Observation of Dynamic Correlations in Collisional Redistribution and Depolarization of Light

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Theory shows that to explain the polarization of light collisionally redistributed from the far line wings of an atomic transition, one must consider correlated events in which absorption *during* a collision, and propagation to the final Zeeman-state superposition at the end of the collision is important. Polarizations of up to about 40% have been measured in the far line wings, substantially confirming this prediction, and showing that scattering experiments *cannot* just be characterized by simple absorption or emission profiles.

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In a series of papers,[1] Burnett and co-workers have examined the validity of factorizing the density matrix when calculating collisional redistribution of radiation; this calculation involves the steady-state density matrix and dipole autocorrelation function of a radiating (or absorbing) atom interacting with a bath of perturbers. It is much easier to obtain the autocorrelation function when one assumes that the density matrix is factorized at all times into an atomic part and a perturber part. This assumes that the timescale which is relevant for the description of the scattering process is much larger than the correlation time for the perturber "bath," i.e., the duration of a collision. When this is true, absorption or emission taking place *during* a strong (close) collision (when atom and perturber are correlated) provide a negligible contribution to the scattering, which can then be characterized by simple emission and absorption profiles. The factorization assumption, or Markoff approximation, is certainly valid in the impact regime,[2] where the detuning, $\Delta\omega$, between the atomic frequency and that of the absorbed (or emitted) photon—corresponding to a timescale of $\Delta\omega^{-1}$—is much less than the inverse of a collision time[1] τ_c. The far wings of the line are, however, determined by evolution on a timescale shorter than τ_c, and in the quasistatic wing absorption of a photon of energy $\hbar\omega_L < \hbar\omega_0$ (Fig. 1), giving rise to fluorescence (rather than Rayleigh scattering), occurs mostly during a collision—when the potential can take up the difference $\hbar(\omega_0 - \omega_L)$ between the energy of the photon and that of the excited atomic state. An absorption (and/or emission) event which takes place during a strong collision is thus called a "correlated event."

The importance of correlated events is illustrated in the following experiment (Figs. 1 and 2). An atomic vapor is irradiated by linearly polarized light of frequency ω_L on the red side of the resonance line (we consider a $J=0$ to $J=1$ transition). The spectrum of the scattered light consists of two peaks, one at ω_L (Rayleigh peak) the other centered at ω_0 (fluorescent peak). The fluorescent component is spectrally resolved from the Rayleigh component and the polarization ratio P is observed (Fig. 2: $I_\parallel$ and $I_\perp$ are the fluorescent intensities polarized parallel and perpendicular to the incident polarization). The polarization of the fluorescence depends on the evolution of the perturbed atom's m_J states between the absorption of a photon of frequency ω_L and the subsequent emission of a fluorescent photon after the

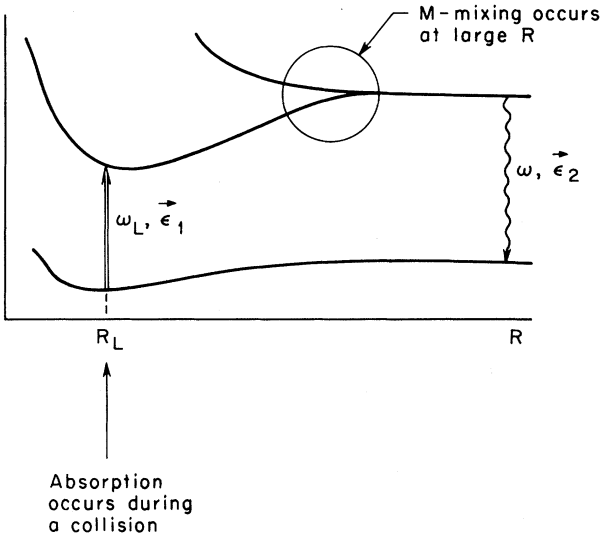

FIG. 1. Polarization dependence of scattering from far line wing.

