presented herein and on their cesium results. The assistance of A. V. Smith in the construction and operation of the heat pipe used in the experiments is gratefully acknowledged.

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Saturation of Two-Photon-Resonant Optical Processes in Cesium Vapor*

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We report observations of saturation of two-photon-resonant third-harmonic generation and of fluorescence following two-photon absorption in cesium vapor. Weakening dependence of these processes on fundamental power and broadening of the resonance line with increased fundamental power are both seen. It is shown that hole burning is important in a certain power range with the resonance becoming predominantly homogeneously broadened at higher power levels.

Two-photon-resonant optical third-harmonic generation[1] and fluorescence following two-photon absorption[1,2] in cesium vapor have been reported previously. We now present observations of saturation in these processes: That is, we find that these processes exhibit a weaker dependence on fundamental power as the fundamental power is increased and this is accompanied by power-dependent broadening and shape changes in the resonance line. Saturation of two-photon absorption in potassium vapor has been discussed (but not observed) by Yatsiv *et al.*[3] and saturation of two-photon-resonant processes in thallium vapor has been investigated by Wang and Davis.[4]

Apparatus used in this work is similar to that described in Ref. 1 with the exception of substantial improvements in the laser spectrum and the data acquisition system. A ruby laser provides 1-MW, 30-nsec [full width at half-maximum (FWHM)] pulses, with wavelength tunable in the region of 693.5 nm where twice the laser photon energy ($2\hbar\omega$) is equal to the energy difference ($\hbar\Omega$) between the $9d\,^2D_{3/2}$ level and the $6s\,^2S_{1/2}$ ground state of the cesium atom. Tuning is effected by control of the rod temperature near 148°K and use of a thermally tuned output etalon. The laser spectrum consists of five adjacent longitudinal modes of the laser cavity, separated by 0.005 cm^{-1} and with about one fifth of the total power in each mode. The center frequency of this spectral distribution can be controlled to within 0.005 cm^{-1}. Except as noted, experiments reported here all use a collimated beam with area 0.03 cm^2. The cesium cell described in Ref. 1 is used at a temperature of about 483°K, selected to give 1 third-harmonic coherence length of cesium vapor which corresponds to a density of 10^{16} atoms/cm^3 and a sample length of 6 cm. The cell also contains 1 Torr of neon to inhibit diffusion of cesium to the windows. Third-harmonic light traveling collinear with the laser beam and fluorescence at 584.7 nm emitted perpendicular to the beam by atoms decaying to the $6p\,^2P_{3/2}$ level following two-photon excitation to the $9d\,^2D_{3/2}$ level are each detected with photomultipliers after suitable spectral filtering. Third-harmonic, fluorescence, and fundamental monitor signals are gated, integrated, digitized, and recorded on punched paper tape. Depletion of fundamental power by interaction with the cesium vapor is negligible (< 3%) and no evidence of self-focusing can be detected in the beam emerging from the cell.

Figure 1 shows log-log plots of third-harmonic power and fluorescence measured as a function of fundamental power P^{ω}. In Fig. 1(a), third-harmonic power with the laser detuned 0.3 cm^{-1} from resonance is seen to vary, unremarkably, as the cube of P^{ω}, but on resonance [Fig. 1(b)] saturation is evident with third-harmonic power now proportional to the square of P^{ω}. In Fig. 1(c), fluorescence for $P^{\omega} < 2 \times 10^4$ W is seen to be linear in P^{ω} rather than quadratic as would be expected in the absence of saturation. At higher P^{ω}, the dependence of fluorescence on P^{ω} further weakens to between $(P^{\omega})^1$ and $(P^{\omega})^0$.

Resonance curves showing fluorescence and third-harmonic emission as a function of laser frequency are shown in Fig. 2 for various fundamental power levels. Improved control of the laser spectrum compared with Ref. 1 allows the ground-state hyperfine-structure doublet (separation 0.3 cm^{-1}) to be clearly resolved. Third-harmonic emission is not detectable for $P^{\omega} < 2 \times 10^4$ W. For $P^{\omega} \leq 10^5$ W [Figs. 2(a), 2(b), and 2(f)] fluorescence and third-harmonic yields have similar line shapes with widths of 0.037 cm^{-1} FWHM, and this is consistent with the combined contributions from laser spectral width (0.02 cm^{-1}) and from Doppler broadening (0.02 cm^{-1} FWHM). At $P^{\omega} = 10^6$ W [Figs. 2(c) and 2(g)], broadening of the resonance becomes evident with the third-harmonic emission broadening more than fluorescence. Figures 2(d) and 2(h) show resonance shapes obtained with a focused fundamental beam which, while not easily comparable with the collimated-beam results, do show qualitatively interesting evidence of increased broadening. Fluorescence, while broadening, maintains essentially the same shape as at lower P^{ω} but, in the case of third-harmonic emission, there is some evidence for *dips* at the

