In this paper, we propose a new method to characterize the temporal structure of arbitrary optical laser pulses with low pulse energies. This approach is based on strong field photoelectron holography with the glory rescattering effect as the underlying mechanism in the near-forward direction. Utilizing the subfemtosecond glory rescattering process as a fast temporal gate to sample the unknown light pulse, the time-dependent vectorial electric field can be retrieved from the streaking photoelectron momentum spectra. Our method avoids the challenging task of generation or manipulation of attosecond pulses and signifies important progress in arbitrary optical waveform characterization.

PACS numbers: 32.80.Rm, 32.80.Fb, 42.40.Kw

Although these recent characterization techniques yield good performance, their requirement of generation or manipulation of broadband isolated attosecond XUV pulses is still very challenging to meet[35–37]. In this work, we propose a new method to extract the waveforms of unknown laser pulses with commonly used strong near-infrared(NIR) table-top laser light as a pump field to irradiate the atoms. Our proposal utilizes facilities from the strong field ionization and strong field photoelectron holography(SFPH) fields[38], and information of the weak test laser pulses is imprinted in the holographic interference fringes of the final photoelectron momentum distribution(PMD).

A strong NIR laser is able to tunnel ionize atoms, and the liberated photoelectron may be driven back and elastically scatter off the parent ion at a later time[39]. Concerning SFPH, strong field tunneling ionization plays the role of an atomic-level beamsplitter: after tunneling, part of the photoelectron wavepacket less impacted by the ionic Coulomb potential forms a reference wave. The other part, termed the signal wave, is steered around and scatters off the atomic core. The hologram stemming from interference of the reference and signal waves at the detector encodes spatiotemporal information about the interaction of the electron-ion system. Recently, the interpretation of SFPH has been improved by the discovery of the glory rescattering effect in strong field ionization[40].

As a demonstration, a fundamental pump laser field with a wavelength of 800nm and an intensity of $1.5 \times 10^{14} W/cm^2$ is used to ionize hydrogen atoms: $E_0(t) = \epsilon_0 \cos^2(\frac{\pi}{T_0}) \cos(\omega_0 t) \hat{x}$, where $T_0 = 3 \times 10^{-15}$, with the time duration only three optical cycles to eliminate multiple rescattering effects. Fig. 1(a) illustrates the PMD in the polarization plane simulated using the time-dependent Schrödinger equation(TDSE), with an orthogonally polarized two-color(OTC) laser field. The test laser pulse

**Subfemtosecond glory hologrammetry for vectorial optical waveform reconstruction**

J. F. Tao¹, J. Cai², Q. Z. Xia³ and J. Liu¹,⁵

¹Beijing Computational Science Research Center, Beijing 100193, China
²School of Physics and Electronic Engineering, Jiangsu Normal University, Xuzhou 221116, China
³National Laboratory of Science and Technology on Computational Physics, Institute of Applied Physics and Computational Mathematics, Beijing 100088, China
⁴Graduate School of China Academy of Engineering Physics, Beijing 100193, China and
⁵CAPT, HEDPS, and IFSA Collaborative Innovation Center of MoE, Peking University, Beijing 100871, China

Probing or manipulation of ultrafast electron dynamics on a subfemtosecond($\leq 10^{-15}$ s) or attosecond($\sim 10^{-18}$ s) timescale necessitates ultrashort laser pulses lasting only a few or near-single optical cycles with controllable waveforms[19, 20]. Developments in frequency comb technology combined with pulse-shaping methods have allowed arbitrary electromagnetic waveforms to be synthesized at optical frequencies[10–15]. Knowledge of the temporal structure of these light pulses is a prerequisite for subsequent applications. Traditional characterization techniques, such as frequency-resolved optical gating(FROG), spectral phase interferometry for direct electric field reconstruction(SPIDER) or dispersion scan(d-scan), have been used to measure the spectral/temporal amplitude/phase or dispersion/chirp of short pulses[16–18]. However, the phase-matching problem of nonlinear crystals and the deficiency in determining the absolute phase(carrier-envelope phase, CEP) both limit their applicability. Instead, direct access to the time-domain electric field $E(t)$ requires a fast nonlinear response that is significantly shorter than an optical cycle[19, 20].

