Comparison of soft-x-ray spectra from optical-field-ionized plasmas generated with linearly and elliptically polarized femtosecond laser pulses

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We compare x-ray spectra from optical-field-ionized plasmas generated by linearly and elliptically polarized 250-fs Ti:sapphire laser pulses. The experiment demonstrates the difference in the spectra resulting from the different electron temperatures under these two conditions. To avoid high-density effects such as electron heating, collisional ionization, or ionization defocusing the plasmas are generated under well-controlled conditions in a low-pressure gas cell. For the gases investigated (He, N_2,O_2, and CH_4) a significantly higher x-ray intensity is observed for elliptically than for linearly polarized light on the majority of lines. This finding is in accord with general theoretical expectations and with the result of simulations. However, for linear polarization, anomalies occur in the relative line intensities of several lines. Possible mechanisms responsible for this effect are discussed. [S1063-651X(97)05108-8]

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I. INTRODUCTION

At intensities above 10^{15} W/cm^2 multiply charged ions are generated directly by the optical field and not by electron collisions as in usual laser plasmas. This effect, known as optical-field ionization (OFI), has been under intense investigation during recent years. The interaction is governed by the Keldysh parameter [1]

$$\gamma=(U_q/2U_p)^{1/2}, \tag{1}$$

where U_q is the ionization potential of the atom or ion with a charge state $q-1$, $U_p=E^2/4\omega^2$ is the ponderomotive potential of the electrons oscillating in the light field, and E and ω are the field strength and the light frequency, respectively. Atomic units are used for all quantities. For $\gamma<1$ one has ionization in the tunneling regime and OFI can be described quasistatically using Ammosov-Delone-Krainov [2] (ADK) or barrier suppression ionization [3,4] (BSI) models.

The observed experimental parameters, in particular the generated ionic states, are, in general, quite well predicted by theory [3,5,6]. The simplest theoretical description, the BSI model, asserts that ionization into a charge state q occurs instantaneously if a threshold intensity

$$I_{\mathrm{th}}=U_q^4/16q^2 \tag{2}$$

is exceeded.

Experiments extracting ions from the interaction volume are carried out at very low pressure and thus plasma effects are irrelevant. The situation is quite different for experiments in which x rays from OFI plasmas are recorded, since a high enough signal is usually only obtained at gas densities exceeding 10^{18} cm^{-3}and therefore high-density effects such as heating of the electrons by inverse bremsstrahlung or stimulated Raman scattering considerably alter the plasma conditions after the initial ionization. In addition, many of the experiments detecting x rays are carried out by focusing the optical pulses into gas jets, resulting in effects due to clustering [7–12]. A further complication, ionization defocusing, arises from the refractive index gradient due to a varying degree of ionization across the beam [13,14].

Because of these complications a difference in the x-ray spectra under illumination with linear and elliptical polarization has, to our knowledge, not been observed. From theoretical considerations, however, the x-ray spectra generated under these two conditions should show significantly different features resulting from the different electron temperatures after field ionization: According to the model of Corkum, Burnett, and Brunel [15], electrons are generated with a very low velocity after tunneling and thus linear polarization should result in a low electron temperature due to the phase shift of $\pi/2$ between the field and electron velocity. On the other hand, for elliptical polarization there is a field component perpendicular to the component extracting the electron that is in phase with the electron velocity and therefore the generated electrons should acquire an energy given approximately by the ponderomotive potential. The possibility to control the electron temperature of the plasma by changing the ellipticity of the optical pulse has important consequences for OFI x-ray lasers using recombination or electron collisional pumping [16–21].

