Hyperfine Structure of the Metastable $^5S_2$ State of ^{17}O Using an AlGaAs Diode Laser at 777 nm

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By exploiting a narrow-linewidth diode-laser source we measure the hyperfine structure of the $^5S_2$ state of ^{17}O. Nuclear parameters can be calculated from the measured hyperfine structure. Recorded collision-free linewidths allow an estimate of the lifetime of the levels involved in a new scheme proposed for the cooling of atomic oxygen.

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Nuclear effects in the spectrum of atomic oxygen are of great interest. Recently, a possible nuclear volume effect on the isotope shift of optical transitions was observed.[1] This effect is usually negligible in light elements but it is enhanced in this case by the "doubly magic" structure of the ^{16}O nucleus which has both proton and neutron closed shells. A deeper understanding of these effects requires more detailed information about the atomic structure. For example, information is required about the electron density at the nucleus which can be obtained from the analysis of the hyperfine structure. ^{17}O is the only stable oxygen isotope which has hyperfine structure since it has one neutron in the $d_{5/2}$ orbital outside the doubly closed ^{16}O nucleus; this produces a nuclear magnetic moment of $-1.894\mu_N$.[2] In this paper we report the first measurement of the hyperfine structure of atomic oxygen performed by high-resolution laser spectroscopy. In fact, the only existing data on the hyperfine structure for this atom were obtained in a paramagnetic-resonance experiment on the ground state.[3]

The level we have investigated is the excited 3^5S_2 level (Fig. 1). The interest of this particular level is twofold. First, it is the lowest in energy (9.14 eV) for the excited configuration $1s^22s^22p^33s$, so stronger nuclear effects can be expected. Since optical transitions start from this level, combined measurements of the isotope shift and the hyperfine structure represent a stringent test for *a priori* Hartree-Fock calculations, which are not easy for this multielectron atom. Second, the 3^5S_2 state is metastable (lifetime $\tau=180$ μs) and is connected to the $3^5P_{1,2,3}$ states by three strong transitions at $\lambda=777$ nm; as we discuss below, radiative cooling of atomic oxygen could be achieved using these transitions and the subsequent decay of the cooled atoms into the ground state would produce very cold ground-state oxygen. This metastable cooling is the only realistic cooling scheme for oxygen since no allowed transition at visible wavelengths is available from the ground state. A critical parameter for cooling is the lifetime of the upper 3^5P state; it determines the time required for the deceleration of the atoms which must be shorter than the lifetime of the 3^5S_2 state. From the present results an estimate for the 3^5P-state lifetime is obtained, which supports this proposed cooling scheme. In addition, GaAlAs/GaAs heterostructure diode lasers emitting at $\lambda\sim780$ nm are now available with enough power to perform nonlinear high-resolution spectroscopy. Interest in diode-laser spectroscopy has increased since the development of several techniques which improve these lasers' spectral purity and tunability[4-8] making them suitable for laser-cooling experiments. In this work a diode laser mounted in an extended cavity configuration is used to perform saturation spectroscopy on the $3s\,^5S_2$-$3p\,^5P_3$ transition at 777.1 nm.

The experimental arrangement is shown in Fig. 2. The laser we used was a commercial AlGaAs/GaAs diode laser emitting at 780 nm at room temperature. Coarse tuning over a range of more than 10 nm was achieved using optical feedback from a 1200-lines/mm

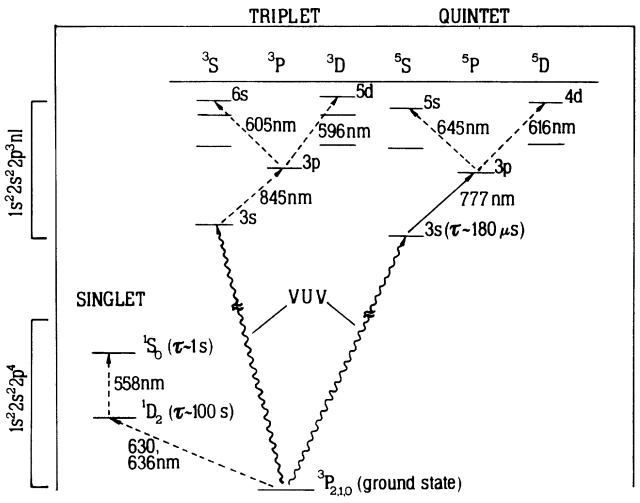

FIG. 1. A simplified energy-level scheme of O I showing the optical transition investigated in this work (solid line) and other transitions of spectroscopic interest (dashed lines).

