Invited Article

Mid IR pulsed light source for laser ultrasonic testing of carbon-fiber-reinforced plastic

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Abstract
A quasi-phase-matched (QPM) optical parametric oscillator (OPO) was developed using a periodically poled Mg-doped stoichiometric LiTaO₃ crystal to generate mid-IR light for excitation of laser ultrasound in carbon fiber reinforced plastic (CFRP). The ultrasound generation efficiency was measured at the three different wavelengths that emanate from the OPO: 1.064 μm, 1.59/1.57 μm, and 3.23/3.30 μm. The measurements indicate that mid-IR 3.2–3.3 μm light generates the most efficient ultrasonic waves in CFRP with the least laser damage. We used mid-IR light in conjunction with a laser interferometer to demonstrate the detection of flaws/defects in CFRP such as the existence of air gaps that mimic delamination and voids in CFRP, and the inhomogeneous adhesion of CFRP material to a metal plate was also clearly detected.

Keywords: optical parametric oscillation, quasi-phase-matching frequency conversion, laser ultrasonic testing, carbon fiber reinforced plastics, MID-IR light source

1. Introduction

Carbon-fiber reinforced plastic (CFRP) is a unique construction material possessing a strength-to-weight ratio forty times that of iron. Its properties provide significant benefits to energy-saving, low-environmental-impact, and reduced-carbon-footprint applications which are in great demand worldwide. Moreover, its potential for a significant reduction in fuel consumption has driven demand for CFRP as a material for constructing major parts of aircraft, cars, boats, and ships. In particular, the first CFRP automobile is now on the market. In fields other than transportation, CFRP is being applied to components such as compressed gas cylinders (hydrogen tanks for fuel-cell vehicles and compressed natural gas [CNG] tanks for CNG vehicles) and wind-turbine blades.

CFRP fabrication is a multi-step process and quite unlike the fabrication of pure metal parts. There is a higher probability of a fabrication error in the making of CFRP. Therefore, from the viewpoint of the manufacturers, priority must
be given to assuring safety and reliability, while guaranteeing the quality of CFRP products. Defects at joints composed of similar CFRP/CFRP materials as well as joints between dissimilar materials [such as CFRP/aluminum or CFRP/titanium alloy] must be accurately detected so that reliability can be assured and product quality guaranteed.

Aimed at satisfying these requirements, various non-destructive evaluation technologies are being developed to diagnose CFRP materials. Each technology has its advantages and disadvantages, and the most suitable choice is based on diverse factors like the shapes and locations of the CFRP components, budget and delivery date, and required precision.

Among the various testing methods, a compact and robust method that is contactless, non-destructive, and can be applied remotely with sufficient spatial resolution would be very desirable. In this paper, laser ultrasonic testing (LUT) as an example of a non-destructive testing (NDT) method is being investigated. LUT generates an ultrasound pulse by irradiating the CFRP surface with a short pulse of laser radiation. The radiation is rapidly absorbed and converted into heat, which generates a pressure wave and an ultrasound pulse. For non-contact inspection, the ultrasound is optically detected with an interferometer, for contact inspection, a microphone could be used [1]. LUT combines the high flaw detectability of ultrasound with the multiple degrees of freedom of an optical system. When the region to be inspected is irradiated with a short-pulse laser, expansions and contractions are locally generated within the bulk of the material, and ultrasound waves propagate both on the surface and inside the volume of the material. If defects are present on the surface or inside the material, the sound propagation properties change, leading to a variation in the vibration amplitude and phase of the propagating waves. It is possible to extract information regarding the interior of the component if this change can be observed. Since LUT utilizes an optical system, a laser interferometer can be used to establish a completely contactless inspection system.

Current state-of-the-art LUT systems utilize legacy carbon-dioxide (CO$_2$) TEA (transverse excited, atmospheric pressure) gas lasers to induce ultrasound in CFRP. These lasers emit at 10.6 micron wavelengths, and, most likely, this light is absorbed by weak molecular vibrations in the IR spectrum of the epoxy matrix. These lasers can be difficult to maintain at their optimum performance level because of the need for a high-voltage excitation mechanism at high gas pressure and the deleterious effects of the high-pressure discharge on the molecular gas composition. These lasers are also large and heavy and can be difficult to maneuver on the shop floor. It is desirable to develop alternative wavelength sources using modern compact, reliable solid-state laser technology.

