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Ultracold Neutral Plasma Produced by One-Color Two-Photon Ionization in Supersonic Nitric Oxide Molecular Beam *

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Nitric oxide (NO) is cooled to 1 K in a seeded Ar supersonic molecular beam. NO molecules are ionized by one-color two-photon resonant enhanced multiphoton ionization (REMPI) process to form an ultracold plasma. The density of the ionized molecules in the illuminated volume approaches to 1.5×10^{13} cm^{-3}. Prompt electrons, plasma electrons and intact NO^+ ions are produced during this process. Experimental results confirm that the lifetime of this ultracold plasma is longer than 18.3 μs. This is the first report of NO ultracold plasma with significant lifetime produced by one-color two-photon REMPI process.

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Normal plasmas contain molecular ions as well as atomic ions, and molecular degrees of freedom affect their properties. Recent techniques on atomic Rydberg gases and ultracold plasmas owe a great deal to development of technology for producing cold atoms and atomic condensates in a magneto optical trap (MOT).[1−12] Researchers[13−15] including our group[16,17] have reported the method to create ultracold plasma in a supersonic nitric oxide (NO) molecular beam.

In this Letter, we present a new experimental approach about one-color two-photon resonant enhanced multiphoton ionization (REMPI) process to generate ultracold neutral plasma in a supersonic molecular beam for the first time, which is valuable for research of strongly coupled phenomena.

The experimental scheme is shown in Fig. 1, a pulsed dye laser (Pulsare Excellent 2400) was pumped by the third harmonics of Nd:YAG laser. The YAG laser was operated at 20 Hz with a pulse width of 7 ns. Either cations or electrons could be detected using time-of-flight (TOF) spectra with different-extraction-voltage polarity applied. Signals were sampled by an oscilloscope, and an SRS DG645 was used to control the time sequence of general valve, Nd:YAG laser pulse and pulsed electronic fields.

A mixture of high purity NO and Ar buffer gases with backing pressure of ∼3 bar was introduced to the source chamber through a general valve with a nozzle of 0.5 mm, and a skimmer was placed at 80 mm downstream from the valve to form differential pumping between the source chamber and the detect chamber, as shown in Fig. 1. A series of extraction plates were located in the detect chamber (with 30 mm separation between adjacent plates). Each extraction plate had a hole in its center and was covered with a copper grid to form a well defined electric field; these grids could be either grounded, or applied with a pulsed voltage. There was a field-free region of about 0.5 m between the last extraction grid and the grounded grid in front of MCP, which was similar to the methods in Ref. [17].

Figure 2 shows the one-color two-photon REMPI spectrum of the NO molecule where laser crossed with the molecular beam 20 mm downstream of grid 1 (shown in Fig. 1). The NO^+ signal was monitored as a function of the excitation laser frequency ω, and a rotationally resolved REMPI spectrum of the $v = 0$ band of the $A^2\Sigma^+$ state could be obtained. By this REMPI spectrum, the rotational temperature of NO in any position could be estimated according to the Maxwell–Boltzmann distribution:

$$N\% = (2N+1)\exp\left\{-\frac{-(B_e - 0.5\alpha_e) \times N(N+1)}{1.4413T}\right\}, \quad (1)$$

where B_e is the rotation constant, α_e is vibration coupling constant, N is the rotational quantum number, and T is the rotational temperature.[18]

All the three peaks in this spectrum originate from the $N' = 1$ state of the $X^2\Pi$ ($v'' = 0$) state. The signal-to-noise ratio (SNR) of the REMPI spectrum is about 200; if the population of rotational excited state is smaller than 0.5%, the signal via those states could not be detected. When the rotational temperature is 1 K, the population of rotational excited state will be about 1.4% according to Eq. (1). Therefore, it is concluded that the rotational temperature is cooler than 1 K at this position. Supersonic expansion cools molecular rotation as well, while the cross section for rotational relaxation is smaller than that for translation. As a result, when collisions cease, the terminal

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rotational temperature exceeds the transitional temperature, that is, $T_{//} < T_R$, so the translational temperature is lower than 1 K.

