CONF-881002--2

CONF-881002--2

DE88 015557

For publication in the Proceedings of the Fourth International Laser Science Conference (ILS-IV) (Advances in Laser Sciences - IV) Mariott Marquis Hotel, Atlanta, Georgia, October 2–6, 1988. to be published by the American Institute of Physics

INFLUENCE OF THE A.C. STARK EFFECT ON STIMULATED HYPER-RAMAN PROFILES IN SODIUM VAPOR*

M. A. Moore, W. R. Garrett, and M. G. Payne**

Chemical Physics Section, Oak Ridge National Laboratory
Post Office Box 2008
Oak Ridge, Tennessee 37831-6378, USA

August 1988

*Research sponsored by the Office of Health and Environmental Research, U.S. Department of Energy under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

**Postdoctoral fellow, University of Tennessee, Knoxville, Tennessee

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Mary Anne Moore,* W. R. Garrett, and M. G. Payne
Chemical Physics Section, Oak Ridge National Laboratory
Oak Ridge, Tennessee 37831-6378

ABSTRACT

When pumping near the two-photon 3d resonance in pure sodium vapor and observing the backward hyper-Raman emission to the 3p substates, an asymmetry in ratios of $3p_{1/2}, 3p_{3/2}$ associated emissions was observed dependent upon the direction of the initial laser detuning from the resonance. It has been determined that this asymmetry can be attributed to the a. c. Stark effect induced by the hyper-Raman emission itself.

Stimulated hyper-Raman (SHR) emission characteristics in pure sodium vapor have been well documented.[1] It has been shown that the three-photon SHR process is suppressed in the direction of the laser photons due to interference with a four-wave mixing (FWM) pumping of the atomic transition that is exactly 180 degrees out of phase with the SHR excitation process.[2,3] However, the backward propagating SHR emission is unaffected by this suppression effect because of the absence of FWM in that direction due to phase-matching considerations.

The SHR process studied in this paper is produced by two laser photons tuned near, but not on the two-photon resonance with the Na 3d states. The fine structure levels (j= 5/2, 3/2) are not resolved by the Lumonics excimer-pumped dye laser used in the experiments. The subsequent SHR emission to the $3p_{1/2}$ and $3p_{3/2}$ states ($\lambda \sim 818.5$ nm and 819.7 nm, respectively) was monitered by a Jarrell-Ashe spectrometer with a resolution of 0.03 nm (see Fig. 1 for an energy level diagram of the process involved).

The gain for the SHR process is inversely proportional to the square of the initial laser detuning, δ, and not sensitive to the direction of that detuning (i.e., whether the laser photons are tuned to the high or low energy side of the 3d resonance). When tuned to the low energy side of the 3d states it was noted that the intensity of the backward directed $3p_{1/2}$ SHR emission was always more suppressed than the $3p_{3/2}$ SHR intensity (Fig. 2b). The opposite would occur when detuned to the high energy side of the two-photon 3d resonance; the $3d - 3p_{3/2}$ signal was reduced in intensity more than the peak at 818.5 nm (Fig. 2c).

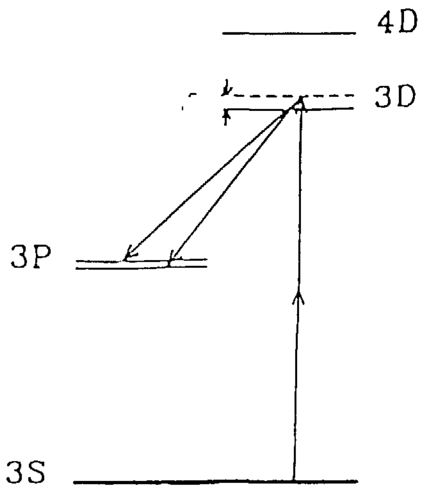

Figure 1. Energy level schematic of hyper-Raman process in sodium vapor.

The asymmetry of SHR peak suppression due to the direction of incident laser detuning was consistently reproducible, and became understandable in light of other work involving the a.c. Stark effect.[3] The a.c. Stark effect causes either the apparent splitting or shifting of atomic sublevels coupled by an electromagetic field. For near resonant processes it is quadratic with respect to the field and proportional to the product of electromagnetic field amplitude and the dipole matrix element connecting two states, and inversely proportional to the energy mismatch of the photon field that couples the states.

Now note that the sign of the energy mismatch (detuning) of the coupling field determines the direction of the shift. If the photon energy, $\hbar\omega$, couples to states at exact resonance, the levels appear to be split into sublevels with equal amplitudes. If the photon has less energy than the transition between levels (i.e., is tuned to the red side) the splitting evolves into a shifting of levels with the atomic states pushed apart. The opposite happens when the photon is more energetic or is tuned to the blue side of the resonance.

