# Note: Higher resolution Brillouin spectroscopy by offset stabilization of a tandem Fabry-Pérot interferometer

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A simple modification to a Sandercock-type tandem Fabry-Pérot interferometer is demonstrated. By adding an independent reference laser with temperature tunability, narrow Brillouin lines that are tens GHz shifted from the Rayleigh line can be recorded with much higher frequency resolution than in the original system. © *2011 American Institute of Physics*. [doi:10.1063/1.3665929]

Light scattering is one of the most useful spectroscopic techniques in science, and it offers a lot of important information on the dynamics of the various elementary excitations in matters. In light-scattering spectroscopy with small frequency shifts, namely, $\lesssim$5 cm$^{-1}$ (150 GHz), e.g., in Brillouin scattering from acoustic sounds or spin waves, Fabry-Pérot (FP) interferometers with a frequency resolution higher than 1 GHz are usually employed. Sandercock has invented a very sophisticated 3+3 pass tandem FP system,[1–3] which has been commercially available since early 1990s, and most scientists engaged in Brillouin spectroscopy use this so-called "Sandercock FP." In FP spectroscopy, the frequency resolution can be increased simply by widening the spacing between the mirrors. In Sandercock FP, however, one can only increase the spectral resolution about the central portion (excitation line) of the scattered spectrum because the excitation light serves also as the reference light for the stabilization of the FP. Therefore, by increasing the mirror spacing, one can resolve a narrow "central peak" better, but cannot easily do it on the frequency-shifted Brillouin lines, which are typically shifted $\gtrsim$10 GHz away from the Rayleigh line.

Here we propose a simple modification in stabilization of a Sandercock FP system by adding a second laser as the reference light. This enables "offset-stabilization" of the system, i.e., one can make best use of the highest resolution of the instrument at arbitrary Brillouin frequency. The proposed modification is completely non-destructive and quickly reversible to the original configuration. Also, it is quite a low-cost upgrade.

Figure 1 shows the high-resolution modification of a Sandercock FP system. The excitation laser is a frequency-doubled diode-pumped solid state (DPSS) Nd:yttrium-aluminum-garnet laser oscillating in a single longitudinal mode at 532 nm (Oxxius SLIM-532 300 mW). The excitation laser is temperature-stabilized by the built-in electronics, and its frequency is nominally fixed. The nominal wavelength stability is less than 1 pm for over 8 h and $\pm 3$ °C. Taking into account the stability of the laboratory temperature, which is approximately $\pm 0.5$ K, the frequency stability is roughly estimated to be $\lesssim$22 MHz/h. The Sandercock FP is usually equipped with a shutter control unit (SC), which consists of a beam splitter and two mechanical shutters. This unit acts as a dynamical notch filter when the FP's scan passes right at the intense elastically-scattered line, and at the same time, the SC unit introduces reference light with moderate intensity from the side for stabilizing the FP. In a normal system, the reference light is picked off from the excitation light by a glass plate (as indicated by dashed path in Fig. 1).

In addition to the excitation laser source, a second weak laser, to which we refer as "reference laser," is also employed to serve as an *independent* reference signal that is used as the stabilization of the FP. The reference laser is a small DPSS Nd:yttrium-vanadate (Nd:$YVO_4$) laser module (Photonic Products 300-0088-01, 4 mW) oscillating in single transverse mode ($TEM_{00}$) and has two to three longitudinal modes separated by 120 GHz. The spectrum of the reference laser is shown in Fig. 2(a). We have found that some of other cheap green-laser modules are even usable if the pumping laser diodes (LDs) are weakly driven. The longitudinal mode spacing in other green-laser modules were found to range from ~50 GHz to 120 GHz.

