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Analysis of Experiments on Energetic Ions from Laser Produced Plasmas with Reference to Hot Electrons and Pulsation

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Summary

The need for highly charged heavy ions from projected particle accelerators has recently led to a re-evaluation of the complex processes of ion production in laser generated plasmas. Possible mechanisms for the production of intense beams of high charge state ions are investigated as is the experimental evidence for these mechanisms. The hypothesis that 20keV ions are driven by 'hot electrons' is not supported by experimental work to date. This work, on the other hand, suggests that 30ps pulsation is the basic mechanism for the acceleration of tantalum ions up to charge state 8+ whose energy increases linearly with charge state up to 24keV . For long pulses and charge states between 8+ and 18+, it appears that there is a secondary mechanism of electron impact ionisation by plasma electrons of approximately 100eV in the plasma in front of the target, resulting in ions whose energy of around 24keV is independent of charge state.

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1) Introduction

Highly charged heavy ions are of interest for the Large Hadron Collider (LHC) project. For example, colliding fully stripped lead nuclei would produce centre of mass energies of 1262TeV [1] and information from such collisions would be important for the understanding of the phase transition [2] from the hadron state into the quark-gluon state of matter at higher densities than in stars or shock waves [3].

Currently, heavy ions are produced quite satisfactorily, for example in electron cyclotron resonance sources (ECR), giving (in accelerated beam pulses) 6×10^8 sulphur ions at 200GeV/u [4]. With present technologies, there would still be a lack of brightness of a factor 30 at the collision points in LHC and it appears, at present, that improving source performance would be more cost effective than adding cooling rings or making other accelerator gymnastics at higher energies.

Of the other techniques for ion production such as electrical discharges, intense electron beams, microwave heating or irradiating surfaces with high power lasers, the last process is interesting because the laser is in a relatively high state of development from research into inertial confinement. In fact, CO_2 lasers with 10kJ pulses of 10μs duration and 100Hz repetition rate have been available for some time [5]. There has, therefore, been some active development of laser ion sources [6-9]. In 1975 CO_2 laser pulses of 25J energy and 2ns duration produced 2MeV Al^{13+} ions with energy conversion efficiencies from laser to ion energy of 90% [10], and highly charged gold ions of 500MeV [5,11] have been measured. More recently, laser ion sources have been in use on both the Dubna (10^9 C^{4+} or 10^8 Mg^{12+} ions per pulse [12]) and ITEP (2.6×10^{13} C^{3+} or 10^{11} Pb^{6+} giving 10^9 Pb^{26+} extracted [13]) synchrotrons. Whilst these experiments were performed with very modest laser intensities and energies, the very interesting range of MeV ions from other laboratory work [14] demonstrated the advantages of high values of $I\lambda^2$ (I intensity, λ wavelength) when using the neodymium (Nd) glass laser.

The questions for future development are whether the recombination rate in the laser produced plasma can be reduced, and by how much the flux of high charge state ions can be increased, by the use of higher power lasers. Some indications from single laser shots are encouraging [10,14].

A better understanding of the non-linearities and anomalies which arise from the interaction of laser and plasma is needed and should profit from the extensive work carried out for laser fusion projects [5,15]. In particular the 'pulsating' or 'stuttering' interaction [5,16] has been explained as a hydrodynamic self-generation of Laue-Bragg reflection and its decay [17] which can be overcome by smoothing techniques [18]. The generation of higher ionisation states of 24keV ions from 100eV plasmas and whether the attribution of these energetic ions to 'hot' or supra-thermal electrons is another topic. Whilst earlier reports indicated a linear dependence of ion energy with charge state [5-10,19,20], this is only true up to 8+ and the energy is nearly constant for the states 9+ to 18+ (for Tantalum).