Recent work has shown that light with a wavelength in the vicinity of 3.2 $\mu$m (namely, close to the strong carbon–hydrogen stretching absorption band in epoxy resin) is suitable for ultrasound generation in various CFRP materials. Moreover, the efficiency and damage thresholds of LUT for CFRP have been reported [4]. A comparison of light sources was carried out using mainly Nd:YAG and CO$_2$ gas lasers[4]. To the best of our knowledge, there is no commercial pulsed solid-state light source in the vicinity of 3.0–3.5 $\mu$m that is suitable for applying LUT to CFRP. We need a mid-IR laser with enough pulse energy (>10 mJ), appropriate pulse width (10–35 ns), relatively high repetition rate (>100 Hz), and portability, with easy handling. In other words, we need a scalable, high average power mid-IR source for LUT, and currently, we have assessed other solid state mid-IR lasers and have determined that they are not sufficiently mature to achieve these desired specifications in the near term.

At NIMS, we are developing a technology that incorporates an efficient wavelength conversion device for generating mid-IR light by pumping an optical parametric oscillator (OPO) using compact Nd:YAG solid-state lasers. These lasers are the most reliable, most well-developed solid-state pump lasers currently available and are ideally suited for LUT applications. We have already produced quasi-phase matching (QPM) wavelength-conversion devices fabricated from a ferroelectric material, stoichiometric lithium tantalate (LiTaO$_3$, hereafter abbreviated as SLT), by means of the periodic poling (PP) technique. The SLT coercive field (the external electric field necessary for the polarization inversion) can be decreased by more than one order of magnitude compared with conventional LiTaO$_3$ crystals by controlling the defect density during crystal growth [5, 6]. This decrease in the coercive field has made it possible to fabricate PPSLT devices [7–9] with apertures as large as 5 mm × 5 mm. PPSLT crystals are also resistant to optical laser damage [10]. Both a wide aperture and damage resistance are prerequisites for a wavelength convertor in any mid-IR LUT application. Compact and robust mid-infrared laser light sources in the vicinity of 3.2 $\mu$m can be realized by combining PPSLT convertor technology with a solid-state Nd:YAG laser as a pump. The reason why PPMgSLT was adopted as a nonlinear material for the OPO laser instead of another periodically poled crystal is based on the demonstrated large aperture capability (as large as 5 mm × 5 mm) needed for high energy per pulse applications and the substantially high laser damage threshold compared with PPLN,PPRTA, PKTP, etc.

In this paper, we describe the system and performance of a mid-IR PPSLT OPO as well as the results of LUT of CFRP material using light generated by the OPO.

2. Experimental methods

2.1. Nonlinear optical material and device for OPO

Figure 1 shows a PPSLT OPO using periodically poled MgSLT (1% MgO doped SLT) placed between two plano–...
plano end mirrors. The QPM grating pitch was designed using equations (1) and (2) below, which describe the energy conservation and phase matching conditions in a QPM parametric convertor device:

\[
\frac{1}{\lambda_p} = \frac{1}{\lambda_s} + \frac{1}{\lambda_i}
\]  
(1)

\[
\frac{1}{\lambda} = \frac{n_p(T)}{\lambda_p} - \frac{n_s(T)}{\lambda_s} - \frac{n_i(T)}{\lambda_i}
\]  
(2)

where \(\lambda_p\), \(\lambda_s\), \(\lambda_i\), and \(\Lambda\) represent the pump wavelength, signal wavelength, idler wavelength, and QPM grating pitch, respectively. \(n_p(T)\), \(n_s(T)\), and \(n_i(T)\) represent the temperature-dependent refractive indices at the pump, signal, and idler. Figure 2 shows the solutions to equations (1) and (2) using the published Sellmeier dispersion equation for SLT [11].

We designed and fabricated two QPM devices with grating pitches of 30.9 \(\mu\)m and 31.1 \(\mu\)m, which correspond to expected idler wavelengths of 3.30 \(\mu\)m and 3.23 \(\mu\)m at 30°C. The lengths and apertures of these devices were the same (35 mm and 3 mm \(\times\) 3 mm).

### 2.2. OPO set-up

Figure 3 is a schematic diagram of the experimental OPO-LUT system. Two Q-switched multimode Nd:YAG lasers were used as the 1.064 \(\mu\)m pump source. A laser beam was selected by using beam switching optics from either of two laser systems: (a) Quantel/Centurion: designated as Laser-A hereafter and (b) Quantel/Ultra: Laser-B, i.e. We used pump laser pulses with a maximum energy of 40 mJ, a pulse length of 7.8 ns (Laser-A)/9.0 ns (Laser-B) FWHM, and a repetition rate of 20 Hz. The beam had a slightly elliptical top-hat shape with a beam quality measure of \(M^2 \sim 15\). The pump energy was controlled using a half-wave plate and polarizer, located upstream of the OPO, so that the pulse profile remained independent of the pump energy. The input pump wave was a collimated beam with a diameter of \(\sim 2.5 \text{ mm} (1/e^2)\) at the OPO resonator. The polarization of the pump laser was set parallel to the \(z\) axis of the PPMgSLT device.

