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Observation of Non-Lorentzian Spectral Line Shapes in Na–Noble-Gas Systems

R. E. Walkup, A. Spielfiedel,[(a)] and D. E. Pritchard
Research Laboratory of Electronics and Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
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This Letter reports observations showing that the spectral line shape in the core region of collision-broadened lines contains a dispersion component in addition to the usual Lorentzian. The physical origin of this component is the finite *duration* of the collisions. For the Na D lines and Xe perturbers, the dispersion component produces an 8% asymmetry 0.1 Å from line center. Calculations based on recent line-broadening theories predict a dispersion component with an amplitude comparable to our experimental results.

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Weisskopf[1] showed that if radiating atoms were subject to collisions which destroy phase coherence then their emission profile would have a purely Lorentzian shape—the shape expected for quenching collisions.[2] Lindholm[3] and Foley[4] showed that this shape was also to be expected—although possibly displaced from the frequency of the unperturbed radiators—if the collisions only changed the phase by small amounts. Recently both Szudy and Baylis[5] and Kielkopf[6] have shown that the finite duration, T_d, of the collision modifies the Lorentzian line shape, the first correction being the addition of a dispersion component to the line shape. We report here a definitive experimental observation of this component[7]; it is in reasonable accord with the theory.

Our experiment determines the line shape in the near-wing region of the line. This is the region where the detuning from resonance Δ is less than the inverse of the collision duration T_d, but several times greater than the Doppler width $\Delta\omega_D$, so that negligibly few atoms interact resonantly. In this region the line shape is

$$I(\Delta)=\frac{1}{\pi}\frac{\gamma(\Delta)}{\Delta^2+\gamma^2}\left[1+\frac{3}{2}\frac{\Delta\omega_D^2}{\Delta^2}+\frac{2\delta_c}{\Delta}+\dots\right], \qquad (1)$$

where $\gamma(\Delta)$ is the amplitude, and the terms in brackets indicate the lowest-order corrections due to the Doppler width $\Delta\omega_D$ (half width at e^{-1} points), and the pressure shift δ_c. A purely Lorentzian line shape would be represented in the near-wing region by Eq. (1) with $\gamma(\Delta)$ constant; we find that $\gamma(\Delta)$ has a term linear in Δ, indicating that the line shape contains a dispersive component.

We have measured the line shape in the near-wing region of the Na D_1 and D_2 lines broadened by collisions with He, Ne, Ar, Kr, and Xe. The line shape is determined by monitoring light scattered out of a monochromatic laser beam versus laser frequency. The laser was a Coherent Radiation Model-599 single-mode cw dye laser with actively stabilized linewidth (±1 MHz) and output power (±0.5%). Light scattered at 90° from the incident laser direction was collected with $f/7$ optics, passed through a 100-Å-wide interference filter centered at 5900 Å, and imaged onto a photomultiplier used in photon counting mode. The incident laser was linearly polarized in the vertical direction, and the collection optics arm contained a linear polarizer rotated 54.7° from the vertical [where $P_2(\cos\theta)=0$], so that the collection optics system was insensitive to variations in the angular distribution of scattered light with laser frequency.[8,9] Frequency detunings were determined relative to the Lamb dip in a reference cell with 20 MHz error.

The target cell contained Na vapor at 390 K and

10 Torr of rare-gas perturber. For these conditions there is negligible absorption of the incident laser (<0.1% for laser frequencies in the near-wing region) and approximately 10% absorption of collisionally redistributed light. With the incident laser power typically 0.2 mW, the scattered light intensity collected by the detection system was 500–10 000 photons/sec. Data points consist of the ratio of scattered light (corrected for background) to laser power and have 0.8% error reflecting independent errors due to counting statistics (0.4%), laser power instability (0.5%), and laser frequency uncertainty.

In Fig. 1 the experimental line shape with Ar perturbers is compared with a superposition (with appropriate weight and displacement to account for hyperfine structure) of line shape functions of the form given by the first term in Eq. (1) with $\gamma(\Delta)=\gamma_0$ (a constant determined by the fit). A significant asymmetry is noticeable. This asymmetry is emphasized in Fig. 2 by plotting the relative deviation from the Lorentzian fit [that is, (data − fit)/fit]. The relative deviation from a purely Lorentzian fit is seen to be a *linear* function of detuning, and may be accurately accounted for by allowing the amplitude $\gamma(\Delta)$ to be a *linear* function of detuning:

$$\gamma(\Delta)=\gamma_N+\gamma_c{}^*(1-b\Delta), \qquad (2)$$

where γ_N is the natural width (due to spontaneous emission), γ_c is the collision broadening width,