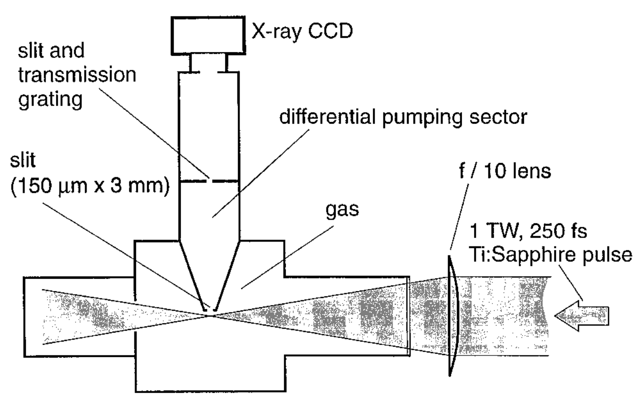

FIG. 1. Experimental setup for recording x-ray spectra from low-density OFI plasmas.

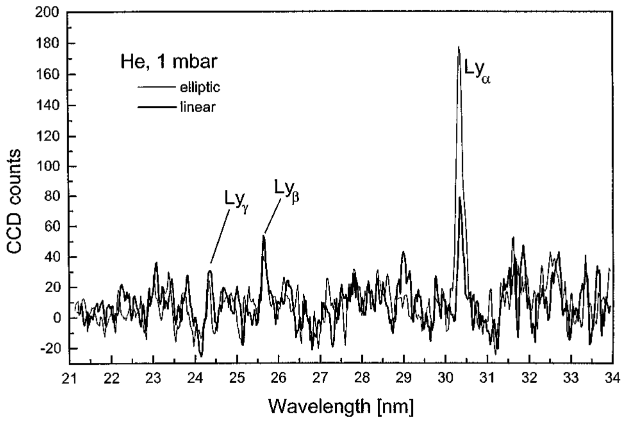

FIG. 2. Measured x-ray spectra of a helium plasma produced by optical-field ionization by elliptically and linearly polarized 250-fs laser pulses focused into 1 mbar of He.

II. EXPERIMENT

In this paper we demonstrate that x-ray spectra from low-density OFI plasmas are distinctly different for linear and elliptical light polarization, as expected from the difference in the electron temperatures. Results showing spectra of nitrogen only have been reported elsewhere [22]. In the experiment soft-x-ray spectra from OFI plasmas are recorded at gas densities considerably lower than those used previously. The experimental arrangement is shown in Fig. 1. Ti:sapphire laser pulses (200 mJ in 250 fs at 10 Hz, 790 nm) were focused into a cell containing the gas under investigation (He, N_2, O_2, and CH_4)at various pressures around 1 mbar. An intensity of 3×10^{16} W/cm^2 was obtained in the focal region. With such an intensity the BSI model predicts fully stripped helium as well as heliumlike nitrogen and carbon to be generated. For oxygen ionization stages up to Be-like are expected.

To avoid absorption of the generated x rays in the residual gas a differential pumping section was used between the gas cell and the spectrometer. The spectrometer consisted of a transmission grating (5000 lines/mm) with a 50-μm slit parallel to the grating lines and an x-ray charge coupled device (CCD) as the detector. A 0.1-μm beryllium filter transmitted soft x rays and rejected visible stray light.

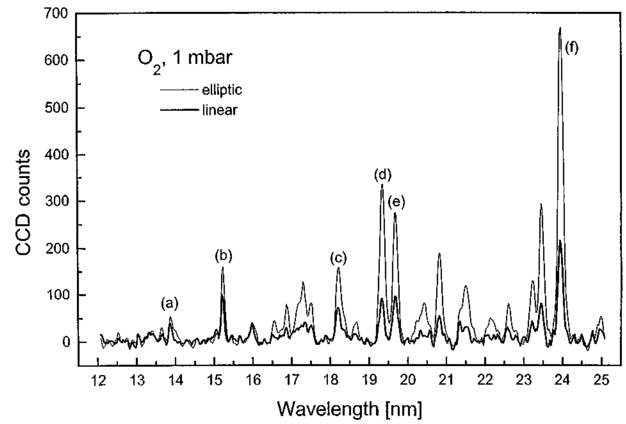

FIG. 4. Soft-x-ray spectra from optical-field-ionized oxygen for elliptical and linear polarization at 1 mbar of O_2. Main lines: (a) O^{4+} $2p$-$5d$, (b) O^{4+} $2p$-$4d$, (c) O^{3+} $2p$-$5d$, (d) O^{4+} $2p$-$3d$, (e) O^{3+} $2p$-$4d$, and (f) O^{3+} $2p$-$3d$.