2) 'Hot' Electrons and Fast Ion Generation

It appears that the irradiation of solid targets produces plasmas whose ions or electrons behave fully thermodynamically. When the laser power exceeded a few megawatts, the behaviour changed. Whilst the plasma temperature remained low, ions with energies in the ten keV range having nonlinear properties were observed and confirmed from detailed evaluation of the temporal evolution of framing camera pictures. Irradiation of a spherical aluminium target showed a fast, nonlinear, crescent shaped plasma with ion energies of up to 10keV with a fully non-linear (nearly quadratic) behaviour moving against the laser light. There remained a spherically symmetric plasma with typical energies and temperatures of 30-100eV that expanded according to the gas-dynamic model [21]. Energy balancing [10] clearly showed that 90% of the laser energy went into ion energy for aluminium charge states 1 to 11 with a linear distribution of energy from 0.15 to 1.7MeV. Since there was confusion over both the very high and the moderate temperatures reported, the detailed x-ray spectrum was measured [22] and showed that there was both a moderate electron temperature of some 100eV and a 'hot' temperature of 20keV or more.

Time-of-flight (TOF) spectra showed sharp peaks corresponding to keV ions [23] whose separation increased linearly with ion energy divided by charge state . These were followed by a broad, nearly thermal ion spectrum. Even peaks corresponding to MeV ions of gold 38+ were identified [10,24]. These highly ionised charge states were confirmed by alternate methods [19,20,23-25]. Spectra for iron 26+ have been measured [26] and for phosphorus 13+ produced with a Nd glass laser irradiation of only 2×10^{15} W/cm^2 where the Doppler shift indicated ion energies of 1.5MeV [27]. This was explained, theoretically, by relativistic self focusing and soliton mechanisms. Some signals evident in the TOF spectra appeared before the energetic ion peaks. These were due to hydrogen or carbon/hydride ions[19] produced in a carbon layer on the surface of the target. This was measured to be 20Å thick whilst theory indicated 19Å [11].

A correlation was found in re-interpreting published TOF measurements with their measured energetic electron energies. Irradiating gold spheres with the HELIOS CO$_2$ laser with 2ns pulses of 7TW, only particles with 2MeV/nucleon were identified and interpreted as protons. The results for ions correspond to 400 to 500MeV gold ions[11]. This correlation was used to suggest that the energetic ions were protons produced by the hot electrons, though the ion energies are 3 to 15 times higher than the electron energies. Later experimental evidence[19] will be used to check whether the fast ions are due to hot electrons.

3) Pulsation (Stuttering) of Laser-plasma Interaction

The phenomenon of the pulsation of a laser-plasma interaction with a fully stochastic 'period' of 10 to 30ps was found in recent experiments. This chapter is devoted to an overview of the phenomenon. Observation of the energetic ions, of the nonlinear expansion and of the hot electrons were repeatedly confirmed before large scale laser fusion projects were initiated in 1972. However, later experiments showed all kinds of confusing anomalies which were frustrating and delaying the hope of an easy solution of the problem of laser induced inertial confinement fusion energy from lasers [5,28,29].

In 1973, detailed computations of the laser/plasma interaction were carried out. These showed that at high laser intensities, nonlinear (ponderomotive) forces [5] pushed the plasma into the nodes of a standing wave field. After the plasma attained a velocity of a few 10^7 cm/s within 2ps, the density ripple acted as a self produced ideal Laue-Bragg grating. This reflected nearly 98% of the laser light in the very low density peripheral plasma corona (Fig. 10.10, ref [5]).

Observations made in 1974 showed that the reflectivity of the irradiated plasma oscillated irregularly from a few percent to nearly 100% within 10-20ps [30]. This pulsating reflectivity confirmed that reflection occurs within the low density outer corona for the majority of cases. The plasma was pushed to a velocity of about $2x10^7$cm/s in a few ps and was not accelerated again until about 15ps later when another impulse came from a further interaction. This clarified why the spectrum of back-scattered radiation had a spectral modulation of about 4Å corresponding to the stepwise plasma acceleration [16]. Numerically, this behaviour could be reproduced exactly in a detailed hydrodynamic model. The computed velocity diagrams [17] show the pulsation of the velocity in step with the 8ps pulsation or stuttering.