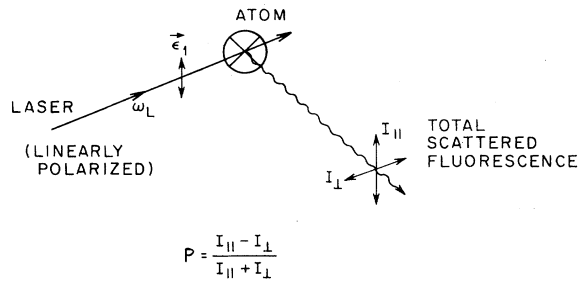

FIG. 2. A schematic scattering experiment.

collision (reemission during the collision is neglected: its spectral distribution is wider than that of the fluorescent peak, and its relative intensity of the order $\gamma_N \tau_c \sim 10^{-4}$, where γ_N is the spontaneous decay rate of the excited level). Two limiting cases correspond to excitation in the impact region and in the quasistatic wing. In the impact region,[3] where correlated events can be neglected, the detailed dynamics of the collisions does not affect the spectrum or the polarization of the fluorescence, which depends exclusively on collisional rates for the relevant atomic multipoles. In the quasistatic wing, on the other hand, correlated events dominate the scattering. As a consequence the spectrum and polarization of the fluorescence depend crucially on the evolution after a correlated absorption *during* the collision. This means[1,3] that the spectrum of the fluorescence *cannot* be characterized by simple absorption spectra (for which the final state is unimportant): The concept of an absorption spectrum has to be generalized.

In the molecular picture, valid for strong collisions, the scattering may be viewed thus (Fig. 1). Absorption takes place at some internuclear distance and then the excited system flies apart, its evolution depending on the geometrical parameters of the collision and on the structure of the potential curves for the excited level. This evolution determines (in the absence of subsequent collisions) the polarization properties of the photon which is reemitted after a time $\sim \gamma_N^{-1}$.

The theory of Burnett and co-workers includes the effects of correlations, and their expressions for the polarization of the fluorescence have been applied to our case, far red wing depolarization. They show that the polarization, P, of the fluorescent light does not go to zero in the far wing, but depends crucially on the nature of the long-range part of the interatomic potential. Some memory of the initial polarization is retained. This result is in disagreement with Behmenburg and Schuller,[4] but was to be expected from calculations by Berman and Lamb[2] showing that a typical strong collision does not completely mix the excited m_J states.

The experiment described above was performed on the strontium resonance line ($\lambda = 4607$ Å), a $J = 0 \rightarrow J = 1$ transition free of hyperfine structure, broadened by argon perturbers. The atomic vapor and buffer gas are contained in a stainless-steel cross whose center is heated to 400–500 °C, and controlled to within 0.1 °C. The ends of the four arms are water cooled such that the strontium vapor condenses on the walls before reaching the windows. Strontium vapor is kept out of the fluorescent light path in the cell by a stainless-steel tube, which is open on its cold side and closed by a sapphire window on its hot side. This eliminates further depolarization by multiple scattering. The laser beam is focused at the center of the cell, typically 100 μm away from the sapphire window. In all measurements this distance is varied and the residual optical depth shown to have no influence.

The laser delivers typically 100 mW cw; its polarization is linear and perpendicular to the detection axis. For measurements at detunings larger than 5 cm^{-1} the laser (Coherent Radiation Model-590 with Stilbene 3 pumped by 3 W of uv power from a Coherent Radiation Model-15 Ar^+ laser) operates with just a birefringent filter, providing a bandwidth of $\lesssim 1$ cm^{-1}. Closer to line center, the bandwidth is reduced to $\lesssim 0.1$ cm^{-1} by a 0.5-mm etalon.

The interaction region is imaged onto the entrance slit of a 1-m McPherson spectrometer which resolves the fluorescent peak from the Rayleigh peak down to detunings of 1 cm^{-1}: The polarizing effect of the grating is eliminated by introducing a linear polarizer, oriented at 45° to both directions of interest, between the spectrometer and the rotating polarizer.[5] All optics between the interaction region and the rotating polarizer were checked to have no effect on the polarization. The response was checked with both linearly polarized and unpolarized light. As an additional check, the polarization of the Rayleigh scattering was shown to be $100^{+0}_{-3}\%$ in both wings.

