Saturated-absorption spectroscopy: Eliminating crossover resonances using co-propagating beams

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We demonstrate a new technique for saturated-absorption spectroscopy using co-propagating beams that does not have the problem of crossover resonances. The pump beam is locked to a transition and its absorption signal is monitored while the probe beam is scanned. As the probe comes into resonance with another transition, the pump absorption is reduced and the signal shows a Doppler-free dip. We use this technique to measure hyperfine intervals in the $D_2$ line of $^{85}$Rb with a precision of 70 kHz, and to resolve hyperfine levels in the $D_2$ line of $^{39}$K that are less than 10 MHz apart. © 2022 Optical Society of America

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Laser spectroscopy in atomic vapor is often limited by Doppler broadening. The standard method to overcome the first-order Doppler effect is to use saturated-absorption spectroscopy. In this technique, the laser is split into a weak probe beam and a strong pump beam, and the two beams are sent in opposite directions...
through the vapor. Due to the opposite Doppler shifts, only the zero-velocity atoms (i.e. atoms moving perpendicular to the direction of the beams) interact with both beams. For these atoms, the stronger pump beam saturates the transition and the probe-absorption spectrum shows a Doppler-free “dip” at line center. Ideally, the linewidth of the dip is limited only by the natural linewidth of the transition.

The above analysis is correct for two-level atoms, i.e. where the other atomic levels are far away compared to the Doppler width. However, in most real atoms, there are several closely-spaced hyperfine levels within the Doppler profile. The presence of multiple levels is a problem in saturated-absorption spectroscopy and results in what are called “crossover resonances”. These spurious resonances occur when the laser is tuned exactly midway between two transitions, so that for some velocity group, the pump drives one transition while the probe drives the other. Since both transitions start from the same ground level, the pump is still effective in reducing absorption from the probe. Crossover resonances are particularly problematic for high-resolution spectroscopy on two levels whose spacing is only slightly larger than the natural linewidth. In this case, the crossover resonance often swamps the real peaks and makes it impossible to resolve the two transitions.

In this article, we demonstrate an alternate technique for saturated-absorption spectroscopy that does not have the problem of crossover resonances. The technique uses to advantage the multilevel structure of the atom in creating differential absorption between the pump and probe beams. The basic idea is to have the two beams co-propagate through the vapor, with the pump frequency fixed on one transition while the probe is scanned over the other transitions. Unlike in conventional
saturated-absorption spectroscopy, both beams have roughly equal intensities, and it is the absorption of the *pump beam* that is monitored. Since the pump is locked to a transition, it is absorbed only by the zero-velocity atoms. The absorption signal remains constant as the probe is scanned since the probe is generally absorbed by a different velocity group. However, as the probe comes into resonance with another transition for the zero-velocity group, the absorption of the pump is reduced and the signal shows a Doppler-free dip.

There are two primary advantages to our scheme. The first, as mentioned earlier, is the absence of crossover resonances. The second is that the signal appears on a flat background. This is different from conventional saturated-absorption spectroscopy where the probe absorption has an underlying Doppler profile which can affect the determination of the peak centers. We have applied this technique to spectroscopy on the \( D_2 \) lines of \(^{85}\text{Rb} \) and \(^{39}\text{K} \). In the case of Rb, we measure the hyperfine interval between the \( F = 1 \) and 2 levels in the \( 5P_{3/2} \) state with a precision of 70 kHz. In K, we are able to resolve all three hyperfine transitions in the \( D_2 \) line, which are completely merged in ordinary saturated-absorption spectroscopy.

The first set of experiments were done with a vapor of \(^{85}\text{Rb} \) atoms. The experimental schematic is shown in Fig. 1. The pump and probe beams are derived from two frequency-stabilized diode lasers operating on the Rb \( D_2 \) line at 780 nm (\( 5S_{1/2} \leftrightarrow 5P_{3/2} \) transition). The intensity in each beam is about 10 \( \mu \text{W/cm}^2 \). The two beams co-propagate through a room-temperature vapor cell such that the angle between them is less than 10 mrad. The pump laser is locked to a hyperfine transition using saturated-absorption spectroscopy in another Rb vapor cell. The probe laser is
scanned by double-passing through an acousto-optic modulator (AOM) and scanning the AOM frequency. The double passing is necessary to maintain directional stability of the beam, which can otherwise lead to systematic shifts of the peak positions. The probe intensity is also stabilized by adjusting the rf power into the AOM in a servo loop.

A normal saturated-absorption spectrum for the $D_2$ line in $^{85}$Rb is shown in Fig. 2. The underlying Doppler profile has been subtracted using a second probe beam that does not interact with the pump. The spectrum is for transitions starting from the $F = 2$ ground level. The $F' = 1$ and $2$ levels in the excited state are barely visible because they are only 30 MHz apart and overlap with the crossover resonance in between. The linewidth of the peaks is 12–15 MHz compared to the natural linewidth of 6 MHz. The primary causes for the increased linewidth are power broadening due to the pump beam and a small angle between the counter-propagating pump and probe beams. In addition, the lineshape of the peaks depends crucially on the intensities in the two beams. At high pump intensities, optical-pumping effects and the effect of velocity redistribution of the atoms from radiation pressure leads to inversion of the peaks or distortion of their Lorentzian lineshape. This is seen in the lower trace of Fig. 2 where the pump intensity has been increased by a factor of 2. The $F' = 1$ and 2 peaks get distorted and the $F' = (1, 2)$ crossover resonance becomes inverted.