Advancements in strong field physics have provided such ultrashort temporal gates. One widely used technique is attosecond streak camera[21–23], isolated attosecond extreme ultraviolet(XUV) pulses generated by higher-order harmonic generation(HHG) processes are used to ionize atoms[24–30]. The ejected photoelectrons are then streaked to different final energies by the test laser field whose waveform is to be measured. The temporal structure of both the test laser and the attosecond XUV pulse can be accurately reconstructed from the streaking photoelectron spectra[31, 32]. Two other all-optical characterization methods, petahertz optical oscilloscope and attosecond spatial interferometry, both utilize the subfemtosecond tunneling-recombination process during HHG generation as the temporal gate to sample the test optical laser field[33, 34].
in which $\mathbf{p}_\perp$ is the asymptotic photoelectron momentum perpendicular to the fundamental laser polarization. $t_r$ is the rescattering time, and $t_0^{ref}$ is the ionization time of the reference photoelectron wave. The intermediate canonical momentum between tunneling and rescattering is $\mathbf{k}_L = -\frac{1}{t_r - t_0^{ref}} \int_{t_0^{ref}}^{t_r} \mathbf{A}_L(t') dt'$ to ensure that the electron travels back to the ion, while $t_0^R$ is the ionization time of the rescattering wavepacket. Generally, for near-forward rescattering with a small transverse momentum $\mathbf{p}_\perp$, the tunneling time for reference and rescattering(signal) quantum paths are approximately the same: $t_0^{ref} \approx t_0^R$. $\mathbf{A}_L(t) = -\int_{t}^{t'} \mathbf{E}_L(t') dt'$ is the vector potential of the weak test laser field.

However, a $\cos(\Re(\delta \phi))$-like peak structure derived from Eqn. 1 for the transverse momentum distribution(black dashed lines in Fig. 1(b1)(b2) for different asymptotic longitudinal momenta $p_z = 0.2, 0.6$) fails to reproduce the TDSE results(blue dotted lines). This problem can be clarified from the semiclassical(SC) perspective of the Feynman path integral method, which dictates that the dominant contributions come from the regions around the classical trajectories. Fig. 1(c)(d) depict the contour plots of the deflection functions $\mathbf{p}_\perp = \mathbf{p}_\perp(\eta_0, \mathbf{p}_{\perp 0})$ obtained by solving Newton’s equation of motion after the electron emerges at $\eta_0 = \omega \zeta_0 = \omega_0 \Re(\delta \phi) \approx 0.3$ [43][44]. Due to Coulomb potential influence, the $p_0-p_0$ plane can be divided into four signal/reference regional pairs: (S1, R1), (S2, R2), (S3, R3) and (S4, R4)(with the latter two not shown). For the final photoelectron momentum originating from inside these pairs, only two classical trajectories are found(Fig. 1(c)); however, infinite classical trajectories stemming from the circle contour dividing the signal and reference regions all contribute to the same asymptotic momentum(Fig. 1(f) depicts four such classical orbits).

This phenomenon is analogous to the (forward) glory effect in quantum scattering theory [35]. The contributions of infinite so-called glory trajectories(GTs) to the final momentum distribution should be summed up. Referring to Eqn. 1 for simplicity, consider the case with only the NIR fundamental pulse; for a small deviation $\Delta p_\perp$ from the forward direction, we have $\Delta(\Re(\delta \phi)) \sim \Delta p_\perp p_{\perp 0}(t_r - t_0) \sim \Delta p_\perp b_g$, where $p_{\perp 0} \neq 0$ is the initial transverse momentum with the Coulomb potential involved. $b_g \sim p_{\perp 0}(t_r - t_0)$ is interpreted as the asymptotic impact factor of GTs(Fig. 1(f))[42]. Then, the transverse momentum distribution in the near-forward direction is $f(p_{\perp}) \propto |\frac{1}{2\pi} \int_{0}^{2\pi} e^{i\Delta p_\perp b_g \cos \theta} d\theta|^2 = J_0^2(b_g \Delta p_\perp)$. In an OTC field, this would result in $f(p_{\perp}) \propto J_0^2(b_g |\mathbf{p}_{\perp} - \mathbf{p}_{\perp 0}|)$. $\mathbf{p}_{\perp}$ is the transverse momenta corresponding to the primary glory interference maxima(GIM) on the circle contour in Fig. 1(c)(d), $|\mathbf{p}_{\perp} - \mathbf{p}_{\perp 0}| \equiv 0$. This result has successfully interpreted the near-forward SFPH interference fringes in PMD [40][42][46].

Using this SC photoelectron trajectory method, $b_g$ can
be retrieved by back-propagation for each $p_x$ [40]. The resulting squared-Bessel-like peak structure (red solid lines in Fig. 2(b1)(b2)) agrees very well with the TDSE simulation. A SC trajectory Monte Carlo simulation also reproduces the position of the GIM oscillating with $\Delta \tau$ (blue dashed arrows indicate the subcycle excursion of the field with the subfemtosecond glory rescattering process).