The OPO resonator was a linear cavity with a mirror separation of 100–160 mm. The input and output couplers were coated plane mirrors with the same dielectric coating: high reflectivity at the signal wavelength (1.5–1.6 \(\mu\)m), high transmission at the pump wavelength, and high transmission at the idler wavelength (3.1–3.5 \(\mu\)m). When the signal wavelength was required, the output coupler was replaced with a mirror coated with \(R \sim 95\%\) at the signal wavelength. The signal and idler waves were first separated from the output light by using a pump-cut filter; then, the idler and signal were separated from each other by a dichroic filter. The idler and signal energy were measured using a power meter with a thermal sensor (Ophir 3A-P). The idler wavelength was measured with a spectrometer consisting of a grating and a pyroelectric sensor array.

### 2.3. LUT experiments

The laser ultrasonic testing system (schematic layout in figure 3) consisted of a pulsed laser for ultrasound generation, a laser Doppler velocimeter (LDV) for ultrasonic measurements, and a sample scanning XY stage.

Three light sources were used for ultrasound generation: a Laser-B (Quantel/Ultra) Nd:YAG laser (1.064 \(\mu\)m), a PPSLT OPO signal wave (1.59/1.57 \(\mu\)m) and a PPSLT OPO idler wave (3.23/3.30 \(\mu\)m). It should be noted that two OPO outputs with different wavelength pairs (1.59 \(\mu\)m and 1.57 \(\mu\)m for the signal wave, 3.23 \(\mu\)m and 3.30 \(\mu\)m for the idler wave) were generated from two separate PPSLT QPM gratings (31.1 \(\mu\)m and 30.9 \(\mu\)m, respectively, and at 30°C). This small wavelength difference has a negligible effect on the generated LUT signals based on the relatively structureless, broad CFRP absorption band. The laser pulse selected by the beam switching device was directed to the surface of the CFP sample through a lens with a beam diameter of 1.0 mm. The laser pulse interacted with the sample surface to induce an ultrasonic pulse that propagated into the sample. Laminated CFRP plates 1.1 mm thick (O-KEI Resin Co. Ltd, Japan) were used for this investigation. It should be noted that commercially available CFRP plates were produced based on lamination of many thin prepreg sheets with thicknesses in the range of 0.02–0.5 mm. These specially made CFRP plates were carefully produced without any known defects, and, therefore, our LUT was not affected by this process.

A commercial LDV system was used as an ultrasonic wave sensor. The system consists of mainly two major parts: (a) a sensor unit (Graphitec AT0023) which includes a helium neon (HeNe) laser light source and a built-in autofocal lens, and (b) an optical heterodyne interferometric signal demodulation device (Thamway T1101077A). The HeNe probe beam from the sensor unit was focused onto the CFRP sample with a beam diameter of around 60 \(\mu\)m (1/e^2 intensity). The probe laser sensing beam and the ultrasound generation beam were aligned to counterpropagate with respect to each other; these two counterpropagating beams were then carefully aligned to overlap each other on the target material. The probe
laser beam reflects off the CFRP surface, is modulated by the ultrasonic soundwave and subsequently interferes with the incident probe beam inside the sensing unit, producing an optical heterodyne interferometric signal. The interferometric signal was then processed and converted to an ultrasonic velocity signal. The ultrasonic velocity signal was analyzed using a 500 MHz oscilloscope (LeCroy). Signal averaging was performed to reduce random noise. We used three scanning methods:

1. A-scan: The temporal response of the ultrasonic signal at a fixed sample point.
2. B-scan: Spatial one-dimensional scanning of the temporal response graphically displayed on a 2D map.
3. C-scan: Spatial two-dimensional scanning of ultrasonic intensity graphically displayed on a 3D map.

3. Results and discussion

3.1. OPO performance

The OPO performance was usually obtained using Laser-A (Quantel/Centurion) as a pump. We found no differences in OPO performance when Laser-B (Quantel/Ultra) was used as a pump. Figure 4 compares the theoretical and experimental temperature dependences of the idler wavelength output from the PPSLT OPO. The straight line and the dashed line represent the theoretical results for QPM grating pitches of 30.9 μm and 31.1 μm, respectively. The squares and circles represent the measured data for the samples with grating pitches of 30.9 μm, and 31.1 μm, respectively, and it is clear that they are in good agreement with theory.

