

OPTICAL DIAGNOSTICS OF LOW PRESSURE OPPOSED FLOW BURNER FLAMES

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ABSTRACT

Our project on understanding the mechanisms of flame suppression using agents more effective than halon 1301 depends heavily on our ability to study the detailed chemistry of diffusion flames. **This** paper presents the details of the experimental design of the laser induced fluorescence spectroscopy equipment as well as detection schemes for **OH**, **H**, **O** radicals and FeO molecules.

INSTRUMENTAL CONSIDERATIONS

The laser induced fluorescence (LIF) experimental apparatus (Fig. 1) is similar to that used by other investigators [1, 2] so only a brief outline of the instrument will be given here. The laser system for all of our measurements consists of a Lambda Physik Compex 102 XeCl excimer laser, a Scanmate 2 dye laser, and a second harmonic generator. In all cases, a filter or monochromator separates the experimental chamber from the detector. For temperature measurements, either a broadband filter centered at *ca.* 308 nm or an Acton Research model 300i monochromator is used. The detectors include a Phillips XP2018 B (for detection of **W** light) and a Phillips XP2017 (for detection of visible) photomultiplier tubes. After passing through a preamplifier, the signal is averaged with a SRS SR250 boxcar integrator/averager, sent to a SRS SR245 A/D board, and stored in a PC. Laser power is measured during wavelength scans with a Scientech AC25HD calorimeter connected to the A/D board. The PC that is interfaced to the A/D Board and the dye laser coordinates the data collection.

TEMPERATURE MEASUREMENTS

The temperature of a flame in the counterflow diffusion burner is a very important measured variable. Frequently, the decrease in temperature of a flame in the presence of an inhibitor is attributable to and proportional to the effectiveness of flame inhibition. Up to this point, most of the work at ARL has used standard techniques to measure flame temperature including the placement of small, inert thermocouples in the flame itself. The LIF instrument that we are currently evaluating is capable of measuring flame temperatures using a multiline spectroscopic thermometry technique based on the well known OH radical **W** spectroscopic structure [3]. Sufficient OH radical concentration to use this technique is expected to be present in all of the flames of interest in our laboratory. This technique is briefly outlined below but has been perfected by others and is described in detail elsewhere. See, for example, reference 4, 5 and references therein.

The temperature of the flame is determined from an analysis of the relative intensities of fluorescent bands obtained by scanning the frequency of the dye laser which has a bandwidth of *ca.*

0.15 cm^{-1} . The **key step** in the procedure involves the determination of the population of ground state energy levels from the relative fluorescence intensities of transitions involving **known** rovibronic levels. The population of a rovibrational level in the ground electronic state is simply related to the signal intensity by the expression

$$I_f = K \Phi_f B_{if} N_i \quad (1)$$

where I_f is the signal intensity, K is a constant including such factors as detector sensitivity, filter transparency, detector solid angle etc. B_{if} is the stimulated absorption coefficient (also **known as** the Einstein B coefficient) coupling the initial state, I , and the final state, f and N_i is the population of the initial rovibronic state. Φ_f is the fluorescence yield which can be calculated from the Einstein stimulated emission coefficient (A coefficient) and rates of nonradiative decay due to quenching and predissociation [4, 6]. Einstein stimulated emission and absorption coefficients as well as ground state rotational energies are all readily available [3, 8, 9, 10] and have been incorporated into a computer program that we have obtained from Luque [11].

Relative populations are obtained from equation (1). Since the hydroxyl radical is usually in a state of rotational thermal equilibrium with the **gas** which constitutes the flame, the relative population along with ground state rotational energies can be plotted and fit to a line using equation (2). Here, g_i is the degeneracy of the initial rotational level ($2J+1$), E_i is the corresponding energy, and N_i is the population of the initial laser-coupled state.

$$N_i = g_i e^{-\frac{E_i}{kT}} \quad (2)$$

From the slope of the resulting Boltzmann plot, rotational temperature is determined. Other methods of treating the data include complete least-square fits of the experimental spectrum to a calculated spectrum with rotational temperature and linewidth being variables [6].

For our temperature measurements, we will be scanning over a region of the R branch of the (0,0) band of the $X^2\Pi - A^2\Sigma^+$ electronic system of the OH radical. **This** follows the work of Crosley and Jeffries [4] and the transitions lie in the **306-308 nm** region.

