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Two-tone frequency modulation saturation spectroscopy for tunable frequency offset locking of a single laser

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ABSTRACT

The two-tone frequency modulation spectroscopy (TTFMS) is widely used for gas trace detection with its low noise absorption signal. In this paper we propose and implement the TTFMS scheme for frequency offset locking a single diode laser to an atomic resonance line by using its dispersion signal. The TTFMS theory is first discussed under the assumption that the intermediate modulation frequency is comparable to the linewidth of the absorption feature, and the dependence of the TTFMS absorption and dispersion features on the intermediate modulation frequency and modulation index are investigated theoretically. Based on a fiber-coupled electro-optic modulator (EOM) with two-tone modulation, we experimentally demonstrated the performance of the frequency offset locking method. The result shows a short-term frequency stability of the frequency offset locked laser reached around 1.1×10^{-11}, with an averaging time of 2 s. This method can find wide applications in fields requiring widely tunable frequency offset locking a single laser to the atomic resonance line, like precision spectroscopy and Raman optics for atom interferometers.

1. Introduction

A frequency offset locking technique can lock a frequency difference between two laser sources to a constant value or lock a single laser source with a frequency difference to an absolute frequency reference like an atomic resonance line. This locking is used for frequency stabilization and also as a means of scanning laser frequency for extending its working frequency to a wider spectral range. These techniques have found applications in many fields requiring wide optical frequency tunability and precise control of the absolute optical frequency, such as precise spectroscopy [1,2], cold atom experiments [3], atom interferometry [4], atomic or optical clocks [5].

Different techniques have been developed for laser frequency offset locking with the offset frequencies ranging from a few megahertz (MHz) to tens of gigahertz (GHz). One commonly used method is based on the master–slave laser scheme, in which a reference laser (master laser) is locked to an atomic resonance line and provides an absolute frequency reference, whereas a probe laser (slave laser) is locked to the master laser based on beat notes between the two lasers. Phase-coherent locking can be obtained by using the optical phase-locked loops (OPLLs) [6], locking to different longitudinal modes of a tunable Fabry–Perot resonator [7,8] or an optical frequency comb [5], or injection locking by an frequency shift beam with an auxiliary acousto-optic modulator (AOM) [9] or EOM [10]. In many spectroscopic or atomic experiments, phase coherency is not required, whereas it is sufficient that the slave laser maintains a high-precise frequency offset from the master laser. In these cases, simple locking schemes can be implemented with different methods to generate the error signal, such as by using an electronic delay line [11], a frequency-to-voltage converter [12], a sharp electronic radio frequency (RF) high-pass filter [13] or frequency modulation on one of the lasers [14]. In these master–slave schemes, an additional reference laser is required and make the system complicated and not cost-effective when only one laser frequency is needed. In addition, this method is limited by the detection bandwidth of the photodiode (PD), which may reach up to tens of GHz; The other method is to lock a laser source directly to a nearby absolute reference frequency like an optical or molecular reference transition with a tunable frequency offset, which can be generated by different atomic spectroscopies [15–20] or a variable-frequency-shifting device such as an AOM or EOM. Both AOMs and EOMs have the advantage that optical frequency tuning is readily accomplished by adjusting the RF of the device driver electronics. The frequency shift and tuning range of AOMs is restricted to typically 1 GHz and 50–100 MHz respectively by the limited phase velocity of the acoustic wave in the acousto-optic crystal. And the diffraction efficiency of AOMs tends to decrease with increasing RF frequency [21]. Using EOMs is a comparatively better choice for frequency shifting over larger

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ranges while maintaining high efficiencies. Frequency offset locking of 40 GHz to a reference atomic transition has been demonstrated by using the fourth-order sideband generated by an EOM and the polarization spectroscopy [22]. But useless sidebands generated by EOMs bring about the problems of phase noise and systematic errors.