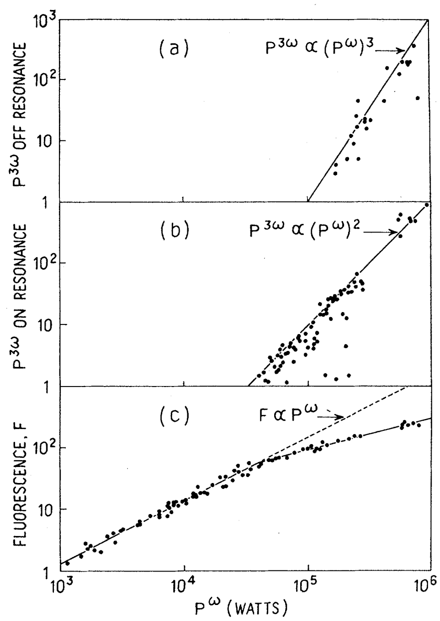

FIG. 1. Log-log plots of the dependence on fundamental power of (a) third-harmonic generation, 0.3 cm^{-1} off resonance, (b) third-harmonic generation, on resonance, (c) fluorescence, on resonance. Each point plotted corresponds to an individual laser shot, and all vertical scales include an arbitrary, multiplicative scale factor. We believe that five or six points in (b) indicating anomalously low third-harmonic generation were caused by unlocking of the laser wavelength- (temperature-) control servo.

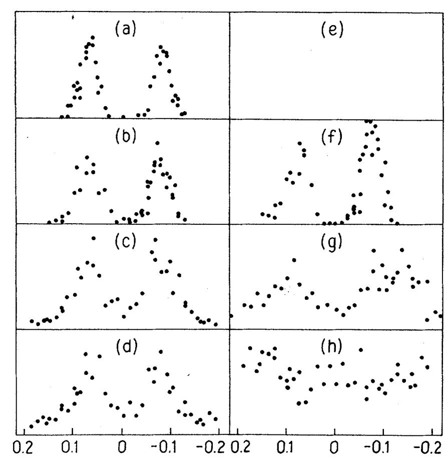

FIG. 2. Resonance curves of (a)–(d) fluorescence and (e)–(h) third-harmonic emission as a function of laser frequency in the region of 14 414 cm^{-1}; $\Delta\omega$ is $\omega - \Omega/2$. Laser power increases from top to bottom: (a), (e) 10^4 W (third harmonic not detectable); (b), (f) 10^5 W; (c), (g) 10^6 W; all were collimated laser beams 0.03 cm^2 in area; (d), (h) 10^6 W focused with 18-cm-focal-length lens. Fluctuations of laser output around these average power levels have been normalized by dividing fluorescent yields by $(P^{\omega})^2$ and third-harmonic yields by $(P^{\omega})^3$ but the choice of normalization does not substantially affect the shape of the curves. Each point plotted corresponds to an individual laser shot and the vertical scale for each plot is linear but otherwise arbitrary.

center of each hyperfine resonance.

For qualitative comparison with the experimental data, we now write expressions for the fluorescence F and the third-harmonic power $P^{3\omega}$ as a function of P^{ω} and laser frequency ω. The expressions were derived by use of the method-of-averages approach to time-dependent perturbation theory,[5] segregating the slowly varying parts of the $6s\,^2S_{1/2}$ and $9d\,^2D_{3/2}$ amplitudes for treatment by Bloch equations[6] with the steady-state approximation. They are written for a single laser mode and without displaying the hyperfine splitting explicitly. A two-photon saturation parameter S may be defined by

$$S = T_1 T_2 / |T^{(2)}|^2 \propto (P^{\omega})^2, \qquad (1)$$

where T_1 and T_2 are relaxation times[7] and $(T^{(2)})^{-1}$ is the Rabi precession frequency:

$$\frac{1}{T^{(2)}} = \frac{1}{2}\left(\frac{e}{\hbar}\right)^2 \sum_n{}' \frac{\langle 9D|z|nP\rangle\langle nP|z|6S\rangle}{\Omega_{np}-\omega}(E^{\omega})^2. \qquad (2)$$

The required relations are then

$$F \propto \tfrac{1}{2}(\gamma\Omega T_2)^{-1} S(1+S)^{-1/2} Z''(x,y) \qquad (3)$$

and

$$P^{3\omega} \propto (\gamma\Omega)^{-2}\,|Z'(x,y) + i(1+S)^{-1/2} Z''(x,y)|^2 (P^{\omega})^3, \qquad (4)$$

or, when homogeneous broadening greatly exceeds inhomogeneous broadening, this can be written

$$P^{3\omega} \propto \left| \frac{(\Omega - 2\omega) + i(T_2)^{-1}}{(\Omega - 2\omega)^2 + (T_2)^{-2}(1+S)} \right|^2 (P^{\omega})^3. \qquad (5)$$

Z' and Z'' are the real imaginary parts of the plasma dispersion function[8] whose arguments are the normalized detuning $x = (2\omega - \Omega)/\gamma\Omega$ and the resonance width $y = (\gamma\Omega T_2)^{-1}(1+S)^{1/2}$. The inhomogeneous broadening has been introduced into Eq. (4) in terms of $\gamma = (2kT/Mc^2)^{1/2}$, a characteristic, normalized velocity for cesium atoms of mass M and temperature T. The results given in Eqs. (3)–(5) are based on a steady-state approximation whose validity is marginal at low powers but improves with increasing saturation.

