

# Giant Momentum Coupling Coefficients from Nanoscale Laser-initiated Exothermic Compounds

Claude R. Phipps\* and James R. Luke†  
*Photonic Associates, LLC, Santa Fe, NM 87508*

Wesley D. Helgeson‡  
*NMT/IERA, Albuquerque, NM 87106*

Darren Naud and Michael Hiskey§  
*Los Alamos National Laboratory, Los Alamos, NM 87545*

Lukas Urech\*\*, Thomas Lippert†† and Alexander Wokaun‡‡  
*Paul Scherrer Institut, Villigen PSI, Switzerland*

**We report here on results obtained with laser-initiated micro-propellants, such as PVN, PVC, GAP, NC, and mixtures of these. All samples were doped with a laser absorbing component. In some cases, this was carbon nanopearls with 10nm mean diameter, while, in others, it was a carbon-based ink with  $\mu\text{m}$ -size particles. We also report results of performance tests for absorbers tuned to the 935-nm laser wavelength.**

## Nomenclature

|                     |                                              |
|---------------------|----------------------------------------------|
| $C_m$               | = laser momentum coupling coefficient        |
| CA                  | = cellulose acetate                          |
| E                   | = short for “10 <sup>^</sup> ”               |
| EFL                 | = effective focal length                     |
| eV                  | = electron-volt                              |
| F                   | = thrust                                     |
| FEP                 | = fluoroethylene polymer                     |
| $g_o$               | = acceleration of gravity at Earth’s surface |
| GAP                 | = glycidyl azide polymer                     |
| I                   | = laser intensity on target                  |
| $I_p$               | = plasma formation threshold intensity       |
| $I_{sp}$            | = specific impulse                           |
| K                   | = DuPont Kapton™ polyimide resin             |
| NC                  | = nitrocellulose                             |
| $\langle P \rangle$ | = average incident laser power               |

---

\* President and General Partner, 200A Ojo de la Vaca Road, [crhipps@aol.com](mailto:crhipps@aol.com). Member AIAA.

† Senior Research Staff Member and Partner, 200A Ojo de la Vaca Road.

‡ Senior Research Associate, 901 University Blvd SE.

§ Research Staff Member, P.O. Box 1663.

\*\* Research Staff Member, Materials Group

†† Head, Materials Group

‡‡ Head, General Energy Research Department; also Professor, Chemistry Department, ETH Zürich

Approved for public release; distribution unlimited.