Similar pulsation was observed for the 3/2 harmonic scattering [31], in double layer potentials [32] and H-alpha emission [33]. Insertion of a marker in the laser beam [34] showed that the reflection was phase-like and not mirror-like. The improved resolution of this experiment showed that the mark turned around with a period of about 30ps. Repeating this experiment with better time resolution would be of interest and the movement of the mark could be used as a test for stuttering.

Suppression of the pulsation was observed using a random phase plate RPP [18] or similar 'smoothing' techniques. The interpretation of smoothing as a suppression of pulsation is new [5] and contrary to the initial idea of using smoothing techniques only for the suppression of lateral beam uniformity [35]. It seems that the major problem in laser-plasma interactions is pulsation and its suppression.

An explanation of the observed keV and MeV ions could be the self focusing of laser beams in plasmas [5,36,37-38], but pulsation was missing to give a complete understanding.

An analogy can be made between the density rippling and relaxation of the pulsation mechanism and nonlinear mechanisms in the beam properties of proton synchrotrons [35]. Contrary to expectations, known fluctuations and inho-mogeities which should cause instability and chaotic decay are stabilised by a mechanism which collects the imperfection effects around certain frequencies and the particle dynamics between these frequencies are unperturbed [39].

4) Experiments on Fast Ion Generation

Evaluation of measurements [19] on tantalum (and other) targets irradiated in vacuum by a neodymium glass laser with fundamental (red) or second harmonic (green) pulses of a few mJ to 10J energy for durations between 30ps and 3ns, clarify many of the problems concerning the extraction and recombination of ions from the plasma plume. There are a large number of empirical results for a wide range of parameters and analysers.

The geometric extension of the focal spot can be determined from the far field pattern of the beam. This is usually the case when high accuracy is not required, it being more important to use the same conditions when comparing ion emission from spots for pulses of different duration and energy. For relative comparisons, errors in the focal dimension are compensated and the following data, with their relatively high comparison accuracy, confirm this.

If the laser beam frequency is doubled for the same spot size, the geometry of the laser pulse and the focusing elements remain unchanged if the frequency doubling crystal is a plane parallel plate inserted into the parallel beam (diffraction limits being determined by the laser fundamental frequency). As the beam is focused by a lens, only chromatic aberrations change the focusing conditions, but this error is usually negligible relative to other errors. It is the aperture of the laser and the focusing lens for red light which determines the dimensions of the final focus. Practically, there is a strong deviation from the diffraction limit (well known in solid state lasers) due to inhomogeneties and birefringence.

For the following evaluations, results were given in plots or tables. The errors were determined in some cases by the rather broad maxima in TOF measurements. In case of doubt, the average value of the broad peaks were used where the width of the peak determined the error (which is sometimes large).

5) Energetic Ions and 'Hot Electrons'

The hot electron mechanism at high laser intensities states that the same interaction conditions exist at the target if the laser intensity I and wavelength λ are related by:-

$$I\lambda^2=\text{constant}$$

Thus, if an intensity I_o at the fundamental frequency produces an effect, then under the same conditions of focusing, duration and irradiated power, a frequency doubled beam would require four times the intensity for the same effect. However, non-linear phenomena could reduce the effective heating or interaction mechanisms.

If energetic ions in the medium irradiation experiments [19] were produced by the hot electron mechanism, doubling the laser frequency, with otherwise identical conditions, should produce ions of half the energy or less. With regard to energetic electrons this would be similar to the observation [11] that the reflectivity of a laser irradiated plasma is reduced for the same intensity at the second harmonic above a threshold intensity. For this reason, most laser fusion schemes use intensities below the threshold which moves to higher values for shorter wavelengths [5].

Figure 1 is a collation of measurements of ion energies and charge states for 30ps laser pulses on tantalum targets for the fundamental (1064nm) and second harmonic (532nm) of the laser [19]. The example had a pulse energy of 30mJ for green light and 135mJ for red (4.5 times higher). Both cases produced nearly the same ion energies of up to 5keV for Ta^{6+}. Since the pulse length and other conditions were equal, it can be assumed that hot electrons are generated in a similar manner, but the green light should produce less energetic hot electrons. Thus, much higher energies would have been expected for the red light if the energetic ions are driven by hot electrons and, because of the higher irradiation, the effect should have been even greater. Similar results could be derived from other data. This seems to indicate that contrary to previous suggestions that the collision with hot electrons is not a dominant process in the generation of fast ions [11].

