Observation of Subnatural Linewidths by Two-Step Resonant Scattering in I_2 Vapor

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Observation of linewidths narrower than the natural width with use of a two-step resonant scattering scheme in I_2 is reported. The 80-kHz-wide hyperfine lines obtained for the $B0_u^+ \to X^1\Sigma_g^+(43,11)$ $R(15)$ transition are used and as high-resolution probes for collisional studies. Applications to frequency standards are considered. Also included are high-resolution measurements of the ac Stark effect in a folded Doppler-broadened system in I_2.

We report the observation of linewidths narrower than the natural width using a two-step resonant scattering scheme in a folded configuration in I_2 vapor. The measured width of the lines at 5828 Å is 80 kHz, with the major contributions to the width stemming from instrumental broadening mechanisms and laser jitter in our present setup. These extremely sharp lines, which, to our knowledge, are the narrowest recorded in the visible region of the spectrum,[1] have a number of important applications. In addition to Doppler-free stimulated-emission spectroscopy of thermally unpopulated levels, these narrow resonances can be used as unique high-resolution probes for the study of collisional effects on specific energy levels and as reference lines for laser frequency standards. In this Letter, we have included preliminary data demonstrating the application of the two-step resonance-scattering method to the measurement of hyperfine structure and collisional broadening of high-lying vibrational levels in the ground electronic state of I_2. In addition, we have also included high-resolution measurements of the ac Stark effect in a folded Doppler-broadened system in I_2.

Two-step excitation for folded and cascade systems in gas cells has been the subject of many recent theoretical and experimental studies.[2-8] In this Letter, however, we are primarily concerned with two-step resonant scattering in a folded, Doppler-broadened three-level system in Fig. 1(a), where Ω_p and Ω_s are the pump and probe frequencies, respectively. If the pump and probe fields are monochromatic and very weak, the linewidth of the probe transition Γ_s can be shown[2-7] to be

$$\Gamma_s = \Gamma_1 + (1 \pm \Omega_s/\Omega_p)\Gamma_2 + \Gamma_3, \qquad (1)$$

where Γ_1, Γ_2 and Γ_3 are the relaxation rates of the populations in levels 1, 2, and 3, respectively. The plus sign is for copropagating beams and the minus sign is for counterpropagating beams. For a stationary atom or an atomic beam Γ_s reduces to $\Gamma_1 + \Gamma_3$. Equation (1) can be easily derived from energy-conserving scattering behavior in a three-level system as long as both fields are weak (i.e., the Rabi frequencies $\mu_{12}E_p/h$ and $\mu_{23}E_s/h$ are much smaller than Γ_2, where μ_{ij} is the dipole matrix element for the transition from level i to j and E_p and E_s are the pump and probe fields, respectively). If levels 1 and 3 are metastable, then Γ_s will approach a δ function in an atomic beam or $(1-\Omega_s/\Omega_p)\Gamma_2$ in a gas cell (at low pressure) for copropagating beams. For the latter case, when $\Omega_s \approx \Omega_p$, Γ_s will also be extremely narrow, certainly much narrower than Γ_2.

Our experimental setup consists of a cw argon-ion laser at 5145 Å as the pump, and a tunable cw dye laser as the probe, collinearly propagating with the same linear polarization in an I_2 vapor cell. Each laser operates in a single frequency[9] and the combined beams are expanded to a 5 mm diameter to reduce transit-time effects. After three passes through the 80-cm-long cell, the probe beam is separated from the pump by a dispersion prism and its intensity detected on a silicon photodiode. To achieve high detection sensitivity, the pump is amplitude modulated electro-optically at 2 kHz and the probe intensity is synchronously demodulated after subtraction of probe intensity fluctuations.

To record the spectra in Fig. 1, the pump laser is held fixed at a frequency within the 21 Doppler-broadened $BO_u^+ - X^1\Sigma_g^+(43,0)$ $R(15)$ $\Delta F = \Delta J$ hyperfine transitions[10] and the probe laser is tuned over the $BO_u^+ \to X^1\Sigma_g^+(43,11)$ $R(15)$ $\Delta F = \Delta J$ transitions at 5828 Å. Figure 1(c) shows 16 of the 21 probe hyperfine lines. The number of lines that can be observed in a particular scan depends on the location of the pump frequency with respect to overlapping Doppler-broadened hyperfine structure associated with the pump transition. Figure 1(b) shows in more detail the hyperfine line terminating on $|J''I''F\rangle = |15,5,20\rangle$ with a width of 80 kHz. The natural width of this transition if observed as a single-step process[11] is expected to be 141 kHz, which includes contributions from both radiative and magnetic predissociation effects.[12] According to Eq. (1), the linewidth for the same transition observed by the two-step process in our experiment should be 16.5 kHz. The measured width of 80 kHz, although narrower than the 141-kHz natural width, is still wider than the theoretically predicted width of 16.5 kHz because of dye-laser jitter and other line-broadening mechanisms in our present setup.

