Velocity Measurements by Modulated Filtered Rayleigh Scattering Using Diode Lasers

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We describe a novel variation on filtered Rayleigh scattering using solid-state near-infrared diode lasers that we call modulated filtered Rayleigh scattering (MFRS). Solid-state diode lasers are simple, rugged, reliable, and relatively inexpensive. Hence an MFRS instrument would be well suited to flight instrumentation and industrial applications. We exploit the tuning capability of the diode laser to set it to the D_2 line of Rb at 780 nm and perform filtered Rayleigh scattering measurements. The continuous tuning is also exploited to modulate the laser frequency rapidly, which provides the MFRS technique with some unique properties. Because the laser operates with a continuous wave, one can obtain continuous velocity and density measurements. In principle, two velocity components can be measured with a single laser beam, and simultaneous multipoint measurements of velocity and density are possible. We present an early demonstration of the technique by making single point velocity measurements in a supersonic jet of CO_2 .

Nomenclature

 $i_{V,S,L}$ = unit vector in the gas velocity, scattered light, and incident laser directions, respectively

V = gas speed

 $\Delta \nu_{\text{Doppler}}$ = Doppler frequency shift λ_0 = laser centerline wavelength

Introduction

THE ability of solid-state lasers to be tuned in operating fre-L quency at megahertz rates by input current modulation, while maintaining a relatively narrow linewidth, has made them useful for spectroscopic measurements.¹⁻⁴ Their other advantages include low cost, reliability, durability, and compact size, making them a good choice for a laser source in many practical applications. For their size they are also very bright. A single GaAlAs diode in a can the size of a pencil eraser is capable of emitting 50-100 mW of continuous power at near-infrared wavelengths. The laser itself is commonly less than a millimeter in size, and monolithic arrays with powers up to 100 W can be fabricated in very compact packages. In a filtered Rayleigh scattering (FRS) experiment⁵⁻⁸ a diode laser can be used to scan across an atomic or molecular absorption line, generating large changes in transmission at the resonances for very small changes in frequency. The hyperfine structure components of atomic lines of alkali metal vapors are closely spaced and very strong, which makes such atomic filters excellent candidates for sensitive Doppler shift detection and therefore for high-resolution velocimetry. In many respects they are superior to molecular absorbers like iodine because of their inherently narrower linewidth and extremely high optical depth at very modest vapor pressures and cell lengths. In the work we describe here we use a rubidium vapor filter, which has the strong D_1 and D_2 lines at 794 and 780 nm, respectively. These transitions are conveniently accessed by near-infrared diode lasers.

The low-power output of infrared laser diodes is their primary drawback relative to other laser systems commonly used for velocimetry. However, the capability to modulate the laser frequency rapidly and continuously helps mitigate this. Using modulation spectroscopy and a heterodyne detection scheme with a lock-in am-

plifier, one can extract submicrovolt signals occurring at a specific frequency from a background that is orders of magnitude stronger. The diode laser modulation is simply achieved by adding a small current modulation to the laser bias current. It may also be swept repetitively in wavelength using an additional lower-frequency current ramp.

Grinstead et al. were the first to describe the use of frequencytunable lasers in FRS experiments.⁹⁻¹² They describe frequencymodulated filtered light scattering (FM FLS) in which they use an electrooptically modulated and electromechanically stabilized Ti:Sapphire laser and use a feedback loop to drive the laser so that the Doppler-shifted light is at the absorption maximum of the potassium filter. 9,10,12 They have also used modulation spectroscopy and first harmonic detection to measure the Doppler shift relative to absorption in a reference cell. 11 However, their stated aim 11 was to move toward the use of a second reference laser, frequency stabilized to the absorption peak without a Doppler shift. The Doppler shift is then determined by beating the two lasers against each other to determine the frequency difference.¹² This has the advantage of making the method independent of fluctuations in the scattering intensity. The disadvantages are that it introduces additional experimental complexity and restricts the method to single point measurements unless one uses an additional laser for each additional point at which a velocity measurement is desired. In our case it was the desire to take advantage of the compact size and low cost of a diode laser that led us to utilize its inherent frequency modulation capability and employ the modulation spectroscopy technique; this would allow us to detect a weak diode laser signal and measure frequency shift by analyzing the transmittance signal through a strongly absorbing alkali vapor cell.

