Ultrashort pulse laser ionization of ions in a plasma

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(Received 16 January 1998; revised manuscript received 4 May 1998)

Theoretical calculations are presented that show that optical field ionization of an ion in a plasma medium, induced by an intense ultrashort laser pulse, is sensitive to the plasma screening of the binding potential of the target ionic electrons by surrounding plasma electrons. Hence experimental measurements of ionization rate versus plasma electron density would provide a direct test of theoretical plasma models for hot, dense matter. [S1063-651X(98)12909-4]

PACS number(s): 52.25.−b

I. INTRODUCTION

In this paper theoretical calculations are presented that demonstrate a new application of optical-field ionization (OFI) induced by an intense, ultrashort laser pulse. When OFI is carried out in a target plasma whose density and temperature are well characterized in a prior backablation step [1,2], then it is possible experimentally to control the laser intensity, which governs the barrier width against OFI [3,4] of a target ion in the plasma, and the plasma density and temperature, which govern the screening length of the effective potential binding the target ionic electrons, such that barrier width and screening length have comparable length scales. The exponential dependence of the OFI tunneling ionization rate on barrier width ([3,4] and Fig. 1) causes a slight change in barrier width due to screening by plasma electrons to significantly affect the ionization rate. We demonstrate this effect by calculating the OFI rate as a function of screening length according the simple Debye-Huckel model [5].

Greater interest, however, lies in regimes of the plasma coupling parameter, $\Gamma = e^2/r_a kT$ [where r_a is the radius of the average ionic sphere, $r_a = (3/4\pi n)^{1/3}$, for an ion density n and plasma temperature T], in which more sophisticated theoretical plasma models [6–9] are appropriate, namely for $\Gamma > 1$. The dependence of more accurate theories of ionization rate (as opposed to the simple tunneling model used in this paper) on plasma-electron density (for a given plasma temperature and laser intensity) should also be invoked to interpret experimental measurements and thus provide a test of the plasma models. Existing experimental tests of theory are based on absorption spectroscopy in a plasma whose density and temperature are well characterized [10].

II. OUTLINE OF THE EXPERIMENT

Neutral high-density metal vapor targets can be fabricated using the laser-induced backablation technique (LIBA) [1,2]. It has been demonstrated that with appropriate masking a highly localized thin slab geometry can be produced. The waveform corresponding to the incident laser pulse can be approximated as a plane wave when the target slab depth is less than the Rayleigh range defined by the focusing optics. Refractive off-axis scattering of laser energy can be suppressed in this way. Mg is a suitable choice of metal substrate for LIBA-produced vapor targets. This is because within the laser intensity range of interest ionization would be limited to the production of Mg^{+1} and Mg^{+2} ions.

OFI rapidly reduces the refractive index within the focal volume of the pulse, causing electron density and refractive-index gradients. The time dependence of the index change follows the time dependence of the rate of growth of plasma electrons generated during this interaction and is manifested as a spectral modulation of the laser waveform predominantly toward shorter wavelengths (blueshifts) [1,14].

It has already been demonstrated [1,2] that the use of polarization-gated frequency-resolved optical gating (PGFROG) can provide time-dependent spectral shift measurements with femtosecond resolution. With this diagnostic it is possible to determine a time-dependent comparison between measured spectral shifts and those predicted by OFI rates. This can be done in both single and double-pulse experiments. For this paper we consider the double-pulse case where preionization is set by the first pulse and can be lim-

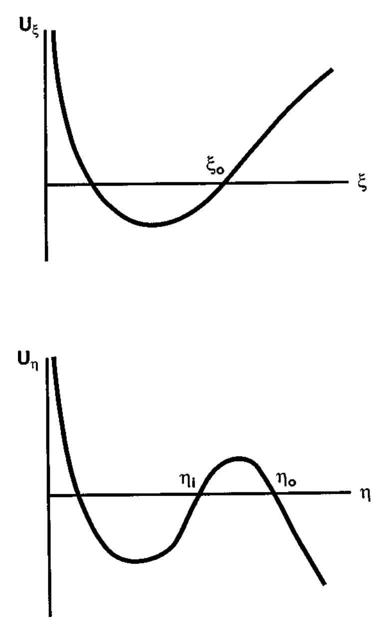

FIG. 1. Typical representation (Ref. [15]) of the unscreened effective potentials in parabolic coordinates $\xi = r + z$ and $\eta = r - z$ for a bound electron in the presence of the electric field realized at a single amplitude and phase of a laser pulse. Then η potential (lower) shows a barrier against ionization.

