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NOBLE-GAS-INDUCED COLLISIONAL BROADENING OF THE 6P1/2-6P3/2 TRA--ETC(U)
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Noble-gas-induced collisional broadening of the $6P_{1/2}$-$6P_{3/2}$ transition of Tl measured by Raman echoes

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(Received 5 November 1981)

We implement the Raman echo for the first time in collision studies to measure the noble-gas-induced broadening of the $6P_{1/2}$-$6P_{3/2}$ transition of atomic thallium. Except in the case of He, the measured broadening cross sections He 59(2), Ne 67(2), Ar 124(4), Kr 167(5), and Xe 204(6) Å^2 ($T=840$ K), agree with those calculated assuming a pure van der Waals collisional interaction. Our measurements, sensitive to collisionally induced changes in axial velocity of $\gtrsim$ 300 cm/sec, reveal that collisional velocity changes of that magnitude are not important in Tl–noble-gas collisions.

As numerous experiments in the last few years have indicated,[1–5] optical echo techniques are very well suited to the study of low perturber-gas pressure and impact-regime collisional broadening measurements. The Doppler-free character of the echo technique facilitates precision measurements of both the phase changing or inelastic aspect of collisions as well as the velocity-changing aspect of atomic collisions. In this paper, we report the results of a Raman echo study of the noble-gas-induced broadening of the electric dipole forbidden 7793-cm^{-1}, $6P_{1/2}$-$6P_{3/2}$ transition of atomic thallium. This experiment marks the first application of the Raman echo to the study of collisional broadening,[6] and demonstrates that echo collisional-broadening-measurement techniques are applicable even when the transition of interest is electric dipole forbidden and in the near infrared. Since other collisional-broadening-measurement techniques are difficult to apply in such cases, it is not surprising that collisional broadening of the $6P_{1/2}$-$6P_{3/2}$ transition in Tl has not been studied previously. For the excitation-pulse separations used in our experiment, no velocity-changing effects associated with Tl-noble-gas collisions are observed. The measured collisional broadening cross sections are, except in the case of He, in excellent agreement with broadening cross sections calculated assuming a pure van der Waals collisional interaction.

The Raman echo[6,7] is the two-photon analog of the ordinary photon echo.[8] The procedure for generating Raman echoes by copropagating excitation pulses is essentially identical to that used for generating two-pulse photon echoes. For Raman echo, each excitation pulse must contain radiation at two frequencies [see Fig. 1(a)] whose difference equals the frequency of the $|0\rangle$-$|2\rangle$ Raman transition [see Fig. 1(b)]. A Raman echo is observed if a probe pulse is applied according to the time sequence of Fig. 1(a). In our experiment, the $|0\rangle$ and $|2\rangle$ states correspond to the hyperfine levels of the $6P_{1/2}$ ground state and the $6P_{3/2}$ metastable excited state, respectively. The

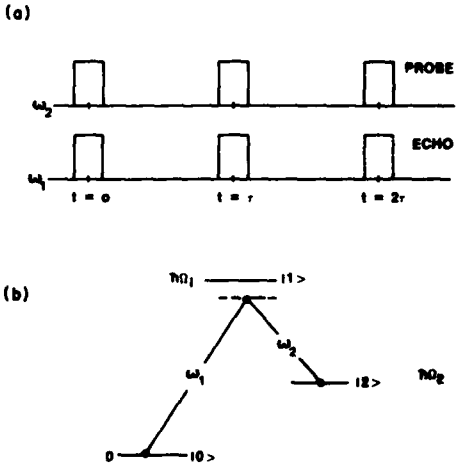

FIG. 1. (a) Two dual-frequency excitation pulses and a single frequency probe pulse are used to create the Raman echo-signal pulse (dashed border). All laser pulses and echo-signal pulse copropagate in our experiment. (b) The two-photon-difference-frequency excitation process, utilized in creating the Raman echo, is depicted in a simple three-level atomic system. The $|1\rangle$ state serves as the "intermediate state" in the excitation and probing processes.

intermediate state $|1\rangle$ corresponds to the $7S_{1/2}$ state.