CONCENTRATION MEASUREMENTS

In addition to using the spectroscopy of OH to measure temperature, relative concentration measurements of **this** radical are being made. We excite the (1-0) band of the A-X transition near **283 nm** and collect the broadband fluorescence centered at **ca. 315 nm** (see Fig. 2) with a wide bandwidth filter (**this is not** the same band as is **used** for the temperature measurements). The relative LF signal collected as the flame is physically moved through the region in which the laser beam is focused will be put on an absolute concentration scale by normalizing it to calculated concentration profiles from computer models described elsewhere [12, 13, 14, 15, 16].

Other radicals of interest include the H, and O radicals. Relative concentrations of all of these will be measured by two photon excitation schemes due to the difficulty of generation of VUV

radiation (Fig. 4). Hydrogen concentrations will be measured by detecting fluorescence that results from pumping the $n=1 \rightarrow n=3$ transition of hydrogen atom with two 205 nm photons. The fluorescence at 656 nm is the result of a $n=2 \rightarrow n=3$ relaxation [17, 18]. Oxygen can be detected by exciting with two 226 nm photons and detecting the resulting fluorescence at 845 nm. These detection experiments have been thoroughly described by others [19, 20, 21] and are described schematically in the energy level diagrams.

MEASUREMENTS ON FeO

One of the model superagent fire suppressants that we are interested in investigating is $\text{Fe}(\text{CO})_5$. It is expected that the presence of this material in the flame will result in significant quantities of FeO in the flame itself. We intend to make spectroscopic investigations of this as well. Our detection scheme for FeO will be identical to that of Zachariah [22]. Briefly, the laser will be tuned to the $X^5\Delta_4 \rightarrow ^5\Delta_4$ transition at 558 nm. and we will measure the resulting broadband fluorescence. Laser scattering will be eliminated with a long pass filter.

CONCLUSION

We have presented a brief discussion of a laser-based spectroscopy instrument which is giving our laboratory additional capabilities in our ongoing investigation of the active mechanisms of flame suppression. We have discussed diagnostic strategies for the determination of temperature and relative concentrations of H, O, and OH radicals. We have also discussed a detection scheme for FeO molecules.

ACKNOWLEDGEMENTS

We acknowledge the ARL sponsored NAS/NRC Postdoctoral Research Associates Program (CKW) and the Next Generation Fire Suppression Technology Program of SERDP.

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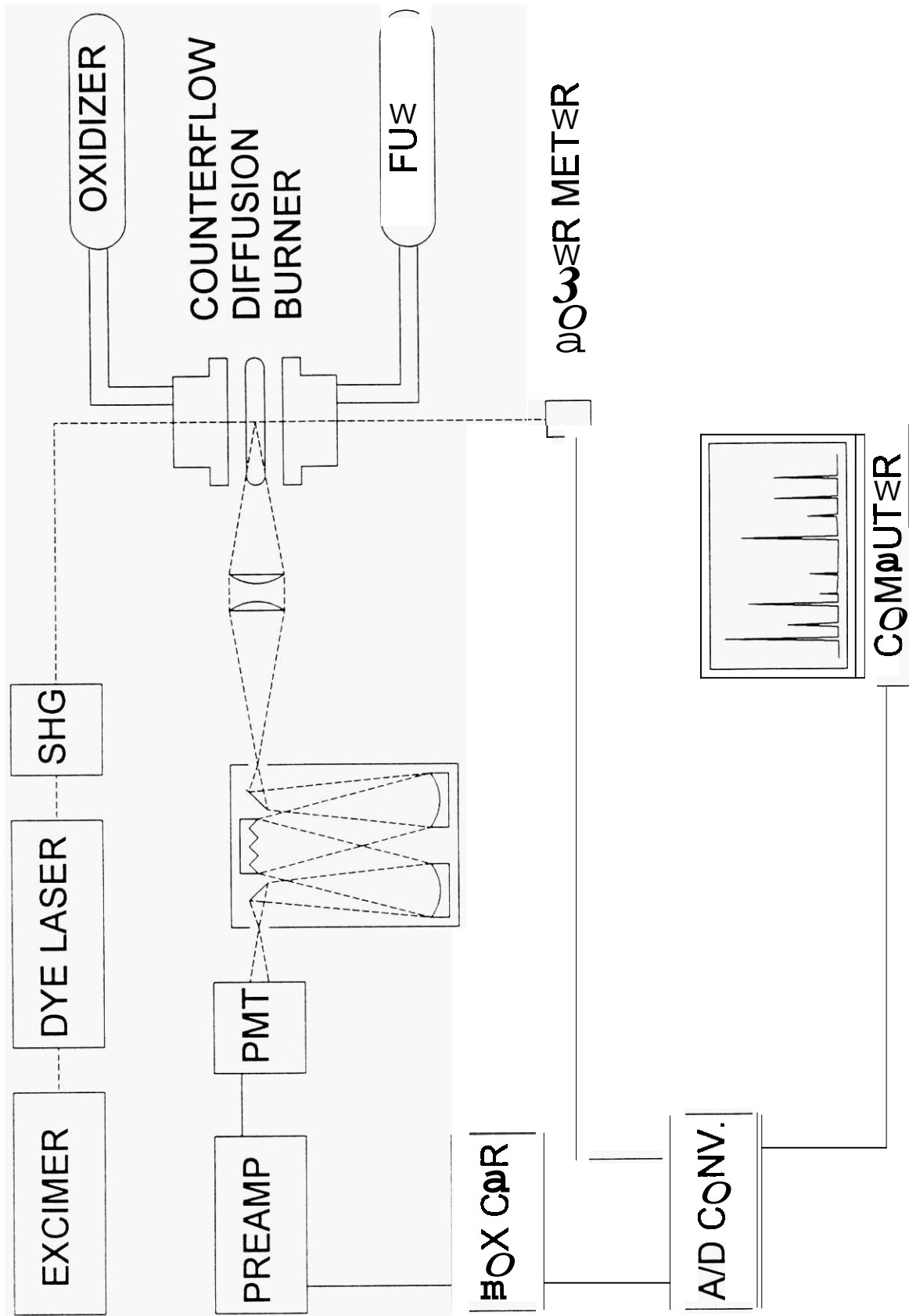


