Supercontinuum Generation in Gases: A High Order Nonlinear Optics Phenomenon*

P.B. Corkum¹, C. Rolland¹, and T. Srinivasan-Rao

¹National Research Council of Canada, Division of Physics,
Ottawa, Ontario, Canada, K1A 0R6

²Brookhaven National Laboratory, Upton, NY 11973, USA

Received by OSTN

APR 02 1990

The recent development of high power, ultrashort pulse sources has created renewed interest in the interaction between intense laser radiation and free atoms and molecules. Not only is it feasible to apply laser fields that approach, or exceed, the strength of the atomic field as seen by the outer electrons, but it is also possible to apply these fields nonadiabatically using ultrashort pulses. Recent experiments have uncovered a number of new phenomena, including energetic electron production from rare gas atoms, [1,2] multiply charged ions, [3] and inner shell excitations.[4]

From a theoretical perspective, when the laser field approaches one atomic unit, it can no longer be considered as a perturbation to the atomic field. Thus non-perturbative theoretical approaches are being developed to treat both atoms [5] and molecules [6] in strong fields.

Until now, experiments have been restricted to isolated atoms. However, because the theories of nonlinear optics and multiphoton ionization are so interrelated, we should expect these new phenomena to have optical signatures. In addition to their intrinsic interest, nonlinear optics experiments can add a new perspective for judging emerging theories of high intensity laser processes. Clearly, there is a new class of experiments to be performed using ultrahigh power, ultrashort pulses.

We describe an experiment performed in high pressure gases ($1 \text{ atm} < p < 40 \text{ atm}$) with a 2 psec or 70 fsec $0.6 \mu\text{m}$ pulse focused to a peak intensity of $I \lesssim 10^{13} \text{ W/cm}^2$. The maximum intensity exceeds that in which multiphoton ionization is observed in longer pulse experiments ($\sim 25 \text{ psec}$) in Xe using either $1.06 \mu\text{m}$ or $0.53 \mu\text{m}$ radiation. It is approximately the intensity at which tunnel ionization is observed with nanosecond $10 \mu\text{m}$ pulses in very low pressure Xe.[7] Furthermore, at such intensities in $0.53 \mu\text{m}$ and $1.06 \mu\text{m}$ experiments, high energy electrons (up to 20 excess photons absorbed) are observed from Xe.[2]

The experiment was performed by focusing the $5 \times 10^9 \text{ W}$, 70 fsec pulse or the $0.4 \times 10^9 \text{ W}$, 2 psec pulse produced by an excimer pumped dye laser [8] with an F/170 lens into a high pressure gas cell. The output was monitored (i) by eye, (ii) on a 10 nm/mm spectrograph and recorded on an OMA or (iii) on a 10A/mm spectrograph and recorded photographically.

Seven gases were investigated, Ne, Ar, Kr, Xe, N_2 , H_2 and CO_2 . Our primary experimental observation was that supercontinua (Figs. 1 and 2) are produced in all of the above gases except Ne. Supercontinuum generation appears to be a near universal response of high pressure gases to high intensity radiation.[5] However, it has never been predicted nor observed previously. (Working independently, P. Sorokin's group at IBM

*This research was funded in part by the U. S. Department of Energy:

Contract No. DE-AC02-76CH00016.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Fig. 1: Continuum Spectra from Xe
 x P = 30 atm, τ = 70 fsec
 o P = 15 atm, τ = 2 psec

Fig. 2: Continuum Spectra
 x P = 38 atm H₂, τ = 70 fsec
 o P = 40 atm N₂, τ = 2 psec

has recently observed supercontinuum from high pressure gases.[9] Their work has expanded on their previous observation of spectral broadening of picosecond UV pulses when focused in room air.[10]) Supercontinuum generation itself is not a new phenomenon.[11] The inadequacy of standard theories of continuum generation [11,12] for the case of high pressure gases is discussed in a related paper and the reader is referred to that work for details.[13]

Comparing Fig. 1 and Fig. 2 we see a remarkable similarity of the spectra for both picosecond and femtosecond pulses and for both molecular and rare gases. (Although the continuum intensity is in arbitrary units in both figures, the units are the same for all of the curves plotted.) The anti-Stokes component is nearly the same for all gases and pulse durations investigated and has a minimum wavelength of $\lambda_{\min} \sim 300$ nm. We have not investigated the Stokes component as thoroughly, but there are significant variations between gases and pulse durations. In most cases, the Stokes component contains more energy than the anti-Stokes. At least with the femtosecond pulse in CO₂ the radiation extends beyond 1.3 μ m, the limit of a S-1 photocathode.

