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Laser-driven inner-shell excitation in high-Z atoms: A shell-selective impact ionization mechanism

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Abstract: A highly selective, *coherent* impact ionization mechanism is proposed for the efficient generation of inner-shell population inversion in laser-driven plasmas. The theoretical analysis is consistent with observed L-shell (2p←3d) emission spectra from laser-excited Xe clusters.
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The efficient generation of inner-shell population inversion in high-Z elements (Z > 30) is the most direct route to the development of an atom-based coherent multi-kilovolt x-ray source (i.e. a ~1Å x-ray laser). In this paper, we describe a shell-selective, *coherent* impact ionization mechanism which is capable of providing a required pumping rate in excess of 1W/atom for the generation of inner-shell population inversion in laser-driven plasmas. The evidence for the existence of such a mechanism is contained in the Xe L-shell (2p←3d) emission spectra (Fig. 1) obtained under laser excitation of 5-20 atom Xe clusters at irradiances of 10^{18}-10^{19}W/cm^2 [1,2]. The spectra display three striking features: (i) the generation of "hollow" atoms [1], including the *inverted* ions $Xe^{27+}(2p^5 3d^{10})$ and $Xe^{28+}(2p^5 3d^9)$, (ii) a ~1000x reduction in the efficiency of Xe(L) emission as the pump-laser wavelength is increased by a factor of ~3 from 248nm to 800nm [2] and, most significantly, (iii) the observation of ionic species with *double* 2p vacancies only under 248nm excitation.

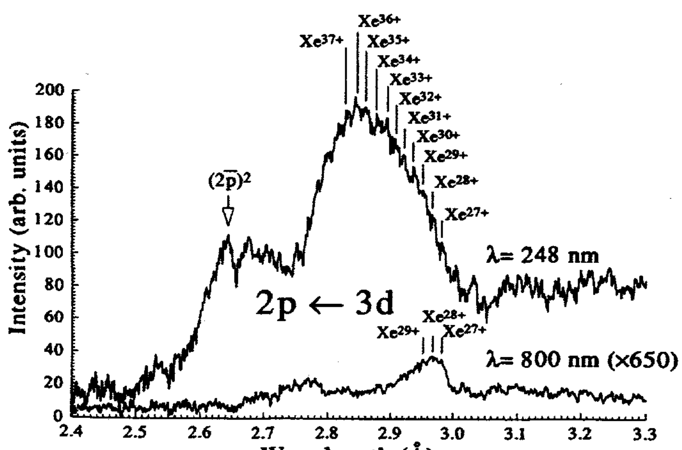

Fig. 1. The kilovolt Xe L-shell (2p←3d) x-ray spectrum emitted by 5-20 atom Xe clusters excited by TW ultraviolet (248nm) and infrared (800nm) sub-picosecond laser pulses. The feature at 2.6-2.65Å is due to a *double* 2p vacancy.

Under the experimental conditions, the observed 2p vacancies cannot be produced by direct photoionization and so must be generated by a collisional ejection process: but *not* by a plane-wave mechanism which would predict the ejection of all 3d electrons before a single 2p vacancy is produced. Instead, we propose that free-electron wavefunctions photoionized by above threshold ionization (ATI) retain the phase and geometric symmetry of their parent bound state – the "sudden" approximation. For Xe at 10^{18}-10^{19}W/cm^2, one of the last photoionized states is the 4p state which, in the absence of dephasing scattering events, will then undergo *coherent* intrinsic quantum mechanical (radial) spreading with a characteristic time $\tau \approx 0.04$fs as it is accelerated by the ponderomotive laser potential. Since the rate of expansion of the ionized state exceeds its internal collisional

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(electron-electron) dephasing rate, the matrix element M_{fi} associated with impact ionization due to the coherent returning 4p electrons exhibits a strong angular momentum (i.e. $l = 1$) selectivity, predominantly ejecting p-state electrons from Xe ions in the parent cluster. Moreover, evaluation of the collision cross-sections (i.e. $|M_{fi}|^2$) for the strongest Coulomb interaction (Fig. 2) reveals (i) an additional strong n-state selectivity in the interaction favoring the ejection of 2p over 3p electrons by a factor of ~1000 and (ii) a spreading-induced factor of ~300 reduction in the cross-section as the pump-laser wavelength is increased from 248 to 800nm – both in agreement with the experimental data (Fig. 1).

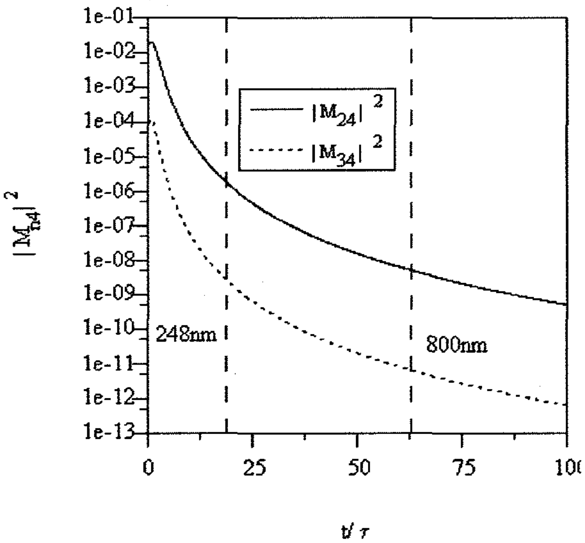

Fig. 2. The modulus square of the calculated interaction matrix element, M_{fi}, as a function of the normalized expansion time t/τ for the collisional ejection of a 2p (solid line) and a 3p (dashed line) in Xe by a photoionized 4p state with a typical incident collision energy of 15keV per electron. The vertical lines indicate the periods and, thus, approximate collision times for 248nm and 800nm irradiation.

The observation of *double* 2p vacancies under 248nm excitation provides further strong evidence for this efficient, shell-selective, *coherent* impact ionization mechanism. The spin-invariant Coulomb interaction allows the coherent action of an ionized anti-symmetric spin-pair state to eject both electrons in the corresponding m-state of the target Xe ion; a selective interaction caused by strong laser-field-induced Stark effects. The absence of double 2p vacancies under longer wavelength 800nm excitation (Fig. 1) is a direct consequence of the fact that a spin-pair state in the ionized wavefunction will be dephased by scattering events at *twice* the rate of a single-electron state.

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