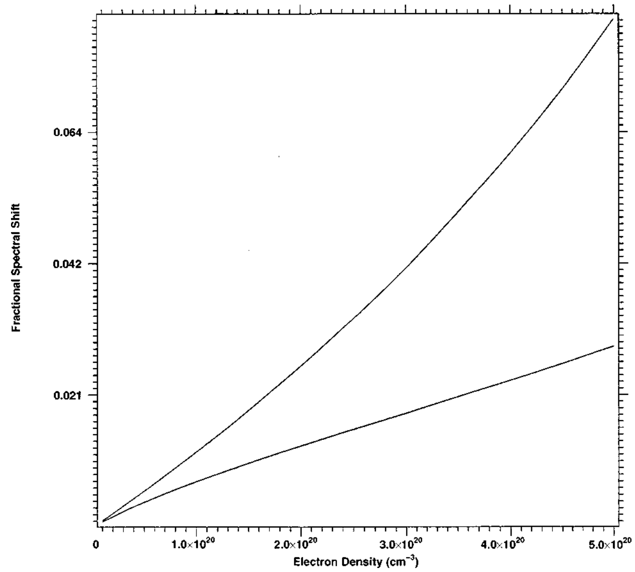

FIG. 2. Fractional frequency shift versus plasma-electron density for the cycle-averaged, hyperbolic-secant-pulse model for an intensity of 1.7×10^{14} W cm^{-2}. Upper curve, unscreened potential; lower curve, screened potential. The nonlinear behavior at higher densities reflects the approach to critical density.

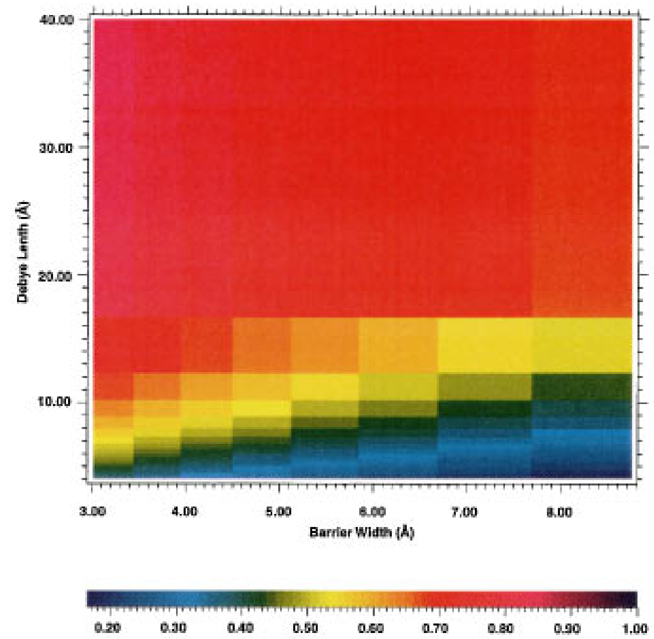

FIG. 3. (Color) Ratio at peak laser field strength of screened to unscreened ionization rates vs Debye length (vertical axis) and barrier width (horizontal axis) measured in angstroms.

ited to production of Mg^{+1} as a target for the second pulse. One could envisage a series of double-pulse measurements in which the preionization step prepares the target plasma at the desired levels of plasma density.

The temperature of the target plasma is expected to be low because laser-induced tunneling ionization is known to produce a cold electron distribution. In fact, we can estimate the electron temperature from calculated photoelectron energy distributions [11]. For example, from Fig. 2(c) of [11], 90% of the electrons have an energy below 10 eV. To produce a similar distribution in the first ''preionization'' step one should use an 800 nm, 1.0×10^{13} W cm^{-2} pulse. We arrive at this estimate for the laser intensity by scaling results of [11] according to the cube of the ionization potential [12,13]. We estimate an average electron energy of a few eV.

The following considerations are also important for the design of such experiments. The first group of electrons produced in the preionization stage are expected to possess a thermalized energy in the range of a few eV before the arrival of the second pulse. When the second pulse arrives it is not expected to increase the temperature of the first group of electrons; there will be a quiver energy along the direction of the laser polarization but this energy is not expected to randomize because at the relevant temperature and density the collision time for randomization is expected to be longer than the duration of the second pulse.

