INTENSITY EFFECTS IN RESONANCE LIGHT SCATTERING*

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When low intensity monochromatic light is incident upon an isolated stationary atom initially in its ground state, the spectrum of the light scattered in a two step excitation-deexcitation process which returns the atom to its ground state must be a $\delta$-function at the incident field frequency. Quantum mechanically, this result follows at once from energy conservation, since in the low intensity limit only one photon is absorbed from the incident field, and only one is emitted under the stated conditions. From a classical point of view, one may understand the $\delta$-function spectrum for the scattered field by thinking of the atom as consisting of one or more harmonic oscillators (a picture which is useful under weak excitation), and recognizing that the induced atomic electric dipole moment must oscillate at the same frequency as the incident field which drives the atom.

Under near resonance conditions, important modifications in the scattered field spectrum appear when the intensity of the incident light is increased to the point where saturation effects begin to appear. These modifications are nonlinear with respect to the incident field intensity, and result from processes in which more than one photon is absorbed and (under near resonance conditions) an equal number emitted, so that though energy is still conserved in the process as a whole, the energy of any one scattered photon, while lying near that of an incident photon, need not equal it exactly. The intensity dependent spectrum contains, in addition to a $\delta$-function or coherent component, broadened incoherent components separated by the Rabi frequency

$$\Omega^* = (\Omega^2 + \Delta^2)^{1/2},$$
where $\Omega$ is the power broadening parameter $\hbar \nu_1/\hbar$ and $\Delta$ is the detuning of the laser from resonance. (One way of understanding the incoherent spectral terms is to think of the field-induced atomic dipole moments are suffering amplitude modulations due to intensity dependent modulations of the atomic populations which in turn are readily understood in the light of the formal equivalence between any two level system and an effective spin system.)

For the case in which the laser field couples the atomic ground state to a single excited state and the damping is purely radiative, the solution has been found in full generality by Mollow\(^1\). The result is valid for arbitrary laser intensity and detuning, subject only to the innocuous restriction that the power-broadened line-width and the detuning be small compared to the optical laser frequency. The spectrum under these conditions is fully symmetrical about the laser frequency, and is given quite generally and exactly by the formula\(^1\)

$$g(v) = g^*(v-w),$$

in which

$$g^*(v) = |\tilde{\rho}_{10}|^2 \pi \delta(v) + \tilde{\rho}_{11} \kappa \Omega^2 (v^2 + \Omega^2/2 + \Delta^2) / |f(iv)|^2,$$

where $\omega$ is the laser frequency, and $\kappa$ is the Einstein A-coefficient for the transition in question. (Theoretical analyses of the same problem have been carried out from a variety of different points of view\(^2\), and all have led to exactly the same results as Mollow\(^1\).)

Ample experimental confirmation of these theoretical predictions has been obtained by Ezekiel and coworkers\(^9\) and Walther and coworkers\(^10\), extending earlier work by Stroud and coworkers\(^11\) on spectral measurements of the sodium D\(_2\) line in an atomic beam.

When the atomic relaxation is due in part to collisional effects which can be treated in the impact approximation, the intensity dependent scattering spectrum can be evaluated by means of simple generalizations of the methods used to treat the radiative case. (One of the earliest treatments of the collisional spectrum, due to Newstein\(^15\), contains limiting forms (corresponding to high saturation) of the solutions for the model treated more fully in Ref. 12.) Reasonable agreement between theory and experiment in the case of the collision-modified spectrum has been