Measurement of Ultrafast Ionization Dynamics of Gases by Multipulse Interferometric Frequency-Resolved Optical Gating

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(Received 1 December 2000; published 5 December 2001)

Ultrafast ionization dynamics of femtosecond laser-irradiated noble and simple diatomic gases were studied using a novel two-color time-domain technique which eliminated significant complications seen in past experiments. Ultrafast depletion of the probing laser pulse was observed strictly coincident with the ionization front and attributed to a previously unobserved nonlinear frequency mixing via the transverse plasma current [F. Brunel, J. Opt. Soc. Am. B **7**, 52 (1990)]. Good quantitative agreement of the measured single-atom ionization rates with Ammosov-Delone-Krainov rates was found, except for O_2 which showed a 200× smaller rate.

DOI: 10.1103/PhysRevLett.87.263002 PACS numbers: 52.25.Jm, 32.80.Fb, 33.80.Rv, 52.38.−r

The ultrafast dynamics of an atom illuminated by a superstrong laser field is rich in both interesting physics and potential applications. A source of both ionization-based tunable radiation [1–4] and high-harmonic UV-VUV-XUV radiation [5], successful "mode locking" of ultrafast ionization may yield the first attosecond pulses [6]. Previous optical studies [1–4] of ultrafast ionization have relied on spectral power density measurements using 100 fs pulses. In such experiments, rapid ionization of the gas leads to an increase in plasma electron density $n_e(t)$ and a reduction in the optical phase $\phi(t)$ impressed upon light copropagating with the ionization front. A time-dependent shift in the frequency of these photons, given by

$$\Delta\nu(t) = -\frac{1}{2\pi}\frac{\partial\phi(t)}{\partial t} = -\frac{\omega_0 z}{c}\frac{\partial n(t)}{\partial t}, \qquad (1)$$

results. Here ω_0 is the central frequency of the optical pulse, $n(t) = \sqrt{1 - n_e(t)/n_{cr}}$ is the refractive index of the plasma [7] with critical density n_{cr}, z is the length of plasma the pulse has traversed, and t is the time coordinate in a speed-of-light frame. While a photon interacting with the ionization front will experience an increase in its frequency and energy (i.e., a blueshift), the detailed structure of the power spectra of an ionization-front blueshifted light pulse depends upon a complicated interplay of the input pulse intensity/phase structure and the ionization dynamics [8]. Moreover, these experiments could not identify and compensate for contributions from cross-phase modulation in the neutral gas, from effects of defocusing from the plasma density profile, and from pump depletion [3,4,8]. Hence, the conclusions made from such experiments have been, at best, well-thought-out and limited inferences.

Modern ultrashort-pulse measurement techniques such as FROG (frequency resolved optical gating [9,10]) and MI-FROG (multipulse interferometric FROG [11,12]) can provide complete measurement of the time-domain intensity and phase of an ultrashort optical pulse, including the constant and linear terms (i.e., a simple shift of the pulse in time) of the spectral phase which are dominant in laser-matter interaction. By using a two-color geometry, we can accurately time resolve the full intensity and phase, clearly identify defocusing effects, and eliminate signal loss in a pump depleted regime.

In addition, a two-color experiment permits the unique opportunity to observe "Brunel mixing" via the transverse plasma current $J_\perp = en_e\mathbf{v}$, where e is the electron charge and $\mathbf{v}$ is the electron velocity [13]. Briefly summarized, for a linearly polarized field oscillating at ω_1, ionization takes place twice per cycle—at the peak and valley of the optical wave—and thus n_e predominantly oscillates at $2\omega_1$. A probe field at ω_2 provides a component of the electron quiver motion at this same frequency ω_2 and thus this mixing process produces harmonics at frequencies $\omega_3 = 2\omega_1 \pm \omega_2$. Similar mixing also occurs via $\chi^{(3)}$ processes in the neutral gas surrounding the plasma [1,2]. However, Brunel mixing should be rigorously confined to the ionization front and should, like n_e, be independent of the relative orientation of the two linearly polarized beams.

