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A Cadmium Photoionization Laser Pumped by Laser Induced Plasma Radiation from a Multi Foci Device

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Abstract. Soft x-rays from a laser-produced plasma were used to perform innershell photoionization of Cd atoms and to generate laser radiation at 442 nm. To achieve longer interaction zones between the Cd vapor and the soft x-ray flux, up to three plasma spots have been applied. In this way a maximum laser energy of 300 μJ with a 600 mJ Nd:YAG laser for the plasma production was achieved. Experimental investigations and corresponding rate-equation calculations indicate, that photoelectrons play an important role in the total laser kinetics.

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Innershell photoionization of atomic gases and vapors by soft x-rays from laser produced plasmas is a potential method for the realization of lasers at short wavelengths. With this technique laser oscillation was first demonstrated by Silfvast et al. [1] for Cd. Later on, systems with Zn [2] and In [3] have been operated. In these systems, the upper laser level is directly populated by soft x-rays from a laser induced plasma. Upon innershell excitation, Auger or shake-up processes may occur [4, 5] resulting in further possibilities for laser transitions. Auger-type laser oscillation in Xe at 108.9 nm has been reported recently [6] and proposals for shorter wavelengths Auger lasers have been published [7]. By excitation with soft x-rays, also high densities of metastable ionic levels can be produced [8]. This is of interest in connection with up-conversion schemes using the anti-Stokes Raman process [9, 10], which should even allow the generation of tunable radiation at very short wavelengths [11]. In order to obtain a high output energy and a good conversion efficiency for the laser, or a high gain to allow oscillation also on weak laser transitions, the excitation and operation parameters have to be carefully optimized and detrimental influences have to be taken into account. In these type of lasers not only optical processes are important, but also electron impact processes have to be considered, because instantaneously with the soft x-ray ionization photoelectrons are produced. In addition, the plasma electrons and ions also have to be considered, in general. While a direct influence of the plasma electrons and ions may be largely reduced by a suitable choice of operation conditions, the photoelectrons can not be avoided, in principle. In so far performed investigations of the excitation and laser processes [1], the influence of photoelectrons has not been discussed in detail. In this contribution measurements of the output energy in dependence of the pump laser energy, the number and the distance of plasma spots (excitation length) have been performed in order to contribute to the understanding of the laser process and, as a consequence, to improve the laser output energy.

It was found that several plasma spots are more favorable than only one as used so far. The optimum number of spots depends on the available pump energy for the plasma production. At a pump energy of 600 mJ (Nd:YAG laser) and with three plasma spots the so far highest output energy (300 μJ) has thus been achieved. All experimental dependences can be adequately described by a rate equation model which includes the influence of photoelectrons.

1. Experimental

For the plasma production and generation of the soft x-ray pump radiation a Nd:YAG laser with a maximum output energy of 600 mJ (8 ns pulse) has been used. The primary laser beam can be divided into up to three beams with equal energies, which are then focussed by lenses with 207 mm focal lengths on a tungsten target in a heat pipe containing Cd vapor. The experimental setup is shown in Fig. 1. The stainless steel heat pipe (diameter 38 mm, length of vapor zone 10 cm) has Brewster windows at the optical axis and two windows in side arms perpendicular to the optical axis for the pump beam and to allow observation of side-off fluorescence. The tungsten target within the heat pipe can be heated to prevent condensation of Cd vapor at the surface and can be rotated to allow a change of the surface area after a number of laser shots.

Typical operation conditions of the heat pipe are a temperature around 450°C (Cd vapor pressure of about 5 mbar) and a buffer gas pressure (He or Ar) around 5 mbar. The focussing optics of the Nd:YAG laser beam allows a variation of the spot size and of the distances between the spots on the target. In this way the pump intensity, which determines the plasma temperature and spectral distribution of the emitted soft x-rays, and the excitation length can be varied.

