Thinking about Saturated Absorption and Crossover Transitions

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Any discussion of saturated absorption spectroscopy must begin with a reprise of the source of the Doppler broadening of absorption features. In absorption spectroscopy, a beam of laser light, which we will call a *probe beam*, is sent through a gas sample, in our case a mixture of ^{85}Rb and ^{87}Rb vapor, into a photodiode detector. The frequency of the light emitted by the diode laser is then modulated. As the frequency of the laser/probe beam sweeps through the frequency equivalent to the energy needed for a particular transition of the gas, photons from the beam will be absorbed and the atoms excited to a higher energy state. Of course, this energy is quickly reradiated as the atoms return to the ground state. The energy, however, is reradiated in all directions. This three-dimensional re-radiation creates a dual phenomenon – a line of fluorescence appears along the path of the photon beam and the intensity of light reaching the photodiode detector decreases significantly. For the TeachSpin Saturated Absorption experiment, the central frequency of the laser sweep is selected to stimulate a transition from the $S_{1/2}$ to $P_{3/2}$ energy state.

Were all the atoms at rest with respect to the beam, a graph of light intensity reaching the detector vs. the frequency of the laser would show a single sharp dip at the exact transition frequency f_0. The axes of an oscilloscope trace of photodiode detector voltage vs. time would be proportional to received light intensity vs. frequency and so would have the same shape. Because, however, atoms are in motion, atoms with a component of velocity toward the laser beam which we shall call $-v_{zA}$ will "see" the photons with a frequency f_A, which is actually an amount Δf below the transition frequency, as having a frequency f_0. These atoms will therefore absorb photons from the beam. In the same fashion, atoms moving away from the laser beam at speed $+v_{zB}$ will "see" photons of frequency f_B, (an equal amount above f_0) as being at the transition frequency. As a result of this phenomenon, the oscilloscope trace we see is a wide, smooth, curve, a Doppler broadening rather than a sharp dip.

Figure 1 offers a way to visualize this Doppler process. In the upper section of the Figure, the vertical axis of the "graph" indicates the magnitude of a particle's velocity in the direction of the laser beam. The horizontal axis is the frequency of the laser. The plotted line shows the velocity that a particle/atom must have in order to absorb the laser photons at that particular laser frequency within the sweep.

The lower section represents an oscilloscope trace of the detector signal vs. time as the frequency of the laser is swept from below to above the f_0 transition frequency of our imaginary gas. The vertical axis thus indicates the amount of laser light reaching the detector while the horizontal axis indicates frequency. On an actual oscilloscope trace, the transition frequency would be hard to determine with much accuracy.

When using saturated absorption spectroscopy (SAS), however, the oscilloscope trace for atoms with a single transition shows a sharp spike within the Doppler dip when the laser frequency matches f_0. The process of SAS, like many great insights, seems obvious in retrospect. The laser beam is split into two unequal portions. The weaker portion, the *probe beam*, with only 10% of the initial intensity, is directed through the gas cell to the detector. The stronger or *pump beam* is directed around the cell and sent back through it, in the opposite direction of but collinear to the probe beam. The upper section of Figure 2 shows the z-velocity vs. frequency plot for particles that will absorb pump photons as a heavy line while the plot for atoms that will absorb probe photons is thinner. Notice that the two lines cross at $v_z = 0$, $f = f_0$.

For most frequencies within the Doppler range, the pump and probe beams interact with groups of atoms moving in opposite directions. When the laser frequency is f_A, for instance, atoms moving toward the probe beam at a velocity we can call $-v_{zA}$ absorb the probe beam photons. However, for the pump beam, it is the atoms moving at $+v_{zA}$ which "see" the pump beam frequency as elevated to f_0. At this frequency, the presence of the pump beam has no effect on the amount of light reaching the detector. As the laser frequency sweeps through f_0, however, atoms with a $v_z = 0$ can absorb photons from either of the beams. The stronger pump beam saturates the transition, leaving far fewer atoms to interact with the probe beam. As a result, the intensity of the probe beam light reaching the detector increases significantly, creating the sharp spike shown in the lower section of Figure 2.

But what happens if there are two or more closely spaced transitions, transitions so close that the oscilloscope signal appears as single wide Doppler dip? As Figure 3 indicates, for a case of two closely spaced transitions, f_{01} and f_{02}, the oscilloscope trace will show not two but three spikes within the Doppler curve. While two spikes are at the expected frequencies, the frequency of the third spike is exactly halfway between the two actual transition frequencies. At this "halfway" frequency, f_{12}, two groups of particles have velocities which allow them to absorb photons from either beam. Because of their motion, atoms with velocity $+v_{z12}$ "see" the frequency of pump beam photons as elevated to f_{02} while the probe beam photons appear to have a frequency of f_{01}. For atoms of velocity $-v_{z12}$, the opposite effect occurs. For both sets of atoms, the absorption of probe beam photons decreases creating a spike in the Doppler curve.