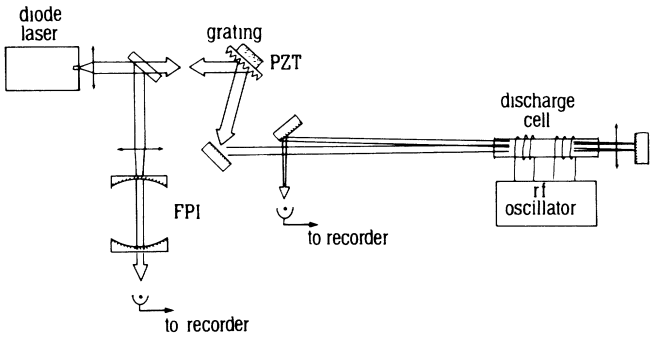

FIG. 2. Scheme of the experimental apparatus. Laser frequency stabilization is achieved by optical feedback from a grating which also acts as the output coupler. The beam is retroreflected through the sample cell to observe sub-Doppler saturation spectra. Double-headed arrows represent lenses. A Fabry-Perot interferometer (FPI) is used to monitor the laser's frequency.

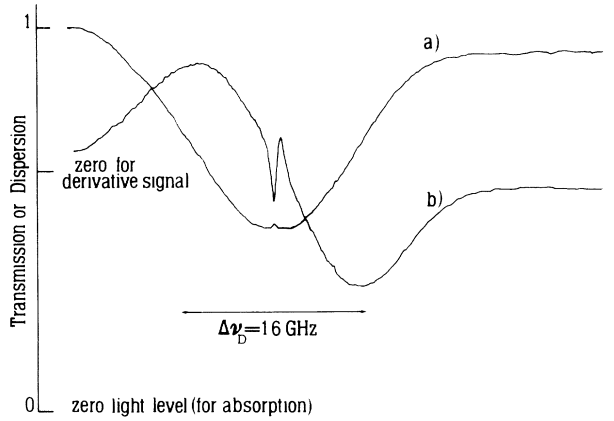

FIG. 3. Absorption profile of the $3s\ ^5S_2$–$3p\ ^5P_3$ oxygen transition at 777.1 nm observed in a natural-abundance sample using the experimental configuration shown in Fig. 2. (a) The saturation dip can be observed on the peak of the Doppler profile. (b) The saturation signal is enhanced relative to the Doppler background with derivative spectroscopy using FM modulation of the laser. The derivative signal is displayed with arbitrary units on the vertical axis.

ruled grating. The first-order diffracted beam was coupled back into the diode laser which had a reduced-reflection coating on the output facet. The optical feedback was strong enough to select a single mode of the cavity. Tuning was achieved by changing the temperature and the injection current of the diode along with the grating angle. The zero-order reflection from the grating was used for the spectroscopy. The power available in the zero order was 6 mW in a single mode (jitter $\sim$1 MHz). Light from an intracavity beam splitter (R =20%) was used for diagnostics. Frequency scans of $\sim$5 GHz were accomplished by synchronously changing the length of the cavity [with a piezoelectric transducer (PZT)] and the injection current of the laser. The calibration of the frequency scan was provided by a Fabry-Perot cavity with a free spectral range of 310 MHz. The low finesse of the cavity and feedback effects on the laser frequency are the main cause of uncertainty in the experiment. Direct absorption could be detected using a photodiode to measure the intensity of the light transmitted through the discharge cell as the laser frequency was scanned over the Doppler profile. The saturation dip could be observed with good signal-to-noise ratio by retroreflecting the laser beam (beam size $\sim$2 mm$\times$4 mm) back through the cell at a small angle. A long-focal-length lens provided a better interaction of the probe beam with the saturating beam in the cell and helped in separating the two after it. This simple configuration has the advantage of exploiting all the laser power to saturate the transition. Derivative saturation spectra were obtained by introducing a 20-kHz modulation on the cavity length by means of the piezoelectric transducer on the grating.

The fast sweep rate and low noise of the diode laser allowed us to detect saturated absorption signals in real time on an oscilloscope (detection bandwidth $\sim$50 kHz). Also, the derivative spectra could be obtained rapidly, limited only by the 1-ms time constant of the lock-in amplifier used for signal demodulation.

The radio-frequency discharge in which atomic oxygen is produced has been described elsewhere.[9] As discussed below, thanks to the higher sensitivity achieved in the present experiment, lower pressure could be used both for oxygen and for the buffer gas (argon in this case). A typical O_2/Ar pressure ratio was 1/20 and sub-Doppler signals could be detected for total pressure ranging from about 150 down to 0.6 Pa. For the measurement of the hyperfine structure, a 50% $^{17}O/^{18}O$ enriched sample was used. Natural abundances are ^{17}O=0.04%, ^{18}O =0.2%, and the enriched sample enhances the relative signal sizes.

In Fig. 3(a) we show the absorption profile for the $3s\ ^5S_2$–$3p\ ^5P_3$ 777.1-nm transition recorded in a natural-abundance sample with the experimental configuration shown in Fig. 2. The small saturation dip can be observed on the top of the broad Doppler profile. This corresponds to the most abundant ^{16}O isotope and is easily observed because of the very low noise on the diode laser. In fact, this feature allowed us to detect oxygen in direct absorption at very low concentrations and with a short ($\sim$2-cm) interaction length. However, observation of sub-Doppler spectra at very low pressures using enriched samples required an increase of sensitivity and resolution. These were obtained by recording the derivative of the absorption profile, as shown in Fig. 3(b). As described before, this was done adding a small dither on the voltage tuning the grating position and using the same dither as the reference signal for the lock-in

