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Observations of Anomalous Transparency in Bound-Free Transitions of Cl_2†*

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We have observed anomalous transparency in bound-free transitions of Cl_2 in pure Cl_2 and in Cl_2-He and Cl_2-Ar mixtures. From the dependence of transmitted laser energy on pressure, we have estimated the following collision diameters for Cl_2-Cl_2, Cl_2-Ar, and Cl_2-He collisions, respectively: 4.9, 4.4, and 1.5 Å.

Self-induced transparency (SIT) has been studied by McCall and Hahn[1,2] and others[3-7] in well-defined two-level systems. We have observed anomalous transparency that is reminiscent of this phenomenon in bound-free transitions of Cl_2.

A schematic diagram of the experimental arrangement is shown in Fig. 1. The input laser pulse (doubled ruby frequency) had a Gaussian distribution, which was found to be unaffected by absorption in Cl_2. In systematic studies with decreased relative power densities (from 1 to 0.0075), we obtained consistent results, thereby indicating that the dominant physical phenomena remained unchanged over the large range of power densities used by us. Representative observed transmission-time data on Cl_2 are shown in Fig. 2; similar results were obtained for Cl_2-He and Cl_2-Ar mixtures. Observed pulse delays (at the intensity maxima) as a function of chlorine pressure for pure Cl_2 and various Cl_2–inert-gas mixtures are plotted in Fig. 3. The bound-free transition of Cl_2, with maximum absorption at room

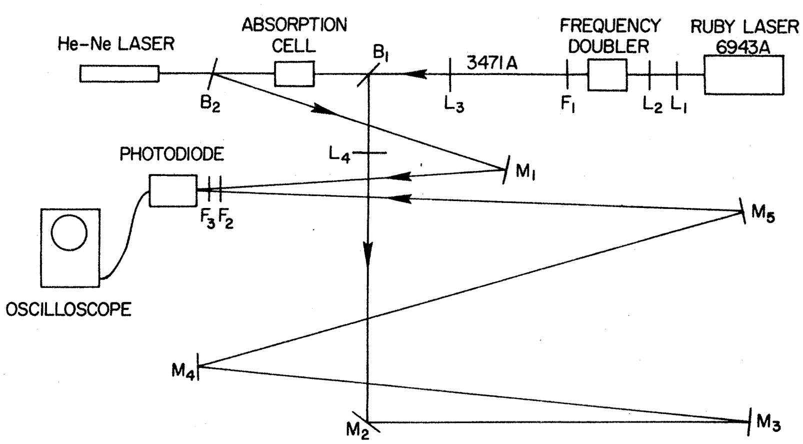

FIG. 1. Schematic diagram showing the experimental arrangement used. The optical components are defined as follows: L_i, lenses; F_i, filters; M_i, mirrors; B_i, half-silvered mirrors. The He-Ne laser was used to facilitate alignment. The optical delay line (B_1-M_2-M_3-M_4-M_5-photodiode) provided a reference beam for intensity measurements.

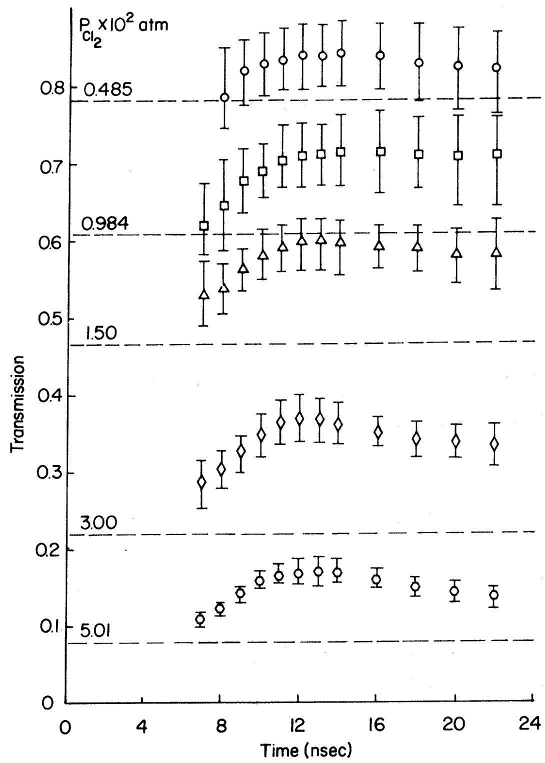

FIG. 2. Observed transmission through pure Cl_2 as a function of time for a peak laser-power output of 14.4 MW/cm^2. The dashed curves indicate the expected transmission for normal absorption.

temperature near 3300 Å in the region of the doubled ruby frequency, has been studied extensively.[8]

Analysis of the experimental conditions has shown that the following phenomena were of negligible importance on the time scale of the experiments and for the laser powers used in producing observed results: thermal changes in the gas along the laser beam, radial diffusion of atomic fragments, three-body collisions and atomic recombinations, laser-saturated absorption or "bleaching waves," self-focusing (which requires much higher powers than were used). Thus, the observed data must be explained in terms of bound-free absorption and dissociation, anomalous transparency involving nonlinear interactions of the bound-free absorbing system with the incident coherent radiation field, and two-body dephasing collisions. The transmission profiles plotted in Fig. 2, and in similar representations for Cl_2-He and Cl_2-Ar mixtures, show marked nonlinear dependence on laser-pulse input energy. If we define the incident, transmitted, and excess energy densities up to the time t, respectively, as

