Final technical report on**Grant N° DE-FG03 - 87ER 60504****P. Lambropoulos, Principal Investigator**

The work on the grant for the entire period of its duration concentrated on two different but related areas, namely coherent control of photoabsorption processes and the calculation of non linear optical processes with short wavelength radiation.

(A) On the first topic, the work dealt with the problem of controlling the population transfer from one to another bound state of a system in a route that passes through a continuum. This question is most important in the context of transferring populations between vibrational states of a molecule through a sequence of two pulses taking the system via the dissociation continuum. Our work was motivated by claims made in the literature that such population transfer can be 100 % efficient.

Building on our work and results obtained during previous periods of the grant we developed the correct formalism for this problem. We were then able to show (1-3) that in general the transfer will not be 100 % efficient. On the contrary under most realistic circumstances, it will be very inefficient due to inevitable losses (decay) into the continuum. After establishing the correct description of the process (1), we extended it to the case where a discrete state may be embedded in the continuum (2) as, for example, is the situation with an autoionizing state. Conditions may be made a bit more favorable in that case. Finally we examined an altogether different scheme which may exploit modern developments in laser technology through which the phase of the laser may be varied as desired. That scheme (2), although technically more demanding, can achieve more controlled trapping and hence population transfer.

(B) On the second topic, our work was motivated by the availability of XUV and soft X-ray coherent radiation sources obtained through high order harmonic generation. In addition, a few other techniques based on schemes of photo-pumped X-ray lasers promise to provide in the near-future similarly coherent sources. It is thus important to have an assessment of the possibility of extending non-linear optical processes to this range of wavelengths. This means assessing the relevant magnitude of the susceptibilities for third harmonic generation, stimulated Raman scattering, two-photon absorption, etc.

We undertook the study of the first two processes in publications 4 - 6. In fact the work for publication number 6 was performed in collaboration with one of the experimentalists (E. Fill) who has developed one X-ray laser. We performed thorough calculations for these susceptibilities in atomic He as it is expected to be a convenient medium for pilot studies of such processes, as well as one with rather small susceptibilities. It is in some sense a worst case scenario.

The results of our calculations (4-6) have provided the first, and only at this point, evaluation of the potential of these sources. In brief, we have calculated the minimum intensities necessary for the observation of such non-linear processes and have also compared the relevant susceptibilities to their counterparts in the optical frequency domain on which much information has been accumulated. The final assessment that has emerged from our

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calculations is that a minimum of $10^{12} - 10^{13} \text{ W/cm}^2$ is necessary. It appears that at this point, such intensities exist only marginally, in the sense that great efforts and demanding technology are necessary for the focusing of the beams, if the above intensities are to be achieved. A few preliminary experiments stimulated by our results are presently under way. Hopefully further progress in the design of these sources will make it easier to study the relevant non-linear processes, not too long from now.

The work on topic A was performed mainly by Dr. T. Nakajima who has now been appointed associate professor at Kyoto University in Japan, while on topic B it was performed mainly by Dr. S.J. van Enk who is presently pursuing further postdoctoral research at the California Institute of Technology in Pasadena, California. They were both supported by the grant while performing the work.

Publications on work supported by the grant

- 1) T. Nakajima, M. Elk, Jian Zhang and P. Lambropoulos 'Population transfer through the continuum', Phys. Rev. A, R 913 (1994)
- 2) T. Nakajima and P. Lambropoulos 'Population trapping through the continuum by phase-switching' Optics Com. 118, 40 (1995)
- 3) T. Nakajima and P. Lambropoulos 'Population transfer through an autoionizing state by pulse delay' Z. für Physik D, 36, 17 (1996)
- 4) S.J. van Enk, Jian Zhang and P. Lambropoulos 'Calculation of $\chi^{(3)}$ in He including configuration interaction for XUV photons' Z. für Physik D, 37, 211 (1996)
- 5) S.J. van Enk, Jian Zhang and P. Lambropoulos 'Third harmonic generation through double resonances: a model in He' J. Phys. B 29, 4041 (1996)
- 6) E.E. Fill, S.J. van Enk, Jian Zhang and P. Lambropoulos 'Stimulated Raman scattering in helium with soft X-ray laser radiation' Phys. Rev. A, 54, 5374 (1996)