To generate elliptically polarized light a low-order retardation plate consisting of a 0.2-mm-thick quartz platelet was inserted into the beam. The maximum ellipticity (defined as the ratio of minimum to maximum electric-field amplitude) induced by the plate was 0.65.

Time-integrated spectra obtained for the gases investigated, using linear and elliptical polarization, are shown in Figs. 2–5. To get a sufficiently high number of counts 6000 shots were accumulated on the CCD. The spectra are corrected for the transmission of the beryllium filter and of the gas between the laser plasma and the detector, a small correction in all cases.

As expected from the BSI model, the spectra exhibit lines emitted from H-like He, Li- and Be-like N and C, and Be- and B-like O ions. The effect of the ellipticity of the laser pulse is quite significant, with the majority of the lines showing considerably higher intensity with elliptical than with linear polarization. This demonstrates, using x rays, that elliptical polarization results in a higher electron temperature and hence in a more efficient excitation than the linear polarization.

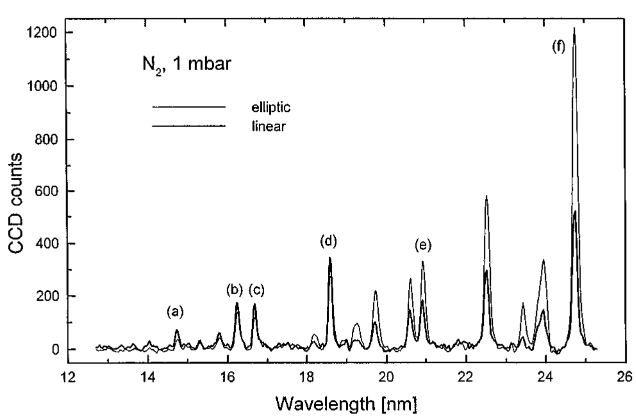

FIG. 3. Soft-x-ray spectra from optical-field-ionized nitrogen for elliptical and linear polarization at 1 mbar of N_2. Main Li-like lines (N^{4+}): (a) $2s$-$5p$, (b) $2s$-$4p$, (c) $2p$-$5d$, (d) $2p$-$4d$, (e) $2s$-$3p$, and (f) $2p$-$3d$. Other significant lines are from Be-like ions (N^{3+}).

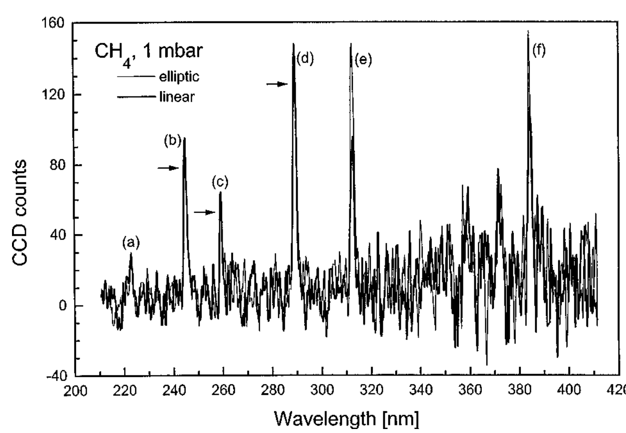

FIG. 5. Soft-x-ray spectra from optical-field-ionized CH_4 for elliptical and linear polarization at 1 mbar. All labeled lines are from Li-like C^{3+}: (a) $2s$-$5p$, (b) $2s$-$4p$, (c) $2p$-$5d$, (d) $2p$-$4d$, (e) $2s$-$3p$, and (f) $2p$-$3d$. Arrows mark the peak of the emission for elliptical polarization.

