New Opportunities in Fiber-Optic Sensors

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Abstract—Several possibilities for new fiber-optic sensors are proposed that are based on two-photon spectroscopic techniques. Examples are chemical sensors, including multiplexed sensors, as well as magnetic/electric sensors. The development of a compact, low-cost fiber-optic method for the accurate measurement of time/frequency is also proposed.

I. INTRODUCTION

In this paper, we will explore the creation of several new fiber-optic sensors based on the more sophisticated two-photon spectroscopic techniques [1] for applications such as chemical sensing, magnetic/electric field sensing, and for the measurement of time or frequency. The unique properties of optical fibers and fiber-optic/integrated-optic components are critical to the development and implementation of such sensors.

Conventional spectroscopic techniques usually involve the excitation of atoms and molecules in a cell using a tunable light source, such as a laser, and measuring the resulting absorption or fluorescence. Fig. 1 shows such a set-up using optical fibers and GRIN lenses to propagate a collimated laser beam through the vapor cell. In this way, it is possible to identify the type of molecule in the vapor, as well as the concentration. This method is generally referred to as a single step or single photon spectroscopy, since the excitation involves a single pair of energy levels.

The resolution in this approach is determined primarily by the linewidth, , for the transition which can be influenced by various broadening mechanisms. Specifically, in a vapor cell, the contributions to the linewidth for a two-level system include: natural broadening, collisions, Doppler, transit-time, laser intensity, and laser jitter. The effects of external magnetic and electric fields can also contribute to the broadening.

In two-step or two-photon spectroscopy [1]–[5], two radiation fields simultaneously interact with the atom or molecule in the vapor. In this case, three energy levels are involved in the excitation. The configuration of these energy levels can be in a "folded," "cascade," or "V" arrangement, as illustrated in Fig. 2. Of interest here is the folded configuration because it offers an inherently high resolution capability [2]. For example, as shown in Fig. 3, if the field exciting the 1–2 transition is held fixed near the 1–2 resonance frequency, and the second field is tuned so that , gain (or reduced absorption) is observed at , with a linewidth that can be extremely narrow, particularly if the lifetimes of levels 1 and 3 are long. The reasons for such a narrow linewidth are listed below.

1) The natural broadening due to the spontaneous relaxation rate of the upper state, , is zero in such a case if the fields are weak [2], [3]. The remaining contributions to the natural linewidth come from the spontaneous relaxation rates of levels 1 and 3, i.e., and , respectively, which are negligible if levels 1 and 3 are long lived.

2) The Doppler broadening is normally reduced by a factor of when the two excitation fields are copropagating and when is well outside the Doppler width of the 1–2 transition, as shown in Fig. 4(a). The Doppler broadening is reduced to when is within the Doppler width of the 1–2 transition, as shown in Fig. 4(b) [2], [3]. In this case, and interact in resonance with only one velocity group of atoms, and we assume that the Doppler width is larger than the homogeneous linewidth for this transition.
3) Collisional broadening is much smaller than in the two-level optical transition and is expected to be similar to the collisional broadening of the 1–3 transition. Particularly, when the 1–3 transition is coupled by magnetic dipole, e.g., in the hyperfine splitting in alkali atoms, the collision broadening is of the order of 50 Hz [6].

4) The broadening due to laser jitter can be virtually eliminated if the jitter in laser \( f_1 \) is correlated with the jitter in laser \( f_2 \) and \( f_1 \) is approximately equal to \( f_2 \). Correlating the jitter in the two lasers can be accomplished by generating one laser frequency from the other using various modulation techniques [7].

Thus, for a folded two-photon excitation of a three-level system with long lived lower levels using copropagating fields, the linewidth becomes primarily determined by transit time broadening, e.g., for a 1 cm laser beam, and an average atomic velocity of \( 10^5 \) cm/s, the transit time linewidth is about 20 kHz.

A simple setup for a two-photon fiber-optic spectrometer is shown in Fig. 5. In this case, the molecules in the vapor are simultaneously excited with two copropagating lasers, at frequencies \( f_1 \) and \( f_2 \), where the frequency difference, \( f_1 - f_2 \), is tuned around the 1–3 transition frequency. Thus, the observed peak amplification (or reduction in absorption) of the \( f_2 \) laser as a function of \( f_1 - f_2 \) determines the frequency of the 1–3 transition which identifies the molecule [4]. Because of the Doppler effect, the two laser beams have to be perfectly superimposed, otherwise the resolution will be reduced by the Doppler broadening caused by this misalignment. One effective way to ensure the superposition of the beams is by transmitting both beams through the same single mode optical fiber. Therefore, the use of a single mode, polarization maintaining fiber and GRIN rod lenses makes such a spectrometer very feasible, very rugged and suitable for deployment in a remote environment.