The objectives of this paper are, therefore, to directly observe the effects of Brunel mixing in an ultrafast ionization front and measure single-atom ionization rates of gases by time-resolved blueshift measurements [Eq. (1)].

By using an experimental setup described previously [11], data was taken with 1–1000 Torr gas pressure [$(3.3–3300) \times 10^{16}$ cm^{-3} neutral gas densities, N_{gas}], of He ($I_p = 24.59$ eV), Ne (21.56 eV), Ar (15.76 eV), Kr (14.00 eV), Xe (12.13 eV), H_2 (15.43 eV), N_2 (15.58 eV), O_2 (12.07 eV), and air at pump-probe increments of 12 fs. The polarization of the 124-fs, 400-nm pump was oriented either parallel (p polarization) or orthogonal (s polarization) to that of the 100× weaker 800-nm, 175-fs, multipulse probe [14].

Figure 1a, the doubly time-resolved probe pulse frequency shift for above-threshold ionization of xenon ($I_{\text{pump}} = 1.5 \times 10^{14}$ W/cm^2), illustrates not only the ionization front itself but also the contributions from neutral-gas cross-phase modulation, defocusing from the plasma density profile, and from pump depletion. For

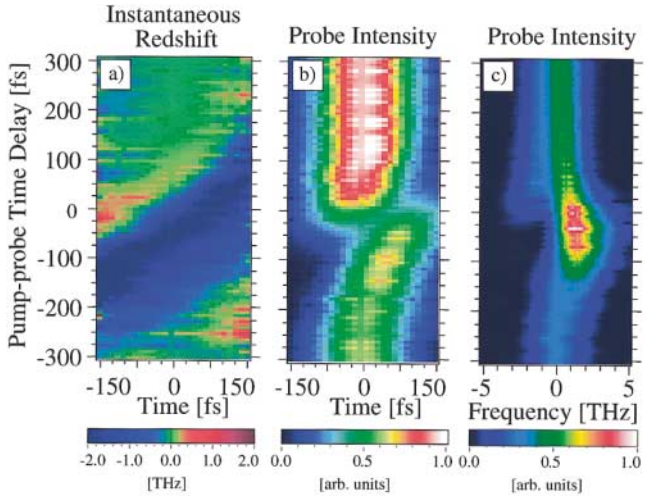

FIG. 1 (color). (a) Doubly time-resolved frequency shift of an 800-nm probe laser pulse in 600 T xenon, with pump and probe orthogonally (s) polarized. Peak pump intensity: $\sim 2 \times 10^{14}$ W/cm^2 (in vacuum). The large dark diagonal corresponds to a strong blueshift, while the upper, lighter diagonal corresponds to a weak redshift. The area of the figure below the blue diagonal corresponds to plasma (i.e., the probe trails the pump); the area above corresponds to preionized neutral gas (the probe leads the pump). (b) Time-resolved probe intensity profile. (c) Time-resolved power spectra.

comparison, Fig. 1c contains the time-resolved power spectra of the same probe pulse. Note that a simple power-spectrum analysis would conclude that only blueshifting was occurring.

The upper, redshifted, diagonal in Fig. 1a was observed even when the pump intensity was reduced to below the ionization threshold and no visual evidence of a plasma was seen. The magnitude of the shift was independent of pump-probe time delay. It was not observed when s polarization was used. Hence, we attribute it to cross-phase modulation via the neutral gas. This feature was strongest when the pump trailed the probe through the focus, an asymmetry suggesting that pump depletion from plasma formation and probe pulse defocusing from the radial plasma density gradient are significant [15]. Defocusing of the probe pulse was independently confirmed by the observed time-resolved drop in transmitted probe pulse energy [16]. Importantly, this reduction persisted for all negative time delays (probe trailing the pump), as would be expected for defocusing from the radial plasma density gradient.

