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Fourier transform infrared spectroscopy approach for measurements of photoluminescence and electroluminescence in mid-infrared

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An improved Fourier transform infrared spectroscopy approach adapting to photoluminescence and electroluminescence measurements in mid-infrared has been developed, in which diode-pumped solid-state excitation lasers were adopted for photoluminescence excitation. In this approach, three different Fourier transform infrared modes of rapid scan, double modulation, and step scan were software switchable without changing the hardware or connections. The advantages and limitations of each mode were analyzed in detail. Using this approach a group of III–V and II–VI samples from near-infrared extending to mid-infrared with photoluminescence intensities in a wider range have been characterized at room temperature to demonstrate the validity and overall performances of the system. The weaker electroluminescence of quantum cascade lasers in mid-infrared band was also surveyed at different resolutions. Results show that for samples with relatively strong photoluminescence or electroluminescence out off the background, rapid scan mode is the most preferable. For weaker photoluminescence or electroluminescence overlapped with background, double modulation is the most effective mode. To get a better signal noise ratio when weaker photoluminescence or electroluminescence signal has been observed in double modulation mode, switching to step scan mode should be an advisable option despite the long data acquiring time and limited resolution.

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I. INTRODUCTION

Fourier transform infrared (FTIR) spectroscopy, with well-developed theory and instrumentation, had been an effective method in numerous fields to distinguish diversified features of materials and devices. Compared to traditional dispersive spectroscopy, the high light flux, low equivalent noise, fast scan speed, and parallel beam features of FTIR make it an optimal option, especially in the mid-infrared (MIR) or far-infrared bands. Those merits have been concluded to the multiplexing and throughput advantages. The FTIR instruments are designed for adapting absorption or reflection measurements originally with the sample mounted inside the spectrometer, adopting an internal infrared radiation source. However, for most subsequent instruments an emission port also has been introduced, through this port the spectral features of materials and devices from external emission source, such as photoluminescence (PL) or electroluminescence (EL), could also be characterized. Based on this arrangement, accessional user configurations for different approaches have to be developed,^{1–16} whereas commercial PL or EL accessory is still rare.

Photoluminescence is a strong nondestructive tool for both scientific research and daily production examination of semiconductor materials. The marriage of PL with FTIR inherits the merits, therefore is becoming popular from near infrared (NIR) extending to MIR bands. Acquiring relatively

strong PL signals apart from the background (BG) using conventional rapid scan (RS) mode of FTIR is valid.^{1,2} Rapid scan mode is also effective for device characterization, such as the narrow band emission spectra of quantum well lasers in NIR (Refs. 3 and 4) and quantum cascade lasers with enough lasing power in MIR,⁵ regardless of continuous wave or pulsed operation. However, because PL signal is often quite weak, and in many cases they are even much weaker than the room temperature (RT) thermal background radiation (mainly distributing at MIR peak around 10 μm) or stray light background (mainly at visible and NIR), so extracting weak PL signal from the background and keeping the convenience of FTIR remains a challenge. Persistent efforts have been taken to find the solution. For example, background subtraction or frame-to-frame subtraction has been used to suppress the thermal background.⁶ Double modulation (DM) mode adopting mechanical modulator,⁶ acoustic-optic modulator⁷ or electro-optical modulator^{8–12} has been used to remove the background more effectively, in which phase sensitive detection or phase sensitive excitation schemes have been adopted. For device in MIR such as pulsed quantum cascade laser with quite weak emission overlapped to thermal BG, DM mode is also preferred since the pulsed device is modulated (normally at tens of kHz) already and no extra modulator is needed.¹³ Step scan (SS) mode adopting mechanical modulator has also been proved to be an impactful method to eliminate the background.^{14,15} These methods are successful on certain aspects, whereas accompanying expenditure of data acquisition time, loss of operation convenience as well as complication

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of the accessional system. In this work, with the comparative analysis of the system, an improved FTIR-PL approach adopting diode-pumped solid-state (DPSS) excitation lasers have been developed. Using this approach, a group of samples extending from NIR to MIR with quite different PL intensities have been characterized to demonstrate the system validity.