One of the consequences is a peak shift of the asymptotic transverse momentum distribution versus time delay are presented in Fig. 3(a1(a2)(a3)) for different final longitudinal momenta $p_x$. The deviation for smaller $p_x$ is due to the Coulomb effects.

Therefore, adding a weak test laser $E_L(\perp E_0)$ introduces an extra factor into the phase difference between the reference and signal photoelectron waves (Eqn. 1) or, classically, slightly perturbs the whole bunch of glory rescattering trajectories (Fig. 1(a1)). One of the consequences is a peak shift of the asymptotic transverse momentum distribution, the same as that in non-dipole strong field ionization [44, 47–51]. Therefore, we can utilize the subfemtosecond glory rescattering process as a fast temporal gate to sample a test laser pulse by varying the time delay between the fundamental and weak light pulses (Fig. 2(a));

$$p_L(\Delta \tau) \approx Re\{-\frac{1}{t_r-t_F^{L}} \int_{t_F^{L}+\Delta \tau}^{t_r+\Delta \tau} A_L(t)dt\}$$  \hspace{1cm} (2)

Fig. 2(b) illustrates the TDSE simulation of PMDs with different time delays; the GIM oscillates with $\Delta \tau$.

The streaking photoelectron spectra of the transverse momentum distribution versus time delay are presented in Fig. 3(a1(a2)(a3)) for different final longitudinal momenta $p_x = 0.4, 0.6, 0.8$. The same test laser light is used as in Fig. 1. TDSE results (black stars) well fit the SC trajectory Monte Carlo simulation results. Blue solid lines are estimated using Eqn. 2.

For all of these complex test light pulses, this approach is also suitable for retrieval of optical waveforms with broad spectral bandwidths. Moreover, even though the proposed procedure
requires that the time delay $\Delta \tau$ be continuously varied, single-shot measurement may be achieved by distributing the atoms spatially and using the spatial dependence of the propagating electromagnetic wave $A(\omega t - \mathbf{k} \cdot \mathbf{r})$ to provide the time delay, where $k = \omega / c$ is the wave vector.

In conclusion, by leveraging the subfemtosecond Coulomb glory rescattering effect as a fast temporal gate, we can sample arbitrary optical waveforms directly in the time domain with electron spectroscopy and reconstruct the temporal structure of the vectorial optical laser pulses. Our method completely avoids the use of attosecond pulses. Our results will facilitate the study of ultrafast electron dynamics in attosecond physics.

This work is supported by the National Natural Science Foundation of China (Grants No. 11674034, No. 11775030, No. 11974057 and No. 11447015) and NSAF (Grants No. U1930402 and No. U1930403).

---

* Electronic address: xiaoqinzhii@iapcm.ac.cn
† Electronic address: jliu@gscaep.ac.cn

[1] M. Drescher, M. Hentschel, R. Kienberger, M. Uiberacker, V. Yakovlev, A. Scrinzi, T. Westerwalbesloh, U. Kleineberg, U. Heinzmann, and F. Krausz, Nature 419, 803 (2002).

[2] E. Goulielmakis, V. S. Yakovlev, A. L. Cavalieri, M. Uiberacker, V. Pervak, A. Apolonski, R. Kienberger, U. Kleineberg, and F. Krausz, Science 317, 769 (2007).

[3] M. Kremer, B. Fischer, B. Feuerstein, V. L. B. de Jesus, V. Sharma, C. Hofrichter, A. Rudenko, U. Thumm, C. D. Schröter, R. Moshammer, and J. Ullrich, Phys. Rev. Lett. 103, 213003 (2009).

[4] G. Sansone, F. Kelkensberg, J. F. Pérez-Torres, F. Morales, M. F. Kling, W. S. Siu, O. Ghafor, P. Johnson, M. Swoboda, E. Benedetti, F. Ferrari, F. Lépine, J. L. Sanz-Vicario, S. Zherebtsov, I. Zhakayev, A. L’Huillier, M. Y. Ivanov, M. Nisoli, F. Martin, and M. J. J. Vrakking, Nature 465, 763 (2010).

[5] A. Schiffrin, T. Paasch-Colberg, N. Karpowicz, V. Apalkov, D. Gerster, S. Mühlbrantd, M. Korhman, J. Reichert, M. Schultze, S. Holzmer, J. V. Barth, R. Kienberger, R. Ernestower, V. S. Yakovlev, M. I. Stockman, and F. Krausz, Nature 493, 70 (2012).