The lower, blueshifted, diagonal in Fig. 1a is predominantly due to the ultrafast ionization front, though comparison of the p- and s-polarized cases indicated that neutral gas contributions were not negligible. As mentioned above, at below-ionization-threshold intensities and for p polarization, both leading red and trailing blue diagonals were seen with frequency shifts of equal magnitude. At higher intensities, the blueshift became much stronger relative to the red. With s polarization the blueshift persisted, though with slightly less magnitude. The time-resolved frequency shift, then, provides a clear and distinct method to identify and account for nonionization-based contributions to the data.

Figure 1b likewise illustrates the corresponding probe pulse intensity profile. It should be stressed that by using MI-FROG a zero of time is well defined, and the presence and apparent motion of the "hole" in the pulse profile is genuine. This feature, seen with *both* polarizations, occurred strictly coincident with the portions of the doubly time-resolved frequency shift that corresponded to the ionization front. Loss due to defocusing from the plasma density would persist after the ionization front. Hence we attribute this observation to Brunel mixing within the ionization front.

We used a 1D numerical model to better compare our results with theoretical predictions. Our model, based on that of Penetrante *et al.* [17], solved Maxwell's equations for the fields and the continuity equations for the electron density and current. A linearly polarized electromagnetic wave, consisting of a linear superposition of two Gaussian pulses representing the intense 400-nm pump and the $\sim 100\times$ weaker 800-nm probe propagating in the z direction, is incident upon a slab of gas of length L. The number density $N_j(z,t)$ of ions with charge state j is followed by a stepwise ionization process in which the probability of an ion being ionized from charge state j to $j+1$ is calculated according to standard Ammosov-Delone-Krainov (ADK) field-ionization rates [18]. At the relatively low gas densities for the data used for comparison, collisional ionization [19] was not found to contribute significantly to the overall ionization rate.

For the conditions of Fig. 1 the 1D model (see Fig. 2) did indeed effectively reproduce the strong hole in the

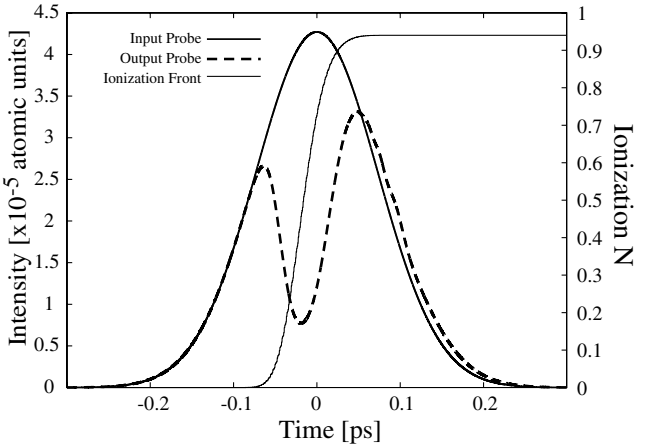

FIG. 2. Calculated 800-nm probe pulse intensity profiles. Solid line: Gaussian input pulse. Dashed line: probe pulse after copropagating with a $100\times$ more intense ($I_{\text{pump}} = 1.5 \times 10^{14}$ W/cm^2) 400-nm pump pulse (not shown) in Xe with a gas density $\times$ interaction length product $\rho z = 8.5 \times 10^{17}$ cm^{-2}.

intensity profile of the probe pulse (Fig. 1b). As well, we found numerically that the third harmonic was saturated and, through cascaded mixing in the plasma current, light at a great many harmonics, both odd and even, was produced (see Fig. 3). Some of these approached 1% of the probe energy [20]. In total, the energy of the harmonics generated was sufficient to deplete the probe pulse on an ultrafast time scale. Although it was clearly accompanied by 2D effects not in our model, the experimentally observed ultrafast depletion, as it occurred only *within* the ionization front and for *both* polarizations, cannot be due to defocusing or neutral-gas effects. We conclude, then, that this work represents the first direct, ultrafast-time-resolved observation of this phenomenon.