In this paper, we present a simple method based on the TTFMS to lock a single laser with frequency offsets a few GHz to tens of GHz from an atomic or molecular reference line. The TTFMS has been demonstrated as a technique for performing sensitive frequency modulation spectroscopy (FMS) at high modulation frequencies with low-frequency detection [23–28], and widely applied for qualitative measurements of gas species with weak absorption [29–37], where the intermediate modulation frequency is small in relation to the spectral variations in an absorption feature, like a typical Doppler halfwidth in the near infrared. For locking a laser, the quadrature dispersion feature in the TTFMS needs to be used as the error signal instead of the absorption feature and becomes apparent only when the intermediate modulation frequency is comparable to the spectral variations, e.g. the sub-Doppler feature (typically a few tens of MHz). The TTFMS avoids the typical limitations of absorption measurements from low-frequency $1/f$ noise by moving the detection band to higher frequencies and using the phase-sensitive heterodyne detection. Therefore, it has the potential to transfer the advantages of high sensitivity, high resolution and high-speed detection of the FMS frequency stabilization [38] to a large offset frequency range. We start with the theoretical analysis on the TTFMS without the assumption of small intermediate modulation frequency, which is commonly used in absorption measurements. The influences of the modulation index and intermediate modulation frequency on the amplitude of a TTFMS signal are investigated theoretically in detail. Then we demonstrate experimentally the TTFMS based frequency offset locking by two-tone phase-modulating the laser via a fiber-pigtailed waveguide-type EOM. The laser frequency locking point can be conveniently and fast tuned at the frequency offset from the resonance line by adjusting the RF driving the EOM over a large range, which can be further extended by using higher-order sidebands of the phase-modulated light. Experimental results show that the frequency offset locking of a single laser has a short-term frequency stability of 1.1×10^{-11} with an averaging time of 2 s, which is comparable with the single-tone FMS locking.

2. Theory

A TTFMS theory has been derived for high-resolution measurements of line parameters or quantitative measurements for gas species [23,25,26,34]. In these applications, the intermediate modulation frequency is chosen such that it is small relative to the widths of the studied absorption features and the dispersion signal is weak and taken into account only when considering its contribution to the line shape distortion. However, for frequency locking a laser, the dispersion signal is suitable with a zero crossing as the locking point. We need to extend the applicability of the TTFMS theory in the direction of performing a frequency offset locking.

A diode laser can be two-tone frequency modulated by imposing two RF signals $\nu_1 \equiv \nu_m + \frac{1}{2}\Omega$ and $\nu_2 \equiv \nu_m - \frac{1}{2}\Omega$ to an EOM. Without considering the amplitude-modulated electric field and the laser linewidth, the phase modulated electric field in terms of Bessel functions $J_n(\beta)$ can be written as

$$E_1(t) = E_0 \exp(i2\pi\nu_0 t) \sum_{n,m=-\infty}^{+\infty} J_n(\beta) J_m(\beta) \exp[i2\pi(n\nu_1 + m\nu_2)t] \tag{1}$$

where E_0 is the amplitude of the laser field, ν_0 is the laser carrier frequency and β is the frequency modulation (FM) index. The frequencies ν_m and Ω are chosen such that the modulation frequency ν_m is far larger than the absorption halfwidth to produce a frequency offset to the carrier frequency and the intermediate modulation frequency $\Omega = \nu_1 - \nu_2$ that is detected is small in comparison with the modulation frequencies

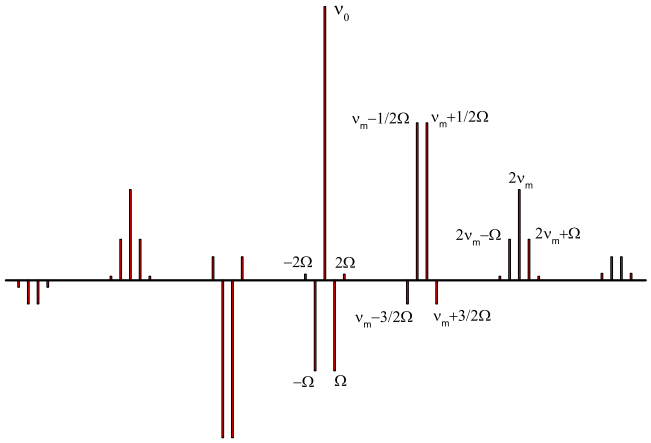

Fig. 1. Spectral distribution of the laser field, two-tone frequency modulated at ν_1 and ν_2, with $\beta = 1$.