| Report Documentation Page                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |                                    |                                     |                            | Form Approved<br>OMB No. 0704-0188             |                                 |
|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------|-------------------------------------|----------------------------|------------------------------------------------|---------------------------------|
| Public reporting burden for the collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to a penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number. |                                    |                                     |                            |                                                |                                 |
| 1. REPORT DATE<br><b>APR 2005</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      |                                    | 2. REPORT TYPE                      |                            | 3. DATES COVERED<br>-                          |                                 |
| 4. TITLE AND SUBTITLE<br><b>Giant Momentum Coupling Coefficients from Nanoscale Laser-initiated Exothermic Compounds</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |                                    |                                     |                            | 5a. CONTRACT NUMBER<br><b>FA9300-04-C-0030</b> |                                 |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |                                    |                                     |                            | 5b. GRANT NUMBER                               |                                 |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |                                    |                                     |                            | 5c. PROGRAM ELEMENT NUMBER                     |                                 |
| 6. AUTHOR(S)<br><b>Claude Phipps; James Luke; Wesley Helgeson; Darren Naud; Michael Hiskey</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         |                                    |                                     |                            | 5d. PROJECT NUMBER<br><b>BMSB</b>              |                                 |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |                                    |                                     |                            | 5e. TASK NUMBER<br><b>R3LA</b>                 |                                 |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |                                    |                                     |                            | 5f. WORK UNIT NUMBER                           |                                 |
| 7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)<br><b>Photonic Associates, LLC, 200 Ojo de la Vaca Road, Santa Fe, NM, 87508</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |                                    |                                     |                            | 8. PERFORMING ORGANIZATION REPORT NUMBER       |                                 |
| 9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                |                                    |                                     |                            | 10. SPONSOR/MONITOR'S ACRONYM(S)               |                                 |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |                                    |                                     |                            | 11. SPONSOR/MONITOR'S REPORT NUMBER(S)         |                                 |
| 12. DISTRIBUTION/AVAILABILITY STATEMENT<br><b>Approved for public release; distribution unlimited</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  |                                    |                                     |                            |                                                |                                 |
| 13. SUPPLEMENTARY NOTES                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                |                                    |                                     |                            |                                                |                                 |
| 14. ABSTRACT<br><b>We report here on results obtained with laser-initiated micro-propellants, such as PVN PVC, GAP, NC, and mixtures of these. All samples were doped with a laser absorbing component. In some cases, this was carbon nanopearls with 10nm mean diameter, while, in others, it was a carbon-based ink with µm-size particles. We also report results of performance tests for absorbers tuned to the 935-nm laser wavelength.</b>                                                                                                                                                                                                                                                                                                                                                                                                     |                                    |                                     |                            |                                                |                                 |
| 15. SUBJECT TERMS                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      |                                    |                                     |                            |                                                |                                 |
| 16. SECURITY CLASSIFICATION OF:                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |                                    |                                     | 17. LIMITATION OF ABSTRACT | 18. NUMBER OF PAGES<br><b>5</b>                | 19a. NAME OF RESPONSIBLE PERSON |
| a. REPORT<br><b>unclassified</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | b. ABSTRACT<br><b>unclassified</b> | c. THIS PAGE<br><b>unclassified</b> |                            |                                                |                                 |

|             |                                              |
|-------------|----------------------------------------------|
| $PVC$       | = polyvinyl chloride                         |
| $PVN$       | = polyvinyl nitrate                          |
| $Q^*$       | = specific ablation energy                   |
| $T$         | = temperature                                |
| $v_E$       | = exhaust velocity                           |
| $W$         | = laser pulse energy incident on test sample |
| $\Delta m$  | = ablated mass                               |
| $\eta_{AB}$ | = ablation efficiency                        |
| $\tau$      | = laser pulse duration                       |

## I. Introduction

The maximum specific impulse available from chemical reactions is about 500s, limited by the heat of formation. For example, the heat of formation for hydrogen-oxygen combustion, one of the hottest chemical reactions, is 57.8 kcal/mole<sup>1</sup>. This translates to a reaction temperature of 2.91E4K, for which the most probable thermal velocity  $(2kT/m_E)^{0.5} = 5.17$  km/s, or  $I_{sp} = 527$  s. However, for example, the Space Shuttle's main engine can produce only 465 s ( $v_E = 4.56$  km/s), because of inefficiencies inherent in the engine<sup>2</sup>. This limit is fundamental, and will not change with improvements in engine materials. Larger  $I_{sp}$  values ( $v_E \gg 5$  km/s) are accessible only by laser ablation, where temperatures can be more than 1E6K for ns-duration pulses, or some other non-chemical process such as ion drives. Specific impulse  $I_{sp}$  up to 7600s has been measured<sup>3</sup> with 20ns-duration pulses from an ordinary KrF laser illuminating Al. This  $I_{sp}$  corresponds to  $T = 9.1E6$  K (780 eV). By combining these two effects, i.e., not only initiating the reaction, but adding significant energy with very intense pulses of laser illumination, the potential exists for attaining combinations of  $C_m$  and  $I_{sp}$  in which  $I_{sp}$  still exceeds what is available from the chemistry alone, and in which  $\eta_{AB} > 1$ . In the work reported here, which deals with ms-duration laser pulses at the modest intensities available from multi-mode diode lasers, we do not expect to see  $I_{sp} > 500$ s, but do expect  $\eta_{AB} > 1$  and the distinct advantage of programmable, discrete target addressability.