Figure 1. Experimental Schematic.

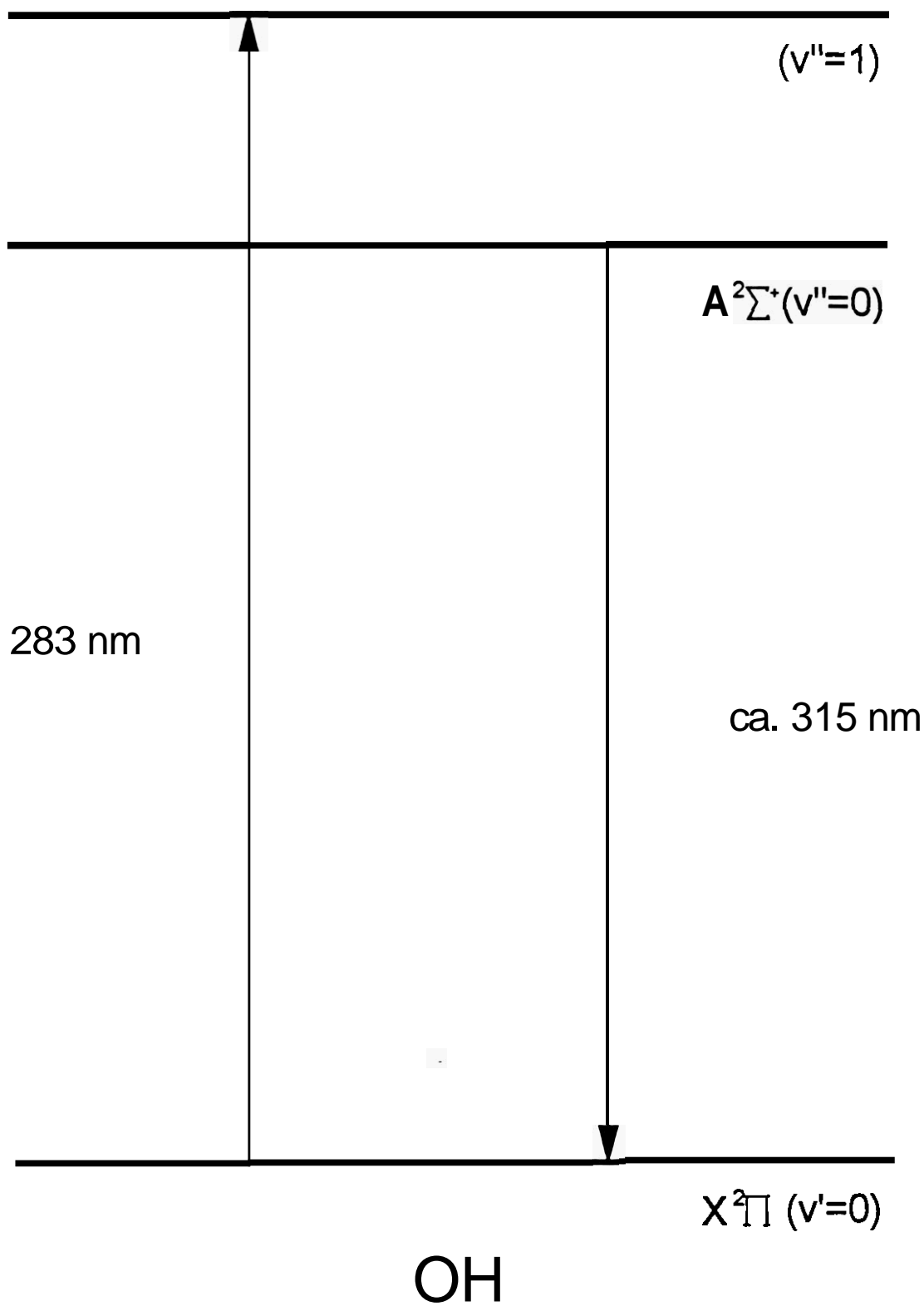


Figure 2. Potential energy level diagram for OH radical.

