FE DOPPLER-FREE SPECTROSCOPY AND OPTICAL HETERODYNE DETECTION FOR ACCURATE FREQUENCY CONTROL OF FE-RESONANCE DOPPLER LIDAR

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ABSTRACT

For a Major Research Instrumentation (MRI) mobile Fe-resonance/Rayleigh/Mie Doppler lidar, we developed a novel approach to accurately calibrate and stabilize the lidar pulse frequency to the peak of 372-nm Fe absorption line. Our revolutionary idea is to achieve the Fe Doppler-free spectroscopy, and then apply such high-resolution spectroscopy and optical heterodyne detection technology to accurately analyze and control the frequency and spectrum of each lidar pulse. This is a significant improvement over the traditional method of controlling the cw seed laser only. Such concept will help produce the first bias-free resonance Doppler lidar for advancing middle and upper atmosphere science. Here we present the Fe Doppler-free spectroscopy and the pulsed optical beat signals that we believe are achieved for the first time in resonance lidar.

1. INTRODUCTION

Ambitiously aiming to improve the temperature and wind measurement accuracy of resonance Doppler lidar to an unprecedented level (~0.1 K and 1 cm/s) for new science endeavors, we started the development of a Major Research Instrumentation (MRI) mobile Fe-resonance/Rayleigh/Mie Doppler lidar about two years ago at the University of Colorado at Boulder [1, 2]. The choice of Fe Doppler lidar came from the combination of high Fe abundance, short UV 372-nm wavelength (thus stronger Rayleigh scattering for larger Rayleigh temperature range), high temperature and wind sensitivity, deep Fraunhofer line in solar spectrum (thus low daytime solar background), and solid-state laser technologies. This lidar is based on the injection-seeded frequency-doubled Pulsed Alexandrite Ring Laser (PARL) technology [3]. Besides developing a unique PARL to produce a single-frequency high-power all-solid-state lidar transmitter, two other major challenges for this MRI lidar are how to achieve accurate absolute-frequency calibration and chirp-influence-free frequency stabilization of each lidar pulse [3]. In this paper we introduce two new approaches – (1) the Fe Doppler-free spectroscopy to provide an absolute frequency reference to the entire lidar, and (2) applying the optical heterodyne detection (OHD) technique to analyze each PARL pulse's frequency chirp, spectral linewidth and power-spectral distribution for accurately controlling the lidar pulse to desired Fe Doppler-free peak. These new concepts can be readily applied to other Doppler lidars.

2. FE DOPPLER-FREE SPECTROSCOPY AT 372 NM AS THE ABSOLUTE FREQUENCY REFERENCE

Iron (Fe) is the most abundance metal species in the mesosphere and lower thermosphere (MLT), providing an excellent tracer for temperature and wind profiling in this MLT region [4]. Fe atoms have four naturally stable isotopes ^{56}Fe, ^{54}Fe, ^{57}Fe, and ^{58}Fe with natural abundance of 91.75%, 5.85%, 2.12%, and 0.28%, respectively. The isotope line shifts relative to the ^{56}Fe line and the relative strengths have been documented in [1] and [2]. An energy level diagram of the main Fe species ^{56}Fe is illustrated in Figure 1. Due to its zero nuclear spin, ^{56}Fe spectrum has no hyperfine structure.