Finally we assume that for the duration of the second pulse the screening is produced only by the first group of electrons and is static. In other words, electrons generated by the second pulse also quiver along the polarization direction of the laser; however, they are born at a laser intensity about an order of magnitude higher than that of the first pulse with insufficient time for energy randomization so that they are not expected to contribute to Debye screening.

In the absence of plasma screening the spectral blueshift increases in a predictable way with plasma-electron density (Fig. 2, upper). However, when the Debye-Huckel model is used to account for the screening of the binding potential by the plasma electrons, the ionization rate is dramatically suppressed within a certain range of parameter space and the blueshift is suppressed (Fig. 2, lower) relative to the unscreened case. For example, Fig. 3 shows how the ratio of screened to unscreened rate varies with Debye length and barrier width in angstroms. Figure 4 shows correspondingly the reciprocal relationship between plasma-electron density in cm^{-3} and laser intensity in W cm^{-2} for the same ratio.

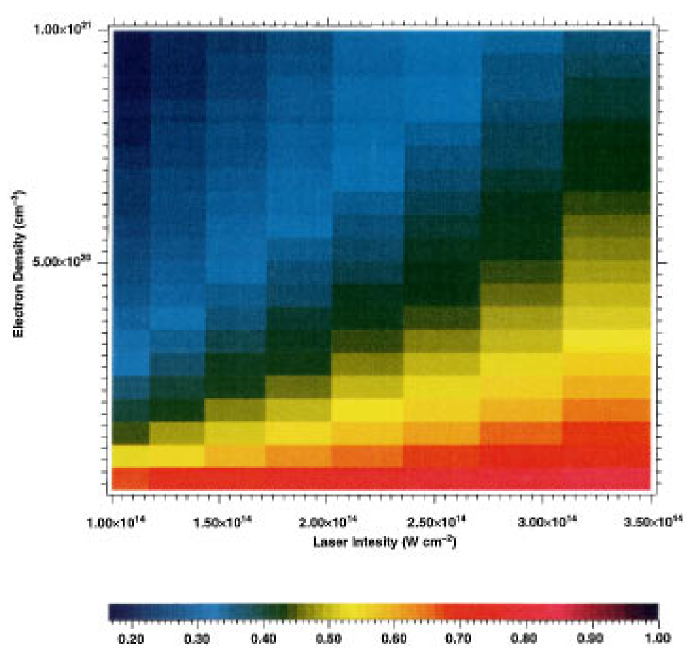

FIG. 4. (Color) Ratio at peak laser field strength of screened to unscreened ionization rates vs plasma-electron density (vertical axis in cm^{-3}) and laser intensity (horizontal axis in W cm^{-2}).

The plasma temperature is 3 eV and the critical density is 1.75×10^{21} cm^{-3} (for a laser wavelength of 800 nm).

The effect of plasma screening on the ionization rate can be understood with reference to Fig. 1. Plasma screening causes the effective potential to be of shorter range than Coulombic. In the presence of the laser field the shorter range causes the barrier against ionization to be widened by simultaneously shifting the inner barrier turning point (η_i) inwards and the outer barrier turning point (η_o) outwards, where Fig. 1 shows a reference barrier for an unscreened Coulomb potential. For example, at a laser intensity of 3.5×10^{14} W cm^{-2} and an electron density of 10^{20} cm^{-3} a 12% lengthening of the barrier width (arising from a 6% change at either turning point) reduces the ionization rate by 30%. This sensitivity arises from the well-known exponential dependence of the tunneling ionization rate on the integral over the electron's local momentum [3,4]. Hence plasma screening can dramatically suppress the ionization rate, as shown in Figs. 3 and 4. This suppression can be observed by measuring the dependence of the blue spectral shift on the plasma electron density (Fig. 2, lower).

We consider a range of plasma electron densities $10^{19}<n_e<10^{21}$ cm^{-3} and laser intensities $10^{14}<I<3.5\times10^{14}$ W cm^{-2}, in which a large effect is observed (Figs. 3 and 4). For lower intensities the ionization rate becomes slower than the pulse duration and for higher intensities the barrier width becomes much shorter than the Debye length.

III. THEORETICAL MODEL

In the remainder of the paper we give the details of the calculations on which our conclusions are based. For mathematical convenience we assume that the plasma-electron ion screening is described by the Debye-Huckel model [5]. For our plasma temperature (3 eV) and density (maximum 10^{21} cm^{-3}) the plasma coupling parameter is well into the intermediate coupling strength range ($\Gamma\approx1$), where more sophisticated models are physically appropriate [6,9]; however, our purpose here is to demonstrate the sensitivity of the tunneling ionization rate to a screened potential having a generic form and to motivate experimental examination of plasma density-dependent ionization rates.