As shown in Fig. 1, we attached a thermoelectric cooler (TEC, Peltier element) underneath the holder of the reference laser, and controlled the temperature within 5 mK/h by a commercial temperature controller (Newport 350). This external temperature control, in turn, offers excellent tunability for the frequency of the reference laser through thermal expansion of the laser cavity. The temperature dependence of the oscillation frequency of one of the reference laser lines is shown in Fig. 2(b). For the present DPSS module of the reference laser, the temperature coefficient of the frequency at around room temperature was measured to be approximately $-7.9$ GHz/K, where the negative sign means that higher temperature of the laser cavity results in longer resonance wavelength due to the thermal expansion effect. The nominal frequency stability of the reference laser is then estimated to be <39 MHz/h. The tuning range of the reference laser solely by changing the temperature is not so wide ($\lesssim$60 GHz) because of mode hopping. However, the reference line can be selected out of the two or three longitudinal modes separated by axial mode interval (120 GHz in our system). Therefore, the effective frequency selectivity is wider than 240 GHz, which covers the Brillouin shifts of most materials on earth. In high resolution measurements, the free spectral range (FSR) of the FP is so narrow ($\lesssim$10 GHz) that the unnecessary lines separated by 120 GHz

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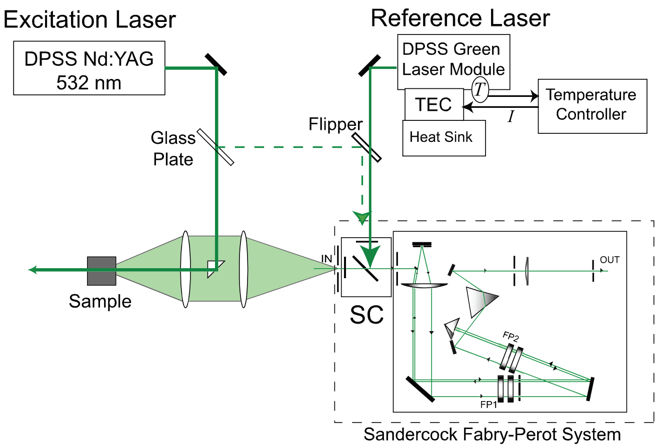


FIG. 1. (Color online) Setup for offset stabilization of a commercial Sandercock Fabry-Pérot system. In addition to the excitation laser, an independent reference laser, which can be temperature tuned, is employed as the reference signal. TEC: thermoelectric cooler (Peltier element) and SC: shutter control unit.

never appear in the spectrum and cause no effect in measurements.

Figure 3 shows the Brillouin spectra from a block of Plexiglas (poly-methyl-methacrylate, PMMA) in the back scattering geometry. Figure 3(a) is a typical Brillouin spectrum measured in original configuration. The mirror spacing of the FP was 2.5 mm, which gave an FSR of 60 GHz. The spectrum includes the Rayleigh line (denoted by R) in the center. In Fig. 3(b), the stokes component of the Brillouin line presented in (a) is simply zoomed in. Measured points are plotted by ○, and the solid curve is a Lorentzian fit (without convolution of the instrumental function), which fails to reproduce the correct line shape. Figure 3(c) shows the spectrum obtained by offset stabilization of a Sandercock FP. The mirror spacing for the offset stabilization was 25 mm, which gave an FSR of 6 GHz, i.e., 10 times higher resolution than that in the case of normal operation shown in Figs. 3(a) and 3(b). The FP was locked at −15.05 GHz shift, which is indicated by a vertical arrow in Fig. 3(c) (the reference laser line is not visible in this figure). It is obvious that the line shape of the Brillouin line from the longitudinal acoustic (LA) phonon is much better resolved in the offset-stabilization configuration. Indeed, the spectrum was successfully fitted by a simple Lorentzian (without using convolution) as shown in the solid curve in Fig. 3(c). The half width at half maximum was directly obtained to be 72 MHz, which was exactly the same value obtained by Voigt fitting (convolution of a Lorentzian and Gaussian). Furthermore, there are much more data points in the spectrum, which should improve the accuracy of analysis. It should be noted that the frequency interval shown in Fig. 3(c) was estimated from the FSR of the FP and the calibrated scan amplitude. The absolute frequency was determined from the peak position of the excitation line recorded in much lower resolution.

In the demonstrations presented above, the absolute frequency has to be separately determined from a lower resolution measurement, that is, the accuracy in the absolute frequency in the present configuration is identical to that in the original system. However, the accuracy in the relative