Theory

The basic principle of FRS is well known.¹³⁻¹⁶ When the laser beam is scattered from a flow, the Rayleigh (and Mie) scattered light is Doppler shifted according to the equation

$$\Delta \nu_{\text{Doppler}} = (V/\lambda_0)[\boldsymbol{i}_V \cdot (\boldsymbol{i}_S - \boldsymbol{i}_L)] \approx V/\lambda_0 \tag{1}$$

The factor containing unit vectors in brackets arises from the geometry of the experiment and is of order unity. If the unshifted beam (V=0) is set on the edge of a particular hyperfine component of Rb, then in a moving gas $(V\neq 0)$ the Doppler effect shifts the scattered light to a new frequency that is attenuated differently by the atomic filter. Velocity fluctuations are thus converted to intensity fluctuations in principle. In practice, a normalizing scheme is needed to account for the fact that the intensity of the scattered signal itself fluctuates because of fluid density fluctuations or because of variations in the density of small particulates carried in the flow. Ambient

Presented as Paper 98-0510 at the AIAA 36th Aerospace Sciences Meeting, Reno, NV, Jan. 12-15, 1998; received March 27, 1998; revision received Feb. 22, 1999; accepted for publication Feb. 25, 1999. Copyright © 1999 by the American Institute of Aeronautics and Astronautics, Inc. All rights reserved.

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light interferences can be a problem because the Rayleigh signal is weak particularly when using a continuous wave (cw) solid-state laser.

To improve detectability in the modulated filtered Rayleigh scattering (MFRS) technique, we modulate the laser frequency and use heterodyne detection. In essence we perform frequency modulation absorption spectroscopy¹⁷ using the Rayleigh scattered light as a light source. Conventional modulation absorption spectroscopy is used to distinguish a weak absorption on a relatively strong laser signal. In our work we use it to detect a relatively strong absorption but on a weak signal. As is well known, if the lock-in amplifier is tuned to extract the *n*th harmonic of the modulation frequency, then an approximation of the nth derivative of the signal will be detected. Therefore, use of a second harmonic detection scheme in our experiments leads to a signal similar to the second derivative of the absorption spectrum (Fig. 1). The resemblance of the nth harmonic signal to the nth derivative of the original signal is dependent upon the modulation current and the frequency response of the synchronous detection electronics. Accuracy improves as the modulation amplitude is reduced, whereas signal strength and distortion increase with modulation amplitude.¹³ Therefore, a compromise must be found between signal clarity vs strength and modulation amplitude. Synchronous detection provides very high rejection of interferences, so that weak signals and ambient light are much less of a problem. Additionally, the modulation of the laser frequency that is needed for MFRS also provides a simple means of stabilizing the laser frequency and improving the measurement precision still further.

Figure 2 is a schematic diagram of an MFRS experimental setup. A small portion of the laser beam is split off to a reference arm containing a Rb cell. The modulation spectrum recorded by the detector (A) in this arm provides an absolute frequency reference. The major portion of the beam is focused into the flow being studied.

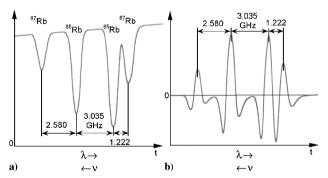


Fig. 1 D_2 resonance line recorded in an absorption cell containing Rb isotopes in natural abundance: a) conventional absorption spectrum and b) spectrum recorded using modulation spectroscopy and second harmonic detection.

The Rayleigh and Mie scattered signal is collected and split into two beams via 50/50 beam splitter. One beam is passed to a reference detector (B); the second part is filtered through another Rb cell before being detected on detector C. The ratio of the two signals gives the fractional transmission and thus removes the dependence on density of scatterers in the flow.

The system may be operated in one of two modes. In the first mode the laser is scanned across several hyperfine components so as to record a portion of the spectrum. The displacement between the derivative spectra, recorded simultaneously in the reference and signal arms, gives the Doppler shift and thus the flow velocity. One advantage of this mode is that one can dispense with the reference detector (B) in the signal arm provided the mean scattering density is constant and the time constant of the detection scheme is long enough to average out density fluctuations. The disadvantage is that the temporal resolution is limited both by the need to average out such fluctuations and by the need to scan the laser. The scan speed must be about two orders of magnitude slower than the modulation frequency so that the dither does not blur features in the absorption signal. It must also be slow relative to the averaging (output) time constant of the lock-in. This is not an insurmountable problem because megahertz modulation frequencies are possible with commercially available high-frequency lock-in amplifiers. One velocity measurement is obtained per scan in this mode.