The $M^2$ of the idler light was estimated to be in the range 15–20 by measuring the propagation of the focused idler beam using a Gaussian beam fitting routine. The idler $M^2$ parameter is strongly dependent on the $M^2$ of the pump wave.

Figure 5 shows the dependence of the output energy for the 3.23 μm and 3.30 μm idler waves (QPM grating pitches of 31.1 μm and 30.9 μm, respectively) on the input pump energy (input–output property) measured at 30 °C. An OPO
threshold energy of \( \sim 5 \) mJ and \( \sim 5 \) mJ for the idler output was obtained for a 35 mJ pump input. Figure 6 shows the dependence of the output energy of idler waves (QPM grating pitches of 31.1 \( \mu \)m) on the input pump energy measured at 50, 100 and 150 \( ^\circ \)C.

3.2. LUT performance

As we previously explained, the LUT performance was characterized using the OPO pumped by Laser-B (Quantel/ Ultra). Figure 7 illustrates a typical temporal ultrasonic waveform (A-scan) measured on a CFRP sample excited by a laser beam with a wavelength of 3.23 \( \mu \)m. A strong negative peak at 0.57 \( \mu \)s from the onset of the exciting laser pulse corresponds to the ultrasonic velocity signal generated at a point on one surface of the CFRP and that propagated on the other surface.

To characterize the ultrasound intensity generated by laser irradiation, we define a parameter (param1) by the following formula (3).

\[
\text{Param1} = -\int_{T_1}^{T_2} V(t) dt / (T_2 - T_1)
\]

Here, \( V(t) \) represents the temporal ultrasonic velocity signal shown in figure 7. \( T_1 \) and \( T_2 \) represent the integration interval endpoints, and in this experiment, \( T_1 = 0.5 \) \( \mu \)s and \( T_2 = 0.65 \) \( \mu \)s. Param1 represents the average voltage of the ultrasonic velocity signal in the integration period.

2D maps of Param1 values were obtained by scanning an area of 10 mm \( \times \) 10 mm of the sample using the ultrasound generation laser at 3.30 \( \mu \)m. The resultant C-scan is shown in figure 8; the bidirectional periodic structure with a cycle of around 2 mm in either direction represents the bidirectional weave of the carbon fibers inside the epoxy matrix (see figure 9). Comparing the C-scan mapping and the CFRP
surface, we can see that more intense ultrasound is generated at parts where the epoxy is exposed on the surface. This result is consistent with the fact that the mid-infrared light is strongly absorbed by the C-H stretching vibration in the \(-\text{CH}_2-(\text{O}-\text{CH}_3)\)– linkage of the epoxy polymer [12].

The thermal expansion of the epoxy resin is considerably larger [13] than the thermal expansion of the carbon fiber, which is close to zero or slightly negative [14]. As a result, very little ultrasound was generated where the carbon fibers were exposed on the surface. Note that the enhancement of ultrasound generation in the epoxy layer was simulated in [15].

Next, we measured the laser wavelength dependence of the ultrasound generation efficiency in CFRP. To reduce the influence of the structure-dependent generation described above, the ultrasound was generated by scanning a 20 mm length with a pitch of 0.2 mm (B-scan), and the ultrasound parameters (param1) were then averaged over the entire region. The comparison was done using the same area of CFRP. For each wavelength, the laser pulse energy and the beam diameter were held constant at 0.2 mJ and 1.0 mm, respectively. Figure 10 shows the variation of the peak ultrasound intensity along the scanning distance with wavelength as a parameter. The figure shows that 3.23 \(\mu\)m wavelength has a stronger capability of ultrasonic generation than 1.064 \(\mu\)m (pump) light and 1.57 \(\mu\)m (signal) light.

A laser scanning experiment was carried out in order to assess the laser’s damage to a rough CFRP surface. The ultrasound generating laser was scanned in an area measuring 3 mm \(\times\) 3 mm with a step of 0.5 mm (total of 7 \(\times\) 7 spots). The irradiation time for each spot was 10 s. The laser energy in the irradiated areas was varied from 0.8 mJ to 2.4 mJ. Figure 11(a) shows photographs of the CFRP sample surface after laser irradiation. Figure 11(b) shows microscope images of the damaged areas. These images were taken at the center of a 2 mm \(\times\) 2 mm area of the CFRP surface. Most of the laser damage occurred in areas where the carbon fibers were exposed on the surface.