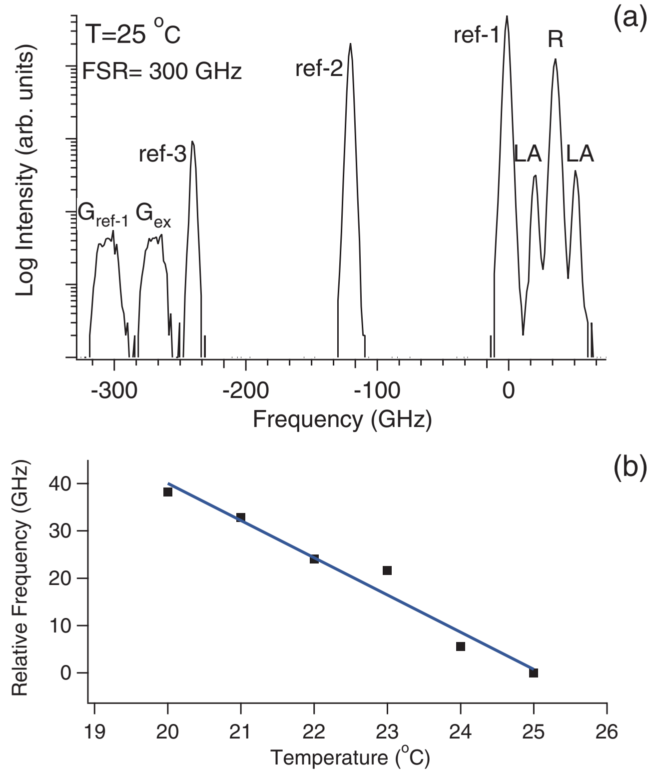


FIG. 2. (Color online) (a) Spectrum of the reference laser recorded with 300 GHz FSR. The Rayleigh and Brillouin spectra from a Plexiglas block excited by the excitation laser are also recorded simultaneously. The lines labeled as ref-1 to ref-3 are the longitudinal cavity modes of the reference laser. The R denotes the elastic Rayleigh scattering and the LAs denote the Brillouin scattering from the longitudinal acoustic phonons in Plexiglas. The $G_{ex}$ and $G_{ref-1}$ denote the first-order transmission lines (ghosts) corresponding to the excitation and ref-1 lines, respectively. (b) Temperature dependence of the oscillation frequency of ref-1 line. The solid line is a least-squares fit to the data.

frequency, i.e., the resolution in linewidth, is drastically improved. For higher accuracy of the absolute frequency shift, one can make use of the frequency-calibration technique employing an electro-optic modulator (EOM) as proposed by Caponi and co-workers.[4] However, since such calibration technique requires a broad band EOM, a microwave oscillator and amplifier with frequency well above several GHz, it would be quite an expensive upgrade.

We should note here the stability of the frequency difference between the excitation and reference lasers. In the present system, we do not actively stabilize the frequency difference because we assume a very simple system that may be easily built by most users of Sandercock FP. Therefore, the relative frequency stability is determined by the *independent* stabilities of the two lasers. The frequency stability of the system can be evaluated by recording the excitation laser line (with reduced intensity) when the FP is locked to one of the reference laser's line. The stability of the present system so recorded for 1 h is shown in Fig. 4. Here the vertical axis is on logarithmic scale, and slight asymmetry is present due to relatively low finesse of the FP when operated with a mirror spacing of 25 mm, which is effectively the longest usable one in the Sandercock FP system. The full linewidths of the reference and excitation lasers shown in Fig. 4 were read

