Ponderomotive electron acceleration by polarization-gated surface-enhanced optical fields

Peifen Lu, Jian Wu,[a] Hongxing Qi, and Heping Zeng[b]
State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai 200062, People's Republic of China

(Received 26 August 2008; accepted 17 October 2008; published online 21 November 2008)

We study ponderomotive electron acceleration with polarization-gated surface-plasmon-resonance enhanced optical fields excited by two counter-incident femtosecond laser pulses. Such a polarization-gated excitation scheme creates an intense linearly polarized high-gradient evanescent field for electron acceleration. The maximum kinetic energy of the accelerated ultrafast electrons is doubled with a symmetric angular distribution about the normal of the metal surface as compared with the conventional one-pulse excitation scheme. © *2008 American Institute of Physics*.
[DOI: 10.1063/1.3020711]

Ultrafast electron pulses with ultrahigh spatial and temporal resolutions are significant to reveal the underlying dynamics in the microworld through experiments such as time-resolved electron diffraction[1–4] and electron microscopy.[5–8] Femtosecond electron pulses with a kinetic energy up to 200 keV can be generated by exciting the photocathode with frequency-tripled femtosecond laser pulses and accelerated with a dc accelerator.[6] Alternatively, ultrafast electron pulses with keV kinetic energy can be produced using surface-plasmon-resonance (SPR) enhanced optical fields excited by femtosecond laser pulses with spatially high-gradient intensity distribution.[9–11] This provides a simple all-optical process for synchronous generation and acceleration of ultrafast electron pulses, which represents the potential for the development of compact optical-pump electron-probe systems. As the incident optical field resonates with the surface plasmon (SP),[12] it is significantly enhanced in intensity close to the metal film and exponentially decreases away from the surface as $E_{\mathrm{SP}}(y,t)=\eta E_l(t)\exp(-\alpha y)$, which leads to a highly spatial gradient evanescent field and can be used for ponderomotive electron acceleration. Here, η is the electric field enhancement factor, $\alpha^{-1}(<\lambda_l)$ is the evanescent decay length into the vacuum side, E_l and E_{SP} are the incident and SPR-enhanced optical fields, respectively. However, the electron acceleration is dramatically influenced by the elliptically polarized feature of the SPR-enhanced optical field, which limits the effective field intensity and thus the kinetic energy of the emitted electron pulses. Recently, it was demonstrated that polarization of the SPR-enhanced optical field can be controlled to be linear by using polarization-gated excitation scheme.[13]

In this paper, we show that the polarization-gated SPR excitation scheme can be used to efficiently accelerate electrons on the basis of ponderomotive force. In contrast with the conventional one-pulse excitation scheme, linear polarization mode can be created as the counter-propagating SPR-enhanced optical pulses with counter-rotating elliptical polarizations are matched in both time and space, which increases the effective field intensity as well. As excited by two synchronized femtosecond laser pulses, the emitted electrons can be accelerated to a doubled maximum kinetic energy with a symmetric angular distribution about the normal of the metal surface.

As shown in Fig. 1, the Kretschmann configuration is considered for the coupling of optical field and SP waves. In order to have an intuitive picture of the polarization-gated excitation scheme under the driving of two synchronized *p*-polarized incident femtosecond laser pulses (femtosecond pulses A and B as shown in Fig. 1), we first consider the simplified formulae[14] for the SPR-enhanced optical fields on the vacuum side of the metal film,

$$
\begin{aligned}
E_{\mathrm{SP},x\_A}(x,y,t) &= \beta E(x,t)\cos[k_{\mathrm{SP}}(x+L/2)-\omega_0 t-\pi/2 \\
&\quad +\phi_0]e^{-\alpha y}, \\
E_{\mathrm{SP},y\_A}(x,y,t) &= E(x,t)\cos[k_{\mathrm{SP}}(x+L/2)-\omega_0 t+\phi_0]e^{-\alpha y}, \\
E_{\mathrm{SP},x\_B}(x,y,t) &= \beta E(-x,t)\cos[-k_{\mathrm{SP}}(x-L/2)-\omega_0 t+\pi/2 \\
&\quad +\phi_0]e^{-\alpha y}, \\
E_{\mathrm{SP},y\_B}(x,y,t) &= E(-x,t)\cos[-k_{\mathrm{SP}}(x-L/2)-\omega_0 t \\
&\quad +\phi_0]e^{-\alpha y}, \qquad (1)
\end{aligned}
$$

where $E_{\mathrm{SP},x(y)\_A(B)}$ is the $x(y)$ component of the SPR-enhanced optical field excited by incident laser pulse A (or