In practice, to enhance signal noise and to eliminate background effects, the \( f_1 \) (or pump) laser is amplitude modulated at a rate \( f_{m1} \) and the \( f_2 \) (or probe) laser is modulated at a rate \( f_{m2} \), as shown in Fig. 6. After passing through the vapor cell, the combined light is detected on a photo detector and demodulated first at \( f_{m2} \) and then at \( f_{m1} \). In this way, the gain on the probe laser can be measured with high sensitivity as the pump/probe frequency difference is tuned over the 1–3 transition. To further increase the signal-to-noise ratio, the pump laser is adjusted to resonate with the 1–2 transition.

Fig. 7 shows a number of probe amplifications in molecular iodine for various rotational states in level 3, obtained using an argon ion laser at 514 nm and a dye laser at 583 nm [2]. The observed linewidth is about 80 kHz, in this case, limited primarily by the frequency jitters in the argon and dye lasers. In the absence of laser jitter, the linewidth would be about 10 kHz, limited by the residual Doppler effect, since there is a large difference between \( f_1 \) and \( f_2 \) in this case. It should be noted that this linewidth is much narrower than the width observed using a two-level configuration [8]. The two-photon method can be easily adapted for remote multiplexed chemical sensing at a number of specific locations, as shown in Fig. 8, by simply using a different probe modulation frequency for each location. The pump modulation frequency remains the same for all locations.

The folded three-level interaction, shown in Fig. 5 is commonly referred to as stimulated Raman gain (SRG) and the technique is called SRG spectroscopy. It is also possible to adapt this setup to perform Coherent Anti-Stokes Raman Spectroscopy (CARS), where the spectrometer before the detector is used to separate out the resulting coherent radiation created at \( 2f_1 - f_2 \) when \( f_1 - f_2 \) is tuned around \( f_{13} \) [9], [10].

We should point out that, in the case when the energy difference between levels 1 and 3 is small so that both states have comparable populations, the SRG process appears as a reduction in the absorption of the probe when \( f_1 - f_2 \approx f_{13} \).

We now consider the use of a three-level system for the creation of a compact, accurate, low cost, sensor of time or frequency [11]. In this case, the frequency of the 1–3 transition is exploited as the stable frequency reference. It should be recalled that in the cesium atomic clock, a hyperfine transition in the ground state of cesium [12] is directly excited by a microwave field at about 9.2 GHz and the frequency of this microwave radiation is locked to the cesium hyperfine transition frequency. By eliminating collisions and employing Ramsey's separated oscillator excitation in an atomic beam to reduce the transit time broadening, a very stable frequency is obtained. The stability can be better than one part in \( 10^{15} \) [13]. In our three-level case, it is the frequency difference between the two optical fields at \( f_1 \) and \( f_2 \) that is locked to the 1–3 transition, as shown in a simplified diagram in Fig. 9. If level 3 is close to level 1, i.e., if the 1–3 transition frequency is

In this case, a population difference is generated between the hyperfine levels by optical pumping or by magnetic state selection.
in the microwave range, we can create a stable frequency at $f_1 - f_2$ which can be easily measured or counted. It should be noted that only optical excitations are used and thus, there is no need for a microwave source or microwave plumbing. For example, levels 1 and 3 could be the hyperfine levels in the ground state of cesium used in the conventional cesium atomic clock.

For good short-term stability, the linewidth of the transition must be as narrow as possible. This requires that the two laser frequencies have very small frequency jitter or have the same (or correlated) jitter so as not to broaden the transit time limited linewidth. To achieve very small jitter in each laser is not easy, but to correlate the jitter in the two lasers is very feasible using modulation techniques [7], as previously discussed. Another technique for generating lasers with correlated frequency jitter uses common cavity stimulated Brillouin lasers [14].

The remaining issue for the linewidth is the transit time. To reduce the transit time linewidth in a vapor, we need to use either low velocity atoms or a large diameter laser beam. Neither is very simple to accomplish in practice. However, by adding a suitable buffer gas at an appropriate pressure, it is possible to substantially reduce the transit time linewidth [15]. Fig. 10 shows a 1.5-kHz linewidth for a stimulated Raman gain set-up in sodium using a helium buffer gas. In contrast, the transit time linewidth would have been about 200 kHz in a 1-mm-diameter laser beam without the buffer gas. If rubidium
In summary, the technique of two-photon spectroscopy can be useful in developing a variety of new fiber-optic sensors. In particular, this approach may be particularly applicable in the creation of remote multiplexed chemical sensors. The measurement of time/frequency and magnetic fields using such techniques have already been demonstrated in our laboratory using bulkoptic components. The availability of diode-pumped fiber lasers and integrated-optic lasers and devices will contribute greatly to the development of two-photon fiber-optic sensors.

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