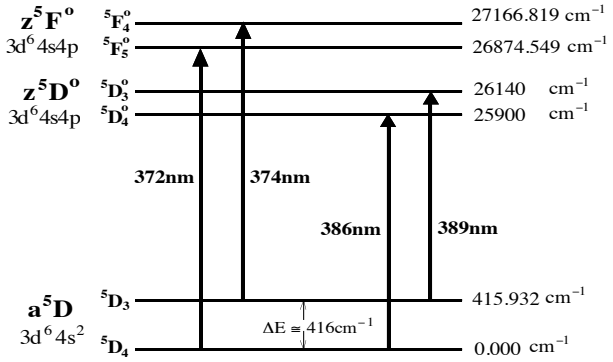

Figure 1. Energy level diagram for atomic ^{56}Fe isotope

A major difficulty for Fe spectroscopy in the past was the lack of a convenient Fe vapor cell and continuous-wave (cw) UV laser source. Intrigued by the pioneering work of *Smeets et al.* [5] and enabled by the commercial Fe-Ar hollow-cathode discharge cell from Hamamatsu and the cw 372-nm External Cavity Diode Laser (ECDL) from Toptica, we were able to achieve the Fe Doppler-free saturation-absorption spectroscopy at the 372-nm absorption line for the first time in resonance Doppler lidar. The experiment setup is shown in Figure 2, where the key component is a see-through Fe hollow-cathode lamp serving as the Fe vapor cell. With its discharging mechanism, this see-through lamp can generate appreciable Fe vapor pressure at relatively low temperature (~600 K, instead of several thousands of Kelvin). Its see-through structure allows two counter-propagating laser beams to go through the volume of Fe vapor, enabling high-resolution saturation spectroscopy. The emergence of UV diode laser at 372 nm makes the entire experiment compact and convenient. The 372-nm laser diode was produced by Nichia of Japan, and the

372-nm external cavity diode laser was put together by Toptica of Germany. Although its specification is only ~7 mW, such power is more than sufficient to produce good Doppler-free spectroscopy signal.

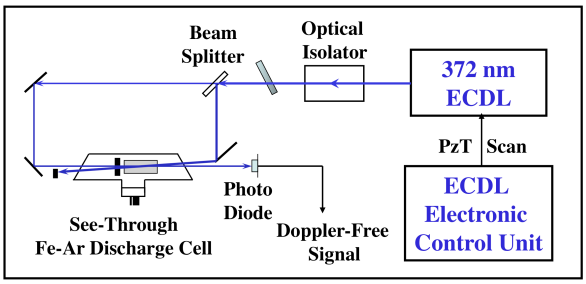

Figure 2. Experimental setup for Fe Doppler-free saturation-absorption spectroscopy at 372 nm with the MRI lidar

The Fe Doppler-free saturation-absorption spectroscopy obtained with our MRI lidar at its first attempt is displayed in Figure 3. Such clear Doppler-free peak was achieved just by the simple setup shown in Figure 2, without any lock-in amplification. The full-width-at-half-maximum (FWHM) of the Doppler-free peak is 36 MHz, which is comparable to *Smeets et al.* [5] results obtained with a Ti:Sapphire laser but much larger than the expected natural linewidth of Fe. The radiative lifetime τ of the upper energy level $z^5F_5^o$ of the 372-nm line is 61.0 ns, which gives a natural linewidth of 2.6 MHz ($\Delta\nu_N = 1/(2\pi\tau) = 2.6MHz$).

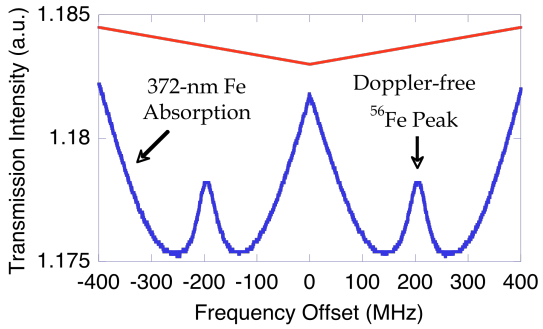

Figure 3. The Fe Doppler-free saturation-absorption spectroscopy at 372 nm obtained with the MRI lidar

Two major factors may have contributed to this line broadening. One is the power or collision broadening inside the Fe vapor cell, and another is the residual Doppler broadening resulting from the fact that two laser beams were not perfectly overlapped due to the traditional setup. We will use a beamsplitter to achieve perfectly collinear beams and reduce the incident power to get narrower Doppler-free peak. Nevertheless, such clean peak enables laser locking to reach sub-MHz accuracy and precision based on *Smeets et al.* [5] results of 0.2 MHz locking precision.