II. FTIR-PL CONSIDERATION AND CONFIGURATION

In FTIR spectrometer, the optical frequency ν (wave numbers, cm^{-1}) is transformed to Fourier frequency of $f_F = 2\nu$ (Hz) through a Michelson interferometer with moving mirror speed of v (cm/s). In general, the moving mirror speed v could be changed 2–3 orders and scaled at wavelength of 632.8 nm of internal He-Ne laser for internal alignment and sampling purpose, the upper is mainly limited by the response speed of the photodetector (hundreds of kHz for quantum type and few kHz for thermal type, with large sensitive area), the lower depending on the mechanical and stable demands for the instruments and circumstance. At low speed, the response is slow and time consuming of data acquisition is long. As mentioned above, if the PL signal was not very weak and located outside the strong background, RS mode could be a preferred option. For conventional RS mode, some advantages and warnings of FTIR-PL have been reviewed.¹⁶ Turning into MIR the thermal background arouses. For 300 K background, the radiation is peaked at $\sim 1035 \text{ cm}^{-1}$, with magnitude greater than 0.1%, 1% or 10% of the peak at ~ 3831 , ~ 3144 , and $\sim 2375 \text{ cm}^{-1}$ from short wavelength side, respectively, the long wavelength tail extends to far-infrared. The PL intensity much weaker than thermal BG is not unusual especially at long wavelength side, hereby for FTIR-PL in MIR ($4000\text{--}400 \text{ cm}^{-1}$) band RS mode should be improved. Double modulation mode adopting phase sensitive detection scheme is developed appending on RS mode using a modulated excitation laser, where the modulation frequency appending on the Fourier frequency forms a double modulation. Through the introduction of lock-in technique, the modulated PL signal could be extracted and unmodulated BG be suppressed effectively. In this approach, the modulation frequency of the excitation laser should be much higher than the Fourier frequency (at least >5 times as a rule of thumb) in the spectrometer, in this case through a lock-in amplifier locked to the modulation frequency with a suitable time constant, the amplified Fourier signal back to the spectrometer could be utilized.

For modulating the excitation laser, mechanical modulator (chopper) is conceivable at first, whereas its maximum frequency is limited to only a few kHz (4 kHz in common). For example, at 4 kHz modulation, the Fourier frequency should be below 800 Hz at least, in this case for wavenumber of $10\,000 \text{ cm}^{-1}$ the moving mirror speed is expected to be below 0.04 cm/s. At this quite low scan speed the phase locking of the lock-in amplifier becomes difficult, the response of the spectrometer is too slow for the alignment of the FTIR-PL setup conveniently; also the data acquisition time will be quite long. For example, at scan speed of 0.0475 cm/s, about 6 min was needed for acquiring a spectrum at $4000\text{--}600 \text{ cm}^{-1}$ span, 16 cm^{-1} resolution with 32 scans in our spectrometer, also in presetting mode the data refresh period was more than

10 s. For electro-optical modulator, the modulation frequency is not a problem (dc to $>1\text{MHz}$), whereas the higher driving voltage ($>200 \text{ V}$), small aperture, and relatively big size are uncomfortable for FTIR-PL application. For acoustic-optic modulator similar situation exists. Besides, the acoustic-optic modulator is operated at a fixed frequency or around a centre frequency, although the driving voltage of acoustic-optic modulator could be lower than those of electro-optical modulator, this frequency is normally too high (tens of MHz) for lock-in amplifier and photodetectors in FTIR. Also, the prices of those additive modulator and driver are quite high. Based on above considerations, the optimal modulation frequency for the excitation laser in DM mode should be tens of kHz, which is high enough for the Fourier frequency of a few kHz for FTIR at normal scan speed ($\sim 0.4 \text{ cm/s}$, at this speed the system response time is adequate), and still low enough for common photodetectors/lock-in amplifiers. Also, an effective, maneuverable, and inexpensive type of laser with convenient electrical modulation scheme is expected. In this case, the instrument settings could be similar, and the data acquiring time is fast enough, as compared to those of conventional RS mode.

Another approach to eliminate the BG is the SS mode. In this mode, the modulating of the excitation laser is still needed, whereas the scan of the interferometer is not continuous but step by step, therefore the restrictions on the modulation frequency are totally released. In SS mode, mechanical chopper could be used conveniently, and the thermal BG could be eliminated effectively.^{14,15} The SS mode has become a standard option for current FTIR spectrometers, the instruments are software switchable between RS mode and SS mode conveniently. However, the SS mode falls across a notable problem, especially in practical operation of the FTIR-PL system. In SS mode, the total scan steps are related to the spectral resolution directly, and the time spending on each step could not be very short mainly because of the mechanical limitations of the instruments. Therefore, at the same spectral span, resolution, and scan times, the data acquiring time for SS mode should be hundreds of times of the RS mode. In this extraordinary long acquiring time, the stability, reliability of the system, and random interference from the circumstance become uncontrollable, the patience of the operator has also been tested. Furthermore, in SS mode, the “instant” response feature of the FTIR was lost, the operator just could not see the effects of operation instantly, such as in the optical path alignment but at least minutes later. Without the help of catching a weak traceable PL signal at other modes, the operation of SS mode independently becomes extremely difficult, especially for a new operator with an unknown sample at hand. These non-theoretical but practical demerits restrict the adoption of SS mode in FTIR-PL remarkably.