All measurements were taken at a temperature such that the rate of Sr-Sr collisions was much smaller than that of Sr-Ar collisions. [γ_c(Sr-Sr) $= 5.6 \times 10^7$ s^{-1} at 530 °C, γ_c(Sr-Ar) $= 1.2 \times 10^9$ s^{-1} at 50 Torr Ar, where γ_c is the impact limit colli-

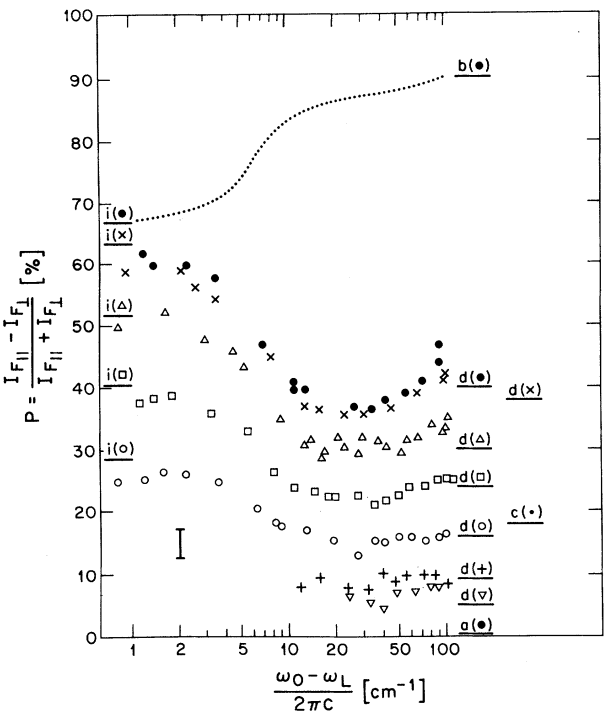

FIG. 3. Polarization ratio P of the fluorescent light (ω_0) vs detuning $(\omega_0 - \omega_L)/2\pi c$ for Sr atoms in Ar buffer gas. Experiment: closed circles, P_{Ar} = 0.8 Torr; crosses, 1.6 Torr; triangles, 5 Torr; squares, 10 Torr; open circles, 20 Torr; plusses, 50 Torr; inverted triangles, 100 Torr. Theory: *a*, *b*, *c*, *d*, and *i* (see text).

sional width.[6]]

The polarization of the fluorescence versus detuning was measured in the range of 1 to 100 cm^{-1} at buffer gas pressures ranging from 0.8 to 20 Torr. All curves show the following behavior (Fig. 3): They are constant in the impact region,[3] decrease in the transition (noting τ_c^{-1} corresponds to 4.6 cm^{-1}) between impact and quasistatic domains, and remain above zero in the far wing (actually, there is a slight increase—see below). The low-pressure curves give the depolarization due to a single collisional event (since there is no further collision in the time γ_N^{-1}). The reduced polarization at higher buffer-gas pressures is due to additional depolarizing collisions between absorption and fluorescence[1] ($\gamma_c/\gamma_N > 1$). The impact limit of P, as well as the effect of increased Ar pressure, is well accounted for by the theoretical values[2,3] (horizontal lines, labeled "*i*" in Fig. 3), assuming that γ_c and γ_N are equal at an Ar pressure of 8 Torr, consistent with our measurements of the ratio of the Rayleigh and fluorescent intensities in the impact region.

Our most important result is that the polarization, while decreasing outside the impact region, stays significantly above zero. This trend should be compared with several theoretical predictions shown in Fig. 3.

(a) Complete depolarization in the far wing (Ref. 4) under the assumption that a strong collision completely destroys the initial anisotropy induced in the excited state by the absorbed photon (see point *a*).