Echo signals depend on the persistence of order[9] (which is introduced into the echo material by the first excitation pulse and is modified by subsequent excitation pulses) until the time at which the echo signal is emitted. Collisions, however, tend to thermalize the sample before the echo signal can be emitted. Since collisions themselves are characteristic of inherent atomic or molecular parameters and independent of the type of echo being studied, it follows that differences in the collisional degradation rates associated with various echo effects can arise only if the nature of the order associated with various echo effects is different. In the case where dispersion is neglected and both frequency components of the Raman echo excitation pulses are copropagating, it is straightforward to show that each dual-frequency pulse affects the sample in the same fashion as would a single-frequency pulse which is directly resonant with the $|0\rangle$-$|2\rangle$ transition and copropagates with the dual-frequency pulse. It can be further shown that under the above conditions the Raman echo inducing order introduced into the sample is identical to that which would be induced in a two-pulse photon echo experiment conducted on the same $|0\rangle-|2\rangle$ transition. It follows that collisions will degrade the Raman and two-pulse photon echoes in the same way. Since the Raman echo experiments described here were conducted with copropagating laser pulses, we can therefore apply the standard treatment[5] of collisional degradation of photon echoes to analyze our Raman echo results. If the frequency components of the Raman echo excitation pulses are not copropagating, the spatial periodicity of the phase information introduced into the sample would not be the same as it would be when induced by direct single-photon excitation. As a result the effect of collisional velocity changes will be different in the two-pulse photon echo and Raman echo cases. In the above discussion, we have neglected relaxation processes occurring during the excitation pulses. These may differ in one- and two-photon excitation cases because of the importance of the intermediate state in two-photon excitation.

As a function of perturber gas pressure P, the Raman echo intensity, I_e^{RE}, is expected to obey[5]

$$I_e^{RE}(P)=I_e^{RE}(0)\exp(-\beta P)\ , \tag{1}$$

where $I_e^{RE}(0)$ is the echo intensity in the absence of perturber gas,

$$\beta=\frac{4v_r\tau}{k_BT}\sigma_{\rm eff}(\tau)\ , \tag{2}$$

$v_r=\sqrt{8k_BT/\pi\mu}$ is the mean radiator perturber relative speed; k_B is the Boltzmann's constant; T is the absolute temperature; μ is the Tl-perturber reduced mass; τ is the excitation-pulse separation, and

$$\sigma_{\rm eff}(\tau)\equiv\sigma_B+\sigma_v(\tau)\ . \tag{3}$$

The effective echo decay cross section[10] $\sigma_{\rm eff}(\tau)$ is comprised of σ_B, which is the τ-independent collisional broadening cross section, and a τ-dependent effective velocity-changing collision cross section σ_v. The broadening cross section σ_B is identical with the broadening cross section measured in traditional pressure broadening studies. At sufficiently long τ, $\sigma_v(\tau)$ becomes constant and represents the total cross section for velocity-changing collisions. At intermediate τ, $\sigma_v(\tau)$ represents, roughly speaking, the cross section for velocity-changing collisions in which $|k\Delta v\tau|\gtrsim 1$, where k is the magnitude of the wave vector associated with the echo transition, and Δv is the change in velocity (along the excitation direction) which occurs in the collision. We note that the

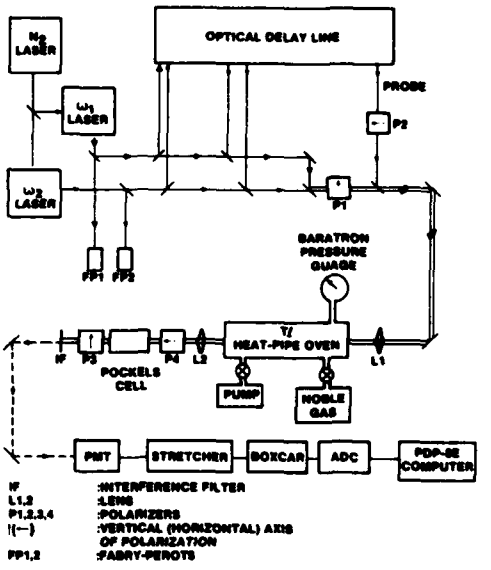

FIG. 2. Experimental set-up for measuring the noble-gas-induced collisional broadening of the $6P_{1/2}$-$6P_{3/2}$ transition of Tl by the Raman echo technique. The Fabry-Perot etalons are used to monitor the dye laser frequencies. A pockels cell optical shutter is introduced to reduce the excitation pulse leakage through the analyzing polarizer $P4$.

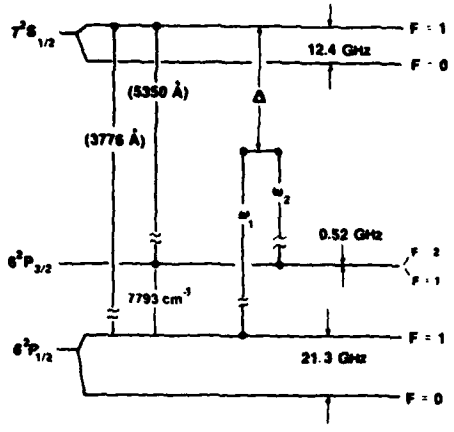

FIG. 3. The energy levels and their separations of interest of thallium. The detuning of the laser fields from the $F=1$, $7S_{1/2}$ intermediate state is denoted by Δ. The difference in hyperfine structure (isotope shift and splittings) between ^{203}Tl and ^{205}Tl is small (see Ref. 12) and is not resolved in our experiment.