[6] N. Ishii, K. Kaneshima, K. Kitano, T. Kanai, S. Watanabe, and J. Itatani, [Nature Communications 5, 3331] (2014).

[7] M. Garg, M. Zhan, T. T. Luu, H. Lakhota, T. Klostermann, A. Guggenmos, and E. Goulielmakis, [Nature 538, 359] (2016).

[8] S. Rozen, A. Comby, B. Bloch, S. Beauvalet, D. Descamps, B. Fabre, S. Petit, V. Blanchet, B. Pons, N. Dudovich, and Y. Mairesse, Phys. Rev. X 9, 031004 (2019).

[9] P. Del’Haye, A. Schliesser, O. Arcizet, T. Wilken, R. Holzwarth, and T. J. Kippenberg, [Nature 450, 1214] (2007).

[10] Z. Jiang, C.-B. Huang, D. E. Leaird, and A. M. Weiner, [Nature Photonics 1, 463] (2007).

[11] S. T. Cundiff and A. M. Weiner, [Nature Photonics 4, 760] (2010).

[12] H.-S. Chan, Z.-M. Hsieh, W.-H. Liang, A. H. Kung, C.-K. Lee, C.-J. Lai, R.-P. Pan, and L.-H. Peng, Science 331, 1165 (2011).

[13] A. Wirth, M. T. Hassan, I. Grguraš, J. Gagnon, A. Moulet, T. T. Luu, S. Pabst, R. Santra, Z. A. Alahmed, A. M. Azzeer, V. S. Yakovlev, R. Kienberger, K. Yakana, N. Karpowicz, M. Schultze, and F. Krausz, [Nature 534, 86] (2016).

[14] A. Schliesser, N. Piqué, and T. W. Hänsch, [Nature Photonics 6, 440] (2012).

[15] R. Trebino, K. W. DeLong, D. N. Fittinghoff, J. N. Sweetser, M. A. Krumbgel, B. A. Richman, and D. J. Kane, Review of Scientific Instruments 68, 3277 (1997).

[16] A. Iaconis and I. A. Walmsley, Opt. Lett. 23, 792 (1998).

[17] R. Weigand, A. L’Huillier, and H. Crespo, Opt. Express 20, 18732 (2012).

[18] S. B. Park, K. Kim, W. Cho, S. I. Hwang, I. Ivanov, C. H. Nann, and K. T. Kim, Optica 5, 402 (2018).

[19] G. Sansone, E. Benedetti, F. Califari, C. Vozzi, L. Avaldi, R. Flammini, L. Polotto, P. Villoreis, C. Attucci, R. Velotta, S. Stagira, S. De Silvestri, and M. Nisoli, [Science 314, 443] (2006).
[22] E. Goulielmakis, M. Uiberacker, R. Kienberger, A. Baltuska, V. Yakovlev, A. Scrinzi, T. Westerwalbesloh, U. Kleineberg, U. Heinzmann, M. Drescher, and F. Krausz, Science 305, 1267 (2004).

[23] J. Itatani, F. Quéré, G. L. Yudin, M. Y. Ivanov, F. Krausz, and P. B. Corkum, Phys. Rev. Lett. 88, 173903 (2002).

[24] R. Boge, S. Heuser, M. Sabbar, M. Lucchini, L. Gallmann, C. Cirelli, and U. Keller, Opt. Express 22, 26967 (2014).

[25] M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L'Huillier, and P. B. Corkum, Phys. Rev. A 49, 2117 (1994).

[26] A. Baltuska, T. Udem, M. Hentschel, E. Goulielmakis, C. Gohle, R. Holzwarth, V. S. Yakovlev, A. Scrinzi, T. W. Hänsch, and F. Krausz, Nature 421, 611 (2003).

[27] T. Witting, F. Frank, W. A. Okell, C. A. Arrell, J. P. Marangos, and J. W. G. Tisch, Journal of Physics B: Atomic, Molecular and Optical Physics 45, 074014 (2012).

[28] M. J. Abel, T. Pfeifer, P. M. Nagel, W. Boutu, M. J. Bell, C. P. Steiner, D. M. Neumark, and S. R. Leone, Chemical Physics 366, 9 (2009).

[29] F. Ferrari, F. Calegari, M. Lucchini, C. Vozzi, S. Stagira, G. Sansone, and M. Nisoli, Nature Photonics 4, 875 (2010).

