Enhancement of spectral purity of injection-seeded titanium:sapphire laser by cavity locking and stimulated Brillouin scattering

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We report improvements to and better characterization of the spectral purity of a diode laser injection-seeded, cavity-locked titanium:sapphire laser that serves as the source for a previously reported rubidium vapor spectrally filtered Thomson scattering apparatus at 780.24 nm. In a detailed set of measurements the spectral purity \( P \) of the laser, defined as the ratio of the narrowband component of the laser output to the total output, has been studied as a function of frequency mismatch between the seed laser frequency and the central frequency of the unseeded cavity. It is found that spectral purity exceeding 0.999 can be obtained for a seed-cavity mismatch as high as ±0.25 nm, corresponding to approximately 950 cavity longitudinal-mode spacings and as high as ~0.9999 for a cavity–seed mismatch in the range ±0.10 nm (380 mode spacings). It is also shown that the addition of an external-cavity stimulated Brillouin-scattering phase-conjugate mirror increases both the spectral purity, to a minimum of 0.9999, and the cavity–seed mismatch range, to ±0.25 nm, for which this maximum effective purity is obtained.

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1. Introduction

Recent advances in solid-state laser technology have enabled a variety of new optical diagnostic techniques based on use of atomic and molecular vapor filters as narrow-bandwidth notch absorption filters and as spectral discriminators. Such filters share the common attribute that elastically scattered radiation from narrow-spectral-linewidth laser sources can be attenuated by many orders of magnitude, whereas Doppler-shifted or Doppler-broadened scattering can be significantly transmitted. The common availability of high-peak-power, narrow-spectral-linewidth pulsed laser sources has enabled a wide variety of such filtered scattering diagnostics, including applications to remote sensing, flow visualization, planar velocimetry, plasma physics, and combustion. In many such low-wave-number shift-based scattering diagnostics, the sensitivity of the measurement is ultimately limited by the spectral purity of the narrow-spectral-bandwidth laser source. The spectral purity is defined as the ratio of the energy (or power) contained within the spectrally narrow component of the laser output to the total energy (or power) output. For tunable pulsed systems, injection seeding is a common method for spectrally narrowing the output to one or two longitudinal modes. In particular, titanium:sapphire has become an extensively utilized solid-state laser because of its versatility and wide spectral tuning range.

In a recent paper we presented a new spectrally filtered Thomson scattering instrument that utilizes an external-cavity diode laser injection-seeded titanium:sapphire laser in combination with a rubidium vapor absorption filter at 780.24 nm. In this system the filter was used to highly attenuate (by as much as \(~10^5\)) elastic scattering (from electrode surfaces) and quasi-elastic Rayleigh scatter from gas medium heavy species (in this case argon), while simultaneously transmitting the wings of the thermally broadened (~3-nm) free-electron Thomson scattering. The emphasis was to demonstrate the utility of the system for measurement of electron temperature and electron number density in weakly ionized plasmas, as well as for other low-wave-number inelastic-scattering diagnostics, such as determination of...
temperature by pure rotational Raman scattering. It should be noted that the system is conceptually similar to both continuous-wave (cw) filtered Raman instruments\(^1\) and a recently reported pulsed dye laser and sodium vapor filter Thomson scattering apparatus.\(^6\) The system is also similar to the Thomson system recently presented by \textit{Zaidi et al.}\(^7\)