## II. Definition of Terms

For the sake of simplicity, we will consider a monoenergetic laser-produced plasma jet with exhaust velocity  $v_E$ . We have shown<sup>3,4</sup> that this approximation will not introduce large errors [ $\langle v^2 \rangle / \langle v \rangle^2 \approx 1.15$ ] for typical laser-produced plasma jets, and the principal points we want to make here will be made more transparently using that assumption. We define  $C_m$  as follows:

$$C_m = F / \langle P \rangle = \Delta m v_E / W. \quad (1)$$

In the ablation process,  $Q^*$  joules of laser light are consumed per kg of ablated mass, and  $C_m$  units of impulse  $\Delta m v_E$  are produced. The product of  $C_m$  and  $Q^*$  is the exhaust velocity  $v_E$  of the ablation stream, given the monoenergetic assumption. This can be seen from the definitions of  $C_m$  and  $Q^*$ :

$$C_m Q^* = (\Delta m v_E / W)(W / \Delta m) = v_E \quad (2)$$

independent of the ablation efficiency. If for example, a significant amount of the incident energy is absorbed as heat in the target substrate rather than producing material ejection,  $Q^*$  will be higher, but  $C_m$  will be proportionately lower, giving the same velocity in the end. In propulsion work, "specific impulse"  $I_{sp}$  is customary notation for  $v_E / g_0$ . In this work, we define  $I_{sp}$  as

$$I_{sp} = C_m Q^* / g_0 \quad (3)$$

Energy conservation prevents  $C_m$  and  $I_{sp}$  from being arbitrary. Increasing one decreases the other. Energy conservation requires that several constant product relationships exist:

$$2\eta_{AB} = \Delta m v_E^2 / W = C_m^2 Q^* = g_o C_m I_{sp} = C_m v_E. \quad (4)$$

In Eq. (3), we introduce the ablation efficiency parameter,  $\eta_{AB}$ , the ratio of exhaust kinetic energy to incident laser energy  $W$ . The definition allows  $\eta_{AB} > 1$  for exothermic targets.

For nonexothermic targets,  $\eta_{AB} \leq 1$ , and the product

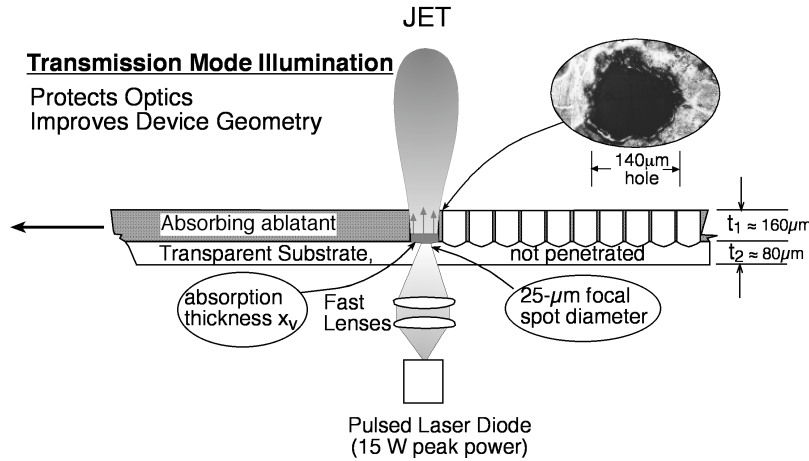
$$C_m I_{sp} \leq 2/g_o = 0.204. \quad (5)$$

Plasma formation threshold is approximately given by<sup>5</sup>

$$I_p = 240/\tau^{0.55} \text{ MW/m}^2. \quad (6)$$

Eq. (6) is a useful guide valid to within factors of 2 for ms to sub-ns pulse durations, for all surface absorbing materials in vacuum, and for visible to infrared wavelengths. It is also a useful guide for predicting plasma initiation in confined ablation.