Based on above considerations, an improved FTIR PL and EL configuration adopting DPSS excitation laser was developed as shown schematically in Fig. 1. With the progress of high power semiconductor lasers, DPSS laser is becoming popular for its high reliability, moderate stability, small size, and lower price. Since this type of solid-state laser is pumped by electrically driven semiconductor diode laser, it could be electrically modulated inherently. The DPSS lasers are also inherently free from the strong non-laser plasma emission

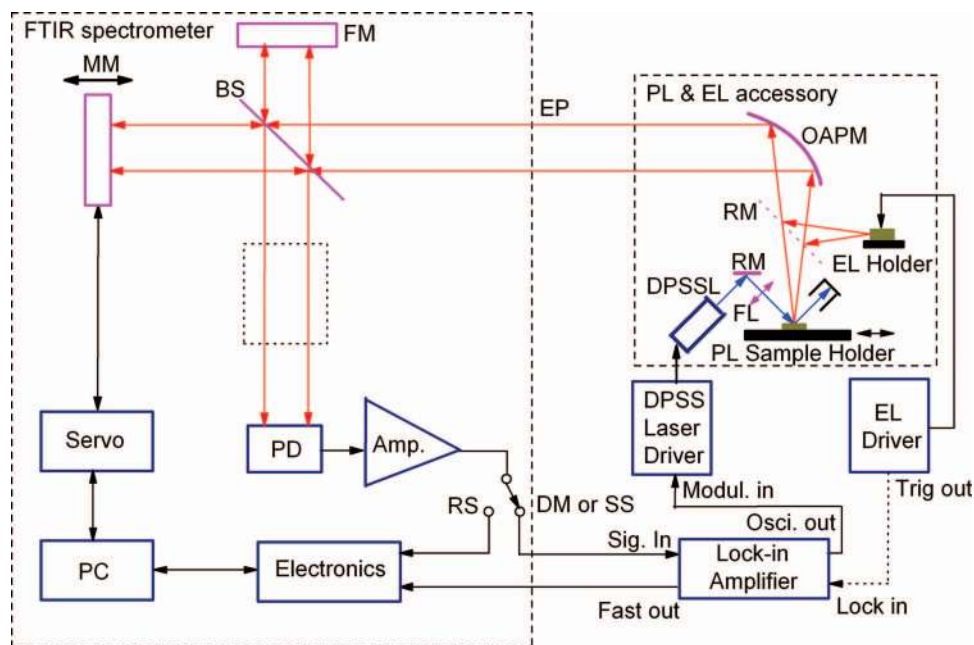


FIG. 1. Schematic diagram of the FTIR spectroscopy setup for PL and EL.

(quite troublesome in PL measurements) of gas lasers. For most DPSS laser products, analog or digital modulation input port of 0–5 V scale has become a standard option, the modulation frequency could be from dc to tens of kHz, just fit the needs. The DPSS lasers have many wavelength options, with different power efficiencies and so on. For numerous wavelengths, the 808 nm diode laser pumped 1064 nm DPSS laser and its frequency doubled 532 nm DPSS laser are preferred for their high power efficiencies and therefore lower price for unit power. The power efficiency could exceed 50% for 1064 nm, and attains 20%–35% for 532 nm DPSS lasers. Normally, the spectral feature and beam quality of low-end DPSS laser products are good enough for PL excitations already. Visible 532 nm DPSS laser could be a direct substitute of the 514.5 nm argon laser frequently used in previous PL system. For reaching deeper excitation depth and higher excitation power, the infrared 1064 nm DPSS laser should be a good choice. Although it is invisible to human eyes, it still could be clearly seen by Si CCD or CMOS image sensors, so the adjustment of the system is still quite easy. Certainly the diode laser itself could also be used for PL excitation more directly, but its beam quality remains a problem, further shaping of the beam is troublesome.

As shown in Fig. 1, our system was based on a Nicolet Magna-IR System 860 FTIR spectrometer (a quite old version) with simultaneous synchronous technique module. The FTIR accessory for PL and EL was built on a 30×30 cm² attachable optical breadboard with beam height match to the spectrometer. The excitation light from the exchangeable DPSS laser head (532 nm or 1064 nm with the same size) was steered by an adjustable reflecting mirror, then focused on the sample by a small focus lens (with focus length of a few centimeters). The sample was put on an adjustable sample holder X-Y stage horizontally. The beam diameter of the DPSS lasers was normally <3 mm, so the spot