Although previous research has demonstrated the utility of our filtered scattering apparatus, in this paper we focus on improvements to and better characterization of the spectral purity of the titanium:sapphire laser source. This research is primarily motivated by the desire to optimize the sensitivity of the new instrument for studies of kinetic processes in high-pressure (of the order of 1 bar) low-electron temperature (of the order of 0.30 eV or lower), nonequilibrium, weakly ionized molecular plasmas.\(^17\) Although the cross section for quasi-elastic scattering of optical radiation by free electrons is of the order of \(10^2\)–\(10^3\) times greater than that for molecular (i.e., Rayleigh) scattering, the free-electron density of typical weakly ionized plasmas is of the order of \(10^{15}\)–\(10^{16}\) cm\(^{-3}\). The scattering intensity, therefore, is inherently low, such that the measurement sensitivity is constrained by the finite filter attenuation of elastic and quasi-elastic scattered light from stray surfaces and heavy species, respectively. For injection-seeded lasers, the realizable attenuation is limited by the small, but not insignificant, broad-spectral-linewidth nonseeded component of the laser output that is superimposed on the more intense seeded output.\(^10\) In this paper we present new measurements of the spectral purity of a pulsed Q-switched Nd:YAG pumped titanium:sapphire laser, injection seeded with a modest-power (8-mW) external-cavity diode laser. We show that incorporation of a cavity feedback loop to lock the pulsed cavity to the seed frequency creates single-longitudinal-mode output with a measured spectral purity as high as \(0.9999\). We show, however, that achieving such high spectral purity requires careful matching of the center frequency of the titanium:sapphire cavity gain profile to the seed laser frequency. Finally, we demonstrate that inclusion of an external-cavity stimulated Brillouin-scattering (SBS) phase-conjugate mirror (PCM) results in a further improvement of spectral purity to a minimum of 0.99999, albeit with a reduction in deliverable laser power by approximately 65%.

2. Experimental

A schematic diagram of the complete apparatus is shown in Fig. 1. The core of the system is the titanium:sapphire laser that is essentially identical to that described by \textit{Rines and Moulton}\(^1\) and employed previously by \textit{Finkelstein et al.}\(^18\) and \textit{Zaidi et al.}\(^7\), but is pumped by as much as 260 mJ/pulse (at 0.532 μm) from a commercial Q-switched Nd:YAG laser. The output from an –8-mW external-cavity cw diode laser is injected into the titanium:sapphire laser cavity through the output coupler using a Faraday rotation optical isolator. Whereas the research reported previously\(^14\) relied on passive injection seeding, in this paper we report measurements obtained incorporating a ramp and lock feedback system,\(^19\) which precisely matches the titanium:sapphire laser cavity to the longitudinal mode closest to that of the diode laser seed frequency, assuring reliable single-longitudinal-mode output of the pulsed cavity. (Note, as discussed in detail below, this is not necessarily the highest-gain longitudinal mode). The ramp and lock feedback loop is essentially identical to that described in Ref. 18. An uncoated flat optical plate is installed in the cavity, reflecting \(\sim 1\%\) of the circulating seed power to a photodiode detector. A few milliseconds prior to each firing of the Nd:YAG pump laser, a piezoelectric actuator is used to translate the cavity high reflector through the equivalent of three or four longitudinal cavity modes. Note that, as described in Ref. 18, some vertical tilt accompanies the desired translation, the effect of which is compensated for by inclusion of a porro prism retro-reflector as the final element in the cavity. The fringes seen by the detector are digitized, and a voltage setpoint is established that corresponds to a defined location on the fringe (which is adjustable by the user). The lock loop then feeds back to the piezoelectric actuator to maintain the cavity spacing to this fringe location. Lock is released just prior to the firing of the pump laser Q-switch (which causes a large intensity spike on the photodiode).

Ignoring the residual, unseeded component, the laser output energy is approximately 60 mJ/pulse with a spectral bandwidth that, although not explicitly measured here, has previously been reported\(^18\) to be close to the Fourier-transform limit of \(\sim 20\) MHz for a pulse duration of 22 ns. The principal differences between this laser system and that reported by \textit{Finkelstein et al.}\(^18\) are use of the diode laser seed source, which substitutes for an argon-ion-pumped cw titanium:sapphire ring laser, and use of the output coupler–optical isolator combination for seed injection, as opposed to the reflection from an intercavity optical flat.

In some experiments, the output of the titanium:sapphire laser is focused with a 150-mm focal-length plano–convex lens into a SBS PCM. The PCM was...
based on that described by Ni and Kung\textsuperscript{19} and consisted of a 15 mm diameter \times 100 mm length cylindrical glass cell, filled with FC-75, a fluorocarbon that is commercially available from 3M. SBS backscattering was separated from the input beam with a second Faraday rotation optical isolator. The net reflection efficiency, including losses from the uncoated cell window and the optical isolator, was approximately 35%, although we did not attempt to optimize this for these studies.