We can see that the tolerance to damage is highest with the mid-infrared laser at a 3.3 \(\mu\)m wavelength. In particular, this wavelength produces no obvious damage at 0.8 mJ and clearly less damage at higher intensities such as 2.4 mJ. A possible explanation for the reason why tolerance to damage is highest with the mid-infrared laser may come from its lowest photon energy. Irradiation of higher photon energy tends to shift to ablation mode easily.

### 3.3. LUT applications for CFRP

The above results confirm the potential for mid-infrared laser induced ultrasound generation in CFRP. Next, we demonstrate two applications of mid-infrared lasers to LUT detection of flaws in CFRP. The first application is detection of voids. We fabricated a sample of CFRP with a void inside it by bonding two CFRP plates with a thickness of 1.1 mm and with an un-bonded area of 7 mm diameter, as illustrated in figure 12(a). The results of the LUT C-scan are shown in figure 12(b). A nearly null signal can be clearly seen, and it corresponds to the location and size of the void.

Figure 13 shows a second application of an LUT C-scan for detecting inhomogeneous adhesion between CFRP and an aluminum plate. Figure 13(a) shows a photograph displaying the inhomogeneous distribution of an adhesive bond (glue) on top of a CFRP plate to which an aluminum plate was bonded. The region where the adhesive bond is missing corresponds to the blue-colored region with a near null signal in the C-scan shown in figure 13(b).
Here, the He-Ne laser sensing beam and the ultrasound generation beam were aligned to counter-propagate with respect to each other. Therefore, the existence of air-gaps, mimicking delamination and voids where the ultrasound cannot penetrate, can be clearly detected.

4. Summary

A QPM OPO using periodically poled MgSLT (MgO 1% doped near-stoichiometric LiTaO₃) was demonstrated. The OPO, pumped by a 1.064 μm Q-switched Nd:YAG laser, was tuned to the wavelength region 3.2–3.3 μm. This is the most promising regime for LUT of CFRP. An experiment was conducted to evaluate the ultrasound generation efficiencies of laser pulses at three different wavelengths (1.064 μm, 1.59/1.57 μm, and 3.23/3.30 μm) that are emitted from the OPO system. We found that the mid-IR laser at a 3.2–3.3 μm wavelength generated the most efficient and least damaging ultrasonic waves in CFRP.

We used mid-IR light in conjunction with a laser interferometer to demonstrate the detection of flaws/defects in CFRP such as the existence of air-gaps that mimic delamination and voids in CFRP, and the inhomogeneous adhesion of CFRP material to a metal plate was also clearly detected.

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References

[1] Green R Jr (ed) 1997 See for example papers, Proc. of the Eighth Int. Symp. on Nondestructive Characterization of Materials: Nondestructive characterization of materials VIII (Boulder, Colorado, June)
[2] Dubois M, Lorraine P W, Filkins R J and Drake T E 2001 Appl. Phys. Lett. 79 1813
[3] Dubois M, Lorraine P W, Filkins R J, Drake T E, Yawn K R and Chuang S-Y 2002 Ultrasonics 40 809
[4] Edwards C, Stratoudaki T, Dixon S and Palmer S 2001 IEEE Proc.-Sci. Meas. Technol. 148 139

[5] Kitamura K, Furukawa Y, Niwa K, Gopalan V and Mitchell T E 1998 Appl. Phys. Lett. 73 3073

[6] Furukawa Y, Kitamura K, Suzuki E and Niwa K 1999 J. Crystal Growth 197 889

[7] Hatanaka H, Nakamura K, Taniuchi T, Ito H, Furukawa Y and Kitamura K 2000 Opt. Lett. 25 651

[8] Kitamura K, Furukawa Y, Takekawa S, Ito H and Gopalan V 2001 Ferroelectrics 253 462

[9] Yu N-E, Kurimura S, Nomura Y, Kitamura K and Tetsumi Y 2004 Appl. Phys. Lett. 85 5134

[10] Louchev O, Hatano H, Wada S and Kitamura K 2013 Appl. Phys. Lett. 103 091114

[11] Lim H H, Kurimura S, Katagai T and Shoji I 2013 Japanese J. Appl. Phys. 52 032601

[12] Nikolic G, Zlatkovic S, Cakic M, Cakic S, Lacnjevac C and Rajic Z 2010 Sensors 10 684

[13] see www.engineeringtoolbox.com/linear-expansion-coefficients-d_95.html

[14] see www.christinedemerchant.com/carboncharacteristics.html

[15] Dubois M, Enguehard F and Bertrand L 1995 Review of Progress in Quantitative Nondestructive Evaluation (New York: Plenum Press) pp 529