In the second mode the laser is operated at a fixed frequency, and the magnitude of the normalized derivative provides a direct measure of the velocity and its fluctuations. Here the reference beam is required to normalize scattering density fluctuations. This signal also provides a measure of Rayleigh cross-section weighted density fluctuations, which gives density directly in flows of fixed composition. Specially tailored mixtures can be used for reacting flows so that the scattered signal directly provides mixture fraction¹⁸ or temperature.¹⁹ In this mode the temporal resolution is limited by the averaging time constant of the synchronous detection scheme. Lock-ins with time constant less than 20 μ s are commercially available giving continuous velocity measurements at the rate of about 50 kHz. In this mode the laser frequency must be stabilized to prevent drifts. However, the modulation scheme offers a simple and convenient way of doing this using a feedback signal generated in the reference arm.

The derivative signal shown in Fig. 1b would be obtained when the scattered light bandwidth is very narrow and reflects the narrow linewidth of the laser. This will be true of particle-ladenflows where Mie scattering dominates. However, in pure Rayleigh scattering the scattered linewidth will reflect the Rayleigh–Brillouin line shape, which is much broader than the laser linewidth and, indeed, broader than the Rb hyperfine multiplet.

The dashed line on Fig. 3 is a plot of an approximation to the Rayleigh signal profile that would be generated from an atmospheric pressure flow of typical combustion products (72.9% N₂, 11.6% CO₂, and 15.5% H₂O) at 1800 K moving at 10 m/s. The

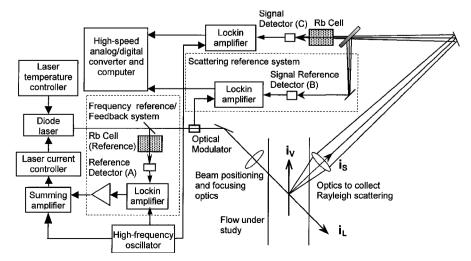


Fig. 2 Schematic diagram of the experimental arrangement for an MFRS experiment.

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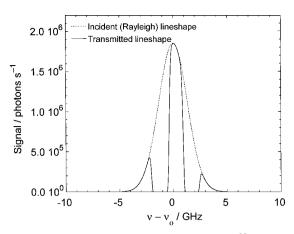


Fig. 3 Effect of a heated cell 10 cm long containing pure 85 Rb heated to 400 K on the Rayleigh scattered signal from typical combustion products at 1800 K and atmospheric pressure moving at 10 m/s. We assume a laser power of 200 mW, a probe volume 2 mm long, and f4 collection optics set at 90 deg to the incident beam. The area under the solid line would be the photon flux collected by the detector for the laser set at this particular frequency and Doppler shift.

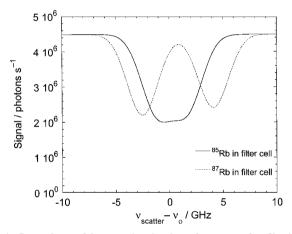


Fig. 4 Dependence of detector signal on laser frequency after filtering through a cell containing a pure Rb isotope heated to $400\,\mathrm{K}$. The assumed experimental conditions are the same as for Fig. 3.

Rayleigh scattering cross section for the mixture was assumed to be 1.44×10^{-28} cm²/sr at 780.24 nm. For this calculation we assume that the incident laser power is 200 mW, the probe volume is 2 mm long, and the collection optics have f number 4 corresponding to a collection solid angle of 0.0485 sr. The flow is assumed to be at 135 deg to the laser beam, and the scattering angle is 90 deg; this aligns the flow velocity vector with $(\mathbf{i}_S - \mathbf{i}_L)$ [see Eq. (1)]. We have neglected small losses in the transmission optics, and the line shape was approximated by a Doppler profile for simplicity. More accurate calculations would use a better scattering line shape that includes Brillouin scattering effects (e.g., the model of Tenti et al.²⁰) and correct for small distortions of the line shape caused by the finite collection solid angle. Figure 3 also shows the profile of the radiation that would be transmitted by a 10-cm cell containing pure ⁸⁵Rb heated to 400 K with the laser set at the center of the hyperfine multiplet of 85Rb. The area under this curve is the signal flux that would reach the detector. We have assumed a 10-cm cell for the calculation, but the same attenuation could be obtained in a much shorter cell and a very modest increase in temperature because the absorption is exponentially dependent on Rb vapor pressure, which itself increases exponentially with temperature.