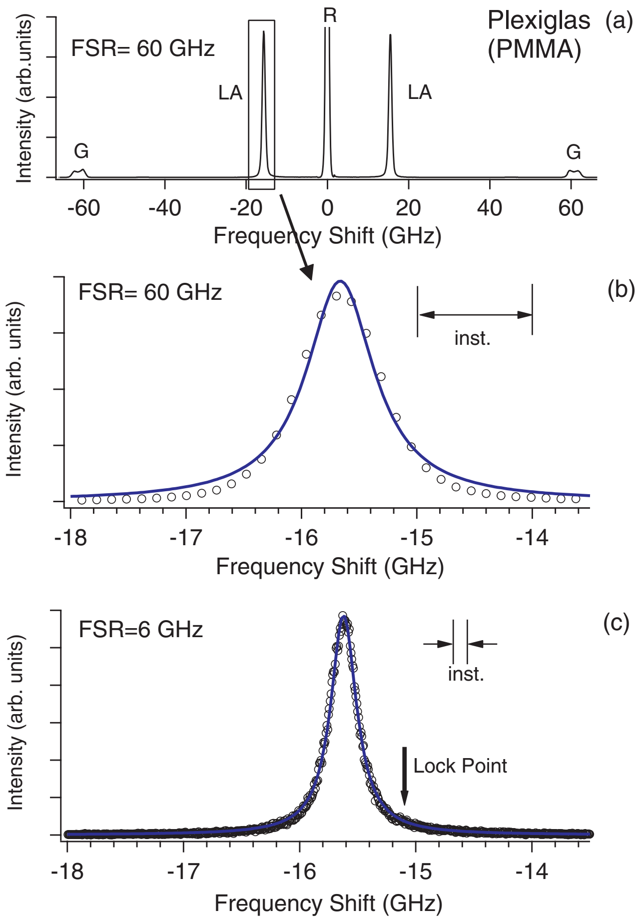


FIG. 3. (Color online) Comparison of spectra obtained by normal and offset stabilization of Sandercock Fabry-Pérot. (a) Spectrum obtained in normal operation. The R, LA, and G denote the Rayleigh (excitation) line, Brillouin line, and first-order transmission of the excitation laser, respectively. (b) Simply zoomed Stokes LA line in (a) (FSR = 60 GHz). (c) Spectrum obtained by offset stabilization method (FSR = 6 GHz). The position of the reference line (where the FP is locked) is indicated by the vertical arrow. In (b) and (c), the solid curves are fitted simple Lorentzian function. Instrumental widths are also indicated in (b) and (c).

to be 173 MHz and 184 MHz, respectively. This means that the line-broadening due to the temperature drift is 11 MHz/h, which is only 6% of the instrumental linewidth (173 MHz). In most Brillouin measurements, such a small broadening may be reasonably neglected or easily de-convoluted as we have presented in Fig. 3(c).

For higher frequency stability, the temperature stability should be improved. If we assume that the temperature instability is due to the reference laser, the measured frequency deviation of 11 MHz corresponds to a temperature drift of 1.4 mK, which is estimated from the measured temperature coefficient shown in Fig. 2(b) (=−7.9 GHz/K). Therefore, a temperature stability better than 1 mK should further improve the performance of the system. For that purpose, in addition to temperature stabilization of the laser cavities, the heat sinks or mounting stages of the excitation/reference lasers may also be stabilized. The current injection circuit for the pumping LD in the reference laser may also be replaced by a high-performance LD driver, which is commercially available at a reasonable cost. Such an LD driver will stabilize the injection current, and in turn, will further stabilize the temperature of the laser cavity.

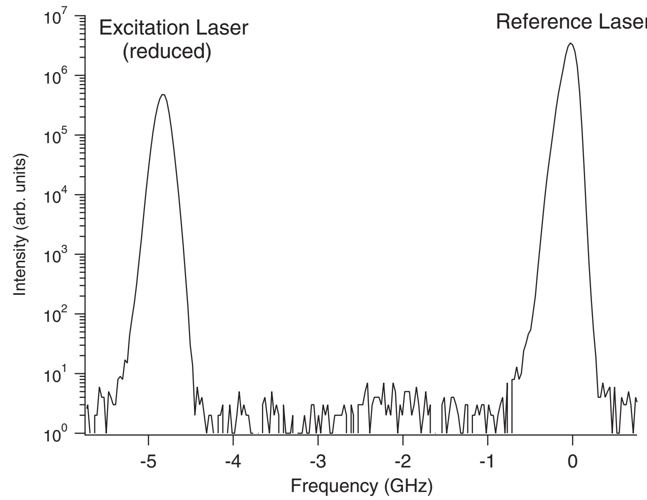


FIG. 4. Frequency stability of the offset-stabilized Sandercock FP. The spectrum of the excitation laser was recorded with the FP locked to a reference laser's line. The acquisition time was about 1 h.

In summary, we demonstrated "offset-stabilization" of a Sandercock FP. The addition of the second laser for the reference light and its temperature tunability achieved Brillouin spectroscopy with the highest frequency resolution available in the system at arbitrary frequency shift. The reference laser can have multiple longitudinal modes (but the transverse mode should be $TEM_{00}$), and such green-laser modules are widely available in quite low cost. The multiple longitudinal modes, in turn, offers very wide frequency selectivity. The present simple configuration was found to be stable enough for most applications in Brillouin spectroscopy, and may be easily constructed from inexpensive commercial products. Even higher frequency stability may be achieved by improved temperature stabilization on both of the excitation and reference lasers.

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