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Collisional Effects in the Saturation Spectroscopy of Three-Level Systems: Theory and Experiment

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We report on a theoretical and experimental study of the influence of collisions on the saturation spectroscopy line shapes associated with three-level gas vapor systems. The study is carried out with the goal of gaining new information concerning (a) the collisional processes that occur in atomic vapors, (b) the nature of the interatomic potential between a ground state and an excited-state atom and (c) the possibility of collision-induced enhancement of the absorption of radiation by an atomic system. In each of these areas, new results are obtained.

Theory Using a model in which collisions are assumed to be phase-interrupting in their effect on level coherences and velocity-changing in their effect on population densities, we calculate the absorption profiles associated with three-level atoms that are subjected to two incident radiation fields while undergoing collisions with structureless perturber atoms. One of the fields (pump) is of arbitrary strength and acts on a given transition while the other field (probe) is weak and acts on a transition sharing a common level with the first. In the absence of collisions, the probe absorption profile can exhibit many well-known features [1], including narrow Doppler-free resonances and strong-pump field induced ac Stark splittings. Collisions distort these profiles and can actually lead to enhanced probe absorption in cases of either large pump detunings or strong pump fields. Moreover the collisional modifications of the profiles may be used to extract information on both total and differential scattering cross sections. Theoretical profiles illustrating these features have been derived.

Experiment The theory is applied to explain the $3S_{1/2}\rightarrow 3P_{1/2}\rightarrow 4D_{3/2}$ excitation spectra that we have obtained for Na atoms undergoing collisions with foreign gas perturbers. A pump laser is detuned either 4.0 GHz or 1.6 GHz below the $3S_{1/2}\rightarrow 3P_{1/2}$ transition frequency and a probe laser beam, counter-propagating with the first, completes transitions to the $4D_{3/2}$ state. The population of the $4D_{3/2}$ state is monitored (via fluorescence) as a function of probe frequency for pump detunings of -4.0 GHz and -1.6 GHz, using various pressures of He, Ne, and Kr perturbers. With a pump detuning of -4.0 GHz, which is greater than the Doppler width ≈1.6 GHz, we are able to systematically study collisional redistribution [2] (resulting from

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collisionally-induced excitation of the $3P_{1/2}$ state) and to obtain a fit to theory containing essentially no free parameters. For a detuning of -1.6 GHz, the pump laser excites a given longitudinal velocity class of atoms. Velocity-changing collisions cause this velocity group to relax back towards equilibrium, and the probe absorption monitors the progress of this relaxation. Attempts to fit the data were made using both the Keilson-Storer and classical hard sphere collision kernels to describe the velocity-changing collisions. The theory includes the effects of $3P_{1/2} \rightarrow 3P_{3/2}$ state-changing collisions, which significantly modify the excitation line shapes.

References

1 See, for example, I.M. Beterov and V.P. Chebotaev, Prog. Quantum Elec. 3, 1 (1974) and references therein.

2 D.L. Huber, Phys. Rev. 178, 93 (1969); A. Omont, E.W. Smith and J. Cooper, Astrophys. J. 175, 185 (1972).