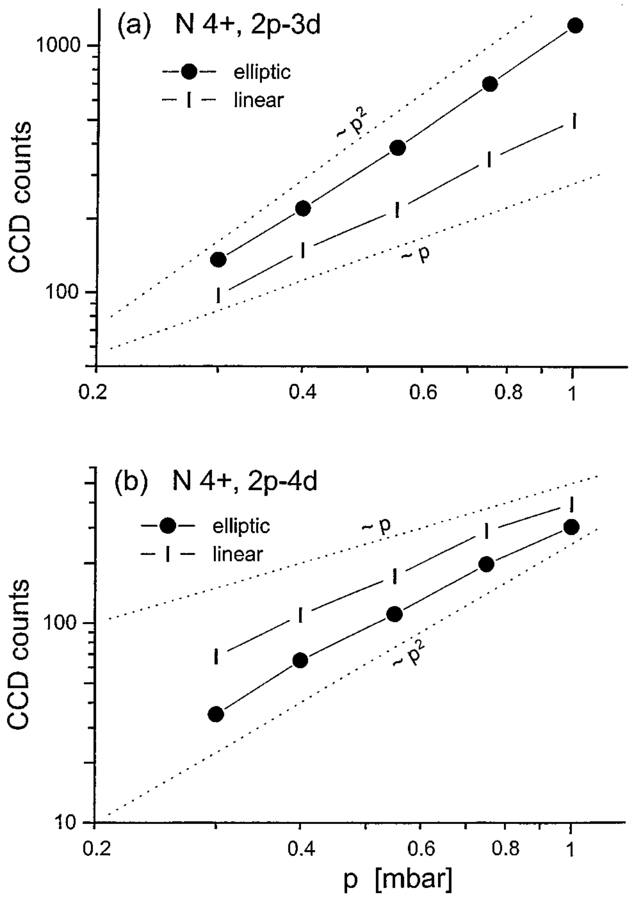

FIG. 6. Li-like nitrogen soft-x-ray emission as a function of pressure in the gas cell. The diagram compares linear with elliptical polarization for (a) N^{4+} $2p$-$3d$ and (b) N^{4+} $2p$-$4d$.

We note that the ratio of intensities at the lines emitted from different ionization stages (e.g., Li-like vs Be-like lines in nitrogen and carbon) is not indicative of the electron temperature as in collisionally dominated plasmas, but reflects essentially the relative volume in which the intensity is appropriate to generate the relevant ionization stage. In fact, at the densities of our experiments recombination and ionization rates are so low that the different ionic stages evolve essentially independently from each other. This is true for linear as well as elliptical polarization, as can be quantified by evaluating, for example, the ionization rate from Be-like to Li-like nitrogen. The rate coefficients for ionization, interpolated from data tabulated by Lotz [23], are 2.12×10^{-9} cm^3/s at an electron temperature of 85 eV and 1.9×10^{-11} cm^3/s at 15 eV (these are the initial electron temperatures obtained from the simulations; see Sec. III below). At an electron density around 2×10^{17} cm^{-3} (corresponding approximately to the conditions for our spectra at 1 mbar of N_2) this results in ionization rates of 4.2×10^8 and 3.8×10^6 s^{-1} for the two cases. Recombination rates are orders of magnitude lower. Thus the time to reach ionization equilibrium by electron collisions is much too long to be of significance in our experiment.

The pressure dependence of selected emission lines of nitrogen is displayed in Fig. 6. It is shown that for the main N^{4+} resonance lines the dependence on pressure is approximately quadratic up to 1 mbar. This proves that at these densities the observed spectra are optically thin excitation spectra since under this condition the excited-state populations are proportional to ion density times electron density. A spectrum arising from three-body recombination would result in a third power density dependence. Radiative recombination can also be ruled out since its rate is much too low to be of significance at the densities in question.

For linear polarization the dependence of the emission on pressure is somewhat below a quadratic one. This indicates that at the low electron temperatures involved effects other than electron collisions come into play for generating excited species (see the discussion at the end of Sec. III).

An unexpected anomaly was observed for linear polarization, viz., several transitions originating from high-energy levels show a stronger emission with linear than with elliptical polarization. This applies in particular to the Ly$_\beta$ line of He^+ and to $2p$-$4d$ and $2p$-$5d$ in Li-like carbon and nitrogen (see Figs. 2, 3, and 5). This feature was found to be independent of the orientation of the linear polarization vector with respect to the direction of observation. From Fig. 6 it can be seen that the anomalous emission is most pronounced at the lowest pressure (0.3 mbar). It gradually disappears at pressures above 2 mbar.