6) Thermal Mechanism

After excluding the 'hot' electron hypothesis, conclusions can be drawn from the above information [19] concerning a thermal mechanism. This process is only relevant to explain the 3-10keV ions since thermal ions in the 100eV range can be expected to be produced in a different way. Figure 2 shows the measured high ion energies for irradiation with fixed wavelength (green, 2Nd harmonic), focusing and intensity. In one case a short 30ps,60mJ irradiation was used and in the second, 100 times longer and more energetic pulses. The result is that the 3-10keV ions have nearly the same energy distribution, despite the higher laser energy.

The analysis of thermal and nonlinear plasmas [21] showed, in line with the gas dynamic models of laser-plasma interactions, that a high energy input results in higher temperatures and in faster thermal expansion. Even if the experimental conditions deviate from the ideal thermodynamic conditions, it is difficult to understand how a change of pulse energy by a factor 100 could result in a less than 20% increase in the quantity of energetic ions.

In practice, the energy of the keV ions depends only on laser intensity. This seems to be an indication of the pulsating interaction mechanism. The 30ps pulses are just sufficient either for ponderomotive filamentation (self focusing) and/or caviton generation [5]. The longer irradiations are divided into consecutive short 20-40ps pulsating interactions each giving similar ion energies. This analysis applies only to energetic ions and not to thermal plasmas.

7) Ion Energies and Charge State

In the preceding section, discussion was based on the fact that the energies of ions up to Ta^{4+} were nearly the same for equal laser intensities although the laser pulse energy, and hence duration, differed by a factor of one hundred. Similar results can be seen in Fig.3 where measured values (table 3.1 of [19]) are presented. However, a significant difference should be noted for higher ionisation states. For those up to 9+, (figures 2 and 3) are comparable (i.e. the ion energy increases linearly with charge state regardless of the irradiation). However, for higher states (up to 18+) ions only appear for the long (3ns) pulse and not at all for the short (30ps) pulse. Additionally, the ion energy is now constant and no longer increases with charge state. The linear dependence has been demonstrated in many laboratories, so the charge independence of the 24keV ions from the 10+ to 18+ charge states must be considered an anomaly.

The data can be explained by the pulsating interaction. As the ion energy (24keV) and linear charge state dependence is the same in both the long and short irradiations, ions up to 8+ are produced by the same short (20-30ps) interaction mechanism. The long pulse process can be explained as a sequence of 30ps interactions in which the interaction stutters along (section 3).

The generation of constant energy higher charge states in long pulses must be due to another mechanism. If the stuttering mechanism is correct, ions will have to pass through the high density plasma produced by the previous interaction. For sufficiently short pulses, obviously there is only one interaction and no dense plasma cloud. It remains to be seen whether the passage of ions through the dense plasma cloud could increase their charge state without a significant change in energy.

Stripping can be excluded since ion energies above 10MeV would be needed in solid targets and gas or plasma targets are even less efficient. It could be asked whether electrons with energies of 100-300eV in the plasma in front of the target could increase the charge state by electron impact ionisation (EII) or related processes. The low mass of the electron would not seriously perturb the ion energy.

