III. ATOMIC RESONANCE AND SCATTERING

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A. TWO-PHOTON HIGH-RESOLUTION SPECTROSCOPY

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We have excited both the 4D and 5S states of neutral atomic sodium by simultaneous two-photon absorption directly from the 3S ground state. This is the first high-resolution two-photon spectroscopy performed in the visible region and the first to utilize continuous wave lasers. An atomic beam of ground-state sodium was excited by a Spectra-Physics System 580 single-mode tunable dye laser, and the resonance was detected by monitoring the decay fluorescence from the excited state. The experimental arrangement is shown in Fig. III-1.

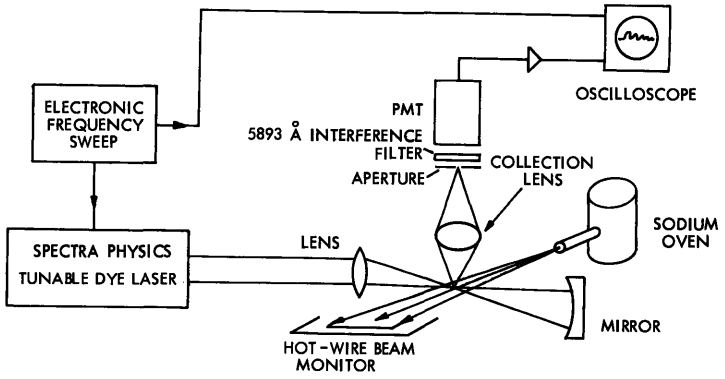

Fig. III-1. Experimental apparatus. A mirror that reflects the laser beam back through the interaction region will be added to quadruple the signal and eliminate Doppler broadening caused by divergence of the atomic beam.

Since the linewidth observed in the experiment was 5×10^{-3} cm^{-1}, we were able to resolve fully the fine structure of the 4 ^{2}D state by electronically sweeping the laser (around 17,274 cm^{-1}, half of the 3S-4D transition energy). The fine-structure splitting

was measured by comparing the spacing between components of the observed spectrum (Fig. III-2) split by the 4D fine structure, Δ, with the spacing arising from the well-known hyperfine splitting of the 3S ground state, Δw. We measured $\Delta_{f.s.} = .0343 \pm .0002\ cm^{-1}$. This error is roughly 1/10 that of any previous measurement of fine-structure splitting.

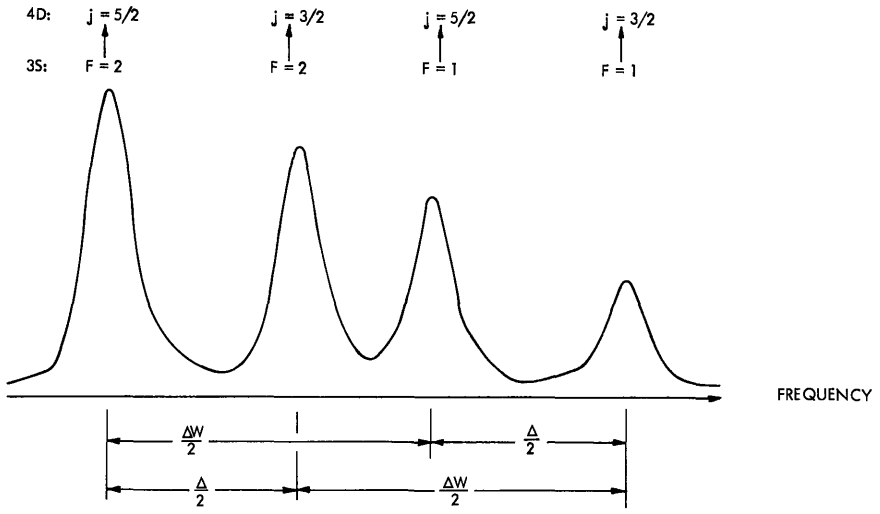

Fig. III-2. Detailed structure of the 3S → 4D two-photon transition (with frequency increasing to the right ~200 MHz/div). The levels involved in each of the four components of the transition are shown above the corresponding peaks and the spacing is shown below.

The transition rate for two-photon absorption is given by second-order time-dependent perturbation theory. If we take the perturbation to be $H = i\frac{e\omega}{c}|\vec{A}|\hat{\epsilon}\cdot\vec{r}$, then the transition probability is proportional to

$$\text{Rate} \propto (\text{Laser Power})^2 \frac{\left|\sum_k \langle f|\hat{\epsilon}\cdot\vec{r}|k\rangle\langle k|\hat{\epsilon}\cdot\vec{r}|i\rangle\right|^2}{[(\omega_{ki}-\omega_{fi}/2)^2+\gamma_k^2/4]},$$

where i and f refer to the initial and final states and the sum is taken over all intermediate states k, and ω_{ki} (ω_{fi}) is the level spacing between the ground and intermediate (final) states.