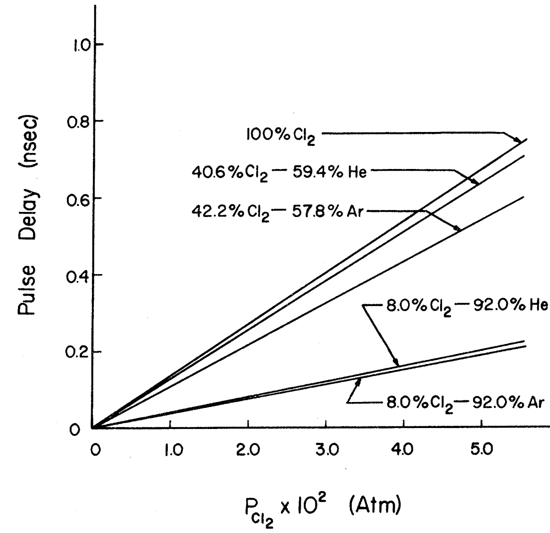

FIG. 3. Observed pulse delay as a function of chlorine pressure in pure Cl_2 and in various Cl_2–inert-gas mixtures.

$$E_{\rm in}(t) = C_0 \int_0^t \varphi(t')dt'.$$

$$E_{\rm out}(t) = C_0 \int_0^t T(t')\varphi(t')\,dt',$$

$$E_{\rm ex}(t) = E_{\rm out}(t) - C_0 \int_0^t T^*(t')\varphi(t')\,dt',$$

where C_0 is an experimentally determined calibration constant, $\varphi(t')$ measures the laser-pulse shape, $T(t')$ is the measured transmission, and $T^*(t')$ is the expected transmission if anomalous transparency did not occur, then we find that plots of $E_{\rm ex}$ as functions of $p_{\rm Cl_2}$, for various input energies, show well-defined maxima. We interpret the occurrence of these maxima as resulting from competition between an anomalous transmission phenomenon and dephasing collisions. Assuming that these competing processes both vary linearly with $p_{\rm Cl_2}$, we have estimated gas-kinetic collision cross sections on the assumption that every collision is a dephasing collision and that the pressure at which $E_{\rm ex}^*(t)$ has decreased by the factor e^{-1} (with the value of $E_{\rm ex}^*$ corresponding to the intersection of asymptotic lines for small and large values of $p_{\rm Cl_2}$) is the pressure at which the dephasing time $\simeq$ pulse duration time $\simeq 10^{-8}$ sec. We find that the average collision diameters obtained in this manner are 4.9, 4.4, and 1.5 Å for Cl_2-Cl_2, Cl_2-Ar, and Cl_2-He collisions, respectively. This ordering, as well as the numerical values, appears to be consistent with findings by Patel and Slusher[6] involving SIT in a two-level system for SF_6.

The pulse-delay times shown in Fig. 3 refer to the intensity peaks; they cannot be accounted for theoretically, even if we were dealing with SIT in a two-level system, since the dephasing collision times and laser-pulse durations were of the same order of magnitude in our studies. We find a decrease in delay time with decreasing chlorine pressure at constant total values of p_{Cl_2} corresponding to a decrease in pulse-delay time with increasing dephasing collision frequency (see Fig. 3).

We did not observe multiple-pulse formation and nearly complete transparency, perhaps because of relatively poor laser-beam quality. Gibbs and Slusher[7] showed in studies of SIT for a two-level system that, for a single-mode laser output, the intensity distribution must be uniform in order to allow observation of multiple pulses and peak amplification.

In conclusion, we re-emphasize the fact that we are dealing with a "pseudo–two-level" system since the upper state in the bound-free transitions is unstable in times of the order of 10^{-12} sec. The fact that anomalous transmission is nevertheless observed must indicate that nonlinear interactions with the coherent radiation field take place on a similar time scale, thereby producing excessive transparency in a kind of lossy system. The detailed nature of this problem clearly requires theoretical study.

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*Much more extensive compilations of experimental findings and of related interpretive work appear in the Ph.D. thesis of R. C. Sepucha, University of California, San Diego, 1971.

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Collisional Excitation of N^+ at 50 keV†

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The excitation spectrum of N^+ has been observed by examining the energy lost by a 50-keV N^+ beam passing through a He target. The spectrum exhibits dramatic features with large cross sections. Determination of the approximate ratio of metastable to ground-state ions in the primary ion beam has permitted measurement of excitation cross sections from both ground-state and metastable N^+ ions colliding with He target atoms.

Energy analysis of fast ions after collision has recently been employed to study excitations of atoms, molecules, and ions.[1-3] Park and Schowengerdt[4] describe the apparatus employed in the present study. Modifications have improved energy resolution and facilitated data handling,[5] but have not altered the basic technique.

The ion N^+ is of interest in astrophysics and atmospheric physics. The forbidden transition $2p^2\,{}^1D \rightarrow 2p^2\,{}^3P$ at 6584 Å is used extensively for determination of electron temperature and degree of ionization in nebulas.[6-8] This transition and the forbidden transition $2p^2\,{}^1S \rightarrow 2p^2\,{}^1D$ at 5755 Å are also observed in aurora and airglow and are employed in understanding these phenomena.[9,10]

Beam-foil spectroscopy measurements of lifetimes of excited N^+ states,[11] electron impact ionization of N^+,[12] and emission lines of N^+ from electron impact on N_2,[13] have been reported.

The apparent differential (energy-loss) cross