High-Frequency Optically Heterodyned Saturation Spectroscopy Via Resonant Degenerate Four-Wave Mixing

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Resonant degenerate four-wave mixing with two close optical frequencies (ω, $\omega+\delta$) is used to perform high-frequency optically heterodyned saturation spectroscopy. Doppler-free spectra of I_2 ($\lambda = 514.5$ nm) are obtained in this way for 20 kHz $< \delta <$ 20 MHz. Lock-in detection for $\delta < 1$ MHz allows relaxation studies and line assignments. At higher frequencies, rf-power detection yields Doppler-free doublets split by $3\delta/2$.

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This Letter reports on a sensitive new scheme of Doppler-free spectroscopy based on a high-frequency heterodyne-detection technique with resonant degenerate four-wave mixing.[1] The advantages of degenerate four-wave mixing to generate phase-conjugate (PC) fields are now well demonstrated, thanks to automatic phase matching. Recently Liao, Bloom, and Economau[2] have shown that this effect can be observed with cw lasers in low-pressure gases if the laser frequency is tuned to an atomic resonance line. Because of the standing-wave character of the pump beam, the Doppler effect is partially or totally canceled.

We used four-wave mixing techniques to perform high-frequency heterodyne Doppler-free spectroscopy. Two schemes may be considered. In the first one [Fig. 1(a)], both pump and probe are standing waves, and the probe frequency, $\omega+\delta$, is shifted from the pump frequency. Thus a PC wave is reemitted opposite to either probe at frequency $\omega-\delta$, and interferes with the return probe wave (serving as local oscillator, LO) to yield a beat at frequency 2δ. Experiments of this type have been performed in Ar discharges and will be published elsewhere. Note that this scheme allows one to vary the LO amplitude without altering the reemitted wave, and in this way, represents an alternative to polarization spectroscopy.[3] We describe here experiments using an alternative scheme [Fig. 1(b)] which consists in overlapping both running probe and standing pump waves in order to increase the signal amplitude. Two PC waves are thus reemitted at frequencies $\omega+\delta$ and $\omega-\delta$, and interfere with the return pump wave to yield a beat signal at frequency δ. Advantages of the above techniques lie in the increased sensitivity of heterodyne detection and the possibility of reaching the signal-shot-noise limit by going out of the laser noise spectrum for large δ's, and eventually using sensitive rf detection.

In a saturated absorption scheme (ω tuned to a one-photon resonance), the configuration of Fig. 1(b) yields Doppler-free doublets separated by $3\delta/2$. Indeed as seen in the diagrams visualizing PC emission at frequencies $\omega \pm \delta$ (Fig. 2), the maximum intensity is obtained when one-photon absorption and three-photon transition to the excited state (by absorption, emission, and absorption, successively) are simultaneously resonant for the same axial velocity group, v. For instance, in the case of Fig. 2, process (A) (emission at $\omega-\delta$) conditions $\omega - kv = \omega_0$ and $\omega - kv - (\omega+\delta-kv) + \omega + kv = \omega_0$ yield $\omega = \omega_0 + \delta/2$ (kv, Doppler shift). Similarly [Fig. 2, process (B)], conditions $\omega+\delta-kv=\omega_0$ and $\omega+\delta-kv-(\omega-kv)+\omega+kv=\omega_0$ yield $\omega=\omega_0-\delta$ for the $\omega+\delta$ emission. A perturbative calculation[4] shows that the beat signal line shape is given (γ, optical linewidth;

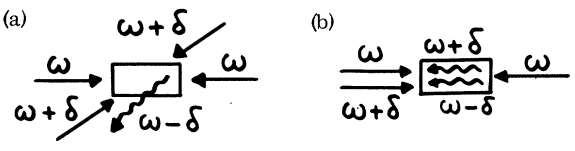

FIG. 1. Schemes for two-frequency degenerate four-wave mixing. In scheme (b), note that, in general, the reemissions at frequencies $\omega+\delta$ and $\omega-\delta$ are not simultaneously resonant for the same frequency ω.