The optical resonator for the laser consists of two spherical mirrors (radii 500 mm), one with high reflectivity (>99%) and the other with different output couplings for the 442 nm radiation. The optical axis of the resonator lies parallel to the target surface in the plane determined by the three pump beams. Its distance to the target surface can be varied and was typically around 5 mm.

The fluorescence and laser radiation was measured and analyzed by means of a spectrometer, photomultiplier and transient digitizer. The absolute laser output energy was determined with a μJ-energy meter. Different filters were used to suppress undesired fluorescence or scattered light.

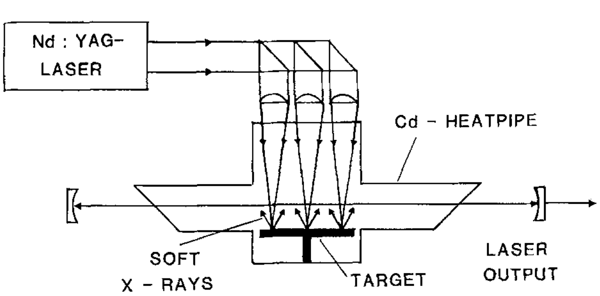

Fig. 1. Experimental set-up

2. Results and Discussion

When focussing laser radiation onto a target, production of a high temperature plasma at the surface of the target is observed for intensities I above about 10^9 Wcm^{-2}. For heavy metal targets the spectral distribution of the emitted radiation can be approximately described by a black body radiation distribution [12]. The corresponding temperature of the plasma thereby changes proportional to $I^{4/9}$ [13]. At intensities well above the threshold intensity for plasma production, typical conversion efficiencies for pump radiation into blackbody radiation are between 10% and 50% [14].

The 442 nm Cd laser transition considered and investigated here occurs between the metastable $4d^9 5s^2\,{}^2D_{5/2}$ and the $4d^{10}5p^2P_{3/2}$ level of the Cd ion. The transition in volves a change of the electron configuration and therefore has a relatively low cross section for stimulated emission of about 10^{-13} cm^2 [1]. The photoionization cross section for a removal of the $4d$-electron and a population of the upper laser level reaches a maximum value of about 15 Mb near 25 nm, while the cross section for a removal of the $5s$-electron and a population of the lower laser level remains below 1 Mb and is shifted to longer wavelengths [15]. Consequently, with black body plasma radiation at a temperature of about 11 eV (peak wavelength of the distribution 25 nm) a high selectivity for the photoionization excitation is given and inversion densities up to 10^{15} cm^{-3} have been achieved [15], allowing laser oscillation even at gain lengths of only a few mm.

For a single plasma spot, the radiation is emitted into a cone of about 90° [16]. Highest optical excitation rates are given near the target surface. However, very close to the target, electrons and ions emitted from the plasma disturb the laser process [1]. Therefore, the optical axis of the laser resonator should lie at a certain distance from the surface. In agreement with former Cd laser experiments [1], we found an optimum distance around 5 mm. At this distance an influence of plasma electrons can practically be neglected because at the given vapor pressure of 5 mbar and within the excitation time these electrons penetrate only about 2–3 mm into the vapor. For a given Nd:YAG pump energy the intensity at the target surface can be changed by changing the distance of the focussing lens to the target. For a maximum output energy of the Cd laser an optimum distance is obtained, corresponding to an optimum pump intensity. For different pump energies, different optimum distances are measured. We found, however, that optimum focussing always leads to approximately the same pump intensity of about 3×10^{10} Wcm^{-2}. This means, that for optimum