III. SIMULATIONS

Simulations of the experiments were carried out for helium and nitrogen using a collisional radiative code that takes into account levels of relevant ionization stages. The code calculates time-dependent level populations and spectra emitted from a plasma specified by a particular initial condition. Hydrodynamic expansion is taken into account by means of a self-similar model [24]. The initial conditions (electron temperature, electron density, and ionic abundances) were calculated by integrating ADK rates of the various ionization stages over a sin^2 pulse with a full width at half maximum of 250 fs. The electron temperatures were spatially averaged over the plasma volume, a procedure justified by the short thermal diffusion times at low electron density. Electron temperatures of 24 and 124 eV were calculated for helium in the case of linear and elliptical polarizations. The corresponding temperatures for nitrogen were 15 and 85 eV. Note that the definition of temperature is an approximation since the initial electron energy distributions are non-Maxwellian and electron-electron relaxation times are of the order of 100 ps.

From the temperatures quoted above one can readily see that the x-ray line emission for both materials should be significantly higher for elliptical data than for linear polarization. For the main ionic emitters the electron temperatures for linear polarization are below the energy of the first resonance level, whereas for elliptical polarization they are considerably above that value. Consider the Ly$_\alpha$ emission of hydrogenic helium: Compilations of experimental and theoretical data for the $1s$-$2p$ excitation (which dominates the 1-2 excitation) show that the collision strength increases almost linearly with increasing impact electron energy, resulting in an excitation cross section that is fairly independent of the electron energy up to at least 200 eV [25,26]. Multiplying the cross section by the electron velocity and integrating over a Maxwellian distribution results in excitation rate co-

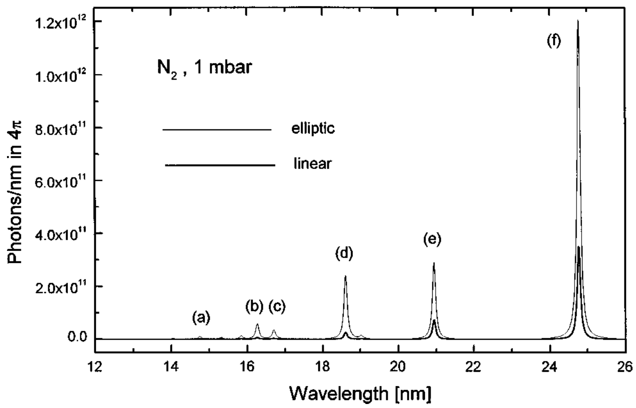

FIG. 7. Simulated time-integrated soft-x-ray spectra from optical-field-ionized nitrogen under conditions of elliptically and linearly polarized laser pulses. The line labels correspond to those in Fig. 3. For details on the simulations see the text.

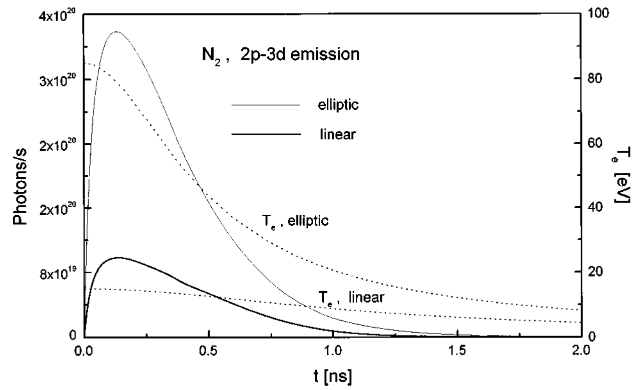

FIG. 8. Time-resolved emission on $2p$-$3d$of Li-like nitrogen for elliptical and linear polarizations obtained from the simulations. The temporal dependence of the electron temperature under the two conditions is also plotted.

efficients that increase monotonically with electron temperature. Specifically, the $1s$-$2p$ excitation rate coefficient is 1.4×10^{-8} cm^2/s at 24 eV and 1.2×10^{-7} cm^3/s at 124 eV and thus the excitation rate is significantly higher for elliptical than for linear polarization. Note that the values of these coefficients cannot be directly related to the Ly$_\alpha$ intensities in the spectra since the electron temperatures quoted above represent only the initial conditions for the plasma evolution. Both the electron density and temperature decrease rapidly after the initial ionization.