diameter through the focus lens on the sample could be below 1 mm. The laser beam reflected from the sample was blocked and absorbed using a covering. The excited PL from the sample were collected and collimated towards the emission port of FTIR spectrometer using a 90° off-axis parabolic mirror (OAPM), its diameter was 2 in. and focus length was 3 in. The OAPM is unselective to wavelength, so this FTIR-PL accessory was suitable from visible to far-infrared. The maximum CW output power of the DPSS lasers was >1 W, with an analog modulation BNC connector jack. The output power of the laser is proportional to the modulation voltage in 5 V range with maximum modulation frequency extending to >30 kHz. In this approach, an EG&G 7265 DSP lock-in amplifier was introduced for phase sensitive detection, the connections were quite simple using only the external jack of the instruments. The input of lock-in amplifier was connected to the detector output jack of the FTIR spectrometer, and its fast output jack was connected back to the external detector input jack A on the simultaneous synchronous technique module. The purpose of using the fast output port was to permit the pass through the signal at Fourier frequency and lock-in the PL signal at modulation frequency more effectively in DM mode, by selecting the time constant related to this port properly. The internal oscillator output of lock-in amplifier with sine wave output voltage adjustable from 0 to 5 V was connected to the laser driver to modulate the DPSS laser at expected lock-in frequency and to control the output power. Other option to modulate the DPSS laser was to use an external pulse generator (square wave was suitable); in this case, the lock-in amplifier was triggered by this pulse generator. By using this arrangement, in this approach the RS, DM, and SS modes could be software switchover freely without the changing of hardware and connections. This attachable PL and EL accessory was rather simple with only 4 optical parts; the operation was also quite easy. In this setup, a removable reflecting mirror

could be simply inserted into the light path for EL measurements as shown in Fig. 1, in this case the trigger output from the EL driver was used as the reference for lock-in amplifier. As illustrated, this FTIR-PL accessory was for RT measurements without temperature control, the sample was simply put on the holder horizontally, the holder X-Y stage was useful to check the PL distribution across sample area. In the measurements, the operator just needed to put the sample on the focus point of the red He-Ne light from the emission port of FTIR spectrometer, and then steered and focused the DPSS laser light on the same point. In this setup, no further adjustment was needed when changing samples, therefore very suitable for daily examination of product. For low-temperature measurements, a similar accessory at a breadboard of the same size was also developed (not shown). Because in low-temperature measurements the samples were normally mounted on the holder of cryostat or Dewar vertically, a 2 in. reflecting mirror was added to turn 90° of the PL path for arranging the setup more conveniently. In case of using 1064 nm DPSS excitation laser, a small net video camera was attached for monitoring the laser beam at the computer screen.

III. DEMONSTRATION AND DISCUSSION

To demonstrate the validity of the approach, a set of III-V or II-VI samples including substrates, bulk epitaxial layers, and device structures were measured using this setup. Those samples were chosen to cover a wider spectral span for the most interested NIR to MIR bands, and with a big difference in their PL intensities. Some details of the samples and FTIR spectrometer settings were listed in Table I. The samples are divided into two groups. The first group of sample, a to f, covers the spectral span of NIR-MIR from 11 500 to 2100 cm^{-1} recommended for the combination of CaF_2 beamsplitter with InSb (77 K) detector, the second group of sample, g and h, covers the spectral span of MIR from 4000 to 600 cm^{-1} recommended for the combination of KBr beamsplitter with MCT-A (77 K) detector. This work was not concentrated on the analysis of spectral details of the samples but the overall performance of the FTIR-PL system, so for easy comparison all the samples were measured at RT and at the same conditions for each group.

Figure 2 showed the measured PL of the first group of samples in RS mode, the BG signal (without sample) was also recorded. All the PL in Fig. 2 were recorded at a rea-

sonable resolution of 16 cm^{-1} with 32 scans at scan speed of 0.3165 cm/s , the data acquiring time for each PL was 25 s using this FTIR spectrometer. At this scan speed, the response time of the spectrometer was still comfortable to the operator, whereas at even lower scan speed the presetting window of the instrument became stagnant. In the measurements, a 532 nm DPSS excitation laser was used, its output power was fixed at about 0.3 W. From Fig. 2 it could be seen that, in RS mode, the shortwave side thermal BG arouses steeply from about 3500 cm^{-1} for this quite sensitive InSb (77 K) detector. The strong CO_2 absorption around 2360 cm^{-1} could be clearly seen. For samples a, b, and c with PL at NIR, normally the influence of BG is not a problem. Therefore, through the appropriate selection of the wavelength span even weaker PL could appear as far as it still exceeds the detection threshold of the FTIR.¹⁶ The zoom in of PL of sample b (a quaternary InAlGaAs layer) with quite weak intensity was also shown in Fig. 1, in this case the PL still could be seen clearly but with a low signal noise ratio (S/N) of only about 2.8. Further increase of the scan times has inconspicuous effects mainly due to the detector/amplifier noise. By using a more sensitive RT InGaAs detector, the S/N of sample b could be improved remarkably (not shown). For samples d, e, and f with PL entered MIR, different situations occurred. For samples d and e with rather strong PL compared to the corresponding thermal BG the PL still appears clearly, but if the PL is weak and submerged into BG the RS mode could not work properly. For sample f of an InAs substrate, its PL intensity was comparable to the BG. In this case, the PL just could be seen but had overlapped with BG. Subtracting the BG from the signal often showed poor results because of the random effects; in this case, the RS mode became inadequate. At even weaker PL intensity the RS mode was invalidated. For a sample with unknown features (peak wavelength, intensity, etc.) as in the case of most scientific research, collecting PL using RS mode in MIR is really a challenge.

