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Report of Progress

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PROGRESS REPORT

MOLECULAR DESIGN CONCEPT FOR X-RAY LASER RESEARCH

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ABSTRACT

The goal of this program is the construction of an x-ray laser in the kilovolt regime. Recent experimental results indicate that a new technique for the generation of strong amplification of x-ray wavelengths is feasible. It involves the combination of (1) a new ultrahigh brightness subpicosecond laser technology and (2) a recently discovered unique mode of strong-field interaction, particularly applicable to molecules. A concept of molecular x-ray laser design emerges from these considerations which matches the mode of excitation to the structure of the molecular system. The molecular approach enables the combination of very highly electronically excited conditions with an environment characteristic of dense cold matter, a general situation exceptionally conducive to x-ray amplification. Both high efficiency and wavelength tunability are intrinsic features of this method.

I. INTRODUCTION

The availability of high peak power ultraviolet (UV) lasers¹⁻⁷ has revolutionized experimental research on the behavior of matter very strong electric fields ($E \geq e/a_0^2$). Moreover, the situation is such that presently available theories are not effective in describing many aspects of the new regime of strong-field coupling being explored. This is particularly true for many-electron systems, a class of materials that includes all molecules. Despite our imperfect understanding of the mechanisms involved in these high-field interactions, recent experimental results suggest that these multiphoton processes can result in a considerable transfer of energy to the irradiated system. For an intensity of $\sim 10^{17}$ W/cm² a total energy transfer exceeding 1 keV has been observed. Consequently, it is expected that total energy transfers considerably above 10 keV will be characteristic of the higher intensities ($> 10^{19}$ W/cm²) that will become available in the near future.

Molecules are emerging as a very interesting class of systems for study for at least two reasons. They are the multitude of species available and the experimental finding that the dynamics of the multiphoton coupling is sensitive to the molecular structure of the system.⁸⁻¹⁰ Accordingly, data concerning several molecules, including N₂, O₂, CO, N₂O, CO₂, and OCS, are under active study.

II. EXPERIMENTAL APPARATUS

The laser system used in these experiments is shown schematically in Fig. (1). Since it has been described in detail elsewhere, its description here will be brief. It has three main components; they are an amplified infrared seed laser, wavelength conversion crystals, and UV excimer amplifiers. A recent modification to the system incorporates a Ti:Al₂O₃ amplifier for amplification of the 745 nm radiation before conversion to the UV. This has resulted in a 40-fold increase (to ~ 40 μ J) in the 248 nm output energy from the crystals, greatly enhancing the overall

performance of the entire system. Railgaps have also been added to the power amplifier to improve its performance.

The system has a final output pulse width of ~ 600 fs and a pulse energy of ~ 500 mJ. Stringent measures have been taken throughout the entire laser system to ensure high spatial quality of the output. This has resulted in a focusability which is within a factor of two of the diffraction limit for a 10 cm diameter beam. Indeed, with the use of a simple F/10 lens, peak intensities approaching a value of $\sim 10^{17}$ can be produced, even in the presence of significant spherical aberration. Higher intensities are now available with the use of an F/6 mirror.

III. DISCUSSION OF EXPERIMENTAL RESULTS

A. Atomic Ionization

The measurement of the ionization of free atoms can be used to give an estimate of the peak field strength produced in the focal zone where the ionized atoms are produced. This can be done with the use of models involving tunneling ionization.^{11–17}

Measurements of Kr have been performed in order to appraise the maximum intensity produced. The result, as presented in the ion time-of-flight spectrum, are illustrated in Fig. (2). Clearly seen are signals from Kr^{8+} and Kr^{9+} and a weak, but discernable, presence of Kr^{11+} is also visible. The O^{2+} peak arising from an impurity of H_2O in the chamber, masks the Kr^{10+} signal that occurs in its vicinity. Note the prominent isotopic splittings that are associated with the Kr^{8+} signal.