The a.c. Stark shift can now be used to explain the backward SHR asymmetry discussed earlier. If one tunes the incident laser to the red side (low energy) side of the two-photon 3d resonance (see Fig. 3), SHR is emitted between the virtual state and both the $3p_{1/2}$ and $3p_{3/2}$ states, labeled ω_1 and ω_2. If one redraws these transitions so that they start from the d state (dotted lines) it is easily seen that while ω_1 is on the red side of both of the $3d-3p$ transitions, ω_2 is to the blue side of $3d-3p_{3/2}$ transition, but to the red side of $3d-3p_{1/2}$ transition. The ω_1 photon pushes the 3d states away from both of the 3p levels, while ω_2 pushes the $3d-3p_{1/2}$ levels even further apart, while pushing together the $3d-3p_{3/2}$ levels. The net effect is to have a total Stark shift which is greater for the $3d-3p_{1/2}$ transition than it is for the $3d-3p_{3/2}$ one. Since the total Stark shift is larger for the $3d-3p_{1/2}$ transition, it has a lower transition rate and will, therefore, be suppressed in intensity compared to the case where the laser is tuned exactly to the two-photon resonance. The exact opposite occurs when tuned to the blue (high energy) side of the resonance. Both ω_1 and ω_2 tend to push the $3d-3p_{3/2}$ states together, while ω_1 pushes $3d-3p_{1/2}$ states apart and pulls the $3d-3p_{3/2}$ together. The net effect is to have the Stark shift greater for the $3d-3p_{3/2}$ transition, and to suppress its transition probability with respect to the $3d-3p_{1/2}$ emission. Put in other terms, when the laser is detuned to the red side of the two-photon 3d resonance, the 818.5 nm emission should be more suppressed than the 819.7 nm emission when compared to intensities at zero detuning. When detuned to the blue side the opposite should occur.

This is exactly what the wavelength scans in Fig. 2 show. One expects the intensity of both peaks to be decreased when detuned from resonance, but if one does not include the a.c. Stark effect it is expected that both peaks are affected by the same amount. It is only when one takes the a.c. Stark effect into account that this asymmetry phenomenon becomes understandable. It is also direct experimental proof that the a.c. Stark effect can suppress the gains for resonant processes such as SHR and amplified spontaneous emission.[3] It should be noted that differences in the shifts at these laser intensities and detunings are small (~10%), and in a region where the gain is still in exponential growth (i.e., not yet limited by saturation effects). Thus, even these small differences can significantly affect peak intensities.

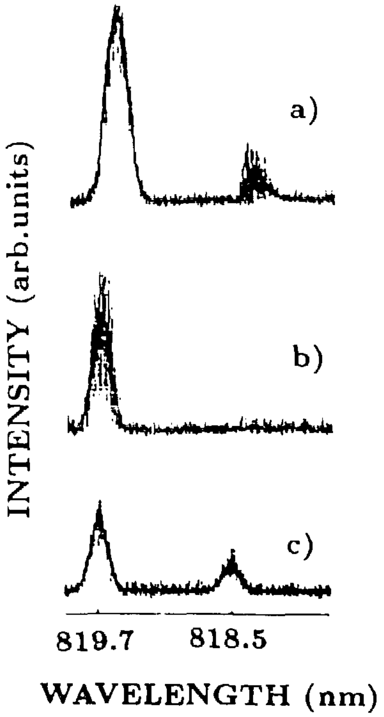

Figure 2. Backward SHR emission from near the Na 3d states to $3p_{3/2}$ and $3p_{1/2}$ states, respectively, at different laser detunings. (a) On resonance, (b) 0.03 nm to the low energy side, $3p_{1/2}$ transition suppressed, (c) 0.05 nm to the high energy side, $3p_{3/2}$ transition suppressed.

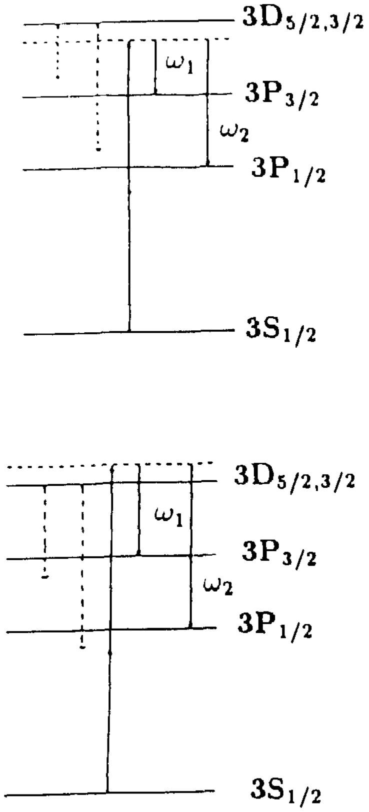

Figure 3. Schematic of backward SHR emission out of 3d states illustrating the radiation responsible for the asymmetric suppression of the 3p states as a function of the direction of pump laser detuning due to the a.c. Stark effect.

Research sponsored by the Office of Health and Environmental Research, U.S. Department of Energy under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc. M. A. Moore is a postdoctoral fellow, University of Tennessee, Knoxville, Tennessee.

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