The Fe Doppler-free spectroscopy provides the absolute frequency reference to the MRI Doppler lidar. Once locking the 372-nm ECDL to this Doppler-free peak, the absolute frequency reference is transferred from the Fe atoms to the cw diode laser, providing a convenient light source for calibrating and stabilizing the PARL pulses as well as the receiver components like etalons.

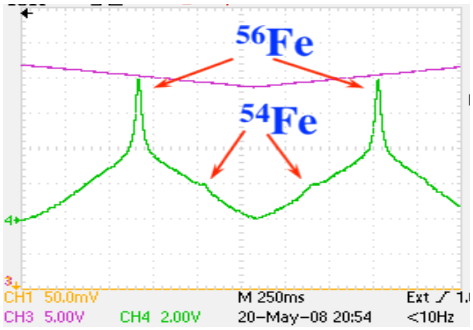

Figure 4. Detection of isotope line shift between ^{56}Fe and ^{54}Fe for the 372-nm absorption line

We have tried to measure the isotope line shifts between ^{56}Fe and other Fe isotopes with a lock-in amplifier and amplitude-modulation spectroscopy using a chopper to chop the laser beam. The ^{54}Fe Doppler-free peak was successfully found as shown in Figure 4. However, we did not see ^{57}Fe and ^{58}Fe peaks at our first try. Two main reasons: (1) the amplitude-modulation method is not as sensitive as the wavelength or frequency modulation because of the jitter of laser intensity and chopper, and (2) the hollow-cathode discharge cell might not have provided sufficient vapor pressure for detection of the very small abundance of ^{57}Fe and ^{58}Fe. Frequency modulation will be our next step to resolve the issue.

3. Optical Heterodyne Detection for Spectral Analysis and Stabilization of Lidar Pulse

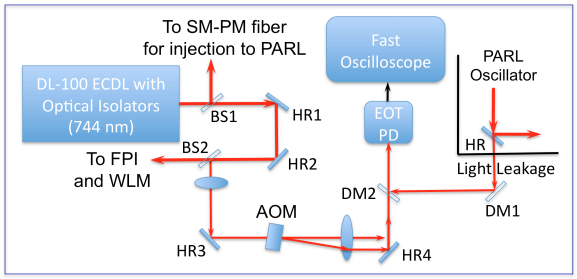

Figure 5. Experimental setup for optical heterodyne detection of PARL pulse with ECDL at 744 nm

As described in a companion paper [3], to achieve the best accuracy for MRI lidar, we must monitor and control each lidar pulse, rather than only controlling the cw seed laser. For pulsed lasers such as alexandrite laser, Nd:YAG laser or pulsed dye amplifier (PDA), the chirp is a major issue leading to unpredictable frequency

variations and offset. It is very challenging to measure the real frequency chirp. Based on an earlier proposal we made in [4] to detect PDA chirp and further inspired by coherent Doppler lidar, we applied a simple but very powerful approach – the optical heterodyne detection (OHD) technique – to the MRI lidar. The experimental setup is illustrated in Figure 5, where an AOM shifts the cw seed laser (ECDL) frequency by 240 MHz, and then the shifted beam is mixed with a tiny portion of the PARL oscillator pulse at 744 nm. The key is to make the laser pulse amplitude comparable with the cw beam amplitude. In our tests, filters with OD of ~7 were used to attenuate the pulse intensity. A few samples of pulsed optical beat signals are plotted in Figure 6.

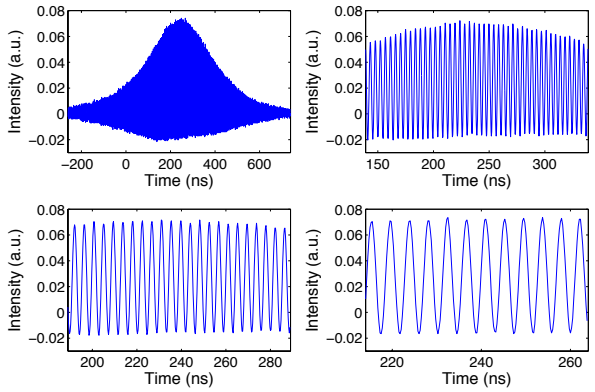

Figure 6. Samples of pulsed optical beat signals

We can infer information like pulse's frequency chirp, line width, power-spectral density as well as central frequency offset from the optical beat signal via Fourier spectral analysis. Initial results are given below.