The dramatic improvement in the spectrum with our new technique is seen in Fig. 3. The pump laser is locked to the $F = 2 \leftrightarrow F' = 3$ transition while the probe laser is scanned over a frequency range of 50 MHz covering the $F' = 1$ and 2 hyperfine levels. The pump-transmission signal shows two well-resolved peaks corresponding to
these levels. The spectrum appears on a flat background and there is no crossover resonance in between. The peaks also have symmetric Lorentzian lineshapes with no significant pulling due to the neighboring peak.

There are two advantages to scanning the probe laser using an AOM instead of the grating angle that controls the optical feedback. The first is that it guarantees linearity of the scan since the voltage-controlled oscillator (VCO) that determines the AOM frequency has a linear transfer function. Secondly, the scan axis has absolute frequency calibration once the voltage-to-frequency transfer function of the VCO is known. This transfer function is readily measured using a frequency counter. We have used the advantage of a calibrated frequency scan to precisely determine the hyperfine interval in Fig. 3. By fitting Lorentzians to the two peaks, we determine the peak centers with a precision of 50 kHz. This yields the value of the interval to be 29.35(7) MHz. The result is in good agreement with earlier values of 29.26(3) MHz and 29.30(3) MHz obtained by us using other techniques. The high level of precision in the current work is a direct consequence of being able to resolve the two transitions without an intervening crossover resonance. One potential source of error in hyperfine measurements is a systematic shift in the lock point of the laser from peak center. However, in our scheme, this is not a problem because both pump and probe beams address the same velocity group. Thus a shift in the pump-laser lock point causes the entire set of hyperfine peaks to shift but the intervals do not change.

We have performed a second set of experiments with $^{39}\text{K}$. For this, the two diode lasers are chosen to operate on the $D_2$ line of K at 767 nm ($4S_{1/2} \leftrightarrow 4P_{3/2}$ transition). The probe laser does not pass through an AOM but is scanned by scanning the grating.
The consequent variation in the direction is negligible for the results presented here. The beams co-propagate through an ultra-high vacuum (UHV) glass cell maintained at a pressure below $10^{-8}$ torr using an ion pump. K vapor is produced by heating a getter source\(^{11}\) with a current of 2.6 A. The UHV environment is necessary to minimize linewidth broadening due to background collisions. The getter source also gives us control over the amount of K vapor in the cell, which is optimized for obtaining the narrowest linewidth.

In Fig. 4, we show the spectrum for $F = 2 \rightarrow F'$ transitions in $^{39}$K. With conventional saturated-absorption spectroscopy, we obtain a single peak containing all the transitions. The underlying Doppler profile is also seen. The lineshape of the peak is a convolution of 6 peaks lying within 30 MHz of each other.\(^{12}\) Indeed, it is hard to identify the location of the peak center with respect to the actual hyperfine transitions. By contrast, the new technique shows the locations of the three levels quite clearly. While the linewidth is still large enough that the peaks overlap, there is no ambiguity in identifying their locations.

In summary, we have demonstrated a technique for saturated-absorption spectroscopy in multilevel atoms using *co-propagating* pump and probe beams. The pump laser is locked to a transition so that it addresses only the zero-velocity atoms. The transmission of the pump shows Doppler-free peaks when the probe comes into resonance with another transition for the same zero-velocity atoms. The main advantages of the technique are the absence of crossover resonances and the appearance of the signal on a flat background. We demonstrate the power of this technique for hyperfine spectroscopy of closely-spaced levels by using an AOM to measure the smallest
interval in the $5P_{3/2}$ state of $^{85}$Rb. We are also able to resolve hyperfine transitions in the $D_2$ line of $^{39}$K. In the current work, we have used two lasers to generate the pump and probe beams, which appears disadvantageous compared to the use of one laser in conventional saturated-absorption spectroscopy. However, it is possible to generate the probe beam from the pump laser using one or more AOMs, which has advantages for scanning the probe in a calibrated manner. Finally, the pump intensity in our scheme has to be kept very small. At higher intensities, we see clear evidence of dressed states created due to the coherent driving by the pump laser. Indeed, we have used this technique as a means of studying dressed states in room-temperature vapor without complications from Doppler broadening.\[\text{[8]}\]

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Fig. 1. Schematic of the experiment. The pump laser is locked to a hyperfine transition in Rb, while the probe laser is scanned by double passing through an AOM and scanning the AOM frequency. The two beams co-propagate through a Rb vapor cell. The angle between them has been exaggerated for clarity.

Fig. 2. Saturated-absorption spectrum in the $D_2$ line of $^{85}$Rb for $F = 2 \rightarrow F'$ transitions. The underlying Doppler profile has been subtracted. The transitions are labeled with the value of $F'$ and crossover resonances with the two values of $F'$ in brackets. The upper trace is the correct spectrum, while the lower trace is obtained when the pump intensity is increased by a factor of 2, resulting in inversion of the $F' = (1, 2)$ peak. Probe detuning is measured from the unperturbed state.
Fig. 3. $^{85}\text{Rb}$ spectrum obtained with new technique. The transitions are the same as in Fig. 2, with a narrow scan around the $F' = 1$ and 2 levels. The frequency scale on the $x$-axis is set by the voltage-controlled oscillator driving the AOM. The Lorentzian fit (solid line) yields a value of $29.35(7)$ MHz for the hyperfine interval.
Fig. 4. Saturated-absorption spectrum in the $D_2$ line of $^{39}\text{K}$. The upper trace shows the probe-transmission signal for $F = 2 \rightarrow F'$ transitions in usual saturated-absorption spectroscopy. The 6 peaks are merged into a single peak since the hyperfine levels lie within 30 MHz, as shown in the inset. The values in the inset are the frequency offset (in MHz) of each hyperfine level from the unperturbed state. The lower trace is the pump-transmission signal obtained with the new technique, clearly showing the locations of the hyperfine levels.