As the laser center frequency is tuned across the Rb multiplet, the photon flux reaching the detector after filtering through the ⁸⁵Rb cell just described varies as shown by the solid line in Fig. 4. The dashed line shows the signal as a function of laser tuning if the cell contained pure ⁸⁷Rb at 400 K. We estimate the signal-to-noise ratio if these photon fluxes were incident on a photomultiplier tube with

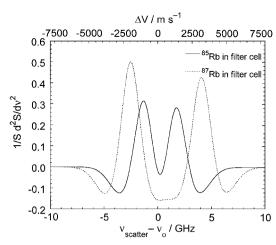


Fig. 5 Normalized second derivative of signals displayed in Fig. 4 showing dependence of derivative signal on laser frequency or, equivalently, gas velocity. Second harmonic detection with a lock-in amplifier produces an approximation to the second derivative.

a responsivity at 780 nm of 10^4 A/W and a dark current of 0.1 nA (typical specifications of a Hamamatsu R636-10). A photon flux of 3×10^6 photons/s at 780 nm corresponds to an incident power of 0.76 pW, which would give a signal current of 7.6 nA and a signal-to-dark current ratio of 76. The photoelectronshot noise corresponding to this signal level becomes comparable to this ratio for detection times of the order of 1 ms. It is obvious that larger collection solid angles and/or a higher-power laser would enable higher temporal resolution.

If the laser is frequency modulated, one again obtains a modulated signal, and a second harmonic detection produces a signal that approximates the second derivative of the curves shown in Fig. 4. The solid and dashed lines in Fig. 5 are the normalized second derivatives of the Rayleigh scattering profiles shown in Fig. 4 for the two filter cells (85Rb and 87Rb, respectively) and show how the signal would vary as a function of laser tuning for zero gas velocity, or equivalently (top axis), the variation of the derivative signal with gas velocity for fixed laser frequency. From the curve for ⁸⁷Rb in Fig. 4, we estimate that, if the laser is modulated at frequency f with an amplitude of 2-3 GHz, then the peak modulation amplitude (at 2f) for a signal filtered by a cell containing pure ⁸⁷Rb will have an amplitude of about 10⁶ photons/s corresponding to a signal amplitude of about 2.5 nA. This is well within the range of commercially available lock-in amplifiers, e.g., the SR-510 from Stanford Research Systems could be operated at its least-sensitive setting (5 nA full scale) for this signal level.

Figures 4 and 5 may be compared to the corresponding profiles in Fig. 1. Note that some of the differences arise because the data shown in Fig. 1b were obtained in a cell containing a mixture of Rb isotopes in natural abundance (72.2% ⁸⁵Rb, 22.8% ⁸⁷Rb). However, the broadening and changes in the shape of the curve are clear. In any case, a modulation signal of this shape can still be used to measure velocity using either method 1 or 2 already described.

Experimental Setup

The experimental setup was shown schematically in Fig. 2. The solid-state laser used in the present experiments is a single diode GaAlAs laser (Hitachi HL7851G) powered by an ultralow noise current source (ILX Lightwave LDX-3620) and cooled by a thermoelectric temperature controller (ILX Lightwave LDT-5910). The rated output of the laser is 50 mW at 780 nm, and its operating characteristics for input current and temperature variation were carefully studied. Both characteristics are linear within the laser operating range. However, like most diode lasers, the one used tends to exhibit mode hop to another linear range of frequencies for given conditions if the input current or temperature are changed too radically. The laser could be made to emit at the frequency of the Rb D₂ hyperfine resonances (780.24 nm) when operating at 5.0°C and a nominal current input of 116 mA. Dry nitrogen is bled into the diode laser head (ILX Lightwave LDM-4420) to avoid condensation.