### III. Target Preparation



**Figure 1. Schematic of Target Tapes.** Each laser pulse passes through the transparent substrate and creates a detonation which begins at the interface between it and an absorbing ablatant layer. The detonation expands to produce the jet. The transparent substrate is not physically penetrated, although some of it may also ablate.

Ablation targets for tests were made in a tape format using a variety of transparent supporting substrates and ablative coatings [Fig.1]. Substrates were chosen for transparency, low outgassing and high optical damage resistance. We found that these requirements were often difficult to satisfy simultaneously. For example, Kapton™ polyimide resin has excellent toughness and outgassing properties, but lower optical damage resistance and adhesivity for some target materials, compared to cellulose acetate. The polyimide has lower damage resistance because of its greater absorptivity at our 935-nm operating wavelength. Among the materials considered for substrates, only cellulose acetate and FEP were excellent in regard to optical damage

resistance. However FEP had the added problem of very poor adhesion to most coatings other than vapor-deposited aluminum, so we settled on cellulose acetate as the best substrates for general use, and polyimide for cases in which the coating application process involves solvents not tolerated by cellulose acetate. Tape substrate thickness ranged from 75 to 125 μm. Coatings ranged from 60 μm to 370 μm.

Coating materials were mixed with a Cole-Parmer A-04737 homogenizer.

Ablative coatings for tapes were usually applied as a spray of the coating material diluted in, e.g., n-butyl acetate, in increments of 5 μm, to attain an ideal total thickness of order 70 μm. In some cases, coatings were applied in one go using a draw-blade applicator.

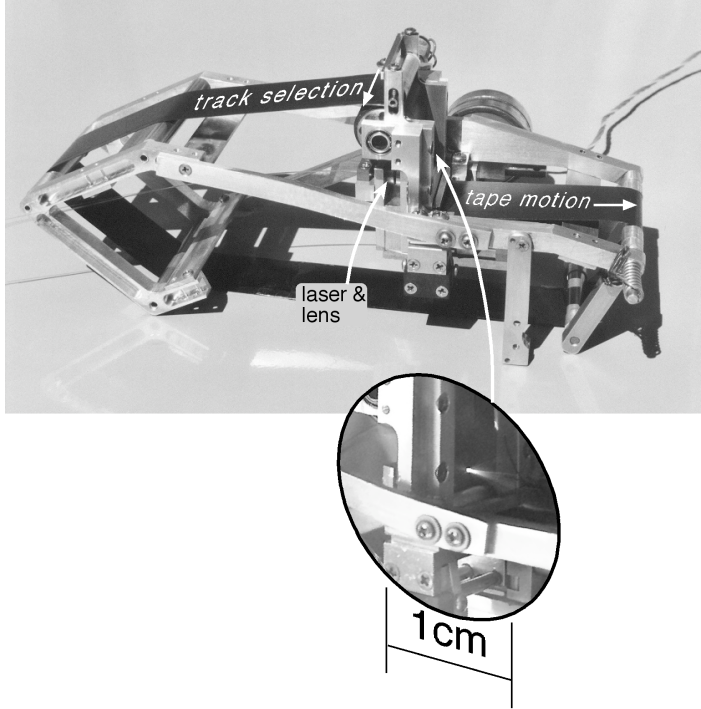
Laser ablatant materials and substrates are listed in Table 1. All proportions are by weight. “Ink” in the Table is the black material dissolved from commercial electric typewriter foil ribbon.

## IV. Experimental Setup

### A. Ablator Test Device

Figure 2 shows the device used to illuminate the target tapes.

One to four repetitively-pulsed, fiber-coupled JDSU type 6380-A/L2 laser diodes inside the device drive the ablation of the fuel tape. Each of these are capable of  $\langle P \rangle = 2.5\text{W}$  output at 910-920 nm wavelength with input current and voltage of 3.37A and 1.71V, giving 43% electrical efficiency at this output level. At 1ms pulse duration, we determined that these could achieve peak delivered power output of 4.75W at 10% duty cycle without damage.