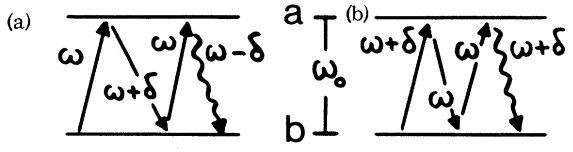

FIG. 2. Processes induced in collinear resonant four-wave mixing. As explained in the text, (a) process (A) is resonant for $\omega=\omega_0+\delta/2$, while (b) process (B) is resonant for $\omega=\omega_0-\delta$.

γ_α, relaxation rate of level α) by

$$S=\sum_\alpha \frac{C_\alpha}{\gamma_\alpha+i\delta}\left[\frac{1}{\gamma-i(\omega-\omega_0-\delta/2)}+\frac{1}{\gamma+i(\omega-\omega_0+\delta)}\right], \quad (1)$$

where C_α is a geometrical coefficient depending on the transition angular momenta and light polarization.[4] In (1), the $(\gamma_\alpha+i\delta)^{-1}$ resonant contributions come from the fact that, in the processes of Fig. 2, populations of levels a and b are modulated at frequency δ. It induces a phase shift in the beat signal. The doublet structure is contained in the resonant behavior of the bracketed quantity of Eq. (1). In three-level systems, crossover resonances[5] are also expected as doublets, but, in (1) the sum over levels α is then reduced to the common level only. Note that, when the frequency difference is very small ($\delta \ll \gamma, \gamma_\alpha$)$S$ reduces to the conventional saturated absorption signal $[\gamma^2+(\omega-\omega_0)^2]^{-1}$.

Our experiments were performed on a 40-cm-long I_2 gas sample with a 5145-Å Ar^+ laser. The procedure to stabilize the laser frequency ω has been discussed by Bordé, Camy, and Decomps.[6] The experimental setup will be described elsewhere in detail.[7] An acousto-optic (A/O) frequency shifter, supplied with two input rf frequencies, $\Delta\pm\delta/2$, allowed us to generate two corunning beams at frequencies $\omega+\Delta\pm\delta/2$, which overlap a counterpropagating beam (frequency ω) inside the I_2 cell. We carefully avoided generation of other optical frequencies. (In the A/O shifter, harmonic rejection was better than 40 dB.) The overall frequency shift, $\Delta\simeq 75$ MHz, has no important effect on the beat signal (the doublet peaks are simply shifted to new values, $\omega=\omega_0-\Delta/2\pm 3\delta/4$) but introduces a noticeable improvement in the optical isolation between the two arms.[7]

The frequency separation δ has been varied between 20 kHz and 20 MHz. For 20 kHz $\lesssim\delta\lesssim$1 MHz, we used a *lock-in detection* of the beat signal. Figure 3 shows a portion of the I_2 spectrum recorded for $\delta=70$ kHz, and two different phases $\Phi_0=29°$ and $\Phi_0+\pi/2$ (the phase origin is taken as the one of the output beam from the A/O shifter). Since δ is smaller than the homogeneous linewidth, the predicted doublet is not resolved,[8] but the overall phase of the signal is shifted because the populations of the molecular levels follow the modulation with some delay (δ is of the order of the γ_α's). Notice that the signal-to-noise ratio is better than 500, for a time constant 0.3 s (close to expected photon-shot-noise limit), and that, without the use of *frequency derivative* techniques, there is no detectable background.[7] An important point is that the phase delays of the various resonances differ, as could be predicted from Eq. (1). This is easily seen around the signal quadrature. At the main lines quadrature ($\Phi=119°$), ground-state crossovers (↓) are still positive (longer phase delay), while excited-state crossovers (↑) are already negative. This property can be easily used for a *simple and di-*

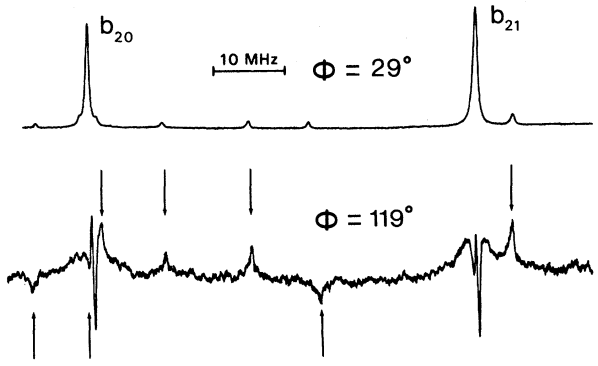

FIG. 3. Spectrum of a portion of the I_2 [$R(15)$, $v=0\rightarrow v=43$] line at 5145 Å for $\delta=70$ kHz and two different phase shifts. Crossover lines due to a commom excited (↑) or ground (↓) level are indicated by arrows ($p=10$ mT). In the lower curve ("quadrature"), scale magnification is 25.