but large enough to avoid low-frequency ($1/f$) noise, i.e., typically 5–20 MHz. Since the modulation frequencies ν_1 and ν_2 are closely spaced, it is assumed that they both induce the same β in the electric field.

Fig. 1 shows schematically the spectral distribution of the two-tone frequency-modulated laser field for $\beta = 1$ and with the intermediate modulation frequency Ω greatly exaggerated for illustration. The central component ($n = m = 0$) is the laser carrier at frequency ν_0 and the sidebands are at frequencies $\nu_0 + n\nu_1 + m\nu_2$, where n, $m = 0, \pm1, \pm2, \ldots$. The sidebands appear in groups symmetrically (in pairs) around the laser carrier frequency, which we refer to as sideband groups. A sideband group contains intermodulation products of different orders, e.g., the first-order sideband group contains a third-order intermodulation product $(2\nu_1 - \nu_2)$.

After the frequency-modulated beam is passed through a sample, each frequency component $\nu_0 + n\nu_1 + m\nu_2$ experiences different attenuations and phase shifts. The electric field after interaction with the sample is

$$E_2(t) = E_0 \exp(i2\pi\nu_0 t) \sum_{n,m=-\infty}^{+\infty} J_n(\beta) J_m(\beta) \times \exp[i2\pi(n\nu_1 + m\nu_2)t] \exp(-\alpha_{n,m} - i\phi_{n,m}), \tag{2}$$

where $\alpha_{n,m} \equiv \alpha(\nu_0 + n\nu_1 + m\nu_2)$ and $\phi_{n,m} \equiv \phi(\nu_0 + n\nu_1 + m\nu_2)$ are the absorption and the phase shift, respectively, induced by the sample at the frequency component $\nu_0 + n\nu_1 + m\nu_2$.

The intensity detected by a photodetector is $I_2(t) = c\varepsilon_0 E_2 E_2^*/2$ or

$$I_2(t) = \frac{c\varepsilon_0}{2} E_0^2 \sum_{n,m,n',m'} J_n J_m J_{n'} J_{m'} \exp\{i2\pi[(n-n')\nu_1 + (m-m')\nu_2]t\} \times \exp[-(\alpha_{n,m} + \alpha_{n',m'}) - i(\phi_{n,m} - \phi_{n',m'})], \tag{3}$$

The time-varying intensity components, arising from the heterodyning of adjacent frequency sidebands at $\pm\Omega$, are obtained for $n - n' = \pm1$ and $m - m' = \mp1$. Thus the beat signal at Ω of Eq. (3) is

$$I_\Omega(t) = \frac{c\varepsilon_0}{2} E_0^2 \exp(-2\pi i\Omega t) \sum_{n,m} J_n J_m J_{n+1} J_{m-1} \times \exp[-(\alpha_{n,m} + \alpha_{n+1,m-1}) - i(\phi_{n,m} - \phi_{n+1,m-1})] + c.c. \tag{4}$$

Separating the absorption and dispersion components in Eq. (4) yields

$$I_\Omega(t) = I_\Omega^\alpha \cos(2\pi\Omega t) + I_\Omega^\phi \sin(2\pi\Omega t), \tag{5}$$

where

$$I_\Omega^\alpha \equiv c\varepsilon_0 E_0^2 \sum_{n,m} J_n J_m J_{n+1} J_{m-1} \times \exp[-(\alpha_{n,m} + \alpha_{n+1,m-1})] \times \cos(\phi_{n,m} - \phi_{n+1,m-1}), \tag{5a}$$