Spectral purity measurements are performed by imaging elastic scattering from a metal rod that is located in an otherwise evacuated cylindrical cell onto the entrance slit of an optical multichannel analyzer (OMA). The OMA consists of a 0.30-m spectrometer and a Gen IV near-IR microchannel plate intensified CCD camera. The spectrometer is operated with an input slit of between 50 and 100 \( \mu \text{m} \). A 5-cm path-length rubidium vapor cell, heated to a temperature between 280° and 320 °C, is placed in the detection path between the scattering cell and the OMA. Figure 2 shows a Fourier-transform transmission spectrum of the rubidium vapor cell at 310 °C, the approximate operating temperature employed for previously reported Thomson scattering measurements,\textsuperscript{14} along with a least-squares fit to a simple spectral model of the \( ^{5}s \ ^{2}S_{1/2} \rightarrow ^{5}p \ ^{2}P_{3/2} \) transition at 780.24 nm (vacuum wavelength). The absorption model, described in more detail in Ref. 14, is a simple sum of 12 individual components, resulting from a combination of hyperfine splitting and natural isotopic abundances.\textsuperscript{20} Spectral purity is determined by numerical integration of the spectrally resolved scattering, with and without the rubidium cell in place, as described in more detail in Section 3.

3. Results and Discussion

A. Laser Spectral Purity Measurements

The spectral purity \( P \) of a seeded laser is defined as the ratio of the narrowband component of the output to the total pulse output, where, in our case, the narrowband component is defined as the total laser energy contained within a single longitudinal cavity mode when the laser is operated in cavity-locked mode, or in one or two modes when the laser is operated in injection-seeded-only mode. As described in detail by Barnes \textit{et al.},\textsuperscript{10} \( P \) is a strong function of the circulating seed pulse energy, scaling as

\[
E_{\text{seed}} \propto P / (1 - P) \tag{1}
\]

when other factors, such as the spatial and spectral overlap between the seed laser beam and the titanium:sapphire cavity modes, are constant. Clearly, in the limit when \( P \) approaches one, the required seed energy increases rapidly.

As a general illustration of the utility of the vapor filter technique, Fig. 3(a) shows a scattering spectrum from a static cell of 500 Torr of nitrogen at room temperature, obtained without operation of the cavity-locking electronics in the laser or insertion of the rubidium vapor filter into the detection path. It appears to be a single central component with a linewidth of 0.20-nm full width at half-maximum (FWHM) corresponding to the spectral resolution of the spectrometer. Figure 3(b) is the spectrum obtained when the vapor filter is employed. Note, in particular, that the absolute intensity axis in Fig. 3(b)
is the same as that in Fig. 3(a). Comparison of Figs. 3(a) and 3(b) illustrates that the peak intensity of the central quasi-elastic-scattering signal has been attenuated by a factor of $\sim 10^5$. Although this clearly represents a significant attenuation, it should be noted that it is substantially lower than that which would be achievable, hypothetically, with an ideal laser source. For example, detailed calculations of rubidium absorption are presented in Ref. 15. For typical vapor cell conditions, the absorption coefficient (the exponent in Beer’s law) is found to be $\sim 10^4 \text{ m}^{-1}$ at the peak of the absorption. Such, essentially complete, attenuation is not realized in practice, however, because of finite spectral purity or other issues such as refuorescence and stray light. It should also be noted that, although not the focus of this paper, the peak quasi-elastic Thomson scattering signal, at our targeted conditions of $10^{13}$ cm$^{-3}$ electron density, will be a factor of approximately 30 lower than the peak Raman signal observed in Fig. 3(b).

Figure 4 illustrates a set of normalized transmitted signals of quasi-elastic scattering that were obtained with the scattering cell evacuated at relatively low circulating seed power in the range 0–0.026 mW. Because the transmission of narrowband scattering is negligibly small compared with the broadband fraction of the laser output, this residual transmitted scattering constitutes a measurement of the laser spectral purity. To obtain a value for $P$, the observed signal is fit to a convolution of an assumed Gaussian spectral profile with the modeled filter transmission at 320 °C. The fraction of laser output contained within the broadband component is given by the ratio of the integrated intensity of this best-fit Gaussian to the integrated signal observed without the filter (see Fig. 5). Note that this fitting procedure accounts for the fraction of the broadband component that is absorbed by the vapor filter. This fraction depends on the temperature of the filter and can be as high as approximately 85% for the highest temperature (320 °C) studied. Figure 5 shows the results of one such fit, and Fig. 6 shows values of spectral purity obtained from this procedure, as a function of circulating seed power. It can be seen that $P$ rapidly approaches 1 as the seed power is increased. More quantitatively, at 2.0 mW of circulating seed power (not illustrated in Fig. 6, but which was the maximum possible), we measure a value of $P$ equal to 0.9996, indicating a total spectrally integrated fractional power of $4 \times 10^{-4}$ in the broadband component.