The spacings between the hyperfine lines of Fig. 1(c) are different from those measured in a single-step process. The line separations in this case depend on the corresponding hyperfine structure associated with the pump transition[10,13] and the ratio Ω_s/Ω_p as discussed elsewhere.[14] We have made a preliminary measurement of the complete hyperfine structure of the $v''=11$ level using the modes of an external Fabry-Perot as frequency markers. The data were fitted by use of a least-squares program with a standard deviation of 690 kHz, resulting in values for the nuclear electric quadrupole ($eQq''=-2455.6\pm 1.3$ MHz) and spin rotation ($C''=7.9\pm 1.0$ kHz) coupling constants for the $v''=11$ level which are consistent with measurements utilizing an optically pumped I_2 laser.[14] A significant improvement in the data can be made by use of a long-term stabilized pump laser[9] and a heterodyne technique for determining the frequency separations.

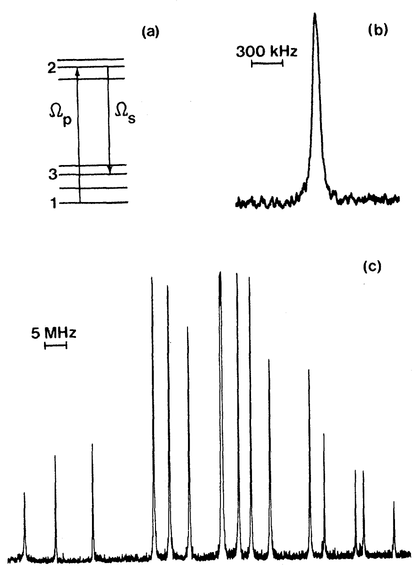

FIG. 1: (a) Schematic diagram of the two-step scattering process in a folded configuration. (b) High-resolution scan of the $|J''I''F''\rangle=|15,5,20\rangle$ $R(15)$ hyperfine transition. Peak intensities: pump 5.2 mW/cm^2, probe 3.6 mW/cm^2. $P=0.5$ mTorr, $\tau=10$ msec, scan rate 600 kHz/sec. (c) Typical scan of (43,11) probe hyperfine transitions for weak fields. Peak intensities: pump 15 mW/cm^2, probe 6.3 mW/cm^2, $P=2$ mTorr, $\tau=10$ msec.

The extreme narrowness of the lines obtained with the two-step scattering scheme suggests many possible applications, one of which is the study of collisional relaxation effects, particularly at very low pressures. Figure 2 shows the hyperfine transition terminating on $|J''I''F''\rangle=|15,1,15\rangle$ for three different I_2 pressures (1.5, 8, and 28 mTorr) recorded with weak pump and probe beams. The line shape in Fig. 2(c) for a pressure of 28 mTorr has a width of 710 kHz which is almost 10 times wider than the present instrumentally limited width and more than 100 times the expected theoretical linewidth[15] at zero pressure. The line shape is indeed Lorentzian as shown by the superimposed fit marked by dots. The increase of the linewidth with pressure yields an estimate for pressure broadening for this two-step process of 22 kHz/mTorr. This can be contrasted with an estimate we have made for the single-step (43,0) $R(15)$ transition of 12 kHz/mTorr based on the data of Camy *et al.*[16] The determination of collisional relaxation rates of individual levels, i.e., Γ_1, Γ_2, and Γ_3, may be computed using linewidth measurements obtained for the single-step process and for copropagating and counterpropagating beams in the two-step process as discussed in Ref. 6.