In order to carry out this program, we use a modified form of the tunneling-ionization model described previously [3,4]. This model is based on the Schrödinger equation for a hydrogenlike atom in a static electric field, which is separable in the parabolic coordinates $\xi=r+z$ and $\eta=r-z$. In atomic units the model is [15]

$$\frac{d^2\chi_1}{d\xi^2}+2(\tfrac{1}{4}E-U_\xi)\chi_1=0, \tag{1a}$$

$$\frac{d^2\chi_2}{d\eta^2}+2(\tfrac{1}{4}E-U_\eta)\chi_2=0, \tag{1b}$$

$$U_\xi=-\frac{Z}{4\xi}+\frac{m^2-1}{8\xi^2}+\frac{1}{8}F\xi, \tag{1c}$$

$$U_\eta=-\frac{Z}{4\eta}+\frac{m^2-1}{8\eta^2}-\frac{1}{8}F\eta \tag{1d}$$

[see Fig. 1 for plots of Eqs. (1c) and (1d) for $m>1$]. In Eqs. (1), Z is the nuclear charge, m is the azimuthal quantum number, F is the strength of an applied static electric field, and χ_1,χ_2 are wave functions in the ξ,η coordinates, respectively. Use of Eqs. (1) leads to the well-known static-field ionization rate formula [3,4]

$$w=4\omega_0\frac{E_i^{5/2}}{F}e^{(-2/3)(E_i^{3/2}/F)}, \tag{2}$$

where $\omega_0=4.13\times10^{16}$ s^{-1} is one atomic unit of binding frequency. In reality the field varies in time for a laser and is written $F=\xi_p(t)\sin(\omega t)$, where ω is the optical frequency and $\varepsilon_p(t)$ is a pulse envelope whose temporal variation is slow compared to ω^{-1}. When ω is small compared to the binding frequency, then one is justified in averaging Eq. (2) over one optical cycle $2\pi/\omega$ [3,4].

The Schrödinger equation is not separable in parabolic coordinates for a screened potential; however, it is approximately separable for large screening lengths, as we show later. The Debye-Huckel potential [5] is

$$V=-\frac{Z}{r}e^{-r/\lambda_D}, \tag{3a}$$

$$\lambda_D=\left(\frac{kT}{4\pi e^2 n_e}\right)^{1/2}. \tag{3b}$$

We assume with others [3,4] that Z is given by the hydrogenic relation $Z^2=E_i$, where E_i is the ionization energy in Rydbergs. We calculate Z for an assumed ionization energy of 15.04 eV, which is close to that for Mg^+. Clearly the further assumption is implied that $\lambda_D\gg r_0$, where r_0 is the average radius of the bound state in the absence of the laser ($r_0\approx1/\sqrt{E_i}$). This condition is easily satisfied here.

In other work on laser ionization in the tunneling regime [3,4] $\lambda_D\rightarrow\infty$, such that V is separable in parabolic coordinates [Eqs. (1)]. For finite λ_D, however, V is clearly not separable,

$$V=-\frac{2Z}{\xi+\eta}e^{-(\xi+\eta)/2\lambda_D}. \tag{4}$$

However, one recognizes (Fig. 1) that the motion in the ξ coordinate is always confined to the region close to the nucleus where screening due to the free electrons outside the ion is negligible ($\xi\ll\lambda_D$). Thus we introduce the separable form,

$$V=\frac{1}{2}\left(-\frac{2Z}{\xi+\eta}-\frac{2Z}{\xi+\eta}e^{-\eta/2\lambda_D}\right), \tag{5}$$

where screening occurs only in the η variable. Equation (5) is simply the average of unscreened and screened Coulomb fields appropriate for the region in which $\xi\ll\lambda_D$ and $\eta\leqslant\lambda_D$, which reduces to the unscreened Coulomb field when $\lambda_D\rightarrow\infty$. A quantitative measure of the accuracy of this approximate form of the potential is obtained by comparing the outer turning point ξ_0 with the average of the barrier inner and outer turning points η_i and η_o, respectively. A sampling of the most important region of parameter space considered here shows us that, although $\eta_i \cong \frac{3}{2}\xi_0$, still $\frac{1}{2}(\eta_i+\eta_o) \cong 3\xi_0$, which is an average range parameter for the tunneling process. The assumption that $\eta \sim$ a few ξ is reasonable. Furthermore, the magnitude of the difference between Eqs. (4) and (5), divided by Eq. (5), to first order in $\eta/2\lambda_D$ is approximately $\eta/4\lambda_D$, which is fairly small compared to unity over the entire range of η.