For nitrogen the situation is similar, except that the collision strength increases somewhat more slowly with increasing electron energy. We use the data of Ref. [27] (which are given for $8\leqslant Z\leqslant 92$) and extrapolate them slightly to $Z=7$. Taking the strong $2p_{3/2}$-$3d_{5/2}$ excitation as an example, the collision strength is 1.64 at 53.2 eV (close to threshold) and 2.18 at an electron energy of 90 eV. The corresponding cross sections are 9.2×10^{-18} cm^2 and 7.2×10^{-18} cm^2, respectively. Evaluation of excitation rate coefficients for a Maxwellian distribution yields a temperature dependence that is still dominated by the exponential factor in the integral, in spite of the slight falloff of the cross section at higher electron energies. Specific values for $2p_{3/2}$-$3d_{5/2}$ excitation are 2.77×10^{-9} and 4.8×10^{-8} cm^2/s at electron temperatures of 15 and 85 eV. We note again that these values represent only the initial conditions immediately after optical-field ionization.

Figure 7 shows simulated spectra for nitrogen, which are temporally and spatially integrated. The entire radiating volume is included in the simulated emission. For linear polarization the volume in which ions are Li-like is somewhat larger than for elliptical one due to the higher peak value of the field and the strong nonlinearity of the ionization rate as a function of the electric-field strength. The spectra are calculated as being optically thin, an assumption justified by the increase in the effective ion temperature due to a Coulomb explosion [28–30]. In comparing the results for linear and elliptical polarization one sees that the simulations, with no adjustable parameters, predict a ratio of Li-like $2p$-$3d$ emissions for the two cases approximately as experimentally observed. The absolute number of the emitted photons predicted by the simulations is within an order of magnitude of the experimental one. Other features of the experimental results are also verified by the simulations, specifically the quadratic increase of the generated line intensities with pressure for elliptical polarization. Similar agreement with experiments is obtained for the helium simulations.

The code results can be used to establish the time dependence of the emission. Figure 8 shows the simulated temporal dependence of the emission on the $2p$-$3d$ line of Li-like nitrogen for linear and elliptical polarization. The corresponding time-dependent electron temperatures are also plotted into the figure. It is obvious that the rise times of the emission under the two conditions are quite similar and are given essentially to be the radiative decay time of the $3d$ level (24 ps). The decay times of the emission, however, are different since they reflect the decay of the collisional excitation rate due to adiabatic cooling of the electron gas. The higher electron temperature at elliptical polarization results in a faster expansion and thus in a higher cooling rate than for linear polarization.

For both helium and nitrogen the simulations fail to agree with experiment for the lines originating from higher principal quantum numbers since they do not reproduce the significantly stronger emission with linear polarization on those lines. The origin of this effect is not clear at the moment. An opacity effect can be ruled out since in this case the $2p$-$3d$ line should show a quite different density dependence from the $2p$-$4d$ line. The same applies to an explanation invoking an enhanced three-body recombination rate resulting from the non-Maxwellian electron energy distribution, which would result in an increase of the emission with the third power of the density. A charge-exchange reaction into excited states has been discussed in a previous paper dealing with nitrogen only [22]. However, in the case of helium such an explanation is not feasible since charge transfer from neutral helium into bare nuclei to form an excited hydrogenic state would be strongly endothermic and thus has a low cross section.