The application of these sharp lines generated by the two-step process to secondary frequency standards is very promising. For example, in

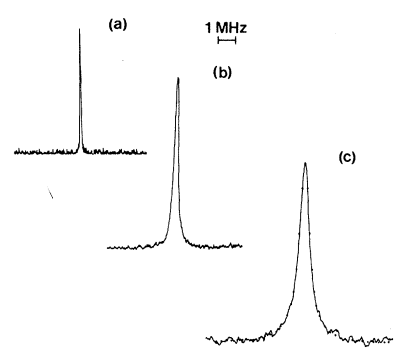

FIG. 2. Effect of collisions on $|J''I''F''\rangle=|15,1,15\rangle$ hyperfine line. (a) $P=1.5$ mTorr, (b) $P=8$ mTorr, (c) $P=28$ mTorr. Peak intensities: pump 7.7 mW/cm^2, probe 3.7 mW/cm^2. Vertical scale is arbitrary.

I_2, the probed rovibrational hyperfine transitions[14] can establish a set of reference frequency markers extending from about 0.5 to 1.3 μm. The stability and reproducibility of a probe laser stabilized to such a reference would be determined primarily by the absolute stability of the pump laser[9] with small contributions from pressure shifts in the I_2 cell.[17] It should also be noted that because these reference lines, whether in atoms or molecules, can be very sharp, the frequency difference between pump and probe (or between two probes using a common pump) may be established extremely accurately. This suggests applications to spectroscopy and frequency standards in the rf microwave/far-ir regions using optical lasers.

We have also examined with high resolution the effect of increasing the pump field intensity on the probe line shape (ac Stark effect) in our folded Doppler-broadened system. The ac Stark effect in three-level Doppler-broadened systems has been the subject of numerous studies both experimentally as well as theoretically.[2-8] Figure 3 shows the resulting change in the $|J''I''F''\rangle = |15,1,5\rangle$ line shape for peak pump intensities ranging from 7.7 mW/cm^2 to 4.7 W/cm^2. Starting from the weak-field case [Fig. 3(a)], the line first broadens [Fig. 3(b)] and then splits [Fig. 3(c)]. Theoretical calculations corresponding to our configuration predict a splitting S which is dependent on the Ω_s/Ω_p ratio, given by[5-7]

$$S = \frac{\mu_{12}E_p}{h}\left[\frac{4\Omega_s}{\Omega_p}\left(1-\frac{\Omega_s}{\Omega_p}\right)\right]^{1/2}. \tag{2}$$

Using Eq. (2) we made an estimate for the pump transition matrix element μ_{12} of 0.057 ± 0.008 D. The Ω_s/Ω_p dependence of Eq. (2) was checked by measuring S with the probe laser tuned to the $v'' = 10$ ($\lambda_s = 5761$ Å) and to the $v'' = 12$ ($\lambda_s = 5879$ Å) levels for the same pump intensity. The predicted 8% change in S was confirmed.

For peak pump intensities greater than about 1 W/cm^2 the line shape also exhibits absorption [Fig. 3(d)] and a further splitting near line center [Fig. 3(e)]. We must emphasize that the details of the line shapes in Fig. 3, especially Fig. 3(e), are apparently not predicted by the closed-form calculations in Ref. 5. The applicability of the numerical approach of Ref. 7 is yet to be examined. A more thorough study of the ac Stark effect in a folded Doppler-broadened system is at present being pursued both experimentally and theoretically.

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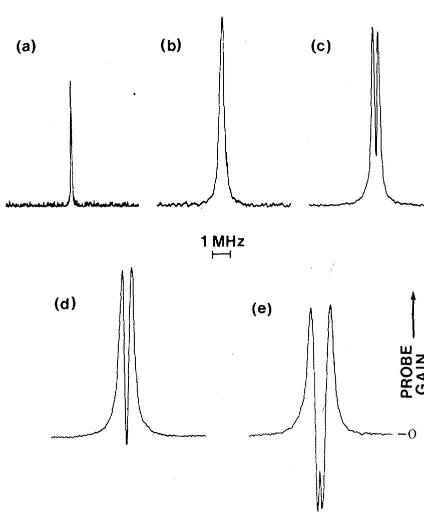

FIG. 3: $|J''I''F''\rangle = |15,1,15\rangle$ hyperfine line as a function of pump intensity. (a) 7.7 mW/cm^2, (b) 68 mW/cm^2, (c) 330 mW/cm^2, (d) 1.0 W/cm^2, (e) 4.7 W/cm^2. Probe intensity 3.1 mW/cm^2, $P = 2$ mTorr. Vertical scale is arbitrary.

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