For the second objective of this paper, the comparison of the measured ionization-front blueshifts with existing models of ionization, we used a reduced frequency-shift parameter δ_ν given by $\Delta\nu/r_e z\lambda N_{\text{gas}}$, where $\Delta\nu$ is the measured shift, $r_e = 2.8 \times 10^{-13}$ cm, z is the interaction length, and λ is the laser wavelength. Thus, δ_ν is determined solely by the single-atom ionization rate. Figure 4 presents the measured δ_ν, obtained by using the confocal parameter of the focus as the interaction length, as well as the results of the numerical model.

For those data sets where the confocal parameter approximated the plasma length (evidenced by no pump or time-resolved probe absorption and by neutral-gas cross-phase modulation features in the frequency shifts.), we found good agreement between the measured ionization rates with those calculated using the 1D plasma fluid model and Ammosov ionization rates [18]. Using Keldysh rates [21] yielded $\sim 10\times$ smaller predicted rates.

In the case of the homonuclear diatomic gases H_2 and N_2 (Fig. 5), we found good agreement with the model, using published molecular ionization potentials (15.43 eV for H_2 and 15.58 eV for N_2, both close to that of argon, 15.76 eV). Thus molecular H_2 and N_2 behave very much like monoatomic argon. However, in the case of O_2 (12.07 eV, close to that of xenon, 12.13 eV), we find a 2 order of magnitude disagreement between experimental data for O_2 and Xe, as well as with the model prediction. Similar results have been seen in ion-yield experiments [22] and their explanation is presently a source of debate [22–25]. Further examination of this discrepancy in homonuclear diatomics, using the techniques of this Letter, will be the subject of a future publication.

Finally, as a potential application of this work, we note that the mixing via the transverse plasma current also produces light near zero frequency (i.e., terahertz pulses). When the ratio of 400-nm light to 800-nm light is closer to unity or even reversed, the primary (not cascaded) mixing process produces $\sim$kV/cm THz pulses directly. This may very well be the origin of the intense THz fields recently seen by Cook and Hochstrasser [26] emerging from copropagating 400-nm and 800-nm femtosecond pulses focused in air, but for which no microscopic origin has been conclusively determined.

In summary, a state-of-the-art time-gated frequency-domain interferometric optical pulse measurement technique was used to study ultrafast field ionization of both noble and homonuclear diatomic gases. The measured single-atom ionization rates were in good agreement with 1D plasma fluid codes based on ADK ionization rates. Ionization dynamics in the homonuclear diatomic gases H_2 ($I_p = 15.43$ eV) and N_2 (15.58 eV) were quantitatively much like that of monoatomic argon (15.76 eV). However, O_2 ($I_p = 12.07$ eV) was found to have an ionization rate 2 orders of magnitude smaller than that of

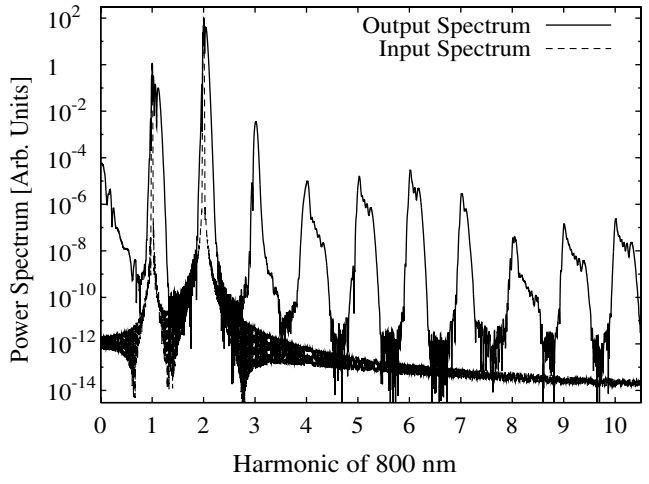

FIG. 3. Calculated output power spectrum (solid line) for an input pulse pair (pump and probe, dashed line) corresponding to the conditions of Fig. 2. For the focal geometry used in the experiment, the 400-nm pump pulse had approximately 100 μJ integrated energy.