The following characteristics of Eqs. (3)–(5) for certain combinations of parameter values may be noted:

Unsaturated ($S \ll 1$).—The resonance line shapes $F(\omega)$ and $P^{3\omega}(\omega)$ are different in general. However, in the unsaturated case, they are similar and become identical if inhomogeneous broadening is negligible. The line shapes are independent of P^{ω}; F varies as the square and $P^{3\omega}$ as the cube of P^{ω}.

Saturated, with hole-burning ($S \gg 1$, $y \ll 1$).—The width of a hole burnt in the atomic velocity distribution is determined by the power-broadened homogeneous resonance width which is y times the inhomogeneous width. Thus, in this case, broadening is predominantly inhomogeneous and the hole is narrow. (We have not included, here, the possibility of significant cross relaxation.) Since y is small, $Z(x,y)$ can be approximated by $Z(x,0)$ which is independent of P^{ω}. Thus while F becomes linear in P^{ω}, it retains its low-power, power-independent line shape. This description coincides with the experimental fluorescence data for the power region $P^{\omega} < 2 \times 10^4$ W. Third-harmonic emission was not detected in this region but Eq. (4) would predict an essentially power-independent line shape dominated by Z' with power dependence changing from $P^{3\omega} \propto P^{\omega}$ on resonance to $P^{3\omega} \propto (P^{\omega})^3$ far from resonance.

Saturated, with predominantly homogeneous broadening ($S \gg 1$, $y \gg 1$).—The S dependence of y ensures that this case of saturation with predominantly homogeneous broadening is reached eventually at sufficiently high power. All atoms are simultaneously accessible to interaction with the radiation and the saturation proceeds uniformly without development of a hole. Equation (3) predicts a resonance linewidth for F which increases as P^{ω} and a peak value independent of P^{ω}. Equation (5) predicts for third-harmonic emission a complicated line shape, broadening with increasing P^{ω}, and having an on-resonance dip. The predicted power dependence ranges from $P^{3\omega} \propto (P^{\omega})^{-1}$ near resonance to $P^{3\omega} \propto (P^{\omega})^3$ far from resonance.

Quantitative comparison between theory and experiment is made difficult by the experimental laser spectrum and the uncertainty in T_2 but a qualitative correspondence can be seen. Estimated parameters ($S \sim 300$ and $y \sim 1$ at $P^{\omega} = 10^6$ W, single mode, with $T_2 = 8$ nsec) indicate that a saturated regime *intermediate* between hole burning and predominantly homogeneous broadening should obtain for $P^{\omega} = 10^5$–10^6 W. This assignment is supported by the onset of line broadening evident in Figs. 2(c), 2(d), 2(g), and 2(h), and the intermediate power dependence of fluorescence shown in Fig. 1(c). The predicted on-resonance dips in the third-harmonic line tend to be obscured by the experimental laser spectrum but some evidence for them survives in the focused-beam data, Fig. 2(h). Numerical calculation taking the laser spectrum into account yields results for third-harmonic emission consistent with the observed $(P^{\omega})^2$ dependence shown in Fig. 1(b). In addition, the fraction of irradiated atoms excited to the $9d\,^2D_{3/2}$ level can be determined from the measured value of F and the result, $10^{-3}\times 10^{\pm 1}$, also indicates significant but not complete saturation.

The experimental results have been explained qualitatively on the basis of saturation involving power broadening and population of the two-photon-resonant state with hole burning being important in a certain range of fundamental power. While we believe these effects are dominant, a significant role may also be played by two-step transitions from the $9d\,^2D_{3/2}$ to $6s\,^2S_{1/2}$ level where one or both steps are stimulated.[9] Our observation of such effects in cesium vapor will be discussed elsewhere. Saturation effects will be important in schemes using resonant nonlinear interactions to enhance frequency-conversion efficiency.[9]

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Two-Electron—One-Photon Transitions in Heavy-Ion Collisions*

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Heavy-ion–atom collisions at medium energies (~0.8 MeV/amu) yield a considerable probability for multiple inner-shell ionization. In the Ni-Ni, Ni-Fe, Fe-Ni, and Fe-Fe cases, distinct lines in the x-ray spectra are observed, which must be interpreted as a correlated two-electron jump into the doubly ionized K shell followed by the emission of only one photon carrying away the total transition energy.

In heavy-ion–atom collisions at medium energies, multiple inner-shell vacancies may be produced. If, for instance, both electrons are removed from the K shell of either the target atom or the projectile during such an encounter, the remaining holes will be filled up by subsequent transitions of two electrons from higher-lying shells. Normally these jumps will be accompanied by the emission of two characteristic x rays and/or the emission of two Auger electrons. But according to the predictions of Heisenberg,[1] Condon,[2] and Goudsmit and Gropper[3] it is also possible that both holes may be filled up by a simultaneous transition of two electrons. In this case