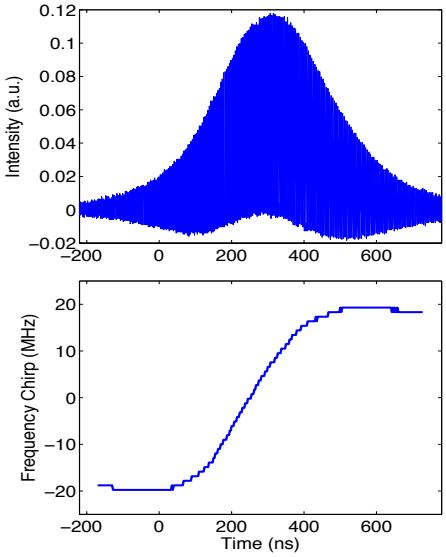

Figure 7. Optical beat signal obtained with the MRI lidar PARL and ECDL, and frequency chirp inferred within the pulse width of PARL oscillator.

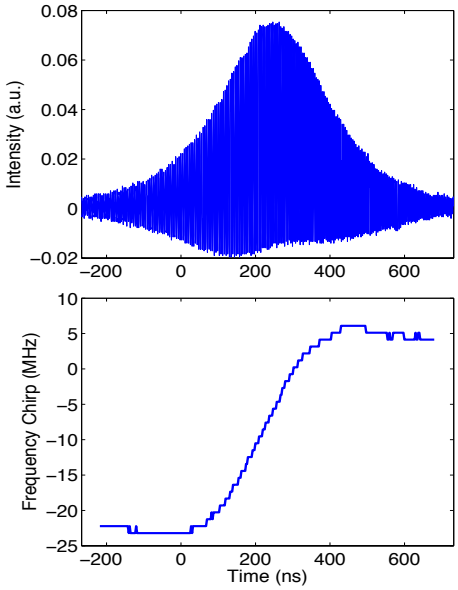

Figure 8. Another example of frequency chirp of the PARL pulse inferred from optical beat signal

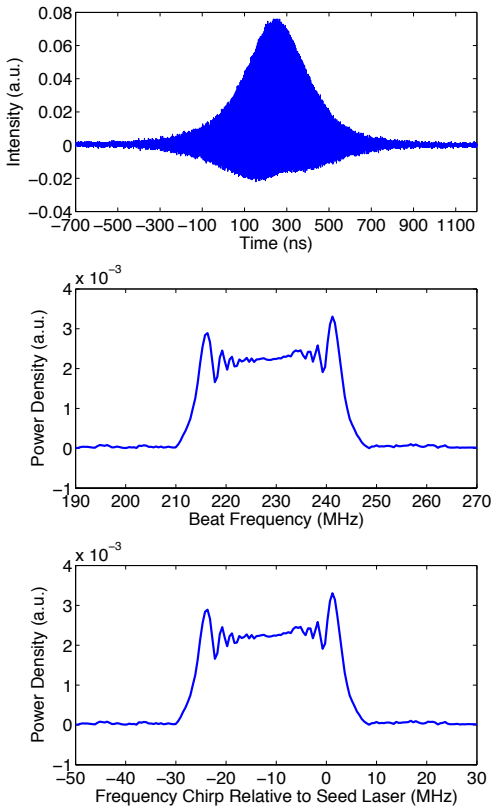

Figure 9. Optical beat signal between PARL pulse and cw seed laser (Top), Power spectral density of PARL pulse inferred from the beat signal (middle), and Power spectral density in terms of frequency chirp, showing line shape (bottom).