Figure 3 showed the measured PL of the same group of samples in DM mode; the BG signal was also shown. All the PL were still recorded at 16 cm^{-1} resolution with 32 scans at speed of 0.3165 cm/s , so the acquiring time for each PL kept unchanged for 25 s. In the measurements, the 532 nm DPSS excitation laser was modulated at about 35 kHz with average output power of about 0.3 W. In this wavelength span, the Fourier frequency was limited to about 7 kHz, so the time constant of lock-in amplifier was set to 80 μs (at slope of

TABLE I. Details of the samples as well as FTIR spectrometer settings.

No.	Sample description	RT-PL peak (intensity)	FTIR setup
a	n-InP substrate	10 761 cm^{-1} (28%)	
b	MBE n-In _{0.52} Al _{0.22} Ga _{0.26} As	8385 cm^{-1} (0.68%)	11 500–2100 cm^{-1}
c	MBE n-In _{0.53} Ga _{0.47} As	5908 cm^{-1} (89%)	InSb PD (77 K)
d	MBE n-In _{0.84} Ga _{0.16} As	3934 cm^{-1} (36%)	CaF_2 beamsplitter
e	MBE p-In _{0.89} Al _{0.11} As/n-In _{0.89} Ga _{0.11} As	3518 cm^{-1} (100%)	R = 16 cm^{-1} , 32 scans
f	n-InAs substrate	2908 cm^{-1} (44%)	
g	LPE HgCdTe	2130 cm^{-1} (5.4%)	4000–600 cm^{-1}
h	MBE p-InSb/n-InSb	1590 cm^{-1} (0.35%)	MCT-A PD(77 K) KBr beamsplitter R = 16 cm^{-1} , 32 scans

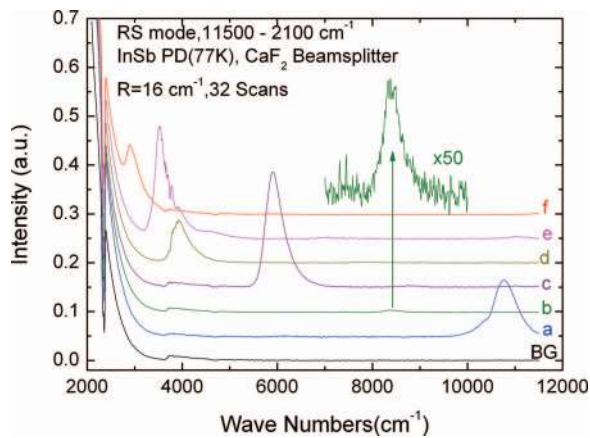


FIG. 2. Measured PL of sample a to sample f and background in RS mode.

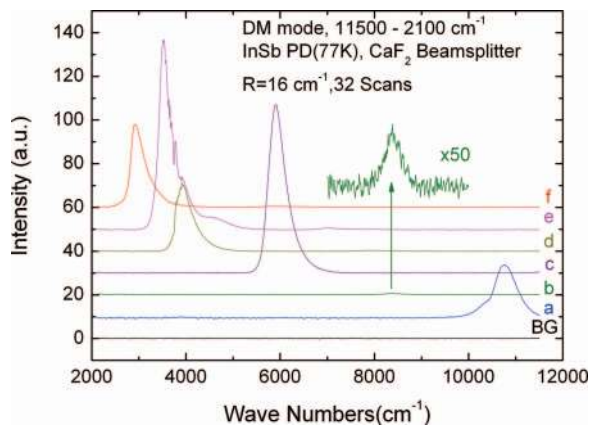


FIG. 3. Measured PL of sample a to sample f and background in DM mode.

