz, and the individual transitions are not resolved. To solve this problem we performed computer simulations of the six transitions within each spectrum. The locations of the peaks are fixed according to the known hyperfine intervals, and the peak heights correspond to those obtained in saturated-absorption spectroscopy. The linewidths of all the transitions are assumed to be the same.

A typical spectrum for $F = 2 \to F'$ transitions is shown in Fig. 2 (open circles); the solid curve is a fit to the measured spectrum. We found that for the best fit the individual linewidths must be $\pm 22$ MHz, which is consistent with the linewidth measured for the $D_1$ line where the transitions are resolved. Using a smaller linewidth for the calculated spectrum leads to a change in the line shape as individual transitions get resolved, whereas using a larger linewidth leads to an increase in the overall linewidth beyond the measured value. With the best fit, we find that the peak in the spectrum is $-2.3$ MHz below the $F = 2 \to F' = (2, 3)$ crossover resonance, and we use this value to determine the center of gravity of the line. The location of the peak is insensitive to slight variations in the heights of transitions that might be caused by intensity variations in the beams. The calculated spectrum, assuming that the linewidth is the natural linewidth of 6.1 MHz, is also shown in Fig. 2. The line shape is clearly different from the measured spectrum, but the location of its peak is only 2 MHz higher. Thus we are confident that errors in our model do not significantly affect the calculation of the center of gravity.

For transitions starting from the $F = 1$ ground level, similar mod-

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**Fig. 1.** Energy levels of $^{89}$Kr. The relevant energy levels of $^{89}$Kr in the ground 4S state and the first excited 4P state are shown. The various hyperfine levels are labeled with the values of the total angular momentum $F$, and the number on each level is the energy displacement (in megahertz) from the unperturbed state (from Ref. 11). The two insets at the left are saturated-absorption spectra on the $D_1$ and $D_2$ lines for transitions starting from the $F = 2$ ground level.

**Fig. 2.** Line shape of the saturated-absorption spectrum for $F = 2 \to F'$ transitions. Probe detuning is measured from the center of gravity of the line. The measured spectrum, shown as open circles, is a convolution of six peaks. The solid curve is a fit to the six peaks with the linewidth as a fit parameter. The best fit yields a linewidth of 22.3(7) MHz. The dotted curve is the expected line shape if the linewidth is the natural linewidth of 6.1 MHz. The peak of the spectrum is shifted from the center of gravity by $+1.5$ MHz for the solid curve and by $+3.5$ MHz for the dotted curve.
The errors are statistical.

We extracted the hyperfine-free energy in vacuum by first correcting for the various hyperfine transitions of the $D_1$ frequency of $384,227,981.877(6)$ MHz. The unknown laser was tuned to the $D_2$ lines in $^{39}$K, as listed. We extracted the hyperfine-free energy in vacuum by first correcting for the dispersion of air and then removing the hyperfine shifts shown in Fig. 1. The errors are statistical 1σ deviations.

The difference in the measured ratio given by $1/cos \theta$, where $\theta$ is the angle between the beams. We try to minimize this error in two ways: In the first method we check for parallelism over a distance of $\sim 3 \text{ m}$ by using the unused output beam of the interferometer as a tracer for aligning the other beam. In the second method we look for a minimum in the measured ratio as the angle of the unknown beam is varied. This works because the measured value is always larger than the correct value, whether $\theta$ is positive or negative, and becomes minimum when $\theta = 0$. We have found this method to be the more reliable, and it quickly guarantees that the beams are perfectly aligned.

As a further check on possible systematic errors in our measured frequencies we repeated the measurement scheme with a different reference laser. This set of measurements was made with a reference diode laser locked to a transition in the $D_1$ line of Rb at 795 nm. The diode laser system is identical to the first one, except that after stabilization it is locked to the $5S_{1/2}, F = 3 \rightarrow 5P_{1/2}, F' = 3$ hyperfine transition in $^{85}$Rb. The frequency of this transition is $377,106,271.6(4)$ MHz. Because this frequency is also known with submegahertz accuracy, we do not expect any decrease in the precision of the measurements. However, because the reference laser is different from that used in the first set of measurements, it requires completely new alignment of the interferometer, which gives us a good check on systematic errors that arise from any misalignment angle between the beams. More importantly, it gives us a check on the reliability of converting wavelength ratios in air to vacuum by use of Edlen’s formula because the reference wavelength enjoys a change from 780 to 795 nm.

The results of the second set of measurements are listed in Table 2. The values for the $D_1$ and $D_2$ lines of K are consistent with the earlier results (Table 1), within the statistical errors. Therefore we are confident that the systematic errors are no larger than the statistical errors. Combining the results from Tables 1 and 2, we obtained the following average values:

$$4P_{1/2} = 12,985.169 \pm 60(13)(15) \text{ cm}^{-1},$$

$$4P_{3/2} = 13,042.875 \pm 95(14)(15) \text{ cm}^{-1},$$

and a precise value for the fine-structure interval:

$$4P_{3/2} - 4P_{1/2} = 57,706.35(19)(15) \text{ cm}^{-1}.$$
The errors are statistical and systematic, respectively. The combined error for the energy levels is \( \sim 0.000 \ 20 \ \text{cm}^{-1} \). Note that the systematic error in the determination of the interval is only \( 0.000 \ 15 \ \text{cm}^{-1} \) because the error that arises due to nonparallelism will shift both lines equally and will cancel in the difference.

The values that we obtain can be compared to the values listed in the K energy-level tables released by the National Institute of Standards and Technology: \( 12 \ 985.170 \ \text{cm}^{-1} \) for the \( 4P_{1/2} \) state; \( 13 \ 042.876 \ \text{cm}^{-1} \) for the \( 4P_{3/2} \) state; and \( 57.706 \ \text{cm}^{-1} \) for the fine-structure interval. Our results are consistent with these values but have significantly higher accuracy, thus demonstrating the power of this technique to improve the accuracy of the existing energy-level tables.

In conclusion, we have used a diode laser stabilized on a Rb transition as an absolute frequency reference to make precise measurements of the energy levels in K. We fed the reference laser and a laser tuned to the atomic transition of interest into a scanning Michelson interferometer to obtain their wavelength ratio. We used this technique to make measurements of the \( D_1 \) and \( D_2 \) lines of K and obtained improvement of \( \sim 1 \) order of magnitude over existing values. Because atomic energy levels are generally known to \( \sim 0.01-\text{cm}^{-1} \) accuracy, this technique has the potential to improve these values by more than an order of magnitude.

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4. The latest energy levels are available from the National Institute of Science and Technology Website: http://physics.nist.gov/PhysRefData.