Similar measurements were performed with the laser operating in cavity-locked mode. As shown in Subsection 3.B, cavity locking improves the spectral purity to $\sim 0.9999$ (or $10^{-4}$ fractional broadband power). This represents an improvement of approximately 30 compared with that reported in Ref. 10 (0.997) for the fundamental and Ref. 21 (0.998) for the third harmonic with cw seeding. It should be noted, however, that Ref. 10 reports a value of 0.999 for pulsed seeding of the cavity with 10-μJ pulses.

B. Effect of Seed and Laser Central Frequency Mismatch

In the course of performing the measurements described above, it was determined that achieving high spectral purity required careful adjustment of the cavity high reflector to precisely match the central
frequency of the unseeded titanium:sapphire laser to that of the seed laser. To explore this in more detail, we performed a series of spectral purity measurements as a function of frequency mismatch. In the absence of seeding, the titanium:sapphire laser output has a spectral FWHM of ~1 nm, which corresponds to approximately 3800 longitudinal modes. Figure 7 shows the titanium:sapphire laser output, as resolved by the OMA alone without use of the vapor filter, when the seed laser is tuned to the center of the rubidium vapor filter resonance at 780.24 nm but the cavity is tuned to a central frequency of ~780.80 nm. Note that tuning the cavity with such precision requires use of a second piezoelectric drive to tilt the horizontal axis of the porro prism retroreflector. Also note that the relatively low spectrometer resolution (0.20 nm) limits the accuracy of the cavity–seed mismatch wavelength to ~0.01 nm, or 5% of the instrumental linewidth. It can be seen from Fig. 7 that, even at this relatively large cavity–seed frequency mismatch, corresponding to ~2000 longitudinal-mode spacings, approximately 50% of the total output power was pulled to the seed frequency (the leftmost peak in Fig. 7) and approximately 50% of the power remains at the unseeded cavity frequency (rightmost peak in Fig. 7).

Figure 8 shows two spectra obtained under conditions similar to Fig. 7 except that the rubidium vapor filter at a temperature of 280 °C was inserted into the path between the scattering cell and the detector. Similar to Figs. 4 and 5, the residual transmitted signal provides a measurement of the spectral purity. In this case, however, the cell temperature was lowered to increase the residual transmitted light to a detectable level. Operation of the cell at 280 °C reduces the absorption band FWHM from the ~1.2 nm illustrated in Fig. 2 to ~0.45 nm. Because the linewidth of the unseeded component is also approximately 1 nm, lowering the cell temperature greatly increases the fraction of the unseeded titanium:sapphire laser component which is not absorbed by the filter and therefore reaches the detector. This improves the accuracy of the spectral purity measurement. Although not directly relevant to the subject of this paper, operation of the vapor cell at a reduced temperature is also critical if Thomson scattering measurements are to be performed at a low electron temperature because, for example, at 0.10 eV the Thomson scattering linewidth, assuming 90° scattering, is ~0.57 nm. Figure 8(a) shows the residual transmitted scattering for a mismatch of ~0.25 nm between the seed wavelength and the unseeded central cavity wavelength. In this case the spectral purity, determined by the integration procedure described above, is 0.9991. Figure 8(b) is identical to Fig. 8(a) except that the central frequency of the cavity was adjusted to match, as closely as possible, the seed frequency. This small adjustment is found to improve the spectral purity by almost 1 order of magnitude, to a measured value of ~0.9999. Also note that, on the basis of a conservative estimate of ten for the signal-to-noise ratio in the spectrum illustrated in Fig. 8(b), we estimate that spectral purity can be determined with an uncertainty of approximately ±0.00001.