The total number of Ta^{q+} ions can be obtained from:-

$$N = n + v.a.t$$

where n is the density of Ta^{q+} ions, v their velocity, a the average cross section of the plasma in front of the target perpendicular to the laser beam axis ($10^{-3}cm^2$) and t is the laser pulse duration (3ns). The density of the Ta^{q+} ions is given by:-

$$n_{Taq+} = S_{Taq+}^{EII} \cdot n_e \cdot n_{Ta(q-1)+} \cdot t'$$

where n_e is the average electron density (assumed to be $10^{18}cm^{-3}$), $n_{Ta(q-1)+}$ the ion density in the initial charge state (e.g. $n_{Ta11+} \sim 10^{15}cm^{-3}$) and t' is the interaction time for ions with the plasma (estimated as 0.6ps). S^{EII} is the ionisation rate obtained by folding the EII cross section σ^{EII} with the Maxwellian velocity distribution of the electrons v':-

$$S^{EII} = < \sigma^{EII}.v'>$$

$$= (8kT/\pi m)^{1/2} \int_{J/kT} \sigma\ (E)(E/kT)e^{(-e/kT)}d(E/kT)$$

where J is the ionisation potential. The EII cross sections for tantalum can be scaled with sufficient accuracy for this exercise from theoretical values of analogous elements [40]. As the exact ionisation potential of Ta^{11+} is not known, it is assumed to be approximately equal to the eigenvalue energy of the 4f subshell which is 35eV [41]. As the average plasma temperature is of the order of 100eV, electron impact ionisation is highly possible. The EII cross section was evaluated using the scaling laws of a given isoelectronic series [40] from the calculated cross sections of Eu^+ [42]:-

$$J^2.\sigma(E) = \sigma_C(X)$$

where $\sigma(E)$ is the cross section for an ion of ionisation potential J for electron energy E, $\sigma_C(X)$ the scaled cross section (the same for all ions in an isoelectronic sequence) and $X = E/J$. This gives an EII cross section of $4x10^{-17}cm^2$ for Ta^{11+}. This value is comparable to those given empirically by the Born approximation [43], the Lotz formula [44] and Donets scaling from EBIS measurements [45]. This cross section gives an estimate of the number of Ta^{12+} ions produced during the laser pulse as $6x10^{11}$ and would seem to indicate that the observed abundances of ions (10^{10}-10^{12})up to 18+ [13] are within the possibilities of EII. The presence of laser radiation in the target plasma will decrease the ionisation potential and modify cross sections [46], but these effects would only slightly change the number of ions which, in any case, is only an estimation.

The charge independence of ion energies show that recombination and electrostatic acceleration do not influence the properties of the energetic ions. Each of these processes would modify the ion energy. Linear charge state dependence for charge states less than 10+ are thus shown to be due to the basic initial mechanism of ion generation and not by space charge, hydromechanical or thermal recombination processes that were claimed (but never proven). The charge state independence above 10+ also excludes thermal and electrostatic paths.

8) Conclusions

Experimental data from laser ion source studies [19,20] were used to analyse basic laser-plasma processes. The data, summarised in figures 1-3 compare fast ion energies (up to 25keV) from tantalum for the carefully selected parameters of equal laser intensity whilst varying pulse duration, energy and laser frequency. According to the hypothesis that energetic ions are accelerated by a 'hot' electron mechanism, the $I\lambda^2$ law would result in much lower energies for shorter wavelengths. The fact that they are nearly equal for much higher power long wavelength pulses excludes the 'hot' electron mechanism.

A further indication that the thermal route cannot produce the measured abundance of 25keV ions from a 100eV plasma is the observation that the much higher energy irradiation on the otherwise same target does not increase the energetic ion energy.

Pulsation (stuttering) can explain that the ion energy is the same for 30ps and 3ns pulse of equal intensity and wavelength. The 3ns interaction being a sequence of shorter pulsating interactions of about 30ps each.

The unexpected result is that there is a range of charge states (10-18+) whose energy is nearly constant at 25keV whereas lower states show a linear relationship. This effect only shows up with long pulses and it is concluded that for the short pulse the pulsation mechanism is predominant. For the long pulses, ions produced in the pulsations are electron impact ionised with little energy change as they traverse the dense plasma in front of the target.