Doppler Broadened Signal

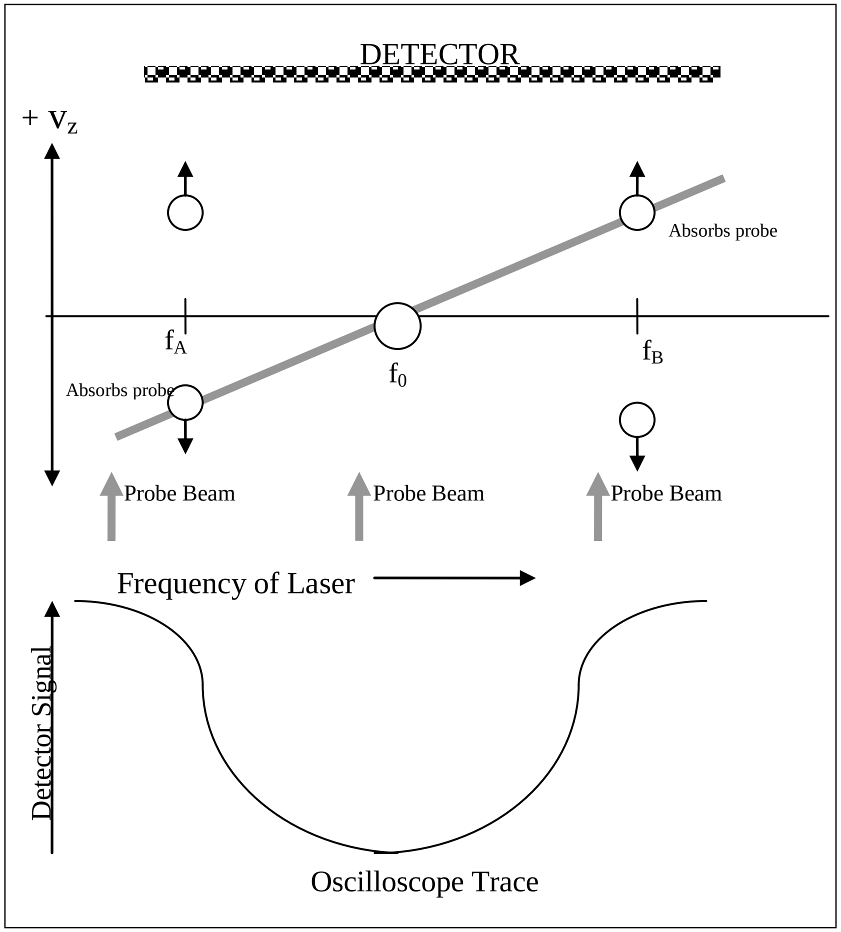

Figure 1

Doppler broadening occurs when the frequency of the laser is swept on either side of the “true” absorption frequency f_0.

When the sweep frequency is below f_0 (as f_A) probe photons are absorbed by gas particles moving towards the probe beam at a velocity which Doppler shifts f_A to f_0. When the sweep frequency is above f_0, probe photons are absorbed by particles moving away at velocities that “see” f_B reduced to f_0. When the laser frequency is f_0, particles at rest or moving perpendicular to the beam absorb photons.