6 dB/octave), a quite critical value for this span and speed. The sensitivity of lock-in amplifier was set to a high but unsaturated value of 5 mV. With those parameters the system still ran properly as demonstrated. From Fig. 3 it could be seen that, in DM mode the thermal BG has been suppressed effectively, all samples showed flat baselines. The PL signal of sample f inundated in the BG in RS mode emerged totally. The S/N in DM mode also improved obviously for the adoption of phase sensitive detection. For sample b with quite weak PL intensity the zoom in shows an increase of S/N from about 2.8 to 4.5, other samples with stronger PL intensities the S/N increased 3-4 times (for sample c from 62 to 200) experimentally. Notice that although from Figs. 2-5 all the y scales are in arbitrary units, their numerical value is quantitative for comparison. The signal intensity increased more than 2 orders in DM mode due to the introduction of lock-in amplifier; therefore detection threshold of FTIR (Ref. 16) was also improved.

For the detectors and lock-in amplifier the modulation frequency could be even higher (>100 kHz) facilely, whereas for this ordinary DPSS laser the modulation frequency was limited to <50 kHz experimentally. Therefore, if the spectral span extends to the shortwave side further, the scan speed still needs to slow down, say 2 times to 0.1571 cm/s. In this speed, the instrument behaved a little stagnant during presetting and data acquiring time increases somewhat, but all were still acceptable. At shortwave side, instead of thermal BG the stray

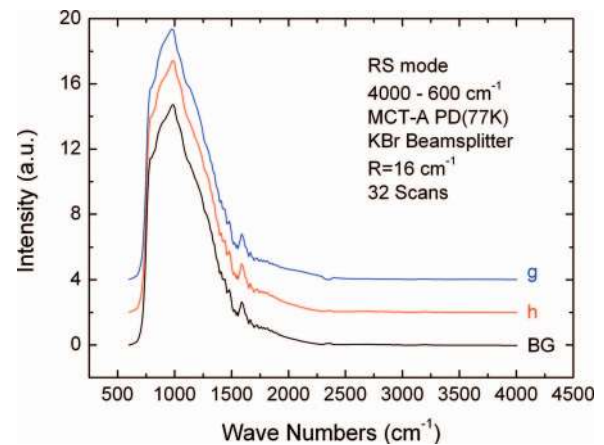


FIG. 4. Measured PL of samples g and h and background in RS mode.

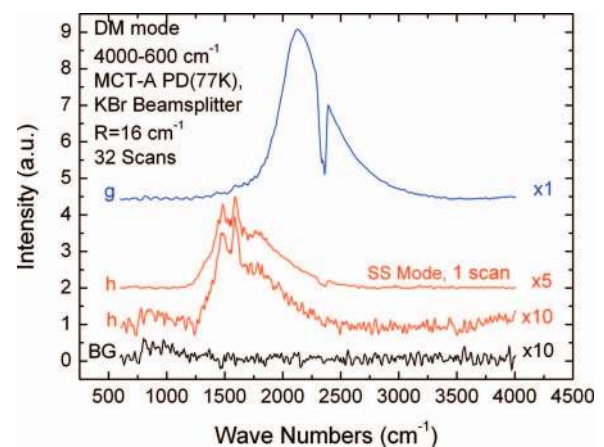


FIG. 5. Measured PL of samples g and h and background in RS mode. The measured PL of sample h in SS mode with one scan was also shown.

light BG might also play an important role. Stray light BG in visible-NIR bands could be from sunlight or illumination, and blocking this BG totally is really a hard work. Also, staying in a full dark room is uncomfortable for the operator. At shortwave side, the stray light BG may form fake signal or even inundating PL signal in RS mode for the more sensitive detectors at this band (e.g., InGaAs or Si), whereas in DM or SS mode with modulated excitation source this unmodulated BG could also be removed effectively (not shown).

Figures 4 and 5 showed the measured PL of the second group samples g and h in RS and DM modes, respectively, at spectral span of 4000-600 cm^{-1} of MIR band. The BG signals were also recorded. All the PL were still recorded at 16 cm^{-1} resolution with 32 scans at speed of 0.3165 cm/s, the acquiring time for each PL is also 25 s. In the measurements, the 532 nm DPSS excitation laser was modulated at about 32 kHz with average power of about 0.3 W. In this wavelength span, the Fourier frequency was limited to about 2.5 kHz, the time constant of lock-in amplifier was still set to 80 μs . In this case, the second modulation frequency is >10 times of the Fourier frequency. The sensitivity of lock-in amplifier was set to a high but unsaturated value of 50 mV. Sample g was a liquid phase epitaxial grown HgCdTe bulk layer in mid-wave infrared band of 3-5 μm , sample h was a MBE grown InSb pn structure with PL at its bandgap wavelength of about

TABLE II. Comparison of the data acquiring time of each mode at certain conditions.