9) References

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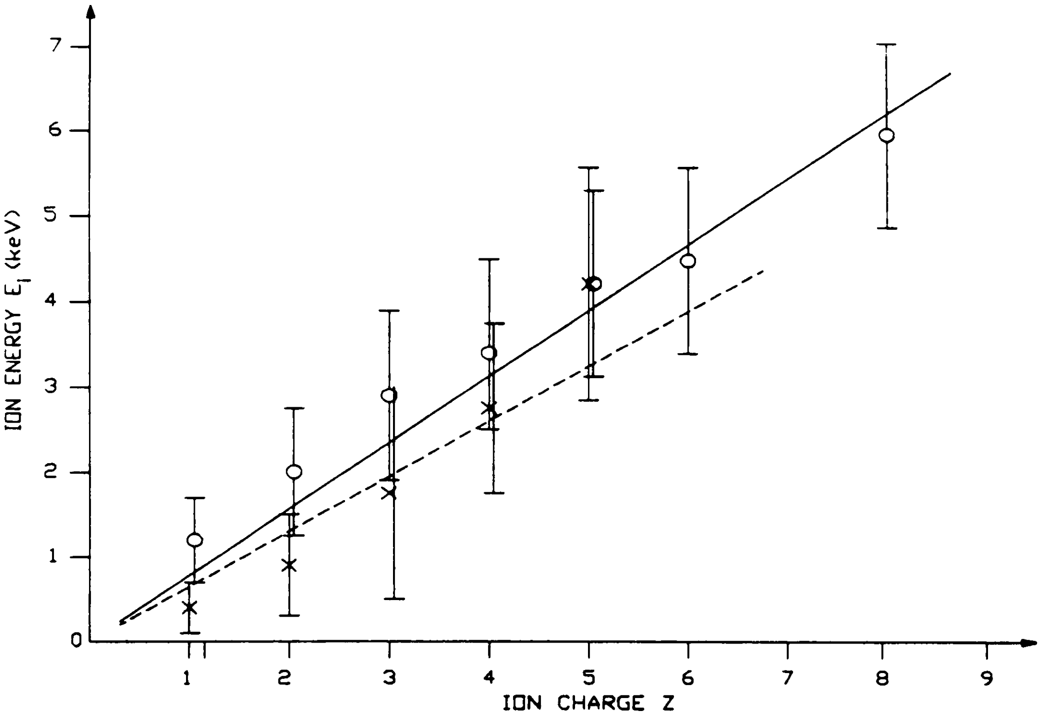

Fig 1. Averaged ion energy of ionisation state Z from tantalum irradiated with 30ps red ° 135mJ and green ^{x} 30mJ Nd-glass laser pulses.