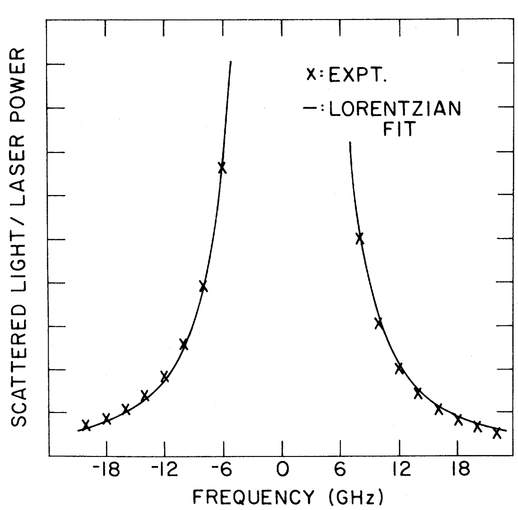

FIG. 1. Experimental data points (×) for the Na $3P_{1/2}$ line perturbed by Ar are compared with a Lorentzian fit (solid curve). "0 GHz" marks the reference Lamb dip for transitions out of $F=2$ in the ground state.

and b is the asymmetry parameter. The term proportional to b adds a dispersion function to the line shape. The asymmetry parameters b shown in Table I were determined by fitting the data with a superposition (as described previously) of line-shape functions given by the leading term in Eq. (1) with $\gamma(\Delta)$ given by Eq. (2). The addition of the dispersion term reduced χ^2 from 100–200 for the perturbers Ar, Kr, and Xe (1–8 for He and Ne) to a value near unity in every case. This tremendous reduction precludes the possibility that an alternative functional form could fit the data as well as the dispersion term. Errors in b arising from use of the known ratio of γ_c/γ_N[10,11] were negligible because γ_N is only (5–10)% of the second term in Eq. (2) at 10 Torr.

Several possible sources of line-shape asymmetry are easily ruled out. The amplitudes for Rayleigh scattering from the two $3p$ fine-structure levels can interfere. This interference affects the degree of polarization of the scattered light but not the total scattered intensity.[8] In addition, for perturber pressures ≃10 Torr, approximately 95% of the total scattered light is collisionally transferred from the virtual level to real levels and the subsequent emission (resonance fluorescence) is incoherent. The two Na D lines are sufficiently separated that one would predict negligible asymmetry from a simple superposition of the two collision-broadened line shapes. However, collisional coupling of the two fine-structure levels can lead to dispersion components[12] of order $(\gamma_I/\gamma_c)(\Delta/\Delta^{FS})$ where γ_I is the rate of inelastic fine-structure-changing collisions and Δ^{FS} is the fine-structure splitting (17.2 cm^{-1}). This would produce an asymmetry <2% in our detuning range; an amount <10% of the ob-

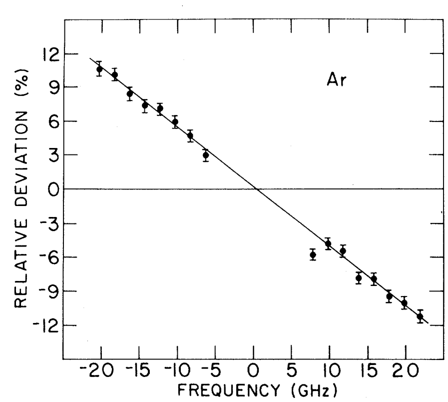

FIG. 2. The relative deviation from the Lorentzian fit of Fig. 1 is displayed vs frequency.

TABLE I. Table of asymmetry parameters: b [Eq. (2)] in units of 10^{-3} GHz^{-1} and a_1 [Eq. (3)]. Theoretical values of b with use of the methods of Kielkopf (Ref. 6) and Szudy and Baylis (Ref. 5) are denoted by b(K) and b(SB), respectively. For the van der Waals C_6R^{-6} interaction the expected value of a_1 is -0.42 (Ref. 5).

	j	b(expt)	b(K)	b(SB)	a_1(expt)
He	1/2	0.1± 0.3	1.14	1.11	-0.04± 0.12
	3/2	0.7± 0.3			-0.20± 0.10
Ne	1/2	1.2± 0.4	2.55	2.52	-0.22± 0.07
	3/2	1.6± 0.3			-0.28± 0.05
Ar	1/2	5.6± 0.3	4.02	4.02	-0.48± 0.03
	3/2	6.5± 0.4			-0.63± 0.04
Kr	1/2	5.3± 0.3	5.01	4.96	-0.41± 0.03
	3/2	7.2± 0.4			-0.58± 0.03
Xe	1/2	6.5± 0.4	5.58	5.61	-0.46± 0.03
	3/2	8.6± 0.4			-0.62± 0.03

served asymmetry for the heavy perturbers. The pressure shift (neglected in the fit) leads to a type of asymmetry because we measure the detuning from an *unperturbed* Lamb dip. From the known values of the pressure shift[11] we make a relative error $\leq 2\%$ in the spectral region which we have examined; in addition, the relative deviation due to such a shift *decreases* with increasing detuning. Thus the type of line-shape asymmetry observed here cannot be due to a line shift. We emphasize that our experiment is conducted well outside the Doppler regime—the excited-state velocity distribution is approximately Maxwell-Boltzmann, independent of laser frequency in the wing region. This eliminates correlations between Doppler and collision broadening. Finally the possibility that instrumental effects produce an asymmetry is ruled out by the observation of symmetric Lorentzian line shapes for both of the D lines perturbed by He and in the case of no buffer gas.