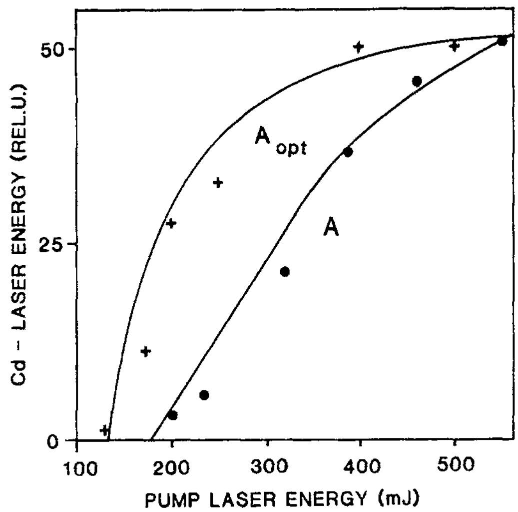

Fig. 2. Calculated (solid lines) and measured dependence of the Cd laser energy at 442 nm as a function of the pump laser energy for the plasma production (single focus, Cd vapor pressure 5 mbar, output coupling 50%). The maximum output energy corresponds to about 70 μJ. For curve A_{opt} the focussing was optimized for each pump laser energy, for curve A focussing is optimum only for the maximum pump energy and constant at lower pump energies

focussing the spectrum of the plasma fits best to the excitation cross section.

When changing the pump energy, the focussing conditions have to be changed as described above in order to ensure maximum output energy. Such an output versus input energy curve for a single plasma spot is given in Fig. 2 (curve A_{opt}). The adjustment of the optimum focussing, however, is a lengthy procedure, and therefore for practical purposes the optimum adjustment is once made for the maximum pump energy and then remains constant at lower pump energies. This leads to the curve A in Fig. 2, which consequently has a higher threshold. Characteristic for both dependences, more for the optimum focussing curve A_{opt}, is the saturation tendency of the output energy at larger pump energies, which implies that the pump energy is not used optimal. In an optically excited system, the saturation may be due to a bleaching of the active material at high excitation rates or due to the onset of quench or intrinsic loss processes generated by the excitation. In the system considered here and for the given parameters, a bleaching of Cd vapor by the soft x-ray flux will not occur at the excitation rates used, because only a few percent of the Cd vapor will be ionized. Saturation due to the onset of amplified spontaneous emission in other directions than the resonator direction may play a certain role but should not be the dominant process here, because for the Cd system the output energy is strongly determined by the optical resonator.

To explain the saturation effect the influence of photoelectrons which are immediately present upon photoionization has to be considered. We therefore established a rate equation model, which, in addition to the soft x-ray excitation and radiative decay processes, considers the formation of the photoelectrons and the interaction of these photoelectrons with all relevant levels, including especially the direct population of the upper and lower laser levels by collisions of photoelectrons with Cd atoms. To calculate the density and energy distribution of the electrons, the following assumptions have been made:

First, the production rate is equal to the photoionization rate. Second, each electron only interacts once, and the production of secondary electrons is neglected.

The energy distribution of the photoelectrons has a maximum around 35 eV and is determined by the photon energy distribution and the wavelength dependence of the photoionization cross section. The electron impact cross section for populating the upper laser level is slightly larger than for the lower laser level above an energy of about 50 eV [17]. Therefore, from the distribution we can estimate, that about 20% of the electrons ("hot electrons") may contribute to the generation of inversion, while the rest can only produce Cd ground-state ions or quench the populations. Compared to the optical excitation this electron contribution to an inversion is expected to be small. With respect to the large quantity of cold electrons (energy below 50 eV) we may therefore conclude that for the system considered here the net contribution of the electrons consists in a reduction of the output energy with increasing excitation rate leading to the observed saturation. This behavior can well be simulated by the model calculations as shown in Fig. 2. The calculated energy curves have been normalized to the experimental energy value for the maximum pump energy. In the model calculations only two fixed electron energies (hot and cold electrons) have been used for simplification. An immediate consequence of the measured and calculated behavior is that an increase of the output energy should be possible by a reduction of the excitation rate (reduction of the electron concentration) and a corresponding increase of the interaction length (interaction volume).