Figure 9 summarizes the results from a set of mea-
measurements as a function of the seed–cavity wavelength detuning in the range of \( \pm 0.40 \) nm. It can be seen that a maximum purity of \(~0.9999\) is obtained over a range of approximately \( \pm 0.10\)-nm relative displacement (or 760 longitudinal modes). It can also be seen that spectral purity exceeding 0.999 is achieved for relative displacement of approximately \( \pm 0.25 \) nm (or 1900 longitudinal modes). It is stressed that this clearly significant difference is completely undetectable by any manual alignment procedure of either the cavity optics or the seed laser or by the locking electronics. To emphasize this point, it is noted that, according to manufacturers’ specifications, the linear translation of the porro prism horizontal axis corresponding to the entire \( 0.40\)-nm range illustrated in Fig. 9 is only \(~6\,\mu\text{m}\) (or \(~100\,\text{V}\) on the piezoelectric transducer).

C. Phase-Conjugate Mirror Measurements

To further increase the effective purity of the system, a SBS PCM was employed. SBS is often employed to filter broadband amplified spontaneous emission that is superimposed on the narrowband output of dye or solid-state lasers. The results of spectral purity measurements incorporating the PCM are plotted as the squares in Fig. 9. It can be seen that the maximum purity is increased and that the cavity–seed mismatch range for which purity exceeding 0.999 can be realized is increased. In fact, for the five data points indicating a spectral purity of 1, the residual transmission is too weak to be detectable, even with the 280 °C vapor cell and 3 min of integration on the CCD detector. This is illustrated in Fig. 10, which is identical to Fig. 8(b), except that the PCM is inserted between the laser and the scattering cell. Because the residual transmitted signal in Fig. 10 is below our detection limit of 0.0001 units, we conclude that a spectral purity of no less than 0.99999 has been achieved, corresponding to a residual transmitted fraction of \(~10^{-5}\). It should be pointed out that, although not essential, from a practical perspective this increased range of high spectral purity was found to greatly reduce the time and effort required to align the laser system for filtered scattering measurements. It should also be pointed out that, although not studied in as much detail, operation of the laser in injection-seeded, but not cavity-locked, mode in combination with the PCM resulted in a maximum measured spectral purity of only \(~0.9999\).

Finally, it should be noted that in a previous study preliminary results incorporating the PCM were reported. In that study, a prism monochromator, identical to that described in Ref. 6, was inserted directly in front of the PCM. The rationale was that, unlike amplified spontaneous emission, the divergence of unseeded output from the titanium:sapphire laser was likely not sufficient to yield a significant spatial discrimination between it and the seeded pump beam in the focal region of the SBS cell. It was thought that providing additional dispersion by means of the prisms would be essential to achieving significant reduction in the unseeded component. However, as illustrated by the data in Figs. 9 and 10, the inclusion of such a dispersing element was not necessary. The most likely explanation is that the increased spectral linewidth of the broadband component is sufficient to provide SBS discrimination. Alternatively, the divergence of the broadband component, which was not measured, may be greater than originally assumed. We plan to study this in more detail in the future.

4. Conclusions

Improvements have been made to a spectrally filtered Thomson scattering apparatus that incorporates an external-cavity diode laser, injected-locked titanium: sapphire laser and an optically thick rubidium vapor filter at 780.24 nm. In a detailed set of measurements the spectral purity \( P \) of the laser, defined as the ratio of the narrowband component of the laser output to the total output, has been studied as a function of circulating seed power. Employing injection seeding alone, \( P \) is found to range from 0.968 to 0.9996 for circulating seed power in the range 0.0012–2 mW. Incorporation of a feedback loop to lock the titanium:sapphire laser cavity to the cw seed...
frequency results in an increase in spectral purity to as high as \(-0.9999\). It is found, however, that achieving such high spectral purity requires careful matching of the seed laser frequency to the center frequency of the unseeded cavity gain profile. Finally, incorporation of a SBS PCM provides a further increase in the effective spectral purity such that residual scattering is too weak to be discernable above the noise floor when the vapor filter is operated at 280 °C. We estimate, on the basis of this measurement, a further improvement in spectral purity to a minimum of \(-0.99999\).

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References and Notes
1. See, for example, Measurement Science and Technology 12(4), 2001, which is a special issue outlining recent progress in molecular filter-based diagnostics.