often used collision broadening rate γ_B, which gives the half-width-at-half-maximum of the collisionally broadened line in units of rad cm^3/sec, is related to σ_B through

$$\gamma_B = v_r \sigma_B \ . \tag{4}$$

Since our Raman echo apparatus (see Fig. 2) is described elsewhere,[11] we present here only the details relevant to our broadening measurements. The Raman echo excitation pulses are supplied by N_2-laser-pumped dye lasers, have an 8-GHz spectral width and a 5-nsec temporal width. Experiments were conducted with the excitation-pulse separations $\tau=20$, 33.5, and 61 nsec. For $\tau=20$ and 33.5 nsec the excitation pulses (at ω_1 and ω_2) were detuned by an amount of $\Delta \approx 43$ GHz from resonance with the $F=1$ hyperfine level of the $7S_{1/2}$ intermediate state (see Fig. 3). The hyperfine structure of the $6P_{3/2}$ state is sufficiently small that our excitation pulses do not resolve it. Measurements have also been made by exciting the $F=0$ hyperfine level of the $6P_{1/2}$ ground state. For the $\tau=61$ nsec case, the loss of laser power introduced by the longer optical delay, made it necessary to set $\Delta=0$ for those measurements. The thallium[12] vapor of natural isotopic concentrations, i.e., 29.5% ^{203}Tl and 70.5% ^{205}Tl, is maintained in a 1.5-inch diameter stainless-steel heat-pipe-type cell[13] which is cooled at the ends. During our experiments, the Tl density is maintained at $\approx 10^{13}$ cm^{-3}, which corresponds to cell temperatures near 570°C. Tl-Tl collisions are expected to be negligible. Perturber gas, whose pressure (ranging from $\sim 0-5$ torr) is monitored by a capacitance manometer, can be admitted to the cell. The echo signal is monitored on a photomultiplier tube (PMT) whose output current is integrated and read by a PDP-8/e computer. The computer also monitors the perturber-gas pressure. In an experimental "run," the echo intensity is monitored while the perturber-gas pressure is first increased and then returned to its original value. Several thousand laser shots (repetition rate ≈ 5 Hz) are measured in each run. We found that the dependence of the echo intensity on perturber pressure was well described by Eq. (1) (see Fig. 4). Values of β and hence $\sigma_{\text{eff}}(\tau)$ were determined by least-squares fits to the data.

The results of our measurements (obtained while exciting the $F=1$ hyperfine level of the $6P_{1/2}$ state) are presented in Table I. The cross sections obtained by exciting the $F=0$ $6P_{1/2}$ level are the same within our experimental accuracy. Since our 5-nsec excitation pulse widths are non-negligible compared to τ, we assume a minimum fractional uncertainty in our σ_{eff} values determined by one-half the excitation pulse width divided by 2τ. The uncertainties presented in Table I represent the larger of the statistical or pulse width induced uncertainties. In separate measurements of σ_{eff} at $\tau=20$ and 33.5 nsec obtained with $\Delta=0$, we found that the cross sections were slightly higher than

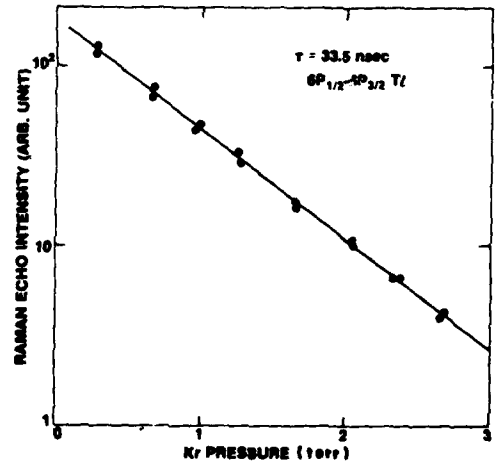

FIG. 4. Raman echo intensity vs Kr pressure. Each point represents an average of 150 or more laser shots. The straight line is a least-squares fit of Eq. (1).

TABLE I. The noble-gas-induced collisional broadening cross sections, $\sigma_{\text{eff}}(\tau)$, deduced from Raman echo studies of the $6P_{1/2}$-$6P_{3/2}$ transition of Tl. Measurements were performed at $T \simeq 570$ °C.

Perturber	$\tau=20$ nsec	$\tau=33.5$ nsec	$\tau=61$ nsec
He	59 (4) Å^2	61 (2) Å^2	58 (2) Å^2
Ne	65 (4)	68 (3)	68 (4)
Ar	121 (8)	126 (5)	126 (5)
Kr	167 (10)	171 (6)	163 (7)
Xe	207 (13)	206 (8)	198 (8)

those obtained with $\Delta=43$ GHz. We infer from this result that collisional relaxation of $7S_{1/2}$-$6P_{1/2}$ and $7S_{1/2}$-$6P_{3/2}$ transitions (superpositions[14]) becomes important when $\Delta=0$. The extent to which such relaxation is important depends on experimental details such as the temporal width, shape, and overlap of the two frequency components of each excitation pulse, and can be minimized by making the components of the excitation pulses short and cotemporal or by using non-intermediate-state resonant ($\Delta \neq 0$) excitation.