Taking this into account, a splitting of the pump beam into two or three beams with equal energy, allowing two or three separated plasma spots at the target, should increase the laser output energy and reduce saturation effects. The calculated dependences for one to three foci devices are shown in Fig. 3. With increasing foci number the saturation should decrease and the optimum output energy should be higher by approximately a factor of two. Corresponding experi-

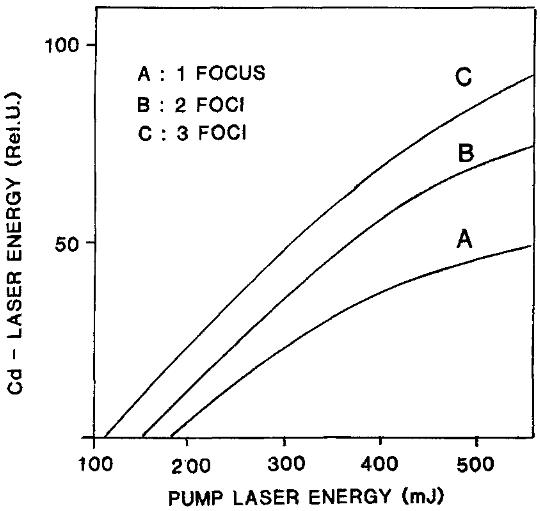

Fig. 3. Calculated Cd laser energy as a function of the total pump energy for different number of plasma foci. For details of the model see text

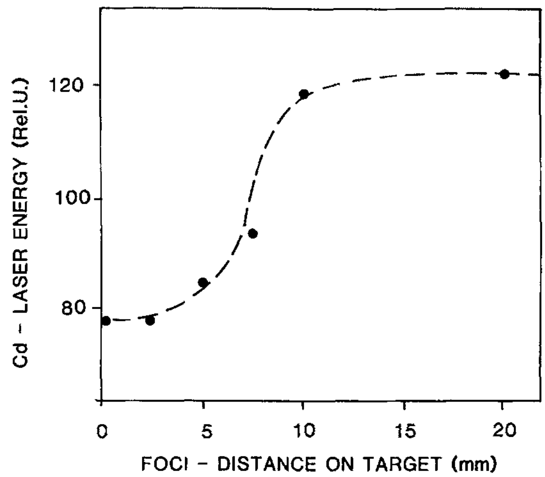

Fig. 5. Cd laser energy (442 nm) as a function of the distance between two plasma spots at the target (parameters as given in Fig. 2). The maximum energy corresponds to the maximum energy of curve *B* in Fig. 4

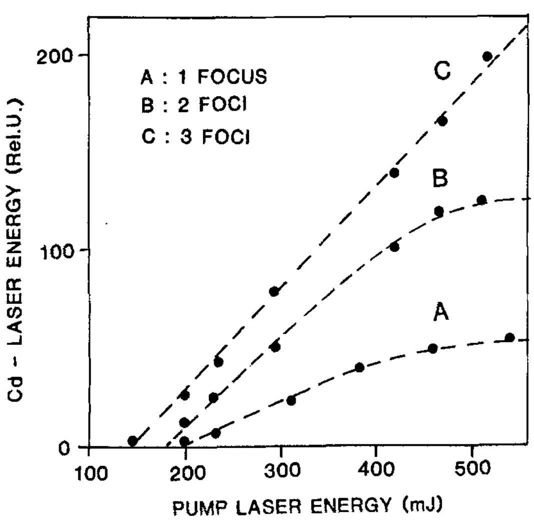

Fig. 4. Measured Cd laser energy (442 nm) as a function of the total pump energy for different number of plasma foci (operation conditions as given in Fig. 2). Focussing is optimum only for maximum pump energy. The relative units for the Cd laser energy can be directly compared with Figs. 2 and 3

mental results are given in Fig. 4. It can be seen that the saturation is reduced for the two foci system and has disappeared for the three foci device. At the same time the output energy increased by nearly a factor of three. With the three foci system the so far highest output energy (up to 300 μJ for an optimized output coupling of 70%) for the Cd-system was achieved. In the experiments, the focussing was always optimized for the highest pump energies as discussed in connection with Fig. 2.