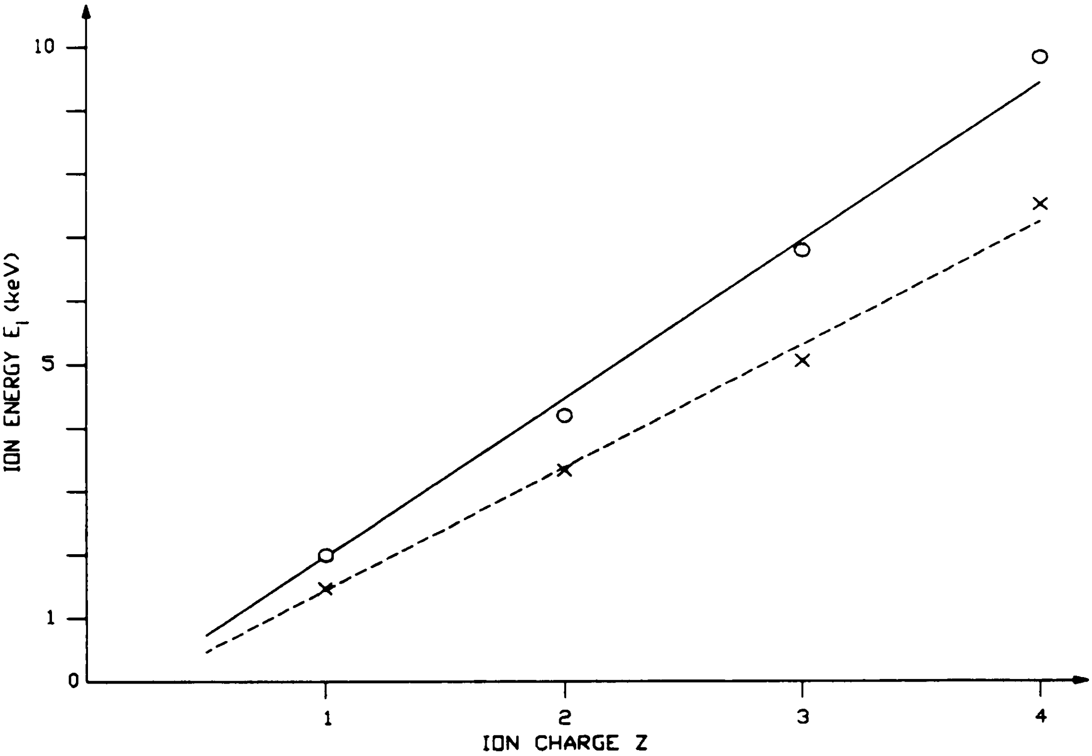

Fig 2. Fast ion energies from a tantalum target of charge state Z for an intensity of 4×10^{13} W/cm^2 green Nd-glass laser irradiation. Pulses 30ps,60mJ ^{x}, 3ns,6J °.

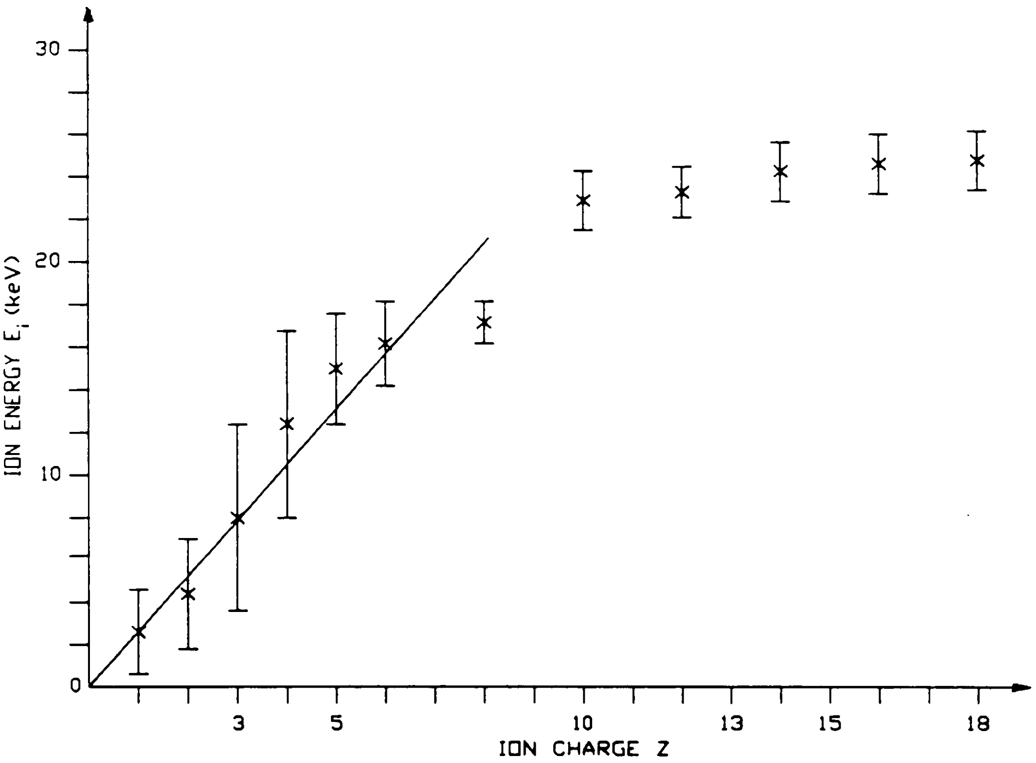

Fig 3. Ion energies and charge state Z for green Nd-glass laser pulses of 3ns, 6J showing linear increse to Z=10 and constant thereafter.