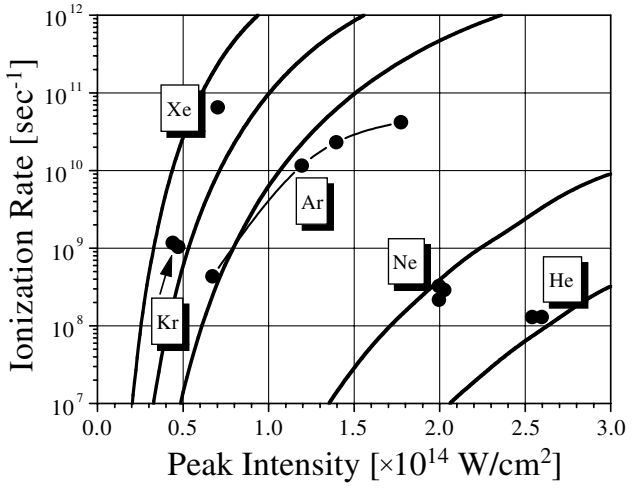

FIG. 4. Measured (•) and modeled (lines) single-atom ionization rates δ_ν for the noble gases, obtained by assuming an interaction length of two Rayleigh lengths. Horizontal error bars are primarily systematic (interaction length and peak intensity), while vertical error bars are on the order of the symbol size.

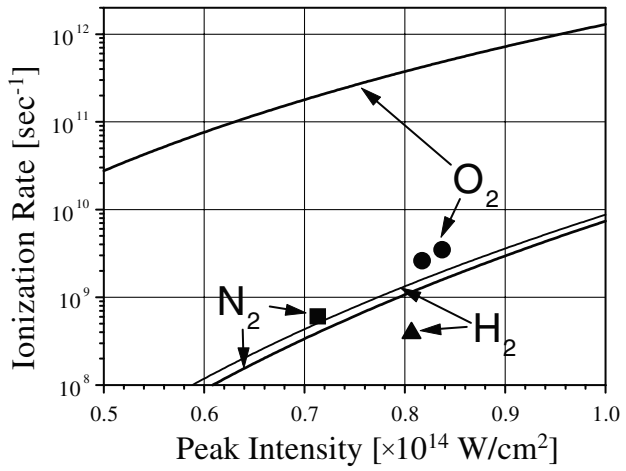

FIG. 5. Measured and modeled (lines) ionization rates for three homonuclear diatomic gases, N_2 (■), O_2 (•), and H_2 (▲).

xenon (12.13 eV). The techniques used in this Letter to accurately measure ultrafast ionization rates can also be used to monitor and provide feedback control for future laser-plasma-based particle accelerators [27,28], as well as other pump-probe studies of ultrafast dynamics. Finally, we have observed, for the first time to our knowledge, the direct effects of Brunel mixing in the transverse current of the ionization front. Such ultrafast time-domain pulse shaping may be useful for the nonlinear design of sculpted pulses impossible to achieve with traditional pulse shaping techniques [29].

This research was supported by the Los Alamos Directed Research and Development Program by the U.S. Department of Energy.