Under these assumptions the static-field ionization rate is [13]

$$w=\omega_0\eta_i Z^3 \times e^{-Z\eta_i-2\int_{\eta_i}^{\eta_0} d\eta \left(\frac{Z^2}{Z}-\frac{1}{4\eta^2}-Z\frac{e^{-\eta/2\lambda_D}}{2\eta}-\frac{1}{4}F\eta\right)^{1/2}}. \tag{6}$$

When $\lambda_D\to\infty$, Eq. (6) agrees with Eq. (2) on using the approximations of [15] to evaluate the integral analytically. Here the integral is evaluated numerically both for the screened and unscreened potentials.

Our results are presented as the ratio of the screened to unscreened static-field rates in Figs. 3 and 4. The ratios are plotted only at laser intensities for which either rate is greater than 10^{12} s^{-1}, and typically at the higher intensities the absolute rates are in the range 10^{14}–10^{15} s^{-1}. We have made similar plots for the cycle-averaged rates where, although their absolute values are smaller, their ratios are not qualitatively different. These plots show that screening can lower the ionization rates in this regime of density and laser intensity substantially; thus our calculations suggest that effect of screening is experimentally observable.

The observation can be made by measuring the blue spectral frequency shift of the transmitted laser pulse induced by the change of refractive index caused by the generation of electrons in the focal volume of the ionizing pulse [12] as a function of the density of plasma electrons surrounding the embedded target ions. Spectral shift measurements at a single plasma density are described in an earlier paper [1].

The frequency shift integrated over the duration of the laser pulse is [1,12]

$$\Delta\omega=\int_0^T dt\frac{\partial\omega(t)}{\partial t}=-\int_0^T dt\frac{\omega(t)\partial n_r}{n_r\partial t}, \tag{7}$$

where we have used $\omega(t)=kc/n_r$, where n_r is the refractive index,

$$n_r=\left(1-\frac{\omega_p^2}{\omega_0^2}\right)^{1/2}, \tag{8}$$

where ω_0 is the line center laser frequency (800 nm), and where

$$\omega_p=\left(\frac{4\pi e^2 n}{m}\right)^{1/2} \tag{9}$$

is the plasma frequency. Thus the frequency shift is expressed in terms of the ionization rate as

$$\Delta\omega=\frac{\omega_{p0}^2}{2\omega_0^2}\int_0^T dt\,\frac{\omega(t)}{n_r^2}\gamma(t)e^{-\int_0^t dt'\gamma(t')}, \tag{10}$$

where $\gamma(t)$ is the cycled-averaged rate and ω_{p0} is the constant plasma frequency as determined by the initial target ion density. The temporal dependence of the rate originates from the time-dependent pulse envelope $\varepsilon_p(t)$ [12] and the upper limit of the integral in Eq. (10) is 125 fs [16]. Figure 2 shows the fractional frequency shift versus density at a laser intensity of 1.7×10^{14} W cm^{-2}.

IV. SUMMARY AND CONCLUSIONS

In summary we have demonstrated the sensitivity of the rate of tunneling ionization of ions embedded in a preionized plasma to the long-ranged screening of the binding potential by the surrounding plasma electrons. Screening is observed by measuring the dependence of the spectral blueshift of the ionizing pulse on plasma electron density.

The Debye-Huckel [5] screening model is characterized by a single screening-length parameter, governed by the plasma temperature and density, which can be compared with the characteristic length scale in the laser-plasma-ion interaction, namely the barrier width against ionization (Fig. 3). The screening effect is found to be important in a regime in which the plasma coupling parameter $\Gamma\approx1$, whereas the Debye-Huckel model is usually considered to be valid in the regime $\Gamma\ll1$. Nevertheless we believe that the model is reliable in a qualitative sense to indicate a strong dependence of ionization probability on plasma-electron density. In the interpretation of experimental data, however, more sophisticated plasma models and theories of OFI should be used.

ACKNOWLEDGMENTS

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract No. W-7405-ENG-48.

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