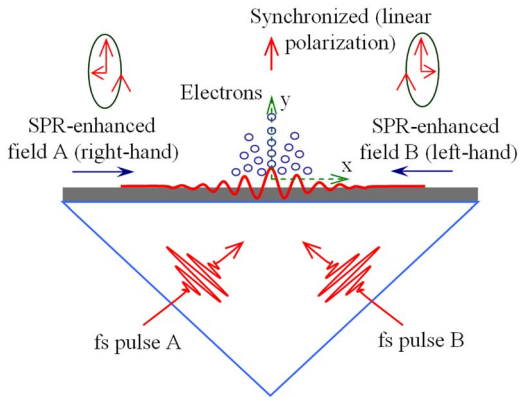

FIG. 1. (Color online) Ponderomotive electron acceleration scheme based on polarization-gated SPR-enhanced optical field.

[a]Electronic mail: jwu@phy.ecnu.edu.cn.
[b]Electronic mail: hpzeng@phy.ecnu.edu.cn.

B), β is the ratio between the amplitudes of the x- and y-components of E_{SP}, $E(x,t)$ is the envelope function of SPR-enhanced fields, k_{SP} is the wave vector of the SP wave, L is the length of the metal film, ω_0 and φ_0 are, respectively, the carrier frequency and carrier-envelope phase of the optical pulse. By assuming that $E(x,t)$ equals to a constant E_0 and $\varphi_0=0$, the SPR-enhanced optical fields close to the surface excited by these two p-polarized laser pulses are

$$E_{SP,x}(x,0,t) = E_{SP,x\_A}(x,0,t) + E_{SP,x\_B}(x,0,t) = 2\beta E_0 \sin(k_{SP}x)\cos(k_{SP}L/2 - \omega_0 t),$$

$$E_{SP,y}(x,0,t) = E_{SP,y\_A}(x,0,t) + E_{SP,y\_B}(x,0,t) = 2E_0 \cos(k_{SP}x)\cos(k_{SP}L/2 - \omega_0 t),$$

$$E_{SP}(x,0,t) = 2E_0 \cos(k_{SP}L/2 - \omega_0 t) \times \sqrt{\beta^2 + (1-\beta^2)\cos^2(k_{SP}x)}. \quad (2)$$

This indicates a linear polarization mode rather than the intrinsic elliptical one, which further periodically evolves along the surface with a period of π/k_{SP}. Meanwhile, it can be derived that using polarization-gated SPR excitation scheme the maximum enhanced intensity can be increased, which is 2 times as large as the one excited by the one-pulse excitation scheme for same incident total intensity. In fact, $E(x,t)$ is determined by the spatiotemporal profiles of the incident pulse. Thus, for the scheme excited by two synchronized femtosecond laser pulses with equal intensity, only the mode at $x=0$ is absolutely linearly polarized with no x-component of E_{SP}, and the modes at the surface away from $x=0$ evolve to be elliptically polarized.

In the following simulations, the full distribution of the enhanced optical field close to the surface is obtained by solving the Maxwell equations for the optical field evolution using the finite-difference time-domain method.[15] Two p-polarized 24 fs laser pulses at 800 nm with equal intensities are steered to excite SP waves on a 50 nm silver film deposited on a prism, which launches SPR at an incident angle of 45°. A peak intensity enhancement factor of 80 (or equivalent 40 relative to the sum incident intensity of the two incident pulses) can be achieved by using the polarization-gated scheme, which is almost doubled as compared with the conventional one-pulse incident case with an enhancement factor of 22 for the case considered here. The test electrons are placed uniformly along the metal film in space and all the temporal contributions for intensities larger than 10% of the pulse peak are included (the electrons excited by the rest fields are negligible for their fairly low energies and observation probabilities) in order to represent all the possible trajectories of the accelerated electrons. The relative weight of the emitted electron is obtained by considering the electron emission as a third-order multiphoton process,[16] i.e., a weight proportional to $I^3(x,y,t)$ is assigned to each accelerated electron where $I(x,y,t)$ stands for the intensity of the SPR-enhanced optical field. Once an electron is ejected from the metal film, the high-gradient evanescent optical fields push it away from the surface and its motion is governed by the classical Lorentz force equation, $d\boldsymbol{v}/dt = q/m(\boldsymbol{E} + \mu_0 \boldsymbol{v} \times \boldsymbol{H})$, where q/m is the charge-to-mass ratio and $\boldsymbol{v}$ is the velocity of the electron. The electron is assumed to have zero initial kinetic energy as it is librated from the metal film by the SPR-enhanced optical fields. At the same time, the electron is considered to be absorbed if it returns to the metal-vacuum boundary and the secondary electron emission is ignored in our calculations. Here, the space-charge effect[17] is negligible due to the fact that it only takes several tens of femtoseconds for the electrons to fully accelerate to their final kinetic energy.