The absorption filters are a pair of 10-cm rubidium vapor cells containing the Rb isotopes in natural abundance: 72.2% ⁸⁵Rb and 27.8% ⁸⁷Rb. Each isotope contributes different frequency resonances of varying strengths to the hyperfine structure—six from each—over a span of 7 GHz. In our experimentation so far, which has been mostly with Mie scattering, we have only seen the absorption spectrum at Doppler limited resolution (Fig. 1) where the components are overlapped into groups of three, giving two peaks per isotope. To increase the attenuation in the scattered arm, the cell is heated to increase the rubidium vapor pressure but not to the point of complete attenuation for the strongest peak.

The laser frequency is scanned repetitively by a 10-Hz triangle wave with an amplitude of 50 mV produced by a function generator (Stanford Research Systems DS345). The laser current source turns this voltage into a 5.0-mA amplitude current modulation that scans the laser across a 10.5-GHz band encompassing the entire hyperfine structure of the D₂ line of both isotopes of Rb. The rapid laser frequency modulation is provided by a 5-mV amplitude sinusoid of frequency of 50 kHz that is also generated by a function generator (Exact 200MSP). It is added to the scanning voltage using a summing amplifier (Stanford Research Systems SR560), summed again with the feedback signal if the frequency locking mode is used, low-pass filtered for noise above 300 kHz, and then input to the current source. The modulation frequency was limited to 50 kHz by the frequency response of the lock-in amplifiers (Stanford Research Systems Models SR530 and 510), which will only detect up to 100 kHz for the second harmonic mode. In addition, the scan frequency was severely limited by the relatively long output time constant of the lock-ins (minimum time constant = 1 ms).

The probe beam is set at 135 deg from the flow direction, while the scattered light collection axis is 45 deg from the flow, i.e., at 90 deg to the laser beam, all in the plane of the optical table on which the experiment is run. Because collection is performed with a lens of large numerical aperture, the detected light is the integral over all scattering angles collected by the lens. The $[i_V \cdot (i_S - i_L)]$ term in Eq. (1) shows that photons collected at an angle shallower than the 45-deg collection axis have a larger Doppler shift than those collected from the opposite side of the axis. However, because of the geometry involved, the contributions are not exactly symmetric with the result that a large collection angle results in a Doppler shift that is systematically a little low. Numerical simulation of the received scattered light gives a good estimation of the error introduced by the large collection optic. In our setup the collection lens had a numerical aperture of 0.24 (fnumber 1.8), and the measured Doppler shift had to be increased by 1.58% to remove this bias.

At the intersection of the laser and the collection optic focus, we could mount various scattering flows, the simplest of which is a disk attached to a motor controlled by an adjustable power supply. Our primary scattering target, however, is a jet of carbon dioxide gas from a pinhole orifice 0.79 mm ($\frac{1}{32}$ in.) in diameter. Carbon dioxide is fed to the orifice through 12-mm- ($\frac{1}{2}$ -in.)-diam insulated tubing at high pressure. The CO_2 flow becomes sonic at the orifice and then accelerates, forming a supersonic barrel shock from 2 to 5 mm in width. The CO_2 is rapidly cooled as it drops to atmospheric pressure, causing it to form the fog that serves as a Mie scattering medium in the flow. The optics visualize a 1-mm segment of the beam for a probe volume of approximately 0.2 mm³.

The scattered light is collected by a 5.66-cm-diam lens operating at a numerical aperture of about 0.24 (fnumber = 1.8) and focused onto an avalanche photodiode (APD) (Hamamatsu C5460). The reference beam is split from the primary beam with a pellicle (or microscope slide for mechanically noisy runs using the supersonic jet) and is detected with a balanced photodetector (New Focus, Nirvana). The signal from the APD goes to the SR530 lock-in, while the reference is input to the SR510. The lock-in outputs are monitored and recorded on a digital storage oscilloscope (Nicolet 3091). A proportionally balanced detection system, in which the absorption signal is ratioed by the instantaneous total scattering signal, has not yet been implemented. (The Nirvana detector has a gainbandwidth product that is too low for use in the Rayleigh scattering arm.) Hence, in the experiments reported here we have to average over scattering density fluctuations, and we use the scanned mode of velocity measurements.