[30] I. J. Sola, E. Mével, L. Elouga, E. Constant, V. Strelkov, L. Poletto, P. Villoresi, E. Benedetti, J.-P. Caumes, S. Stagira, C. Vozzi, G. Sansone, and M. Nisoli, Nature Physics 2, 319 (2006).

[31] Y. Mairesse and F. Quéré, Phys. Rev. A 71, 011401(R) (2005).

[32] M. Lucchini, M. Brügmann, A. Ludwig, L. Gallmann, U. Keller, and T. Feurer, Opt. Express 23, 29502 (2015).

[33] K. T. Kim, C. Zhang, A. D. Shiner, B. E. Schmidt, F. Legaré, D. M. Villeneuve, and P. B. Corkum, Nature Photonics 7, 958 (2013).

[34] P. Carpeggiani, M. Reduzzi, A. Comby, H. Ahmadi, S. Kühn, F. Calegari, M. Nisoli, F. Frassetto, L. Polletto, D. Hoff, J. Ullrich, C. D. Schröter, R. Moshammer, G. G. Paulus, and G. Sansone, Nature Photonics 11, 383 (2017).

[35] H. Wang, M. Chini, S. D. Khan, S. Chen, S. Gilbertson, X. Feng, H. Mashiko, and Z. Chang, Journal of Physics B: Atomic, Molecular and Optical Physics 42, 134007 (2009).

[36] F. Krausz and M. I. Stockman, Nature Photonics 8, 205 (2014).

[37] M. Chini, K. Zhao, and Z. Chang, Nature Photonics 8, 178 (2014).

[38] Y. Huismans, A. Rouzée, A. Gijsbertsen, J. H. Jungmann, A. S. Smolkowska, P. S. W. M. Logman, F. Lépine, C. Cauchi, S. Zamith, T. Marchenko, J. M. Bakker, G. Berden, B. Redlich, A. F. G. van der Meer, H. G. Muller, W. Vermin, K. J. Schafer, M. Spanner, M. Y. Ivanov, O. Smirnova, D. Bauer, S. V. Popruzhenko, and M. J. J. Vrakking, Science 331, 61 (2011).

[39] P. B. Corkum, Phys. Rev. Lett. 71, 1994 (1993).

[40] Q. Z. Xia, J. F. Tao, J. Cai, L. B. Fu, and J. Liu, Phys. Rev. Lett. 121, 143201 (2018).

[41] M. Li, H. Xie, W. Cao, S. Luo, J. Tan, Y. Feng, B. Du, W. Zhang, Y. Li, Q. Zhang, P. Lan, Y. Zhou, and P. Lu, Phys. Rev. Lett. 122, 183202 (2019).

[42] S. Brennecke and M. Lein, Phys. Rev. A 100, 023413 (2019).

[43] J. Liu, Classical Trajectory Perspective of Atomic Ionization in Strong Laser Fields (Springer-Verlag, Berlin, 2014).

[44] J. Danék, K. Z. Hatsagortsyan, and C. H. Keitel, Phys. Rev. A 97, 063409 (2018).

[45] K. W. Ford and J. A. Wheeler, Annals of Physics 7, 259 (1959).

[46] S. D. López and D. G. Arbó, Phys. Rev. A 100, 023419 (2019).

[47] J. F. Tao, Q. Z. Xia, J. Cai, L. B. Fu, and J. Liu, Phys. Rev. A 95, 011402(R) (2017).

[48] A. Ludwig, J. Maurer, B. W. Mayer, C. R. Phillips, L. Gallmann, and U. Keller, Phys. Rev. Lett. 113, 243001 (2014).

[49] M.-X. Wang, H. Liang, X.-R. Xiao, S.-G. Chen, W.-C. Jiang, and L.-Y. Peng, Phys. Rev. A 98, 023412 (2018).

[50] P.-L. He, D. Lao, and F. He, Phys. Rev. Lett. 118, 163203 (2017).

[51] A. Hartung, S. Eckart, S. Brennecke, J. Rist, D. Trabet, K. Feher, M. Richter, H. Samm, S. Zeller, K. Henrichs, G. Kastirke, J. Hoehl, A. Kalinin, M. S. Schöffler, T. Jahnke, L. P. H. Schmidt, M. Lein, M. Kunitski, and R. Dörner, Nature Physics (2019), 10.1038/s41567-019-0653-y.

[52] The carrier frequency is $\omega_L = 0.038$ a.u., corresponding to a wavelength of 1200 nm, with a time duration of $T_L = 4 \times \frac{2\pi}{\omega_L}$, $T_d = T_L/2$. And $\epsilon_L = 0.08 \times \epsilon_0$. 