All continua showed a distinct threshold. Below the continuum generation threshold there was little or no broadening. The threshold value of the gas pressure multiplied by the laser power required for supercontinuum generation is plotted in Fig. 3 as a function of the inverse of the laser power. The plotted data are for femtosecond pulses.

Self-focusing has a similar threshold dependence to that shown in Fig. 3. For self-focusing the pressure-power product is given by:

$$(\underline{P} \ p)_{TH} = \pi \epsilon_0 c \lambda^2 \eta_2^{-1} , \quad (1)$$

where $\underline{P}$ is the laser power, p the gas pressure, and η_2 is the second order nonlinear refractive index at atmospheric pressure. Equation 1 indicates that each gas has a characteristic pressure-power product at the self-focusing threshold. Figure 3 shows a similar behaviour for the threshold of continuum generation. Taking the ratio between the values of $(\underline{P} \ p)_{TH}$ in Fig. 3, we find that all ratios agree within 20% with the ratio

continuum generation in nearly all cases. We propose that self-focusing allows the field to be applied in a less adiabatic manner and therefore increases the nonlinearity of the interaction.

References

1. P. Agostini, F. Fabre, G. Mainfray, G. Petite and N.K. Rahman, Phys. Rev. Lett. 42, 1127 (1979).
2. L.A. Lompré, A. L'Huillier, G. Mainfray and C. Manus, J. Opt. Soc. Am. B2, 1906 (1985).
3. A. L'Huillier, L-A. Lompré, G. Mainfray and C. Manus, in Laser Techniques in the Extreme Ultraviolet, edited by S.E. Harris and T.B. Lucatorto, AIP Conference Proceedings No. 119, AIP, New York (1984), p. 79.
4. C.K. Rhodes, Science 229, 1345, (1985).
5. See for example Multiphoton Ionization of Atoms, edited by S.L. Chin and P. Lambropoulos (Academic Press, New York, 1984).
6. See for example T. Tung Nguyen-Dang and A.D. Bandrauk, J. Chem. Phys. 79, 3256 (1983); J. Chem. Phys. 80, 4926 (1984); A.D. Bandrauk and T. Tung Nguyen-Dang, J. Chem. Phys. 83, 2840 (1985).
7. S.L. Chin, F. Yergeau and P. Lavigne, J. Phys. B: At. Mol. Phys. 18, L213 (1985).
8. C. Rolland and P.B. Corkum, accepted for publication, Opt. Commun.
9. P.P. Sorokin - private communication.
10. J.H. Glowia, G. Arjavalingam, P.P. Sorokin, and J.E. Rothenberg, Opt. Lett. 11, 79 (1986).
11. J.T. Manassah, M.A. Mustafa, R.R. Alfano and P.P. Ho, IEEE J. Quantum Electron. QE-22, 197 (1986).
12. N. Bloembergen, Opt. Commun. 8, 285 (1973).
13. P.B. Corkum, C. Rolland and T. Srinivasan-Rao, submitted for publication, Phys. Rev. Lett.
14. H.J. Lehmeyer, W. Leupacher and A. Penzkofer, Opt. Commun. 56, 67 (1985).
15. P. Lambropoulos, Phys. Rev. Lett. 55, 2141 (1985).
16. In preliminary experiments we have unsuccessfully attempted to produce supercontinua with our psec pulse using small F# optics while maintaining the pressure-power product below threshold.
17. J.H. Marburger and W.G. Wagner, IEEE J. Quantum Electron. QE-3, 415 (1967).

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.