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Mestastable State Population in Laser Induced Plasmas

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ABSTRACT

Laser induced plasma has been used as a source of neutrals and ions in the study of astrophysical plasmas. The purity of state of this source is essential in the determination of collision parameters such as the charge transfer rate coefficients between ions and neutrals. We will show that the temperature of the laser induced plasma is a rapidly decreasing function of time. The temperature is initially high but cools off rapidly through collisions with the expanding plasma electrons as the plasma recombines and streams into the vacuum. This rapid expansion of the plasma, similar to a supersonic jet, drastically lowers the internal energy of the neutrals and ions.

1. Introduction

Laser induced plasma has been used as a source of neutrals and ions. The purity of state of this source is essential to the measurement of collision parameters such as the charge transfer rate coefficients between ions and neutrals that are used in the modeling of astrophysical plasmas. We will address the purity of state by reviewing theoretical and experimental evidence on the expansion dynamics of laser induced plasmas. Furthermore, we will also examine several pieces of experimental evidence available to us that can review the metastable fractions in both the neutrals and ions in the laser ablation source.

2. Charge transfer of O^{2+}, C^{2+} and H_2

Laser induced plasma was used as an ion source for O^{2+} in the measurement of the charge transfer rate coefficient for O^{2+} ions and H_2 in an ion trap (Fang and Kwong 1995). O^{2+}, however, has several low lying metastable states. The $2p^2\ ^1D$ metastable state, for example, is 2.5 eV above the $2p^2\ ^3P$ ground state. The mean lifetime of this state is 37s, which is much longer than the storage time during the measurement. Significant amounts of

the metastable ion, if they are present in the laser induced plasma, can also be stored in the trap and react with the H_2 target gas. The measured rate coefficient can therefore reflect the convolution of two independent charge transfer processes involving the ground state and the metastable state O^{2+} ions. The contribution of the metastable state ions can be assessed by comparing our measurement with the known charge transfer rate coefficients of the ground state and the metastable state.

The ground state and the metastable state rate coefficients have been measured by Church and Holzscheiter (1989). Their rate coefficients are 1.71 ± 0.15 x 10^{-9} and 9.6 ± 0.6 x 10^{-9} for the $2p^2$ ^3P ground state and the $2p^2$ ^1D metastable state respectively. Our measured charge transfer rate coefficient of 2.36 ± 0.22 x 10^{-9} is within 30% of the measured value for the $2p^2$ ^3P ground state. This suggests that the ions produced by laser ablation and stored in the trap are mainly in the ground state.

In the charge transfer cross section measurement of C^{2+} and H_2 with a reflection time of flight mass spectrometer, we examined the metastable content of the laser ablation ion source $20\mu s$ after the ions were produced. Metastable state ions with lifetimes greater than $20\mu s$ can be present in significant amounts since the electron temperature of the laser induced plasma is expected to be very high during laser ablation.

C^{2+} has low lying $2s2p$ $^3P_{0,1,2}$ metastable states with energy ≤ 6.5 eV above the ground state. These low lying metastable states have a charge transfer cross section about 6 times larger than that of the ground state (Unterreiter et al. 1991). The $2s2p$ $^3P_1$ metastable state lifetime was measured to be 8.26ms (Kwong et al. 1993) and the $2s2p$ $^3P_{0,2}$ metastable states lifetimes are 10^4 times longer. If these metastable state ions are present, changing the population ratio between the metastable state and the ground state will alter the measured cross section significantly. The mean electron temperature of the laser induced plasma is raised from 9 eV to 12 eV by increasing the energy of the ablation laser from 10 mJ to 20 mJ. The estimated population ratio of the metastable state to the ground state should increase from 0.5% to 1.9% (Wang 1997). Because of that change in ratio, one would expect to see the observed charge transf er cross section increase by as much as 60%. Our measured cross section, however, remains unchanged (6.90 ± 0.78 x 10^{-16} cm^{-2}) within the experimental uncertainty (see Fig.1). This suggests that the internal temperature of the laser ablation ions must be cold and the metastable ion fractions in the beam are negligibly small (Wang and Kwong 1997).

3. Expansion dynamics of metastable fractions in laser induced plasmas

This unique characteristic of low metastable fractions in both neutrals and ions in the laser induced plasma can be explained by the cooling of plasma electrons in the rapidly expanding plasma (Rumsby and Paul 1974). During laser ablation, a hot and high density plasma is formed and expands rapidly into the vacuum. The estimated initial density is

$\approx 10^{21}$ cm^{-3}. At such a high density, the collision time between ions and electrons is orders of magnitude shorter than the laser pulse. Local thermodynamic equilibrium is established within the time duration of the laser pulse. The temperature of the plasma has been estimated to be in excess of 10^5 K (Kwong 1979). At such a high temperature, we can safely assume complete dissociation and ionization of the material inside the ablation plume.