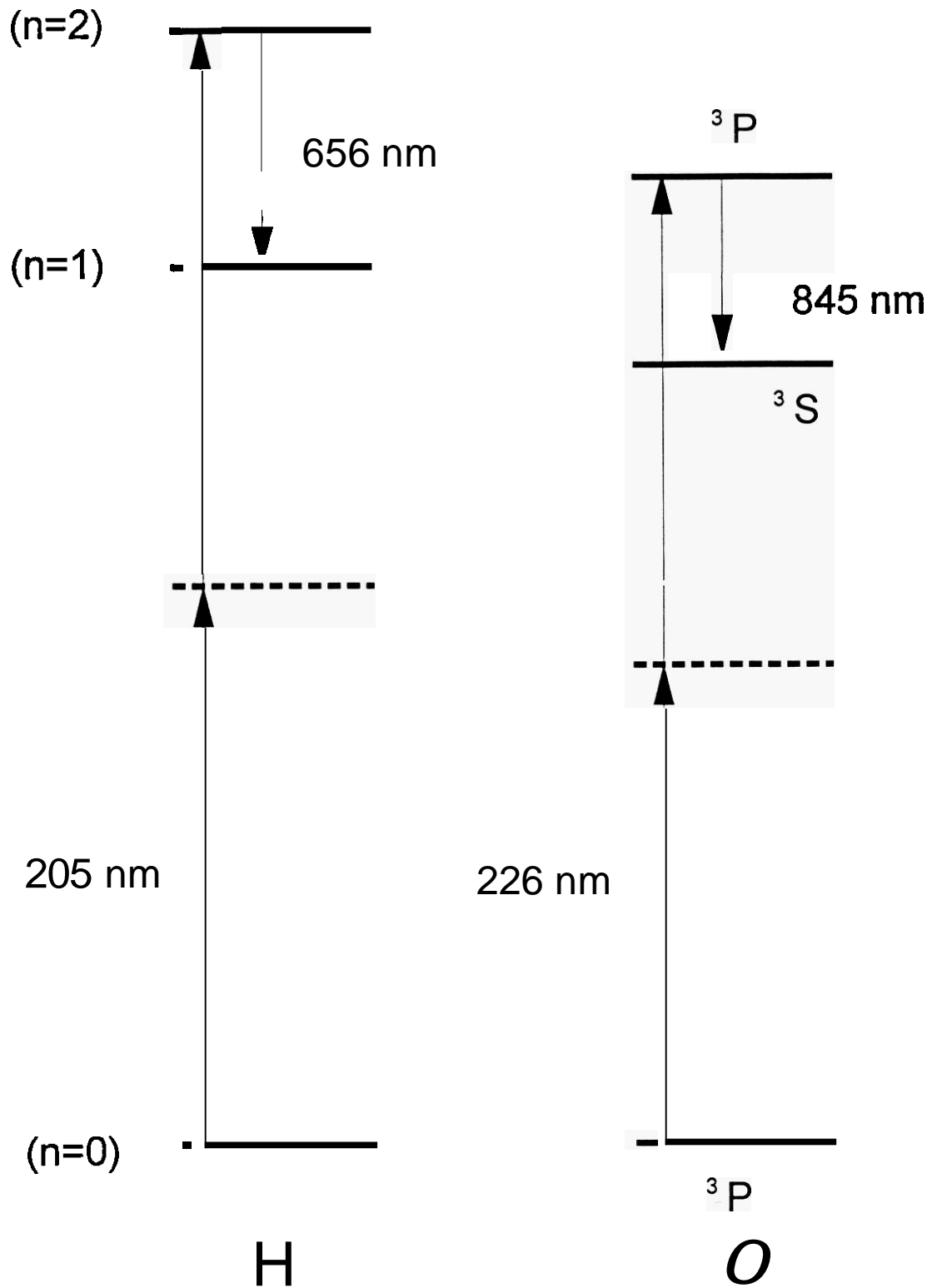


Figure 3. Potential energy level diagram for O and H atoms.