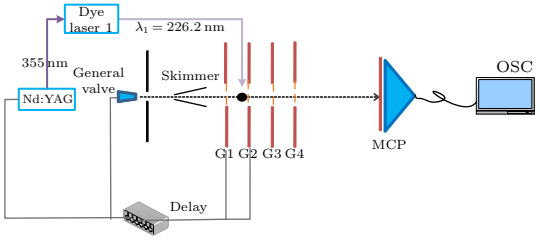

Fig. 1. Experimental scheme showing the molecular beam flight path. Ions (electrons) flew from the differentially pumped source chamber through a skimmer to a system of four grids, and were then detected by a micro-channel plate (MCP). The laser beam was introduced to vacuum chamber and photoionized NO molecules at 20 mm downstream of grid 1.

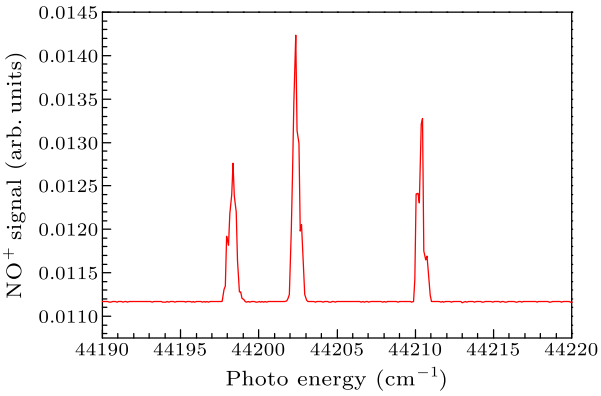

Fig. 2. Resonant 1+1 ionization spectrum of NO seeded 1:8 in the supersonic beam of Ar via $A^2\Sigma^+$ ($v' = 0$) state. The intensities fit a ground-state population distribution corresponding to a rotational temperature $T_R < 1$ K.

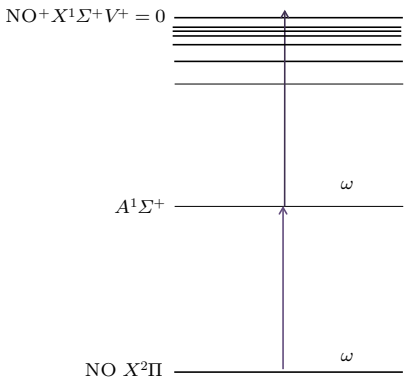

Fig. 3. Excitation scheme of NO by one-color resonant transitions via the $A^2\Sigma^+$ state.

When the wavelength of laser is tuned to 226.2 nm (44202 cm^{-1}), NO^+ signal will reach the maximum. This should correspond to the maximum ionization cross section, and this wavelength was used throughout our experiments. Figure 3 shows the energy diagram for excitation of NO. A laser of 226.2 nm pumps the NO molecules from the ground state ($X^2\Pi$) to rotational substates of the $v = 0$ vibrational level of the first electronically excited state, $A^2\Sigma^+$. Then the $A^2\Sigma^+$ NO absorbs the second photon and is ionized.

In this experiment, a pulsed voltage of −1200 V was applied on grid 1 while a −921 V pulsed voltage was applied on grid 2 (Wiley-McLaren TOF scheme[19]). The pulsed voltage was adjusted to synchronize with laser radiation. The electrons' TOF signal was collected by an MCP and recorded by an oscilloscope, which was exemplified by Figs. 4 and 5. Figures 4 and 5 show the TOF spectra of electrons with time delay between laser radiation and pulsed extraction voltage being 0 and 1.3 μs, respectively. This waveform consists of prompt electron signal (P1), followed by a broader pulse of plasma electron (P2).

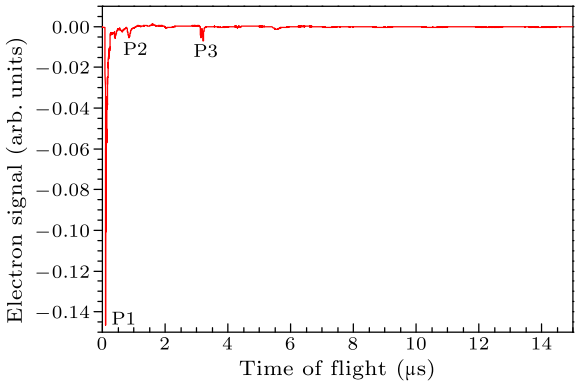

Fig. 4. TOF spectrum of prompt electrons and plasma electrons when supersonic NO beam is ionized by one-color two-photon process, and there is no time delay between laser radiation and pulsed extraction voltage.