**Figure 2. Ablation tape test unit.** The inset shows the laser-produced plasma jet.

The ends of the four 100- $\mu\text{m}$ -core-diameter fibers were stripped of their buffer layers for closest spacing, and mounted in a fiber vise. Their light output was collimated with a Lightpath, Inc. GPX5-5 axially graded refractive index aspheric lens and focused with a Thorlabs C330-TM-B aspheric. The EFL's of the two lenses are 5.16 mm and 3.10 mm, respectively. Both lenses are AR-coated. Intensity on target is about  $30\text{GW}/\text{m}^2$  when 15W peak are incident from all four lasers. A stepper motor drives the closed-loop fuel tape in the longitudinal direction at speeds of 5 to 20 mm/s. A second stepper motor slowly advances the beam delivery system (fiber ends and two lenses) across the tape at a rate of one track width per revolution of the tape loop, resulting in a continuous helical path that eventually accesses the entire tape.

### B. Micronewton Test Stand

The test unit was mounted to one end of a counterbalanced bar suspended by a 250- $\mu\text{m}$  steel fiber inside a vacuum chamber<sup>6</sup>. Thrust  $F$  generated by the test unit was determined by measuring the rotation of the bar. Ablation mass loss  $\Delta m$  was determined by weighing the

ablation test tape before and after a test run using a microgram Mettler balance. Ambient pressure during tests was typically 50 $\mu\text{torr}$ .

To eliminate external torsion inputs to the thrust measurement, power is supplied to the testbed thruster by onboard Li-ion batteries (which, with electronics, constitute the counterbalance), and test unit operation is controlled by an external computer via an optical link.

Since the publication of reference 6, these measures have improved the resolution of our torsion pendulum to  $\pm 2\mu\text{N}$  [Figure 3].

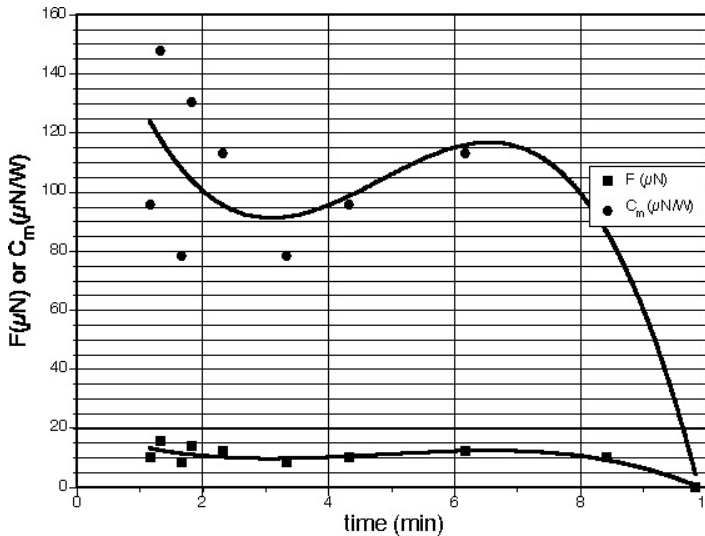
## V. Results

Results we obtained are given in Table 2.

## VI. Conclusion

Our results showed the largest coupling coefficient  $C_m$  ever reported for some target materials, almost  $2\text{mN}/\text{W}$ .

As expected from its definition,  $\eta_{AB} > 1$  for the GAP test samples, but much lower for the less exothermic materials.



