[4](a) E.J. Schimitschek, J.E. Celto, and J.A. Trias, Appl. Phys. Lett. 31, 608 (1977); (b) J.G. Eden, Appl. Phys. Lett. 33, 495 (1978).
[5]K.Y. Tang, R.O. Hunter, Jr., J. Oldenettel, and D.L. Huestis, J. Chem. Phys. (to be published).
[6]R.W. Waynant and J.G. Eden, Appl. Phys. Lett., 33, 708 (1978).
[7]J.G. Eden and R.W. Waynant, J. Chem. Phys. 68, 2850 (1978); J.G. Eden, R.W. Waynant, S.K. Searles, and R. Burnham, Appl. Phys. Lett. 32, 733 (1978).
[8]J. Maya, J. Chem. Phys. 67, 4976 (1977) and references quoted therein.
[9]J.R. Murray and H.T. Powell, in *Electronic Transition Lasers II*, edited by L.E. Wilson, S.N. Suchard, and J.I. Steinfeld, (MIT Press, Cambridge, Mass., 1977).
[10]*Handbook of Chemistry and Physics*, edited by R. Weast (Chemical Rubber, Cleveland 1973).
[11]G. Herzberg, *Molecular Spectra and Molecular Structure: I. Spectra of Diatomic Molecules* (Van Nostrand Reinhold, New York, 1950) p. 537.
[12]W.T. Whitney, 10th Int. Quantum Elect. Conf., Atlanta, 1978, paper U.9 (unpublished).

Intensity dependence of harmonic generation in 10-μm laser-produced plasmas

H. A. Baldis, N. H. Burnett, G. D. Enright, and M. C. Richardson

National Research Council of Canada, Division of Physics, Ottawa, K1A 0R6, Canada
(Received 18 September 1978; accepted for publication 27 November 1978)

Integral harmonic intensities in the radiation scattered from nanosecond 10.6-μm laser-target interaction have been studied as a function of laser energy and target orientation. Measurements suggest a power-law dependence of scattered harmonic intensity with increasing laser intensity, $I_{n\omega 0} \propto I^n_{\omega 0}$, up to $\sim 1 \times 10^{14}$ W/cm^2 incident with evidence of saturation above this level. The scattered harmonic light at $7\omega_0$ was found to be fairly isotropic in front of the target.

PACS numbers: 52.25.Ps, 52.50.Jm, 52.40.Db, 42.65.Cq

The existence of an extended series of integral harmonic lines in the spectrum of radiation scattered from laser-target interaction with $I\lambda^2 \sim 10^{16}$W μm^2/cm^2 has been reported for both 10.6- and 1.06-μm experiments.[1,2] In the former case, harmonic lines up to $11\omega_0$ have been observed, while for the 1.06-μm experiments, observations up to $5\omega_0$ have been made. The remarkably similar spectra and intensities of the harmonic lines in the two cases are suggestive of a wavelength scaling characteristic of phenomena associated with the electron oscillation energy. In this letter we would like to present some additional data on the intensity and target orientational dependence of harmonic emission in 10.6-μm target experiments.

The intensity dependence of harmonic generation in direct backscatter was studied with the experimental setup shown in Fig. 1. A nanosecond ($\sim$ 1.5-ns (FWHM) pulse from the COCO-II laser at N.R.C. was focused in an $f/2.5$ beam onto massive Al targets in vacuum. The prepulse contrast ratio was $\sim 10^{-6}$ and the target focal spot size had a measured half-energy diameter of 110 μm at best focus. The collimated backscattered radiation was sampled with a NaCl beamsplitter in the incident beamline and directed onto a detector-filter array which monitored the second-, third-, fourth-, and tenth-harmonic levels. A Ge :Au photoconductive detector and narrow-bandpass dielectric filter was used for the second harmonic. In : As and Hg : Cd : Te photovoltaic detectors with suitable filters sampled the third and fourth harmonics and an S1 photomultiplier with narrow-bandpass filter responded to the tenth harmonic. In addition to those harmonic lines monitored in the collimated backscatter, seventh-harmonic emission was monitored in the four directions shown in Fig. 1 (directions 2 and 3 being in the plane of incidence and 45° to the plane of incidence, respectively). For this purpose, Ge avalanche photodiodes were used in combination with a Si window which served as a long-wavelength pass filter to isolate $7\omega_0$. This combination