4000–600 cm ⁻¹	RS mode (0.3165 cm/s)		DM mode (0.3165 cm/s)		SS mode	
	R = 8 cm ⁻¹	R = 16 cm ⁻¹	R = 8 cm ⁻¹	R = 16 cm ⁻¹	R = 8 cm ⁻¹	R = 16 cm ⁻¹
1 scan	<2 s	<1 s	<2 s	<1 s	12 m	7.4 m
32 scans	38 s	25 s	38 s	25 s	6.2 h	3.8 h

7.3 μm . Resolution of 16 cm^{-1} was just enough for most PL samples without fine spectral structure especially at RT. For a higher resolution of 8 cm^{-1} , the acquiring time increased to 38 s correspondingly. Normally with the increase of wavelength in long wavelength side of MIR the PL intensity decreases dramatically, observation of PL at RT at longer wavelength becomes more difficult. From Fig. 4 it could be seen that, in RS mode with MCT-A detector at MIR band, the whole RT thermal BG appeared with peak around about 1000 cm^{-1} . For samples g and h at RT, no noticeable PL signal could be found except the strong thermal BG. The recorded BG and “PL” curves were identical. Zoom in or subtracting the BG from the signal still resulted in no trace of the PL, implying that the PL of those samples were much weaker than the RT BG. By adopting DM mode, the PL signals were clearly extracted as shown in Fig. 5; the thermal BG was suppressed effectively. In this group of samples, the PL of sample g of a liquid phase epitaxial grown MCT material in mid-wave infrared was relatively “strong” with moderate S/N, in the curve a sharp absorption band of CO₂ around 2360 cm^{-1} still occurred clearly. Through the purge of the FTIR spectrometer with dry nitrogen, those strong CO₂ or H₂O related absorption bands could be weakened, whereas for this open-path FTIR-PL accessory they still could not be totally removed. Therefore, in case of those bands overlapped with useful PL information severely, a hermetic PL accessory box connected to the emission port of FTIR spectrometer should be developed air tight despite the inconvenience in operation. The PL intensity of sample h of a MBE grown InSb pn structure on GaAs substrate at RT was more than one order lower than sample g; however, in this case, the PL with some fine structures still could be differentiated clearly in DM mode but with lower S/N. Notice that for those type of samples in MIR the PL were only observed at low temperatures;^{1,6–12} therefore, the overall performance of DM mode using DPSS excitation laser modulated at appropriate frequency was favorable. Observing the PL of sample h and BG data in DM mode, a very weak residual BG hill (seems around the position of thermal BG) with its amplitude comparable to the noise floor could be noticed. We find that this noise hill was related to the electromagnetic interference between the adjunctive lock-in amplifier/laser driver and FTIR spectrometer. Through appropriate grounding this interference had been weakened effectively. In experiments, it is also noticed that, due to the introduction of second modulation frequency, some extra interference or noise at unpredictable wavenumber zone might be presented. Those interference or noise was most possibly caused by the weaker cross modulation components of the second modulation frequency with the Fourier signal frequency in the lock-in amplifier though it was set out off the Fourier frequency far away.

Through the fine adjustment of the excitation laser modulation frequency, those interference or noise could be removed or suppressed adequately. In experiments, the DPSS excitation laser modulation frequency was adjusted finely around the setting value to see the cleanness (free of interference peak or strong noise, without sample) of the monitor wavenumber window in presetting mode of the FTIR spectrometer, or the wavelength span was shifted/narrowed to keeping it away from interference peak or strong noise zone. In this case, the better S/N could be reached for the acquired data.

In Fig. 5, the PL of sample h in SS mode was also shown, these data were acquired still at resolution of 16 cm^{-1} but only in one scan. The DPSS laser was still modulated at the same frequency. From this trace it could be seen that, even using only one scan the S/N is better than that in DM mode. In SS mode, adopting this approach the excitation laser modulation frequency is more appropriate for the lock-in amplifier to reach lower noise, also the Fourier frequency is “zero” so have no possible cross interference with the excitation laser modulation frequency, and therefore a better S/N was achieved even at only one scan. However, the data acquiring time for one scan in SS mode at the same spectral span was 7.4 min, a still acceptable but much longer value than those of 32 scans in RS or DM modes. For 32 scans in SS mode, data acquiring time was increased to an unpractical value of 3.8 h. Table II compared the data acquiring time of each mode at certain conditions on this spectrometer.

It could be noticed that, the time consumption of SS mode was an obvious practical demerit. For FTIR-PL measurements at this lower resolution, at least several minutes should be needed in SS mode, this longer time period is not only a challenge to the stability of the system including laser and spectrometer, but also a perilous factor for data acquiring failure of catching a random strong electromagnetic interference from power line and so on, despite the patience of the operator. Depending on those situations, the SS mode could be an effective spare mode for FTIR-PL in this approach. That is, when a weak PL signal has been caught in DM mode but with poor S/N, switch the FTIR spectrometer to SS mode directly with a few scans that could be attempted to acquire better data. In all the RS, DM, and SS modes, the data acquiring time is related to the spectral span and resolution directly. For device characterization using emission port, such as the lasing spectrum of semiconductor laser, the highest resolution (0.125 cm^{-1} in this spectrometer) but limited spectral span was recommended commonly for differentiating its lasing modes and line shape in a narrower wavelength range. At this highest resolution, the repeatability of different scans and stability of device under test could not be guaranteed, therefore normally only one scan is used even in RS or DM modes.