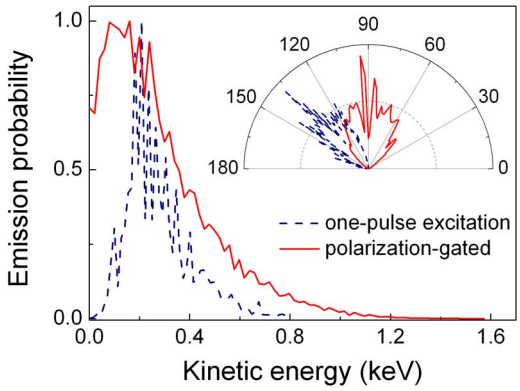

FIG. 2. (Color online) Kinetic energy spectra and angular distributions (inset) of accelerated electrons by SPR-enhanced optical fields. The navy and red lines stand for the results by using one-pulse excitation scheme with a maximum E_l of 0.4×10^{11} V/m ($E_{SP}\sim1.87\times10^{11}$ V/m), and polarization-gated scheme with a maximum E_l of 0.28×10^{11} V/m for each incident femtosecond laser pulse ($E_{SP}\sim2.63\times10^{11}$ V/m), respectively.

Figure 2 shows the kinetic energy spectra of emitted electrons produced respectively by one-pulse excitation and polarization-gated excitation schemes. A similar overall feature is observed for these two cases with a peak emission probability at a certain kinetic energy. For the conventional one-pulse excitation scheme (pulse A as shown in Fig. 1), the peak of the emission probability is located at 208 eV with a full width at half maximum (FWHM) of kinetic energy distribution as 150 eV; while for the polarization-gated excitation scheme, the peak emission probability is shifted to 160 eV with a much broader FWHM distribution as 360 eV. However, even the incident pulses have the same intensity in total, the maximum kinetic energy of the emitted electron is doubled by using the polarization-gated SPR excitation scheme. Such a high-kinetic-energy electron emission mainly originated from SPR-enhanced optical fields with an increased effective enhancement factor and gated linear polarization. During an optical cycle, the earlier the electrons emitted from the same point of the metal surface, the higher the kinetic energy of the electrons can be obtained with a lower emission probability. This results in a higher maximum kinetic energy with a lower emission probability as shown in Fig. 2.

The angular distributions of the emitted electrons for these two different schemes are shown in the inset of Fig. 2. Here, all the electrons with different kinetic energies are included. The electrons emit in an angle range spanning from 90° to 180° with a peak at 137° and an angular FWHM of 30° for the one-pulse excitation scheme, while the angular distribution for polarization-gated scheme appears symmetrically about the normal of the metal surface and spans from 0° to 180° with a favorable acceleration direction around 90° and an angular FWHM of 50°. The symmetric angular distribution of electron emissions result from the symmetric SPR coupling geometry as illustrated in Fig. 1. The sharp peaks near the normal of the metal surface come from the

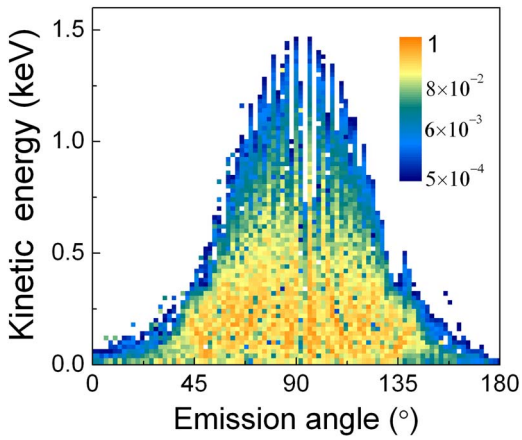

FIG. 3. (Color online) Angle-resolved energy distribution of the accelerated electrons by polarization-gated SPR-enhanced optical field, where the false color stands for the observation probability.