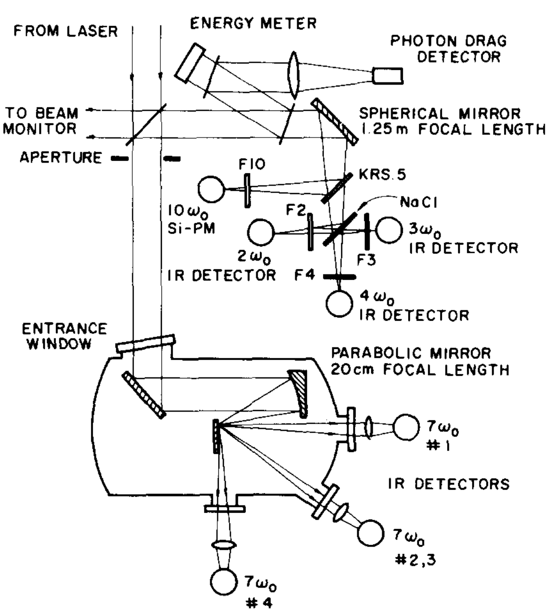

FIG. 1. Experimental setup to study harmonic emission.

 0003-6951/79/050327-03$00.50

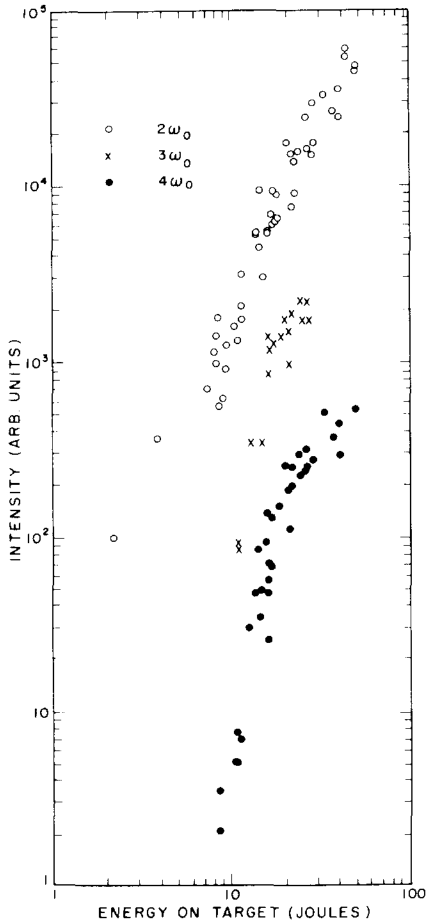

FIG. 2. Energy dependence of the second-, third-, and fourth-harmonic energies in backscatter.

had sufficient sensitivity to detect $7\omega_0$ harmonic radiation scattered from the target in a solid angle $< 10^{-3}$ sr allowing these detectors to view the interaction through external target chamber windows. All detectors used in these studies had time resolution in the nanosecond range, allowing easy identification of harmonic and plasma continuum signals.

As discussed in Ref. 1, previous measurements using incident laser energies in the range from 25 to 50 J indicate that from the third to the eleventh harmonics an intensity ratio of ~ 6 exists between successive harmonic energies $E_{n\omega_0}/E_{(n+1)\omega_0}$ in direct backscatter with only a weak dependence of the higher harmonic levels on incident laser energy in this range. In the present study, incident laser energy was varied systematically over the range from 1 to 40 J using a propylene gas cell or Mylar attenuators at the laser output. Care was taken to ensure that the laser operated on a single rotational line (P20) in the 10.4-μm vibrational band and that the laser pulse shape remained constant within the 700-ps resolution of the photon drag monitor. The incident radiation was P polarized at a mean incident angle of 20° to target normal.

The relative levels of second-, third-, and fourth-harmonic energies in backscatter with incident 10.6-μm energy in the range 1–40 J are shown in Fig. 2. The tenth-harmonic signal exhibited only a weak power dependence ($\lesssim 2$) on incident energy in the range $20 < E_{\omega_0} < 40$ J but fell even more rapidly than the fourth harmonic as the incident energy was reduced below 20 J and became buried in the plasma continuum at incident energies $\lesssim 15$ J. In an attempt to eliminate that part of the scatter due to incident energy calibration error and undetected variations in laser pulse shape from shot to shot., the third- and fourth-harmonic levels were plotted as a function of second-harmonic signal for the same shot, with the results shown in Figs. 3(a) and 3(b).

It is apparent that the scatter in the data points is somewhat reduced and that low-energy points fall along the power laws $E_{3\omega_0} \propto E_{2\omega_0}^{1.5}$ and $E_{4\omega_0} \propto E_{2\omega_0}^{2}$. In the upper range of incident energies a saturation is evident with a much weaker increase in the third and fourth harmonics with increasing second-harmonic emission. This transition occurs at an incident energy of approximately 20 J and is also evident in Fig. 2. The data of Fig. 2 can be reasonably well described for incident energies E_{ω_0} below 20 J by $E_{n\omega_0} \propto E_{\omega_0}^{n}$ for the second, third, and fourth harmonics, although the best fit slopes are slightly in excess of these values.