A hint of a possible mechanism is given by the fact that the time-resolved helium emission was reported to exhibit an initial spike [31]. This observation had been tentatively attributed to an initial excited-state population in helium after field ionization. Excited-state populations after short-pulse multiphoton ionization have been observed previously in xe-

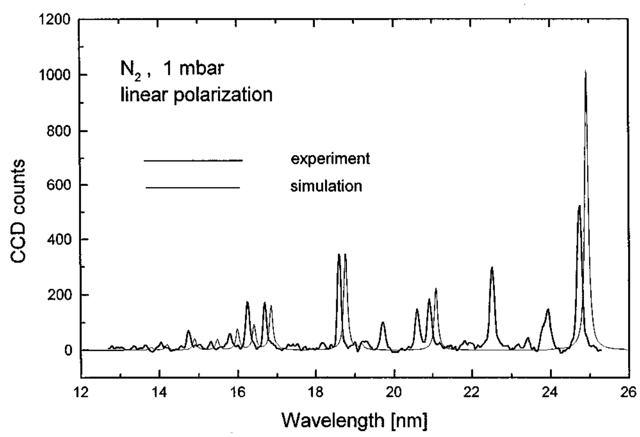

FIG. 9. Attempt to improve the agreement between experimental and simulated nitrogen spectra for linear polarization by modifying the initial excitation temperature of the ions. An excitation temperature of 600 eV is assumed for the initial Li-like nitrogen population. To allow a better comparison the spectra are normalized at the Li-like $2p$-$4d$ emission. The simulated spectrum is laterally displaced by 0.2 nm for clarity of presentation.

non [32]. The population of excited states during field ionization is conceivable by collisions of the electrons with the original nucleus [33,34], an effect that is only possible for linear polarization. The observation that the anomaly becomes less pronounced at higher density is consistent with this explanation since the probability of the electron to return to the nucleus is reduced. Note that this effect results in a linear dependence of the emission on pressure.

An attempt to use the concept of initially excited states for improving the match between simulated and experimental spectra for linear polarization is shown in Fig. 9. For generating this spectrum the initial condition of the OFI plasma was altered by assuming for the initial Li-like ion population an excitation temperature equal to the ponderomotive potential. The exact value of that temperature was found to be not critical. It is seen that the agreement with experiment is considerably improved, but the $2p$-$4d$ to $2p$-$3d$ ratio is still not as high as the experimental one. Arbitrarily reducing the initial population in $3d$ would result in even better agreement, but is hard to justify theoretically. Further experimental and theoretical studies are required to clarify the mechanism involved.

IV. CONCLUSION

We have shown that x-ray spectra from low-density optical-field-ionized plasmas exhibit features characteristic of the ellipticity of the pulse generating the plasma. For a variety of ions the x-ray spectra obtained with elliptically polarized optical pulses are considerably more intense than the ones with linearly polarized pulses. This observation is in keeping with the theoretical expectation of a considerably higher electron temperature at elliptical polarization taking into account that a pure excitation spectrum is observed at the low densities used in the experiment. Simulations of the experiment assuming initial conditions obtained by ADK theory agree with many of the experimental features. An anomaly is seen in the spectra with linear polarization, viz., transitions originating from high principal quantum number states are enhanced. This effect is not reproduced by the simulations. The fact that the anomaly is also observed in helium excludes an explanation invoking a charge-exchange reaction. A clarification of the effect is expected from future investigations.