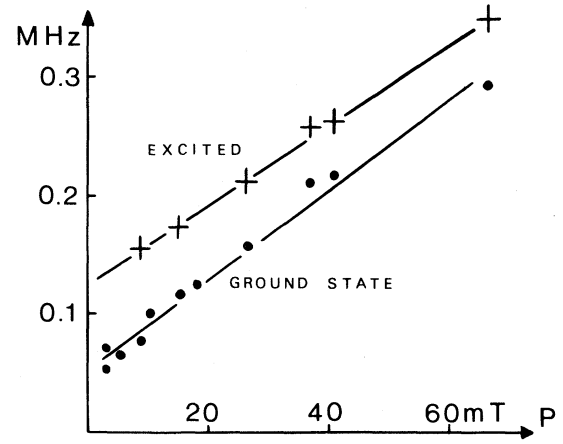

FIG. 4. Relaxation of the ground (closed circles) and excited state [hyperfine level b_{20} (crosses)], as obtained from the crossover phase delays.

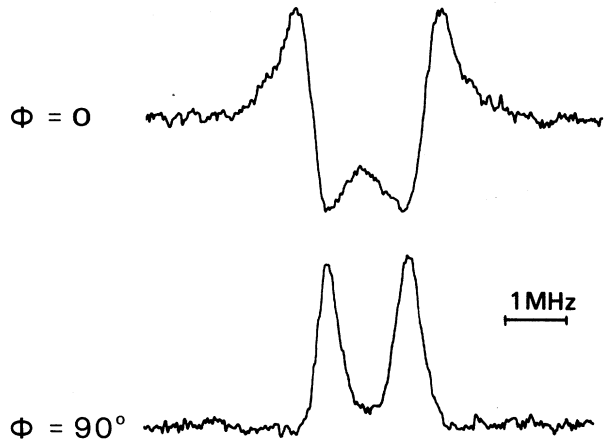

FIG. 5. Resonance line shape for $\delta = 1$ MHz [$P(13)$, line a_{11}; $p = 3$ mT]. Depending on the phase, the detected heterodyne beat yields either saturated dispersion (zero phase delay) or saturated absorption signals ($\pi/2$ phase delay).

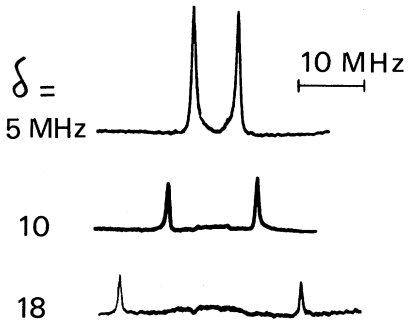

FIG. 6. Power detection of the beat signal for various values of δ.

rect assignment of the crossovers. Also note the presence, in the main lines quadrature, of a pedestal of width (5–10 MHz) intermediate between homogeneous ($2\gamma \sim 1$ MHz) and Doppler linewidths (~450 MHz). From its pressure variations, we can attribute it to the effect of elastic velocity-changing collisions. In spite of its relative weakness (~1%), this pedestal may be observed because its phase delay is larger than the one of the main lines (the molecules which contribute to it have a longer lifetime).[9]

The phase delays of the various crossovers allow us to measure the relaxation rates of ground and excited states (Fig. 4). The pressure broadening is roughly identical for both states (3.4–3.8 MHz/T). The present work on the I_2 ground state, to our knowledge, is the first measurement of quenching collisions in gas phase. The large value of the ground-state pressure-broadening rate represents an important step towards the comprehension of the relaxation mechanisms involved in cw molecular iodine lasers.[10] In the excited state, the fact that the pressure broadening is larger than the one observed in fluorescence decay (2.5 MHz/T)[11] might be attributed to a residual effect of weak elastic collisions. The difference between the zero-pressure decay rates comes from the excited state natural width (~77 kHz for the b_{20} hyperfine level[6]). The measurement technique resembles the phase-shift method in modulated fluorescence,[11] but is much more powerful because the frequency resolution allows the selection of a *single state* and of a *single velocity group.*[12]