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[1] C. W. Siders *et al.,* J. Opt. Soc. Am. B **13**, 330 (1996).
[2] S. Backus *et al.,* Opt. Lett. **21**, 665 (1996).
[3] W. M. Wood, C. W. Siders, and M. C. Downer, Phys. Rev. Lett. **67**, 3523 (1991).
[4] S. P. Le Blanc, R. Sauerbrey, S. C. Rae, and K. Burnett, J. Opt. Soc. Am. B **10**, 1801 (1993).
[5] M. Gavrila, *Atoms in Intense Laser Fields,* vol. 1 of *Advances in Atomic, Molecular, and Optical Physics* (Academic Press, Boston, 1992).
[6] I. P. Christov, M. M. Murnane, and H. C. Kapteyn, Phys. Rev. Lett. **78**, 1251 (1997).
[7] Other contributions to the refractive index, such as from bound electrons in the neutral atoms and ions cores within the plasma are small enough to neglect [19].
[8] B. Rau *et al.,* J. Opt. Soc. Am. B **14**, 643 (1997).
[9] D. J. Kane and R. Trebino, Opt. Lett. **18**, 823 (1993).
[10] D. N. Fittinghoff *et al.,* Opt. Lett. **21**(16), 1313 (1996).
[11] C. W. Siders, A. J. Taylor, and M. C. Downer, Opt. Lett. **22**, 624 (1997).
[12] C. W. Siders, J. L. W. Siders, F. G. Omenetto, and A. J. Taylor, IEEE J. Quantum Electron. **35**, 432 (1999).
[13] F. Brunel, J. Opt. Soc. Am. B **7**, 521 (1990).
[14] Our heavily automated apparatus acquired FROG and MI-FROG traces, as well as power spectra of the initial laser pulse and the light pulses entering and exiting the chamber, all in the same spectrometer and on the same detector. The energy of the pump pulse after the plasma was also measured. This set of measurements was repeated both with and without the laser-produced plasma at a large number of pump-probe time delays (256 typical) and for both polarizations. This large self-consistent set of data allowed the unambiguous identification of defocusing and pump depletion, provided time-resolved intensity profiles of the probe, as well as "differential" measurements of the time-resolved phase and frequency shifts produced by the ionization front.
[15] Pump depletion is expected from energy considerations of the ionization: 12.13 eV ionization potential for Xe and 10^{-6} cm^3 ionized volume would require 40 μJ for complete single ionization (the peak intensity is, in this case, approximately twice the single-ionization threshold). A 15 μJ energy loss in transmitted pump energy was observed for the data in Fig. 1.
[16] The integral of the power spectra (proportional to the energy) was seen to change from 100% when the probe led the pump (and hence did not "see" the plasma), to 80% as the pump moved through the probe.
[17] B. M. Penetrante *et al.,* J. Opt. Soc. Am. B **9**, 2032 (1992).
[18] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Sov. Phys. JETP **64**, 1191 (1986).
[19] W. M. Wood, C. W. Siders, and M. C. Downer, IEEE Trans. Plasma Sci. **21**, 20 (1993).
[20] In the experiment, light at the third harmonic was clearly visible exiting the focal region and was present for both polarization combinations, though extremely weak for the orthogonally polarized case. The residual ellipticity of our beams, however, made it impossible to quantify how much of this 267-nm light could be attributed to the ionization front as opposed to the neutral gas [1,2].
[21] L. V. Keldysh, Sov. Phys. JETP **20**, 1307 (1965).
[22] A. Talebpour, C.-Y. Chien, and S. L. Chin, J. Phys. B **29**, L677 (1996).
[23] G. N. Gibson, G. Dunne, and K. J. Bergquist, Phys. Rev. Lett. **81**, 2663 (1998).
[24] C. Guo, Phys. Rev. Lett. **85**, 2276 (2000).
[25] J. Muth-Böhm, A. Becker, and F. H. M. Faisal, Phys. Rev. Lett. **85**, 2280 (2000).
[26] D. J. Cook and R. M. Hochstrasser, Opt. Lett. **25**, 1210 (2000).
[27] T. Tajima and J. M. Dawson, Phys. Rev. Lett. **43**, 267 (1979).
[28] C. W. Siders *et al.,* Phys. Rev. Lett. **76**, 3570 (1996).
[29] A. M. Weiner, Rev. Sci. Instr. **71**, 1929 (2000).