The representation given in Fig. (3) combines experimental data on the threshold intensities for ion production in Kr with the corresponding predictions of several theoretical models. The measured datum for Kr^{11+} ($Z = 11$), which has¹⁸ $E_p \approx 318$ eV and is explicitly shown, gives $I_{th} \approx 2 \times 10^{17}$ W/cm². This

experimental value is within approximately a factor of two of that predicted by the 1-D picture.¹¹ This would give a peak field of $\sim 2.4 (e/a_0^2) = 1.2 \times 10^{10}$ V/cm produced by the ultraviolet wave.

B. Molecular Studies

A series of experiments has been begun on molecular systems. Data have been taken for N_2 , O_2 , CO , N_2O , and CO_2 and we are currently in the process of evaluating this information. In order to present certain features of these new data, we now explore some of the results for CO_2 .

In previous preliminary work,¹⁹ certain aspects of the problem of fragmentation exhibited by CO_2 have been discussed. These earlier data¹⁹ were taken with a peak intensity of $\sim 3 \times 10^{16}$ W/cm² and the observed kinetic energy distribution for the O^{3+} channel is shown in Fig. (4A). In this case, most of the observed ions had kinetic energies below ~ 65 eV.

The intensity dependence⁹ of the distributions of kinetic energy can reveal important properties of the molecular coupling. The production of Kr^{11+} , as discussed in paragraph III.A above, demonstrates that the improved focusing system currently being used produces a maximum intensity $I_m \sim 2 \times 10^{17}$ W/cm², a value considerably higher than available in the earlier work.¹⁹ Figure (4B) illustrates the kinetic energy spectrum for the O^{3+} channel observed from CO_2 with this higher value for the maximum intensity. Under these conditions, the peak in the kinetic energy distribution occurs at ~ 65 eV and a discernable signal exists even in the 90–100 eV range. A comparison of these data with the results appearing in Fig. (4A) clearly illustrates a significant shift of the distribution to higher kinetic energies. The mean energy of the O^{3+} ions is roughly doubled in going from $\sim 3 \times 10^{16}$ W/cm² to $\sim 2 \times 10^{17}$ W/cm² in the maximum intensity.

This result indicates that considerably more energy has been transferred to the molecule prior to fragmentation by the stronger external field.

IV. CONCLUSIONS

A program of molecular studies has commenced for the evaluation of a new concept for x-ray amplification. Data on several systems, including N_2 , O_2 , CO , N_2O , and CO_2 , have been obtained and are currently being analyzed. We plan to continue this line of inquiry and extend it to other molecular systems such as SF_6 and UF_6 .

Fig. (1): A schematic of the high intensity KrF laser system.

Fig. (2): Time-of-flight ion spectrum of Kr. (A) The region showing Kr⁸⁺ and Kr⁹⁺ with appropriate isotopic mass numbers identifying the peaks. Ionization with $q \geq 9$ must involve the 3d shell. The Kr¹⁰⁺ signal is obscured by the O^{2+} signal arising from H₂O present as an impurity. (B) The region showing Kr¹¹⁺. O^{2+} , O^{3+} , and C^{2+} signals stemming from impurities are also identified. The ionization energies associated with the production of Kr⁸⁺, Kr⁹⁺, and Kr¹¹⁺ species are shown as ~ 133 eV, ~ 219 eV, and ~ 318 eV, respectively, in these panels.

Fig. (3): Scaled threshold ionization intensities I_{th} as a function of ionization potential E_p for Kr along with calculated results from various theories. The parameter Z represents the final ionic charge. The theoretical curves corresponds to a 1-D model, 3-D simple atom picture, a 3-D complex atom model and the Keldysh theory, as discussed in Ref. (17). Experimental data are also shown, in particular for the point corresponding to Kr^{11+} ($E_p = 318$ eV).

Fig. (4): Kinetic energy spectra for O^{3+} from CO_2 . (A) Ion distribution obtained with a maximum UV intensity $I_m \sim 3 \times 10^{16} \text{ W/cm}^2$. Most ions have an energy below $\sim 65 \text{ eV}$. (B) Ion distribution observed with a maximum UV intensity $I_m \approx 2 \times 10^{17} \text{ W/cm}^2$. The peak in the kinetic energy distribution occurs at $\sim 65 \text{ eV}$.

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