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Time-resolved investigation of transient charges in laser-produced nanoplasmas

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Synopsis We report on the observation of a transient C^{4+} ion charge state in nanoplasmas produced by the interaction of intense near-infrared (NIR) laser pulses with CH_4 clusters. The underlying dynamics are studied by pump-probe spectroscopy, which reveals that the ion charge states are lowered by electron-ion recombination. Furthermore, we present direct evidence that autoionization of multiply-excited ions plays an important role in expanding nanoplasmas, in contrast to models that neglect quantum phenomena.

In recent experiments, transient charge states were observed following the interaction of intense extreme-ultraviolet (XUV) pulses with rare-gas clusters using fluorescence spectroscopy [1, 2]. The results were explained by the generation of highly charged ions that recombine with electrons in the nanoplasma after the XUV laser pulse has ended. In order to reveal the underlying processes involved in the nanoplasma relaxation dynamics, we have developed an intense XUV source based on high-order harmonic generation, and we have shown that highly excited Rydberg atoms are formed in the nanoplasma by charge recombination [3]. We were able to time-resolve these dynamics, and we demonstrated that similar processes take place following XUV and NIR ionization of clusters [4, 5].

Here we study the interaction of intense 32 fs NIR laser pulses with CH_4 clusters, where we observe the formation of a transient C^{4+} ion contribution in pump-probe experiments that is hardly visible in the ion time-of-flight (TOF) spectrum obtained by the pump pulse only (Fig. 1(a)). In the electron spectrum, we find a peak at ≈ 1 eV (not shown) that is explained by the reionization of excited states following charge recombination using the time-delayed probe pulse.

When using 400 fs NIR pump pulses at the same intensity, the cluster becomes strongly ionized, and exhibits a dominant C^{3+} ion contribution (Fig. 1(b)). Nevertheless, the C^{4+} ion yield using the pump pulse only remains small. It is strongly enhanced though by the time-delayed weak probe pulse, demonstrating that it results from reionization of a C^{3+} Rydberg state that has been formed by charge recombination of C^{4+} that must have been transiently formed during the cluster excitation. Furthermore, the electron spectrum in Fig. 1(c) exhibits a peak at an energy of 7 eV that is surprisingly identified as electron emission following autoionization of doubly-excited C^+ and C^{2+} ions.

Our results clearly demonstrate that charge recombination is responsible for the observation of transient charges in nanoplasmas. Moreover, the sharp ion charge state distribution that is dominated by a C^{3+} peak can be explained by extensive autoionization of multiply-excited atoms [6] and ions. The results are in stark contrast to existing models of nanoplasmas that treat electrons and ions as classical particles.

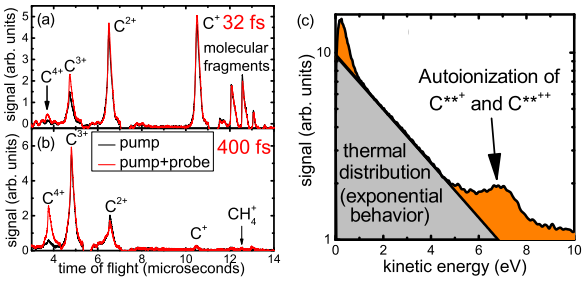

Figure 1. Ion TOF spectra from CH_4 clusters obtained using a 1×10^{14} W/cm^2 NIR pulse with a pulse length of (a) 32 fs and (b) 400 fs. When using an additional weak NIR probe pulse (I=5 $\times$ 10^{12} W/cm^2) at a time delay of 50 ps, the C^{4+} ion signal is significantly increased. (c) The corresponding electron spectrum using the NIR pump pulse exhibits a clear autoionization peak.

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