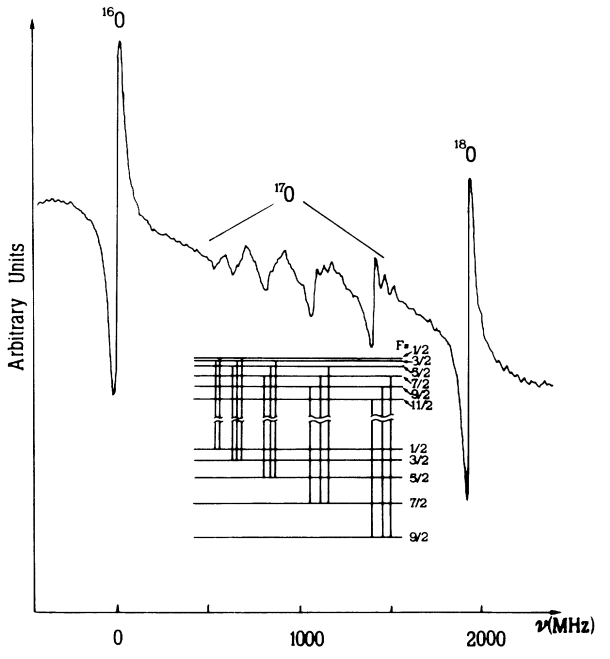

FIG. 4. Resonances of the three stable isotopes of oxygen as observed in an isotopically enriched sample. Hyperfine structure of the $^5S_2$ level is fully resolved for ^{17}O. Each of the five expected peaks also shows finer structure due to the upper-state hyperfine splittings, which are partially resolved. These data represent a single scan taken with a 1-ms time constant. Inset: The ^{17}O hyperfine levels (not to scale).

amplifier. The amplitude of the dither was optimized to reduce the amplitude of the broad Doppler signal relative to the saturation dip. In Fig. 4 we show such a spectrum observed in an enriched sample. Besides the individual resonances of ^{16}O and ^{18}O, the spectrum shows that the five hyperfine components of the lower $^5S_2$ level are fully resolved for ^{17}O. The hyperfine structure of the upper $^5P_3$ level is also almost resolved in this case, while in previous measurements on transitions starting from this level the hyperfine structure was unresolved for the ^{17}O resonance.

The measured splittings for the $^5S_2$ level are reported in Table I. Comparison of these data with the theoretical expression $E_F = E_J + \frac{1}{2}hAK$, with $K = F(F+1) - I(I+1) - J(J+1)$, gives a value for the magnetic coupling constant of $A = 74.3(4)$ MHz. The quadrupole interaction is neglected because of the insufficient accuracy of the data.

The results we obtained are of interest for a deeper understanding of nuclear effects on the atomic spectra. From the measured A constant we can estimate the electron density at the nucleus $|\Psi(0)|^2$. We obtain $a_0^3 \times |\Psi(0)|^2 = 0.12$, where a_0 is the Bohr radius. A more refined approach would require Hartree-Fock calculations, which are not presently accurate enough. Our experimental results provide a useful test for such theoretical calculations.

TABLE I. Measured hyperfine splittings of the $^5S_2$ level of ^{17}O obtained by averaging over ten different recordings. Uncertainties correspond to 1 standard deviation.

$\Delta_{F,F'}$	Measured hf splittings (MHz)
$\Delta_{1/2,3/2}$	118.3 ± 18
$\Delta_{3/2,5/2}$	172.0 ± 22
$\Delta_{5/2,7/2}$	253.6 ± 20
$\Delta_{7/2,9/2}$	342.0 ± 18

The resonances recorded in this experiment are nearly free from collisional broadening and correspond to a linewidth of 60 MHz FWHM, a factor of 2 or 3 narrower than those previously obtained for different transitions by intermodulated optogalvanic[10] or polarization[1] spectroscopy with a dye laser. From measurements of the linewidth as a function of pressure down to 0.1 Pa we can give a lower limit to the radiative lifetime τ of the 3^5P_3 level of 3 ns. Taking into account the effect of broadening due to residual pressure and saturation effects, we estimate a radiative lifetime of 6 ns. In anticipation of the radiative cooling of oxygen this value is of fundamental importance. In the scheme we propose, in fact, it is necessary that the deceleration time t_{decel} is shorter than the lifetime of the lower 3^5S_2 state. Assuming a starting velocity for O atoms of 4×10^2 m/s and a final velocity near zero, we get the necessary number of cycles of laser absorption and spontaneous emission, $N_{\text{ph}} = 12000$. The deceleration time is then $t_{\text{decel}} = 2N_{\text{ph}}\tau \sim 150$ μs, which is within the time available of 180 μs. This demonstrates the possibility of cooling a thermal oxygen beam by resonant scattering of 777-nm radiation. Very cold oxygen atoms could be produced in the ground state with a nonequilibrium distribution among the fine-structure levels populations. This opens the possibility of performing experiments of metrological interest since the ground state is connected by visible transitions to the $^1D_2$ state, whose lifetime is ~ 100 s. Also of interest are the magnetic-dipole fine-structure transitions in the ground state at 63 and 145 μm.

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