The fact that $\sigma_{\text{eff}}(\tau)$ is τ independent has two possible explanations. One is that we are already, at $\tau=20$ nsec, in the long-τ region for $\sigma_v(\tau)$. This implies that essentially all velocity changes which occur in Tl-noble-gas collisions satisfy

$$|k\Delta v\tau| \gtrsim 1 \text{ or } \Delta v \gtrsim 1000 \text{ cm/sec} .$$

However, based on results obtained in studies of Na–noble-gas[5] and Li–noble-gas[10] collisions and on the fact that Tl is quite massive, it appears unlikely that Δv is so large. The other explanation for the τ-independence of σ_{eff} is that $\sigma_v(\tau)$ is negligibly small compared to σ_B even for $\tau=61$ nsec. For this to be true there must be an insignificant number of velocity-changing collisions (compared to those which are primarily phase disrupting) in which $\Delta v \gtrsim 300$ cm/sec. This result is reasonable in light of the large mass of Tl; accordingly, we equate our σ_{eff} with σ_B.

If we assume that the Tl–noble-gas interaction is exclusively of the van der Waals type, we can calculate the expected collisional-broadening cross sections. Following Ref. 15, σ_B is found to be given by

$$\sigma_B = 4.08(C_6/\hbar)^{2/5} v_r^{-2/5} , \tag{5}$$

where C_6 is the van der Waals coefficient and is given by

$$C_6 = e^2 \alpha_d (\langle r_u^2 \rangle_{\text{av}} - \langle r_l^2 \rangle_{\text{av}}) . \tag{6}$$

Here, e is the electronic charge, α_d is the polarizability of the noble-gas atom, and $\langle r_u^2 \rangle_{\text{av}}$ ($\langle r_l^2 \rangle_{\text{av}}$) is the average value of the radius squared of the electron in the upper (lower) state of the broadened transition (in our case the $6P_{1/2}$-$6P_{3/2}$ transition). An approximate expression for $\langle r_k^2 \rangle_{\text{av}}$ is given by

$$\langle r_k^2 \rangle_{\text{av}} = \tfrac{1}{2} a_0^2 n^{*2} [5n^{*2} + 1 - 3l(l+1)] , \tag{7}$$

where $a_0 = 5.292 \times 10^{-9}$ cm is the Bohr radius, n^* is the effective principal quantum number of state k, and l is the orbital angular momentum quantum number of state k. We determine n^* through the relation

$$E_k = R_\infty / n_k^{*2} ,$$

where $R_\infty = 109\,737.3$ cm^{-1} is the Rydberg constant, and E_k is the energy of state k. We find $n^*(6P_{1/2}) = 1.492$ and $n^*(6P_{3/2}) = 1.627$. Then, assuming a temperature of 840 K, we arrive at the values of σ_B presented in Table II. The polarizabilities used[16] are also given in the Table. Comparing the calculated value of σ_B with $\bar{\sigma}_{\text{eff}}$ [i.e., $\sigma_{\text{eff}}(\tau)$ averaged over the three values of τ] we find that agreement is exceptional except for He, and in light of the simplicity of the model perhaps accidental. However, at least for the three heavier noble gases a primarily van der Waals interaction is reasonable.

TABLE II. A comparison of the noble-gas-induced collisional broadening cross sections of the $6P_{1/2}$-$6P_{3/2}$ transition of Tl as measured using Raman echoes and as calculated (see text). The cross section $\bar{\sigma}_{\text{eff}}$ represents $\sigma_{\text{eff}}(\tau)$ (see Table I) averaged over the three values of τ. A temperature of 840 K was assumed in the calculation of σ_B. The noble gas polarizabilities α_d were obtained from Ref. 16.

Perturber	$\bar{\sigma}_{\text{eff}}$	σ_B (Calc.)	α_d
He	59 (2) Å^2	37 Å^2	2.05×10^{-25} (cm^3)
Ne	67 (2)	65	3.95
Ar	124 (4)	129	16.4
Kr	167 (5)	171	24.8
Xe	204 (6)	222	41.2

Self-broadening of the electric-dipole forbidden transition in Tl can be studied by the measurement

of the Raman echo intensity as a function of excitation pulse separation τ. To make the self-broadening important, the measurement must be performed at high Tl number density, and using a nonintermediate-state resonant ($\Delta \neq 0$) Raman echo to reduce the problem of absorption.[11] It would be interesting to see such measurements performed.

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