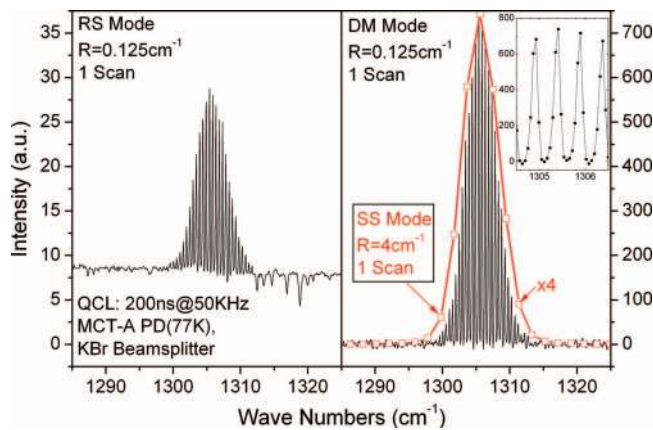


FIG. 6. Measured EL spectra of a room temperature quantum cascade laser around $7.7 \mu\text{m}$ in RS, DM, and SS modes. The inset was the zoom in of the spectrum in DM mode.

In SS mode, the highest resolution is normally lower than those in RS mode (0.5 cm^{-1} in this spectrometer) because of the oppressive data acquiring time at high resolution and servo precision of the spectrometer. If the highest resolution in SS mode was still enough for device characterization, the spectral span should be as narrow as possible for decreasing the total data acquiring time, in this case the SS mode still could be a useful spare mode.

As a complementary, EL spectra of a quantum cascade laser¹⁷ at room temperature around $7.7 \mu\text{m}$ were also measured in RS, DM, and SS modes, respectively, at the same driving condition, as shown in Fig. 6. This sample was driven electrically below its threshold using pulses of 200 ns duration at repeat frequency of 50 kHz. The cavity length was 3 mm. The EL intensity is normally much weaker than the lasing intensity; at MIR band it is often comparable to the thermal BG. As shown in the left side of Fig. 6, in RS mode at the highest resolution of 0.125 cm^{-1} of this instrument with one scan, the EL signal could be seen clearly but overlapped on the thermal BG, some absorption features of the water vapor on the thermal BG also appeared. The acquiring time for this spectrum was 15 s at scan speed of 0.9495 cm/s . In this case, the resolution was almost enough but the BG is really too high, therefore the data were unsuitable for extracting the gain/loss features of the cavity using Hakki-Paoli¹⁸ or modified methods. The right side of Fig. 6 showed the spectra in DM mode at the same acquiring time, the thermal BG was totally restrained and the data quality improved remarkably. This data with appropriate contrast as shown in the inset of Fig. 6 was good enough for extracting the gain/loss features. The EL spectrum was also measured using SS mode at 32 times lower resolution of 4 cm^{-1} with one scan as also shown in the right side, in this case the S/N was good but the acquiring time increased dramatically to 1268 s at the same wavelength span. Notice that the highest resolution in SS mode was limited to a lower value of 0.5 cm^{-1} in this instrument for the oppressive long acquiring time ($>2.5 \text{ h}$ at 0.5 cm^{-1} resolution, 100 cm^{-1} wavelength span with one scan), the chirp of the sample and random effect during this long acquiring time were uncontrollable, therefore the SS mode was unsuitable for acquiring fine spectral features.

IV. CONCLUSION

In conclusion, an improved FTIR spectroscopy approach for measurements of PL and EL in mid-infrared has been developed, in which DPSS laser was adopted as PL excitation source. In this approach, three different modes of RS, DM, and SS were software switchable without changing of the hardware or connections. The advantage and drawback of each mode were discussed in detail. Using this approach a group of samples extending from NIR to MIR with quite different PL intensities have been characterized to demonstrate the validity of the system. The weaker electroluminescence of quantum cascade lasers in mid-infrared band was also surveyed at different resolutions. Results show that for samples with relatively strong PL or EL out off the BG, RS mode is preferable. For weaker PL overlapped with BG, DM is the most effective mode. To get a better S/N when weaker PL or EL signal has been observed in DM mode, SS mode should be an advisable option despite its long data acquiring time and limited resolution.

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