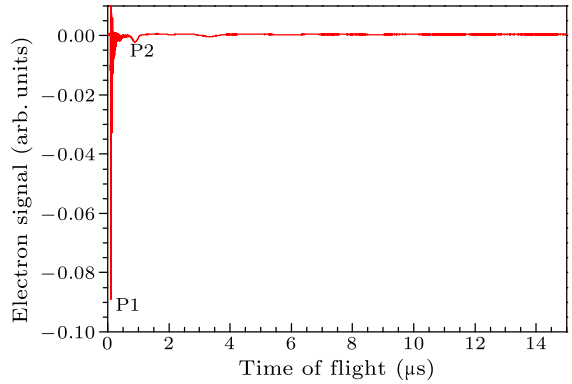

Fig. 5. TOF spectrum of prompt electrons and plasma electrons with time delay of 1.3.μs between laser radiation and pulsed extraction voltage.

The electro-ionization discrimination field equation in the adiabatic process is

$$6.5\sqrt{F} \geq E_n, \quad \left(E_n = \frac{R}{n^2}\right) \tag{2}$$

where F is the discrimination field; R is the Rydberg constant and n is the principal quantum number.[5] The pulsed voltages applied on the two grids are −1200 and −921 V, the pulse field F = 93 V/cm,

which could ionize Rydberg NO with $n > 40$. Therefore, the high-Rydberg NO molecules are ionized immediately after the pulsed voltage applied on the extraction grids. The lifetime of Rydberg state NO varies with n^5,[20,21] the low-Rydberg-state NO could relax to the ground state very quickly, while high-Rydberg NO could be ionized in a few nanoseconds when an electronic field is applied, so P2 could not be due to Rydberg electron either. Therefore, P2 must be plasma electron. The M/C ratio of P3 is 11.18. There were no reports about anion generation under similar experimental conditions. The origin of P3 is still unclear; it might be due to the electrons from the bombardment of ions onto the copper grid.

Around P2, there are some weak peaks; they are likely signals due to different velocities of electrons at the moment the plasma formed. As the delay time between laser radiation and pulsed extraction voltage increases, kinetic electrons would escape from plasma, and could not be detected, therefore 1.3 μs after laser radiation, there are no such peaks around P2 (shown in Figs. 5 and 6).

Researchers have used the two-color two-photon REMPI process to produce ultracold plasma consisting of prompt electrons and plasma electrons, and the lifetime is longer than 40 μs.[13–15] We also produced ultracold plasma with significant lifetime by one-color two-photon REMPI process for the first time. In our experiment, there were no direct means to measure the absolute density of excited NO molecules. From the nozzle diameter and backing pressure, the initial centerline density of particles in the molecular beam was estimated to be $8 \times 10^{19}\,\mathrm{cm}^{-3}$. The density of molecule beam at laser irradiation area should be more than $9.46 \times 10^{14}\,\mathrm{cm}^{-3}$ according to

$$\frac{\rho}{\rho_0} = \left(\frac{1}{1 + \frac{1}{2}(\gamma - 1)M^2}\right)^{\frac{1}{\gamma - 1}}, \tag{3}$$

where ρ is the molecular density and ρ_0 is the initial molecular density; M is the Mach number, $M = A(x/d)^{\gamma-1}$ with $A = 3.26$.[22] The seeding ratio of NO is 0.125. At rotational temperature of 1 K, about 98% of these molecules populate the two parity components of the rotational ground state according to Eq. (1). Because at the interaction region the number of photons are much more than NO molecules, this saturated excitation makes 25% of NO molecules be excited to the $A^2\Sigma^+$ ($v'' = 0$) state, and half the NO molecules in the $A^2\Sigma^+$ state would be ionized by the second photon.[13] In combination, these factors predicting the density of ionized NO molecules available for plasma are about $1.5 \times 10^{13}\,\mathrm{cm}^{-3}$. On the assumption that the electrons are all fast electrons with the kinetic energy of around 1.7 eV (19000 K), the plasma Debye radius is[5,16]

$$\lambda_D = \sqrt{\frac{\varepsilon_0 K_B T_e}{e^2 \rho_e}} = 69\sqrt{\frac{T_e}{\rho_e}} < 3\,\mu\mathrm{m},$$

which is much smaller than the laser irradiating volume (0.4 mm, the initial plasma volume), so the plasma was generated right after the photo ionization process. Here K_B is Boltzmann's constant, T_e is the temperature of electrons in units of Kelvin, and ρ_e is the density of electrons in units of m^{-3}.