When δ is increased, the doublet is clearly resolved, as seen on Fig. 5, for $\delta = 1$ MHz. The overall phase delay is nearly $\pi/2$ [cf. Eq. (1)], and then the part of the beat signal in phase with the A/O shifter output beam exhibits dispersion-type features (Fig. 5, $\Phi = 0$). Note that now in-phase and out-of-phase components have the same amplitude. At higher frequencies, we used another detection method in which the *signal power*, after mixing with a local oscillator, was monitored at the intermediate frequency of 30 MHz. In Fig. 6, doublets corresponding to a single line (a_3) are shown for various values of δ. Even at 18 MHz, the beat is easily observable in spite of the attenuation introduced by the nonresonant population denominator of Eq. (1). The doublet splitting, $3\delta/2$, can be used as a precise frequency-calibration technique.

This set of result obtained on I_2 by high-frequency optically heterodyned saturation spectroscopy shows the power of this technique to get new information on the line assignment, the decay rate of the levels, and the analysis of the relaxation dynamics in gas phase (elastic collisions, etc.). This method is generally applicable to any nonlinear spectroscopic schemes (such as polarization spectroscopy,[3] etc.) where it allows an optimization of the signal-to-noise ratio by an adequate choice of the detection frequency. This is particularly promising for two-photon and Raman spectroscopy, because the signal is thus nearly insensitive to the detection frequency.[4]

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[8]However, the narrow structure observed on the main lines quadrature (Fig. 3, $\Phi = 119°$) comes from the fact that δ is not completely negligible compared with γ. [Relative amplitude of the quadrature signal, $3\delta/8\gamma$ (Ref. 4).] On the other hand, the width of the in-phase signal is $\Delta\omega \simeq 2\gamma + 9\delta^2/16\gamma$.

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[12]It should be noted that, in some sense, high-freqency heterodyne saturation spectroscopy is the Fourier transform analog of time-resolved saturation spectroscopy [M. Ducloy, J. R. R. Leite, and M. S. Feld, Phys. Rev. A 17, 623 (1978)].

Subnatural Linewidth Spectroscopy by Double Optical Resonance with Two-Photon Pumping

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A variant of the double-optical-resonance method is proposed in which the pumping step is a two-photon transition, either stimulated Raman scattering or two-photon absorption. It is shown that the probe linewidth can be minimized by proper choice of pump frequencies and assignment of pump beam propagation directions relative to the probe beam. The minimum linewidth is smaller than the value obtained by the usual one-photon pumping technique, and the method does not require any relations among level spacings.

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Linewidths narrower than the natural width have been observed by the use of Ramsey fringes,[1-7] time-delayed fluorescence,[8-10] and double optical resonance.[11-17] The last method was recently used by Hackel and Ezekiel[18,19] to obtain subnatural linewidths by two-step resonant scattering in I_2 vapor, with the level scheme shown in Fig. 1(a). In their experiment the transition $a \rightarrow b$ is driven by a pump field E of frequency Ω, and the transition $b \rightarrow c$ is probed by a collinearly propagating field E' of frequency Ω' which satisfies $|\mu_{bc}E'| \ll |\mu_{ba}E|$. In this Letter we present a variation on this idea in which the three levels are chosen so that the pumping transition $a \rightarrow b$ can be a two-photon process, either stimulated Raman scattering, Fig. 2(a), or two-photon absorption, Fig. 2(b). The principal advantage of this scheme is that the two pump frequencies are only constrained by one resonance condition; thus one frequency is freely adjustable. This freedom can be used to reduce the theoretically predicted linewidth below the minimum value possible for conventional double resonance with single-photon processes. In the interests of definiteness we will only describe the analysis for the level scheme of Fig. 1(a), but we should point out that two-photon pumping schemes, with the advantage just mentioned, cal also be devised for line-narrowing experiments suing the level schemes of Figs. 1(b) and 1(c).

The calculation of the probe linewidth for a two-photon pumping scheme is easily done by exploiting the formal similarity between the theory for Fig. 1(a) and the theory for Figs. 2(a) or 2(b). The object in each case is to find the steady-state solution for the three-level density matrix, this in turn determines the nonlinear susceptibility which is measured in experiments. This was