Comparing the experimental with the calculated curves, the agreement is good for the one focus system. For the two and three foci systems the measured output energies are even higher as expected. This discrepancy may be due to an overaccentuation of the quenching influence of the electrons at lower concentrations but can also have its origin in the change of the geometry and symmetry of the interaction volume itself, which is not adequately described by the present model. Attempts to improve the model are presently being persued. In any case, the tendency to use lower excitation rates and longer interaction zones is obvious and implies that a further considerable improvement of the output energy should be possible by applying still more plasma spots. Estimations give an optimum number around 8 for a maximum pump energy of about 600 mJ and a further doubling of the output energy.

In the above investigations, the distance between the foci at the target was chosen to 10 mm. At this distance the interaction zones of the individual plasma spots just starts to overlap as can be seen from Fig. 5, where for two plasma spots the output energy has been measured as a function of the spot separation at the target. At large distances, the excitation regions of the two plasma spots are totally separated and the output energy stays constant at the value of the two foci device (Fig. 4). With decreasing distance the interaction length (volume) decreases and with it the output energy. This decrease is not compensated by the increase of the excitation rate as the system operates in a region of saturation. This decrease may also be considered as a proof that the observed saturation does not occur in the plasma itself but in the excited volume due to the formation of the photoelectrons. Finally, at short spot distance the excitation conditions and with it the output energy remains constant. The final output value, where the spots are almost overlapping is larger than for the one spot system. This implies that plasma formation using a higher energy and larger spot diameter is worse than using lower energies and smaller closely separated spots at the same intensity. This effect, which is not understood at present, may explain the discrepancy between calculations and experiments presented in Figs. 3 and 4. Again this observation supports the generally observed tendency for individual plasma spots at lower pump energies to give better results.

3. Conclusion

In the photoionization process with soft x-rays from a laser-produced plasma large numbers of photoelectrons are also produced. These photoelectrons may influence and modify an initially pure optical excitation and inversion generation process. Whether the photoelectrons support or diminish such a process depends on the actual system and the corresponding electron excitation and ionization cross sections. For the Cd-photoionization laser considered here, a saturation of the output energy at high excitation rates and the observed dependence of the output energy on the number of plasma spots and their distance may be mainly caused by a net quenching influence of the produced photoelectrons. An established rate equation model supports this conclusion and allows a satisfying description of most of the observed dependences. It is hoped that discrepancies between theory and experiments can be reduced by improving the model. Therefore, more and better information about the radiation spectrum emitted from the plasma under different formation conditions would be helpful.

The experiments have shown that longer excitation lengths and lower excitation rates are favorable to obtain higher output energies and conversion efficiencies. Larger gain lengths are also of interest, if weaker laser transitions are considered or in connection with the anti-Stokes up-conversion technique, where longer interaction lengths are favorable [18]. In the Cd system, the laser operates between higher lying ionic levels, which can only be populated by relative hot electrons, while the colder electrons populate the ionic ground state or contribute to a quenching of the level populations. There exist a number of systems that would allow much shorter wavelengths than the Cd system, where, however, the final level of the laser or of the anti-Stokes process is the ionic ground state. Candidates for this scheme are the alkali metals (for example Li [11]) or mercury. In these systems the influence of photoelectrons may be more severe than in the Cd system [19]. Calculations for a Hg-anti-Stokes laser operating between the levels $5d^9 6s^2\,{}^2D_{5/2}$ and $5d^{10} 6s^2 S_{1/2}$ (intermediate level $5d^{10} 6p^2 P_{3/2}$) indicate that photoelectrons have to be largely avoided. This is possible with pump pulses for the plasma production shorter than the collision time of the photoelectrons which is in the order of about 100 ps. Short pump pulses ($\approx$100 ps) have already been applied in the Cd and the Xe systems, leading to higher gain in comparison with former experiments [19].

By means of subnanosecond pump pulses in combination with longer excitation zones, it should be possible to realize further direct photoionization lasers and also to operate tunable short-wavelength anti-Stokes Raman laser systems.

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