We have compared our results with predictions of scalar theories of line shapes[5,6] which do not make the impact ($\Delta T_d \to 0$) approximation. In order to make simple calculations for comparision with experiment, we assume that the difference potential between excited and ground electronic states behaves as C_6R^{-6}, using Mahan's[13] values of C_6. (Ironically the Na-rare-gas systems studied here are among the few systems for which the relevent difference potentials are accurately known.) Calculations were performed with use of both an impact-parameter approximation to the quantal theory of Szudy and Baylis[5] and the semiclassical theory of Anderson[14] as developed by Kielkopf[6] in which the line shape is calculated by taking the Fourier transform of the dipole autocorrelation function (calculated numerically on our PDP-11/34 computer). The results, showing $<2\%$ discrepancies between the two theories, are presented in Table I. Reassuringly the smallest fractional discrepancies with experiment are found for the heavier rare gases, where the long-range portion of the potential is known to be more influential (relative to the short-range repulsion). In view of the crudeness of the approximation to the potentials, the agreement between theory and experiment is satisfactory.

Our results can be expressed in an intuitively appealing form which explicitly includes the collision duration T_d. The collision duration may be defined by $T_d = R_b/v_{rel}$ where v_{rel} is the relative speed between the active atom and the perturber and R_b is a characteristic interaction distance for collision broadening. We define R_b in terms of the Lorentzian width γ_c, half width at half maximum (HWHM), or equivalently in terms of the collision broadening cross section σ_b by the relations $2\gamma_c = n_p \sigma_b v_{rel} = n_p \pi R_b^2 v_{rel}$. Then we write

$$\gamma_c(\Delta) = \gamma_c(1 + a_1 \Delta T_d + \ldots), \tag{3}$$

where a_1 is a numerical coefficient. For potentials of the form $C_m R^{-m}$, a_1 depends *only* on m; the collision duration contains all of the information about the strength of the collisional interaction (the magnitude of C_m) and the speed or temperature dependence. From the work of Szudy and Baylis,[5] we obtain $|a_1| = 0.517$, 0.420, 0.300 for $m = 4$, 6, or 12, respectively (a_1 is negative if the excited-state potential is more attractive than the ground-state potential). For more realistic potentials we expect a_1 to be slightly temperature dependent because the shape of the potential sampled during a collision depends on the relative velocity. Comparing Eqs. (2) and (3), we find $a_1 = -bT_d^{-1}$. Values for a_1—presented in Table I—were derived from the measured asymmetry parameters b with T_d calculated from the known values of γ_c[10,11] according to the definitions above. The inverse of the collision duration varies from 70 GHz for Na-Xe to 300 GHz for Na-He; thus our data (detunings $\lesssim 20$ GHz) are well inside the impact region. For the heavier perturbers and the D_1 line, a_1 is approximately the value expected for a C_6R^{-6} potential. The values for the D_2 line are considerably larger probably because of the role of the $B\Sigma$ potential.

The lighter perturbers He and Ne have rather small values of a_1 indicating the importance of the repulsive parts of the potentials for these systems.

Our measurements confirm the prediction that the collision-broadened line shape manifests a significant dispersion component in the core region in addition to the well-known Lorentzian. The dispersion component is due to the finite duration of collisions (T_d) and produces an asymmetric line shape with a red-blue asymmetry of order $|\omega - \omega_0| T_d$. The dispersion component accurately accounts for the difference between observed line shapes and a Lorentzian. Hence the core-region line shape can be characterized by three parameters—the Lorentzian width and shift, and the asymmetry parameter. The asymmetry parameters have been determined with less than 10% error, but it has not been possible to check corresponding theoretical predictions to this precision. The scalar theories[5,6] do not contain prescriptions for including the three excited-state potentials dissociating to the Na $3p$ fine-structure levels, hence accurate theoretical calculations have not been possible even where the potentials are accurately known. Calculations based on more sophisticated theories[15] would be most welcome, and would have the advantage of being immediately testable at the (5–10)% level.

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[(a)]Current address: Centre d'Etudes Nucléaires de Saclay, Service de Physique Atomique, F-91190 Gif-sur-Yvette, France.

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Subcritical Transition to Turbulence in Plane Channel Flows

Steven A. Orszag and Anthony T. Patera
Department of Mathematics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
(Received 15 July 1980)

A linear three-dimensional instability mechanism is presented that predicts Reynolds numbers for transition to turbulence in plane channel flows in good agreement with experiment.

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While experiments[1] show that incompressible plane Poiseuille and plane couette flow may undergo transition to turbulence at Reynolds numbers R of order 1000, linear stability analysis of these plane parallel flows gives critical Reynolds numbers of 5772 for plane Poiseuille flow[2] and ∞ for plane couette flow.[3] This discrepancy between theory and experiment suggests that the mechanism of transition is not properly represented by parallel-flow linear stability analysis. In this Letter, we present a new linear three-dimensional mechanism that predicts transition at Reynolds numbers in good agreement with experiment for both plane Poiseuille and plane couette flows. Here we present the theory applied to plane Poiseuille flow, defined as flow between