The $7\omega_0$ detectors shown in Fig. 1 indicated a fairly isotropic emission at this wavelength over the four directions monitored. The signals did not vary consistently by more than a factor of 2 in any of the four detectors, with the highest signals on detector No. 1 (nearly direct backscatter). In

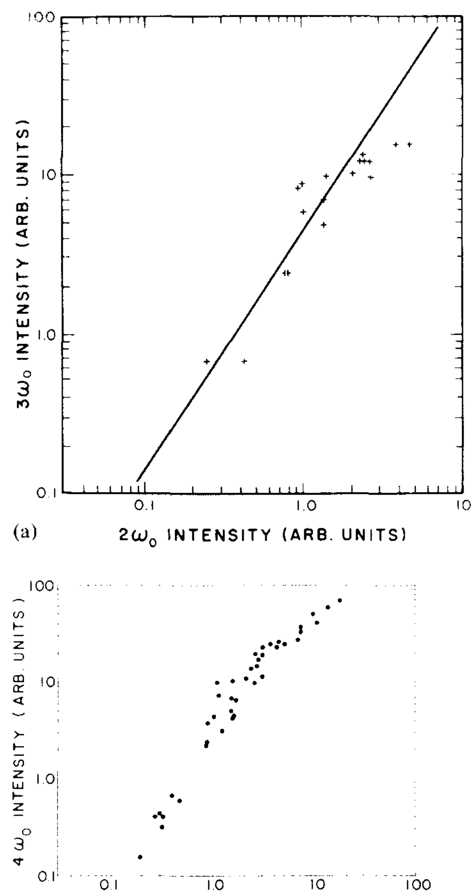

FIG. 3. (a) Third-, and (b) fourth-harmonic levels as a function of second-harmonic signal for the same shots. Straight lines have slopes of 1.5 and 2 respectively.

this location the $7\omega_0$ emission was observed to be strongly (>75%) polarized paralled to the E vector of the incident 10.6-μm radiation.

The effect of target orientation and laser polarization on harmonic production was also examined. With the laser output fixed at 30 J the target normal was varied from 20° to 45° with respect to the incident focal cone (P polarized), the backscattered harmonics were observed to decrease with increasing target angle by an amount roughly corresponding to that expected with the reduced intensity on target. At angles closer to normal than ~15°, observations were not possible due to the onset of target-induced prelase which drastically quenched harmonic production. Finally, the polarization of the 10.6-μm laser beam was rotated by 90° by means of a half-wave plate inserted in the beam line after the oscillator and switch-out stages. The substitution of S- for P-polarized laser output had no observable effect on the levels of any of the harmonics or their dependence on target angular orientation.

An apparent saturation or transition from power-law behavior in harmonic conversion is observed in these measurements at an incident energy of 20 J (peak intensity $\sim 10^{14}$ W/cm^2). We estimate that at this incident intensity approximately 1% of the total incident laser energy is converted to integral harmonics (mostly second harmonic). It is evident, however, that if all harmonics up to the eleventh continued to increase with incident laser energy as power laws $E_{n\omega_0} \propto E^n_{\omega_0}$, then total harmonic conversion would approach 100% at incident energies of ~100 J, so that some form of saturation must be expected. The transition intensity of 10^{14} W/cm^2 is well above the onset of strong profiled modification $(v_{os}/v_{th})^2 \sim 1$, so that assuming the harmonic production observed here is related to a resonant field enhancement at critical density (resonance absorption) saturation may be related to the effect of profile modification on the resonant field.[3,4] The absence of a strong effect on harmonic conversion in changing from P to S polarization is consistent with x-ray studies of these plasmas[5] where the suprathermal emission temperature is also observed to be relatively insensitive to polarization and target orientation. Because of the relatively long pulse and high $I\lambda^2$ encountered here, distortion of the critical density front occurs, and it is not possible to associate an "angle of incidence" with the original target angle. This effect is clearly seen in interferometric observations of these plasmas.[6]

[1]N.H. Burnett, H.A. Baldis, G.D. Enright, and M.C. Richardson, Appl. Phys. 31, 173 (1977).
[2]E.A.McLean, J.A. Stamper, B.H. Ripin, H.R. Griem, J.M. McMahon, and S.E. Bodner, Appl. Phys. Lett. 31, 825 (1977).
[3]K.G. Estabrook, E.J. Valeo, and W.L. Kruer, Phys. Fluids 18, 1151 (1975).
[4]D.N. Forslund, J.M. Kindel, and K. Lee, Phys. Rev. Lett. 39, 284 (1977).
[5]G.D. Enright, M.C. Richardson, and N.H. Burnett, Bull. Am. Phys. Soc. 22, 1112 (1977).
[6]R. Fedosejevs, G.D. Enright, I.V. Tomov, and M.C. Richardson, Bull. Can Assoc. Phys. 34, 6 (1978).