The interesting feature of this expression is the resonance denominator, which allows high transition rates when one of the intermediate states is nearly midway between the initial and final states. In the case of our 3S → 4D transition, the 3P state is 350 cm^{-1} away from being halfway between the 3S and 4D levels (the sodium term diagram is shown in Fig. III-3), thereby allowing saturation of the two-photon transition with a relatively low power (2×10^3 W/cm^2). (No direct excitation of the 3P state is produced by

the laser when operating near the frequency $\omega_{fi}/2$, since this is roughly 5×10^5 linewidths away from the resonance of the 3S → 3P transition.

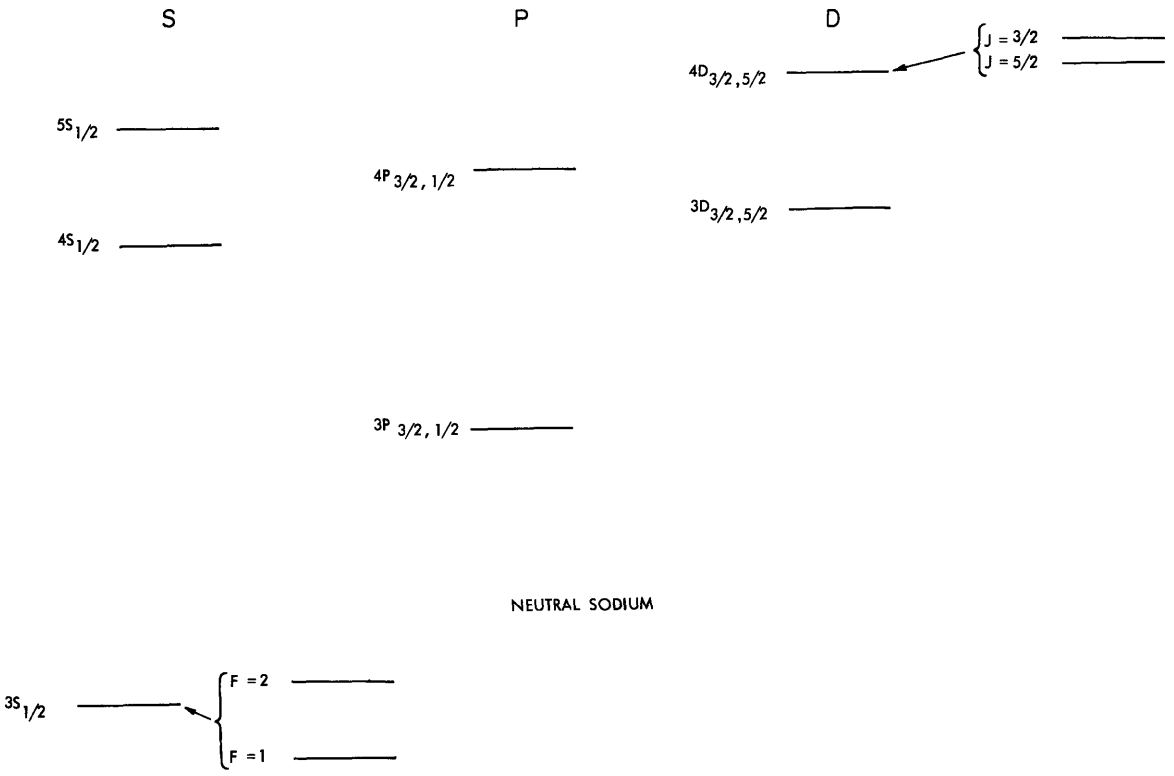

Fig. III-3. Energy-level diagram for neutral atomic sodium. The main diagram is roughly to scale, as are the enlargements of the 4D fine structure and the 3S hyperfine structure.

Two-photon spectroscopy is applicable, with improved optics and possibly signal averaging techniques, to systems in which the intermediate state is 1000-3000 cm^{-1} from being halfway between the initial and final states. Thus this technique is well-suited to the precise spectroscopy of many systems in which levels have previously been inaccessible when using single-photon dipole transitions. Unlike stepwise excitation, this technique requires only a single laser, although a second cω laser (not tunable) at a different frequency can be utilized to extend the accessible frequency range considerably.