The internal temperature of the atomic and ionic species, however, is closely coupled to the temperature of the plasma electrons through rapid collision. Three body and radiative recombination occur with the formation of neutral atoms from plasma ions. As the temperature of the plasma electrons drops, their initial energy is converted into directed energy of expansion. The internal temperature of the atomic and ionic species follows and decreases as well. The time dependence of the electron density, n_e, and electron temperature, T_e, is proportional to t^{-3} and t^{-1} respectively (Rumsby and Paul 1974). Therefore the internal temperature of the atomic and ionic species freezes out when n_e drops below the threshold density to maintain collision equilibrium.

This rapid decrease of electron temperature in a laser induced plasma has been demonstrated by Drewell (1979) in plasma diagnostic experiments seeded with neutral Cr. In Drewell's experiment, Cr plasma was produced through laser ablation of a pure Cr target. The population ratio between the a^{5}S$_2$ metastable state and the a^{7}S$_3$ ground state of neutral Cr was measured by the intensity ratio of the laser induced fluorescence linked to these states. The a^{5}S$_2$ metastable state and the a^{7}S$_3$ ground state have an energy difference of 0.94 eV. The time dependence of the population ratio is shown in Figure 2 (Drewell, 1979). The population ratio freezes out 4μs after laser ablation. This ratio is $\approx 10^{-3}$, which gives a freeze-out temperature of $\approx$1000K.

4.   Conclusion

Since the power density of the ablation laser used in our measurement is similar to that used by Drewell (1979), we expect that the internal temperature of the ions and neutrals in our laser ablation ion source to be similar. At this low temperature, it is highly unlikely that the metastable state atoms with energy greater than 1 eV above the ground state will be present in a measurable quantity in the pulsed atom or ion beam. This is consistent with all our measured rate coefficients and cross sections which correspond to the ground state. On the other hand, the freeze-out temperature of the plasma electron ($\approx$1000 K) limits the purity of state of stored ions and neutrals with low lying metastable states $\leq$ 0.4eV above the ground state.
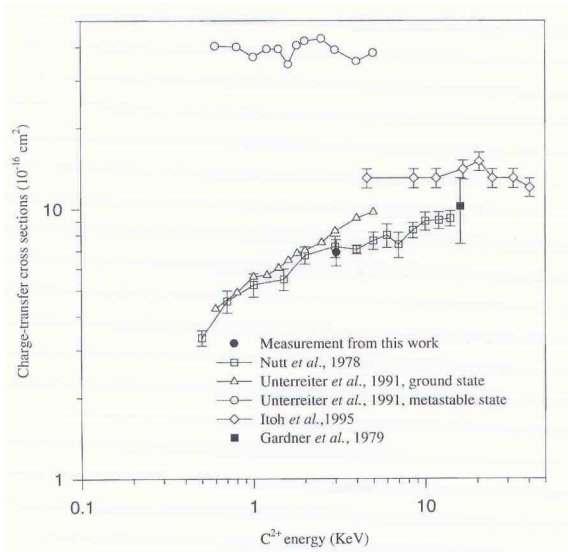
FIG. 1. Single electron charge-transfer cross sections for $C^{2+} + H_2$
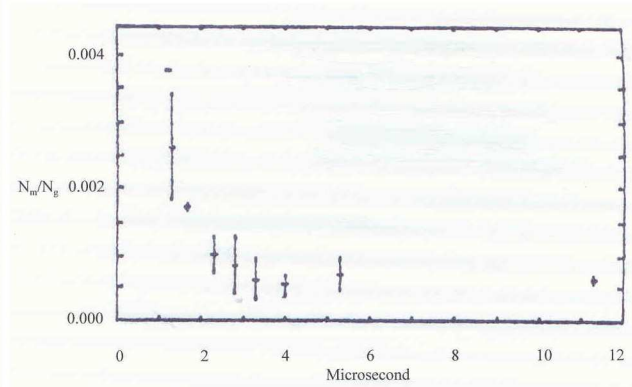at low energies reported by several research groups.

Fig. 2. Population ratio between the a^5S_2 metastable state and the a^7S_3 ground state of
neutral chromium in laser produced plasma after laser ablation.

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