(b) A unified theory of redistribution where correlations are neglected by the factorization approximation (points *b*). The scattering can here be characterized using simple profiles (see Nienhuis and Schuller,[7] Nienhuis[8]; also Cooper[9]). The frequency-dependent rate for the destruction of optical coherence has been taken from experiments of Carlsten, Szöke, and Raymer,[6] whereas the ratio, $\gamma_c^{(2)}/\gamma_c^{(1)}(0)$, of the collisional rates for the destruction of alignment and optical coherence is taken from Ref. 2.

(c) An estimate of the far-wing depolarization based on the theory of Burnett and co-workers[1] which includes correlated events. The m_J mixing in the excited state that occurs as the atom-perturber pair flies apart has been calculated by integrating the equations of motion along classical trajectories in a van der Waals potential, using an adiabatic approximation for the Σ and Π states. The expected polarization is then of order 20% in the region of the far wing where the van der Waals interaction holds. [Point *c* for zero pressure is plotted.]

(d) A statistical mixing estimate. Beyond 20 cm^{-1}, P is above the van der Waals prediction. In like manner, Carlsten, Szöke, and Raymer[6] found that the cross section for destruction of optical coherence only agreed with a van der Waals calculation less than 20 cm^{-1}. This similarity indicates that beyond 20 cm^{-1} the Sr-Ar potential curves cease to be accurately described by the dipole-dipole interaction alone. A simple statistical argument has suggested that a factor of order $\frac{1}{3}$ be associated with the evolution from the initial coherence to that at the end of the collision. These values are plotted as points *d*. The agreement must be considered tentative since the Sr-Ar potential curve at small R is not even qualitatively known. Systematic measurements of the polarization beyond 100 cm^{-1} are needed and will be undertaken.

To conclude, there is substantial evidence that our basic picture for the depolarization of the fluorescent light in the quasistatic wing is correct

and that correlated events, together with incomplete m mixing for strong collisions, play an important role. One of the most encouraging aspects of the results is that a considerable amount of extra information about the dynamics of an atomic collision is needed to describe far-wing depolarization and, conversely, a great deal of information may be obtained from its study. They confirm[1,3] that scattering of light *cannot* be characterized by simple absorption (or emission profiles).

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Ion Thermal Conductivity in a Helical Toroid

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Numerical calculations of the ion thermal conductivity in helical toroidal (torsatron) magnetic configurations show the presence of a plateau regime extending over two orders of magnitude in collision frequency, ν. The value of the ion thermal conductivity is approximately equal to the neoclassical plateau value for an equivalent torus without helical modulation. The predicted adverse $1/\nu$ behavior due to ripple trapping is not seen.

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The family of plasma confinement devices that includes stellarators, torsatrons, and heliotrons is of considerable interest for controlled-fusion applications. These are toroidal devices in which closed, nested magnetic surfaces are generated in the vacuum magnetic field by a helical configuration of the external windings. A major feature of this family is the presence of a strong helical modulation, or ripple, of the field strength on the flux surfaces. Until now, neoclassical transport theory has associated large transport coefficients with this modulation, due to particles which are trapped in the helical magnetic wells.[1,2] These particles are subject to a vertical drift resulting from the toroidal curvature of the magnetic field and can, in some circumstances, make large excursions from their initial flux surfaces.

An estimate of the ion thermal conductivity resulting from this mechanism has been given for stellarators by Connor and Hastie[2]:

$$\chi_i \simeq 11.6\epsilon_h^{3/2}\rho_i^2 v_{ti}^2/\nu_{ii}R^2 \quad (1)$$

(in SI units), where ϵ_h is the helical modulation of the field, ρ_i is the ion gyroradius, v_{ti} is the ion thermal velocity, R is the major radius, and ν_{ii} is the ion-ion collision frequency. This expression is presumed to be valid when the collision frequency is small enough for particles to complete bounce orbits in the helical modulation but large enough that these trapped particles do not complete their poloidal drift orbits.

This result is of particular concern for plasma conditions appropriate to fusion reactors. In fact, the $1/\nu$ dependence of the transport coefficients has led recent torsatron reactor design studies to concentrate on low-temperature, high-density plasma regimes where collisionality is large and χ_i is acceptably small.[3,4]