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[1] L. Y. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)].
[2] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Zh. Eksp. Teor. Fiz. **91**, 2008 (1986) [Sov. Phys. JETP **64**, 1191 (1986)].
[3] S. Augst, D. Strickland, D. D. Meyerhofer, S. L. Chin, and J. H. Eberly, Phys. Rev. Lett. **63**, 2212 (1989).
[4] B. M. Penetrante and J. N. Bardsley, Phys. Rev. A **43**, 3100 (1991).
[5] S. Augst, D. D. Meyerhofer, D. Strickland, and S. L. Chin, J. Opt. Soc. Am. B **8**, 858 (1991).
[6] M. D. Perry, A. Szoke, O. L. Landen, and E. M. Campbell, Phys. Rev. Lett. **60**, 1270 (1988).
[7] A. A. Offenberger, W. Blyth, A. Dangor, A. Djaoui, M. H. Key, Z. Najmudin, and J. S. Wark, Phys. Rev. Lett. **71**, 3983 (1993).
[8] T. Ditmire, T. Donnelly, R. W. Falcone, and M. D. Perry, Phys. Rev. Lett. **75**, 3122 (1995).
[9] A. McPherson, T. S. Luk, B. D. Thompson, K. Boyer, and C. K. Rhodes, Appl. Phys. B **57**, 337 (1993).
[10] W. J. Blyth, S. G. Preston, A. A. Offenberger, M. H. Key, J. S. Wark, Z. Najmudin, A. Modena, A. Djaoui, and A. E. Dangor, Phys. Rev. Lett. **74**, 554 (1995).
[11] J. K. Crane, H. Nguyen, S. C. Wilks, T. Ditmire, C. A. Coverdeale, T. E. Glover, M. D. Perry, and Zakharenkov, J. Opt. Soc. Am. B **13**, 89 (1996).
[12] E. Fill, S. Borgström, J. Larsson, T. Starczewski, C.-G. Wahlström, and S. Svanberg, Phys. Rev. E **51**, 6016 (1995).
[13] P. Monot, T. Auguste, L. A. Lompré, G. Mainfray, and C. Manus, J. Opt. Soc. Am. B **9**, 1579 (1992).
[14] E. E. Fill, J. Opt. Soc. Am. B **11**, 2241 (1994).
[15] P. B. Corkum, N. H. Burnett, and F. Brunel, Phys. Rev. Lett. **62**, 1259 (1989).
[16] N. H. Burnett and P. B. Corkum, J. Opt. Soc. Am. B **6**, 1195 (1989).
[17] P. Amendt, D. C. Eder, and S. C. Wilks, Phys. Rev. Lett. **66**, 2589 (1991).
[18] B. E. Lemoff, C. P. Barty, and S. E. Harris, Opt. Lett. **19**, 569 (1994).
[19] Y. Nagata, K. Midorikawa, S. Kubodera, M. Obara, H. Tashiro, and K. Toyoda, Phys. Rev. Lett. **71**, 3774 (1993).

[20] B. E. Lemoff, G. Y. Yin, C. L. Gordon, III, C. P. Barty, and S. E. Harris, Phys. Rev. Lett. **74**, 1574 (1995).
[21] T. E. Glover, T. D. Donelly, E. A. Lipman, A. Sullivan, and R. W. Falcone, Phys. Rev. Lett. **73**, 78 (1994).
[22] G. Pretzler and E. E. Fill, Opt. Lett. **22**, 733 (1997).
[23] W. Lotz, Z. Phys. **216**, 241 (1968).
[24] R. A. London and M. D. Rosen, Phys. Fluids **29**, 3813 (1986).
[25] R. J. W. Henry, Phys. Rep. **68**, 1 (1981).
[26] M. Basu, P. S. Mazumder, and A. S. Ghosh, Can. J. Phys. **61**, 1297 (1983).
[27] H. L. Zhang, D. H. Sampson, and C. J. Fontes, At. Data Nucl. Data Tables **44**, 31 (1990).
[28] L. J. Frasinski, K. Codling, P. A. Hatherly, J. R. M. Barr, I. N. Ross, and W. T. Toner, Phys. Rev. Lett. **58**, 2424 (1987).
[29] K. Codling and L. J. Frasinski, Contemp. Phys. **35**, 243 (1994).
[30] M. Ivanov, T. Seideman, P. Corkum, F. Ilkov, and P. Dietrich, Phys. Rev. A **54**, 1541 (1996).
[31] S. Borgström, E. Fill, T. Starczewski, J. Steingruber, S. Svanberg, and C.-G. Wahlström, Laser Part. Beams **13**, 459 (1995).
[32] M. P. de Boer and H. G. Muller, Phys. Rev. Lett. **68**, 2747 (1992).
[33] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[34] T. Brabec, M. Yu. Ivanov, and P. B. Corkum, Phys. Rev. A **54**, R2551 (1996).