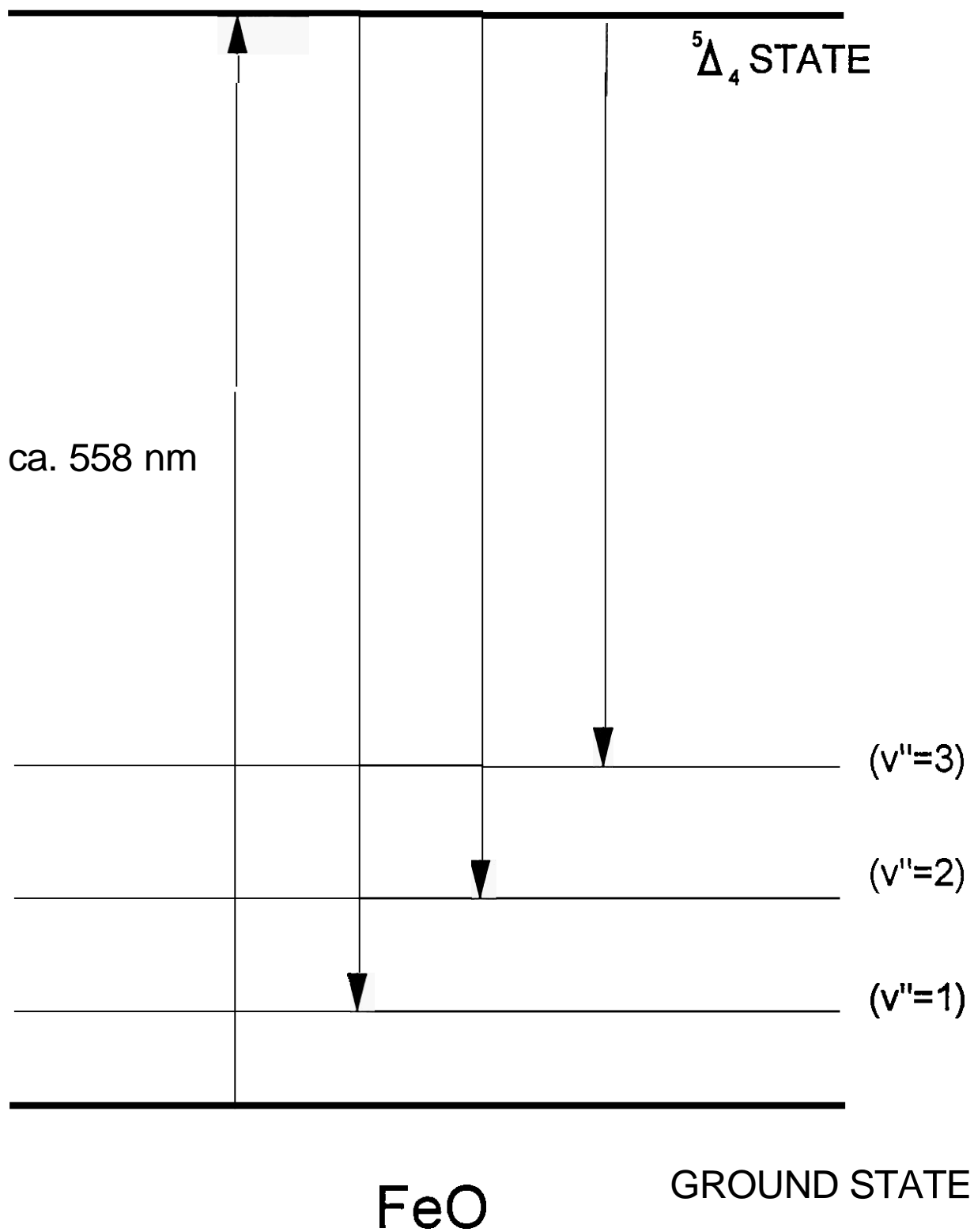


Figure 4. Potential energy level diagram for FeO.