polarization-gated optical fields with minimum x-component and maximum y-component of E_{SP}, while the broader shoulders are mainly due to the periodic spatial evolution of the polarization modes of the enhanced optical fields. Accordingly, by using a particular surface morphology of the metal film (for example, by depositing a dielectric layer on the metal surface with a suitably wide opening or coating the prism surface with a quite narrow metal film, where only the perpendicularly linear polarization mode is excited), it is possible to produce an ultrafast electron pulse with a quite narrow emission angular distribution of several degrees in the forward direction.

The sources of the emitted electrons with different kinetic energies can be seen more clearly by taking the angle-resolved kinetic energy distribution of the accelerated electrons as shown in Fig. 3, which describes the relationship of the emission angle, kinetic energy, and emission probability. In an optical cycle, electrons excited at different times have different kinetic energies. Consequently, the gated optical field with polarization perpendicular to the metal film creates electrons with a large range of kinetic energy distribution and an almost doubled maximum, which shows a normal emission direction. The enhanced optical fields at various positions along the surface with different polarization modes lead to electron emission with relatively low kinetic energies at the corresponding angles.

In summary, we numerically demonstrate that the polarization-gated SPR excitation scheme can be used to efficiently produce ultrafast electrons based on its improved polarization mode and enhanced effective intensity, which doubles the maximum kinetic energy of the accelerated electrons and leads to a symmetric angular emission distribution. Well-behaved electron pulses with particular kinetic energies can be obtained along various emission angles.

This work was funded in part by National Natural Science Fund (Grant Nos. 10774045, 10525416, and 10804032), National Key Project for Basic Research (Grant No. 2006CB806005), Projects from Shanghai Science and Technology Commission (Grant Nos. 06JC14025, 06SR07102, and 08ZR1407100), Program for Changjiang Scholars and Innovative Research Team, and Shanghai Educational Development Foundation (Grant No. 2008CG29).

[1] B. Siwick, J. Dwyer, R. Jordan, and R. Miller, Science **302**, 1382 (2003).
[2] P. Baum, D. Yang, and A. Zewail, Science **318**, 788 (2007).
[3] D. Yang, C. Lao, and A. Zewail, Science **321**, 1660 (2008).
[4] F. Carbone, P. Baum, P. Rudolf, and A. Zewail, Phys. Rev. Lett. **100**, 035501 (2008).
[5] V. Lobastov, R. Srinivasan, and A. Zewail, Proc. Natl. Acad. Sci. U.S.A. **102**, 7069 (2005).
[6] H. Park, J. Baskin, O. Kwon, and A. Zewail, Nano Lett. **7**, 2545 (2007).
[7] P. Baum and A. Zewail, Proc. Natl. Acad. Sci. U.S.A. **104**, 18409 (2007).
[8] A. Gahlmann, S. Park, and A. Zewail, Phys. Chem. Chem. Phys. **10**, 2894 (2008).
[9] J. Zawadzka, D. Jaroszynski, J. Carey, and K. Wynne, Appl. Phys. Lett. **79**, 2130 (2001).
[10] J. Kupersztych, P. Monchicourt, and M. Raynaud, Phys. Rev. Lett. **86**, 5180 (2001).
[11] S. Irvine, A. Dechant, and A. Elezzabi, Phys. Rev. Lett. **93**, 184801 (2004).
[12] H. Raether, *Surface Plasmons on Smooth and Rough Surfaces and on Gratings* (Springer, Berlin, 1988).
[13] J. Wu, H. Qi, and H. Zeng, Appl. Phys. Lett. **93**, 051103 (2008).
[14] P. Dombi and P. Rácz, Opt. Express **16**, 2887 (2008).
[15] A. Taflove, *Computational Electrodynamics* (Artech House, Boston, 1995).
[16] S. Irvine and A. Elezzabi, Phys. Rev. A **73**, 013815 (2006).
[17] B. Siwick, J. Dwyer, R. Jordan, and R. Miller, J. Appl. Phys. **92**, 1643 (2002).