Saturated Absorption

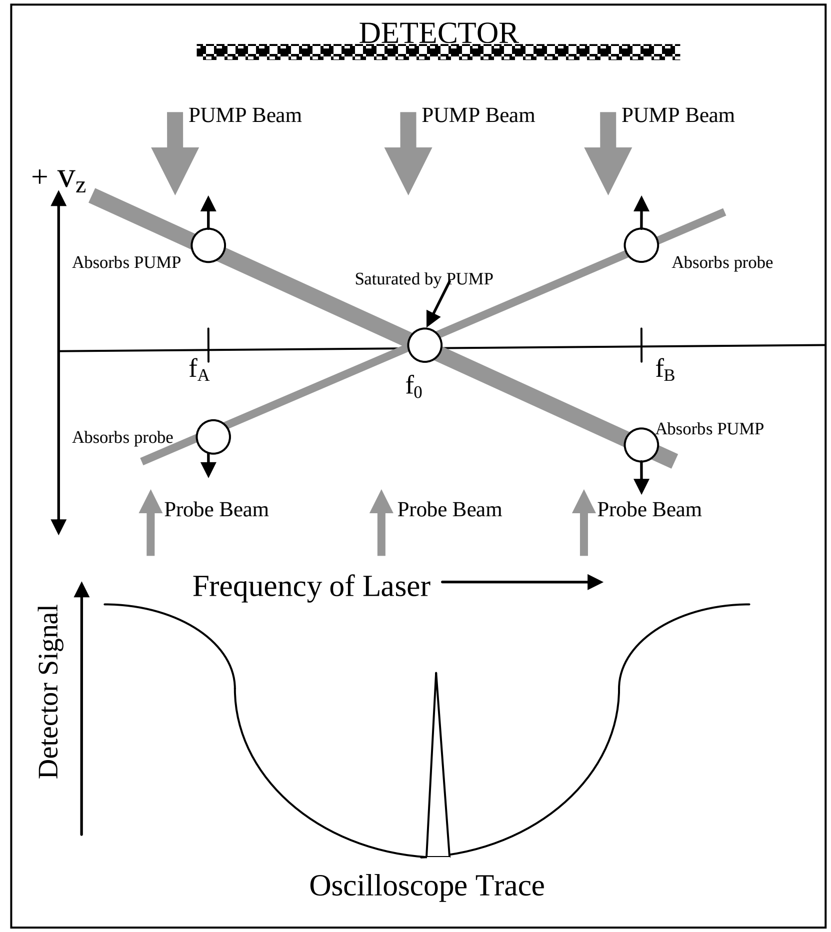

Figure 2

At frequency f_A, (below true absorption frequency f_0) probe and pump photons are absorbed by particles moving towards the each beam. These are completely different sets of particles so the probe signal is unchanged. With the laser frequency at f_B, which is above f_0, photons are absorbed by particles moving away from each beam.

When, however, the laser frequency passes through f_0, particles at rest or moving perpendicular to the beams can absorb photons from either beam. The PUMP beam ‘saturates’ the transition. Since fewer particles are available to absorb probe photons, the intensity of the probe beam reaching the detector is increased and the signal “spikes”.

Crossover Transition

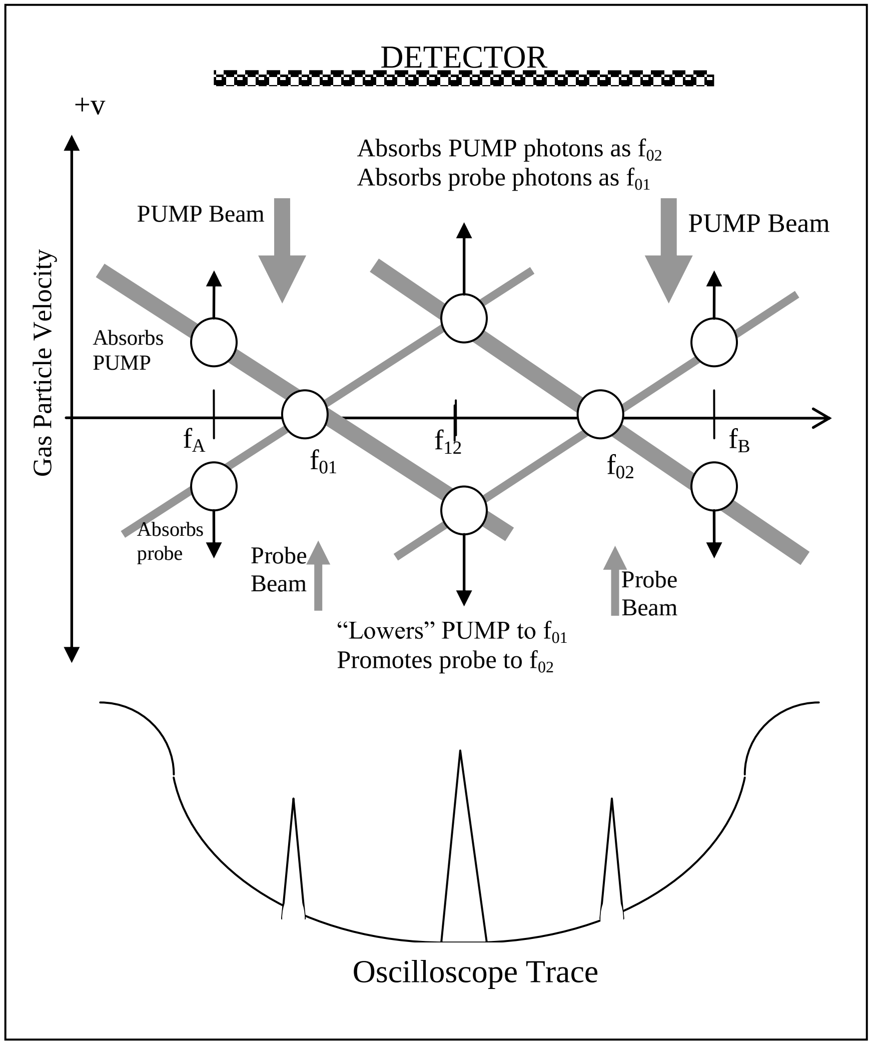

Figure 3

When a transition can be made from the ground state to two upper levels, adding the pump beam creates three "spikes" within the Doppler curve. Increases in transmission occur, as expected at the two actual absorption frequencies, f_{01} and f_{02}. The third spike, called the "crossover" transition, occurs exactly halfway between the two. At this frequency, an atom moving with the proper velocity in the $+v_z$ direction can either Doppler shift the probe beam photons down to f_{01} or the pump beam photons up to f_{02}. Thus at this crossover frequency f_{12} the atoms available for absorbing probe beam photons is reduced. More probe beam photons pass through the vapor and the detected intensity rises. Similar reasoning describes the fate of atoms moving at the proper velocity in the $-v_z$ direction.

When three possible transition frequencies fall within a Doppler curve, as in our rubidium vapor experiment, six spikes are produced. In addition to spikes at f_1, f_2, and f_3, there will be crossover spikes at f_{12}, f_{23} and f_{13}.