Figure 6 shows the TOF spectra of pump electrons and plasma electrons when the delay time between laser radiation and pulsed electronic field is adjusted to 0.3 μs, 1.3 μs, 5.3 μs, 8.3 μs, 13.3 μs and 18.3 μs (there is 2 μs offset time between adjacent TOF spectra for clarity). The signals of 8.3 μs, 13.3 μs and 18.3 μs are multiplied 6, 10 and 10 times separately for clarity. It could be observed that the plasma electrons exist for 18.3 μs, and after 18.3 μs there is little electron signal detected. If the photoelectrons produced by the one-color two-photon process all are fast electrons (with a speed of about 18000 m/s), they would leave the excitation region within 100 ns. Therefore the lifetime of plasma generated by the one-color two-photon process is expected to be less than 100 ns scale. However, in fact, electrons from NO by the one-color two-photon process could be divided into three groups:[23–26] fast electrons with kinetic energy of about 1.7 eV (∼20000 K), slow electrons with nearly zero kinetic energy (<0.1 eV[25,26]) and Rydberg electrons. Slow electrons and Rydberg electrons contribute to the longer lifetime of plasma.

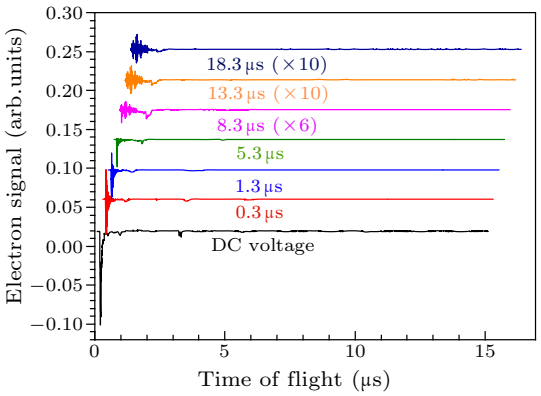

Fig. 6. TOF spectrum of prompt electrons and plasma electrons with different time delays between laser radiation and pulsed extraction voltage.

In the previous experiment,[13–15] a two-color two-photon REMPI process was used to excite NO molecules to a selected high-Rydberg state below the ionization threshold.[9,10] The expansion rate of ultracold plasma depends principally on the kinetic energy of its electrons.[11] Pohl and co-workers recognized the collisional ionization of Rydberg atoms

as a cooling mechanism for electrons in ultracold plasma.[28] Therefore, in the experiments of Refs. [13–15], the initial electron temperature could be maintained in a range of a few tens of Kelvin, the lifetime of plasma was longer than 40 μs. Under one-color two-photon ionization conditions, the majority of electrons were fast electrons, and there were also portions of slow electrons and Rydberg electrons. Fast electrons would evaporate and escape from plasma in tens of a nanosecond and cause partial charge imbalance, therefore the expansion of plasma was quite fast. More ions escaped from the plasma than electrons; this process would not continue until the number density of electrons and ions were almost the same. After sometime longer than 18.3 μs, the plasma criteria were no longer satisfied, therefore there were no plasma signals detected. Because the number density of slow electrons plus Rydberg electrons was smaller in comparison with the experiments of Refs. [13–15], the lifetime was shorter than plasma evolution from Rydberg gas.

In summary, the ultracold NO plasma has been produced by a one-color two-photon REMPI process for the first time and the lifetime is longer than 18.3 μs. This lifetime is significant shorter than the experiments of Refs. [13–15]. This is because the one-color two-photon REMPI process produces a large population of fast electrons and a fraction of slow electrons (including Rydberg electrons); fast electrons would cause the NO ultracold plasma to expand and lifetime to be shorter than the two-color two-photon REMPI process.

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