We have also tested the improvement in laser frequency stability (and hence velocity precision) possible with feedback stabilization. Our preliminary feedback stabilization system consists of a coaxial cable connecting the output of the reference arm lock-in to the current modulation input on the diode current source with a variable resistor attenuator. First, another attenuators moothly brings the amplitude of the frequency scanning ramp to zero, so that the diode does not have a mode hop to a different frequency at the sudden change of input current. (The scanning ramp is usually used at startup to facilitate finding the resonance lines.) Then, with only the high-frequency modulation on the laser current, the laser bias current is adjusted to tune the laser to the edge of an absorption line. Because the laser characteristics are such that frequency decreases with an increase in driving current, an edge that increased in voltage with an increasing frequency was selected. Once near the locking frequency, the output of the reference lock-in is attenuated by a variable attenuator (set to maximum attenuation) and fed back to the laser driving current by a summing amplifier that adds it to the high-frequency modulation input. Decreasing the attenuation increases the feedback gain and increases the laser's frequency stability until there is so much gain that a feedback resonance begins to grow.

Results

Velocity measurements using the Doppler displacement technique were performed to validate the MFRS concept. Even without the balanced detection system, the results are very promising. Doppler-shifted signal from the jet-scattered laser is recognizable (Fig. 6) with a single scan measurement. Distortion in the signal made single-scan peak-to-peak measurements of the Doppler shift uncertainto $\pm 9\%$ at jet velocities of 280 m/s (± 25 m/s). This could be improved by using a cross-correlation analysis of the entire recorded spectra. For mean velocity measurements multiple sampling could greatly increase the precision. The condensing jet of CO_2 used in these preliminary experiments was itself not very stable with unsteady flow fluctuations easily visible to the naked eye, and some of the observed distortions in the signal arise from these fluctuations.

Despite its primitive nature, feedback stabilization of the laser was quite effective. The frequency stability as well as its long-term drift was measured by setting the laser to a frequency on the edge of a narrow Rb peak and monitoring the value of the reference lock-in output (Fig. 7). Without feedback the frequency of the laser was found to jitter by as much as ± 11 MHz and drifted by 78 MHz over a span of 8 s. These translate to an uncertainty in velocity of 9 and 43 m/s, respectively. Even at maximum attenuation on the feedback signal, there was immediately a marked improvement in laser stability. Increasing the gain to the optimum decreased the jitter to ± 290 kHz with a drift of 1.2 MHz over 8 s. Therefore, the laser instability would only contribute an uncertainty in velocity of ± 0.16 m/s under these conditions. A carefully designed feedback control loop should improve the precision by at least two orders of magnitude,

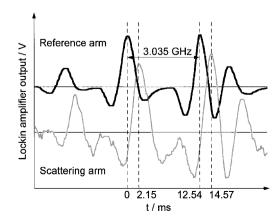


Fig. 6 Measurement of velocity by direct measurement of Doppler shift between modulation spectrum of FRS and one in a reference cell. The spacing of the hyperfine components provides an internal frequency calibration. Data were obtained in a supersonic jet of condensing CO₂.

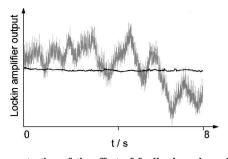


Fig. 7 Demonstration of the effect of feedback on laser frequency stability. Both short-term jitter and longer-term drift are greatly reduced: _______, feedback loop open, and _______, feedback loop closed.

and we can probably reduce the velocity error associated with laser frequency drift to negligible levels. Frequency stabilization of diode lasers via feedback is a well-established technology,^{21–23} and diode lasers have been stabilized to less than 1 kHz (Ref. 24).

Conclusions

Velocity measurements with an inexpensive prototype MFRS system have been made in a jet of condensing CO₂. The use of modulation spectroscopy techniques and precise feedback stabilization of the diode laser show promise of providing high-resolution continuous velocity measurements with a diode laser system.

Future work will involve implementing the fixed frequency technique to obtain continuous velocity measurements and extension to unseeded flows. The addition of a second detection arm opposite the first will allow for simultaneous measurement of two velocity components with a single laser beam, which would be significantly less complex than most other two-axis laser velocimetry techniques. Finally, in collaboration with Linne of the Colorado School of Mines, we wish to extend the technique to simultaneous multipoint velocity measurements along a line using the demodulating linear detector arrays they are developing.

Acknowledgment

This work was sponsored by the Texas Advanced Technology Program under Grant 003658-454.

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