**Figure 3. Very low thrust data demonstrates precision of our  $\mu\text{N}$  thrust stand.** The scatter is not due to measurement error, but rather the periodic motion of the torsion balance. The solid lines are a third-degree polynomial fit to the data. American Institute of Aeronautics and Astronautics thrust stand with a simple probe-laser readout is  $\pm 2\mu\text{N}$ . Even better precision has been demonstrated with an interferometric readout and will be reported in a subsequent paper

**Table 1. Laser Ablatant Materials**

| Ablator | Absorber                       | Substrate |
|---------|--------------------------------|-----------|
| PVN     | 10% carbon nanopearls          | CA        |
| PVC     | 5% ink                         | CA        |
| GAP     | carbon nanopearls              | K         |
| GAP     | 5% ink                         | K         |
| GAP     | 1% Epolin 2057 935-nm absorber | K         |
| PVN     | 1% Epolin 2057 935-nm absorber | K         |
| NC      | 2% carbon nanopearls           | CA        |

The specific impulse values in the table, computed from measured mass loss of the ablated tapes, correspond to exhaust velocities up to 2.5km/s. These values do not exceed what is available from chemistry. The advantage of using the combination of laser and chemical energy in the present work is discrete, programmable thrust output.

We reported  $\pm 2\mu\text{N}$  sensitivity in our torsion-balance-based thrust

**Table 2. Results of Measurements**

| Ablator                   | I(MW/m <sup>2</sup> ) | $\tau$ (ms) | C <sub>m</sub> ( $\mu\text{N/W}$ ) | F( $\mu\text{N}$ ) | I <sub>sp</sub> (s) | $\eta_{\text{AB}}$ (%) |
|---------------------------|-----------------------|-------------|------------------------------------|--------------------|---------------------|------------------------|
| PVN/10% carbon nanopearls | 403                   | 1           | 310                                | 113                | 137                 | 21                     |
| PVN/10% ink               | 403                   | 1           | 256                                | 94                 | 155                 | 19                     |
| PVN/1% Epolin 2057        | 175                   | 1           | 103                                | 35                 | 76                  | 4                      |
| GAP/ carbon nanopearls    | 175                   | 1           | 1960                               | 392                | 188                 | 181                    |
| GAP/5% ink                | 150                   | 1           | 1970                               | 394                | 190                 | 183                    |
| GAP/1% Epolin 2057        | 230                   | 1           | 1574                               | 167                | 256                 | 198                    |
| PVC/5% ink                | 570                   | 2.5         | 94                                 | 137                | 189                 | 8.7                    |
| NC/2% carbon nanopearls   | 400                   | 1           | 471                                | 179                | 183                 | 42                     |

stand, a figure which is substantially improved relative to earlier reports.<sup>6</sup>

In nearly identical circumstances, nanopearl carbon as a laser absorber gave similar or better performance compared to ink (which contains  $\mu\text{m}$ - rather than  $\text{nm}$ -size particulates), while the tuned absorber dye gave similar or somewhat worse performance. Although the advantage is slight, we saw a definite benefit to using nanosize absorbing particles. We believe this is not due to better light-absorbing efficiency of nanosize particulates, nor, for ms-duration pulses to higher transient temperatures, but rather to more intimate interaction with the exothermic components of the absorbing mix.

## References

- <sup>1</sup> Weast, R. (ed.), *CRC Handbook of Chemistry and Physics*, Chemical Rubber Publishing Co, Cleveland, 1978, p D-71
- <sup>2</sup> Johnson, R.D. & Holbrow, C. (eds.), *Space Settlements, a Design Study*, NASA report SP-413, 1977
- <sup>3</sup> Phipps, C. and Michaelis, M., "Laser Impulse Space Propulsion", *Journal of Laser and Particle Beams* **12** no. 1, 1994, pp. 23-54
- <sup>4</sup> Phipps, C., Reilly, J. and Campbell, J., "Optimum Parameters for Laser-launching Objects into Low Earth Orbit", *J. Laser and Particle Beams*, **18** no. 4, 2000, pp. 661-695
- <sup>5</sup> Phipps, C. R. and Luke, J. R., "Diode Laser-driven Microthrusters: A new departure for micropropulsion," *AIAA Journal* **40** no. 2 2002, pp. 310-318
- <sup>6</sup> Phipps, C., Luke, J., Lippert, T., Hauer, M. and Wokaun, A., "Micropropulsion using a Laser Ablation Jet," *J. Propulsion and Power*, **20** no. 6, 2004, pp. 1000-1011