Optical heterodyne detection can be expressed as the following. The frequencies of AOM-shifted (f_{AOM} = 240 MHz) cw beam and laser pulse are given by Eq. (1):

$$f_{CW} = f_{Seed} - f_{AOM}$$
$$f_{Pulse} = f_{Seed} + chirp \quad (1)$$

The difference between these two frequencies gives the optical beat frequency:

$$f_{Beat} = \left| f_{Pulse} - f_{CW} \right| = chirp + f_{AOM} \quad (2)$$

Therefore, the chirp between the PARL pulse and the ECDL seed laser is derived as

$$chirp = f_{Beat} - f_{AOM} \quad (3)$$

where positive chirp means the PARL pulse frequency is higher than the seed laser, i.e., blue shifted.

Applying Fourier transform to the optical beat signals, we can acquire the spectral information of each pulse. Fourier analysis can be done for each portion of the pulse, thus we can infer how the frequency varies through the pulse width. Two examples are plotted in Figures 7 and 8. It is clear that the gain-induced chirp goes blue shift in the central portion of the pulse, while mechanical chirp going red shift evident on two wing portions. The edge-to-edge linewidth is 30-40 MHz for PARL oscillator at its fundamental wavelength 744 nm.

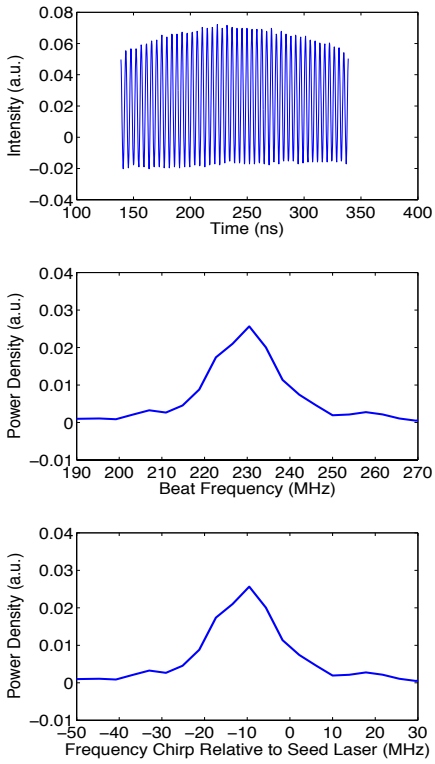

Figure 10. Same as Figure 9 but for pulse central portion only

Fourier analysis to the entire pulse provides the power-spectral density, and Figure 9 confirms the linewidth inferred above but the "ear" structure is striking. Further investigation shows that two ears come from the leading and trailing edges, respectively, of the PARL pulse. If only looking at the central portion of the beat pulse, we obtain a near-Gaussian spectral peak in Figure 10, whose FWHM is less than 20 MHz.

Such "ear" structure had never shown in wavelength meter or FPI measurements. This is most likely due to the insufficient spectral resolution of these instruments. Since the ECDL seed laser has less than 1 MHz spectral width, the optical heterodyne detection can well resolve sub-MHz spectrum, enabling the measurement of true frequency chirp and spectral structure of laser pulses for resonance Doppler lidars.

4. Conclusions and Outlook

To remove chirp influence on measurement accuracy and provide absolute frequency stabilization, our idea is to lock the lidar pulse frequency onto the Fe 372-nm line peak. First, the 372-nm ECDL is locked to the peak of Fe Doppler-free spectroscopy at 372 nm, thus transferring the absolute frequency reference from Fe atoms to the 372-nm ECDL. Second, we apply the optical heterodyne detection technique to the 372-nm cw and pulsed lasers. The optical beat signal tells whether the 372-nm pulse frequency differs from the Fe peak. If discrepancy occurs, we then tune the PARL frequency via tuning the 744-nm seed laser to make the lidar pulse frequency match the absolute frequency reference. Such scheme is expected to produce a bias-free resonance Doppler lidar for the first time in the world.

Acknowledgements

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