Revealing the effect of atomic orbitals on the phase distribution of an ionizing electron wave packet with circularly polarized two-color laser fields

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Abstract: We theoretically study the interference of photoelectrons released from atomic \( p_{\pm} \) orbitals in co-rotating and counter-rotating circularly polarized two-color laser pulses consisting of a strong 400-nm field and a weak 800-nm field. We find that in co-rotating fields the interference fringes in the photoelectron momentum distributions are nearly the same for \( p_{\pm} \) orbitals, while in counter-rotating fields the interference fringes for \( p_{+} \) and \( p_{-} \) orbitals oscillate out of phase with respect to the electron emission angle. The simulations based on the strong-field approximation show good agreement with the numerical solutions of the time-dependent Schrödinger equation. We find that different phase distributions of the electron wave packets emitted from \( p_{+} \) and \( p_{-} \) orbitals can be easily revealed by the counter-rotating circularly polarized two-color laser fields. We further show that the photoelectron interference patterns in the circularly polarized two-color laser fields record the time differences of the electron wave packets released within an optical cycle.

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1. Introduction

Strong-field atomic ionization is one of the most important processes in ultrafast physics, which initiates a series of interesting phenomena, such as high-order harmonic generation [1–6], photoelectron interference and holography [7–12], laser-induced electron diffraction [13,14], and nonsequential double ionization [15–17]. Strong-field ionization and those strong-field phenomena have attracted increasing interest and have been studied continuously during the past decades. Generally, noble-gas atoms were used as the experimental targets in those studies due to their simple and stable structures. Most of those experimental results were interpreted without considering the effect of the magnetic quantum number of the initial orbital [18–20]. However, except for helium atom, the noble-gas atoms naturally carry \( p_{\pm} \) orbitals, which have a non-negligible effect on the electron dynamics in the strong-field ionization process [21–25]. It has been shown that the ionization rates and photoelectron momentum distributions (PMDs) depend sensitively on the magnetic quantum number of the initial orbital in a rotating laser field [21–23,26–31]. In a right circularly polarized laser field (defined from the point of view of the source) of nonadiabatic tunneling regime, the ionization rate of \( p_{-} \) orbital (counter-rotating with the laser field) is higher than that of \( p_{+} \) orbital (co-rotating with the laser field), while the released electron from \( p_{+} \) orbital can achieve higher kinetic energy than that from \( p_{-} \) orbital. Recently, it
has been further predicted that the initial $p_\pm$ orbitals are deformed and polarized in elliptically polarized laser fields, leading to different angular shifts in the PMDs [31].

Due to the spin-orbit couplings in the ground states of atoms and molecules, the selective ionization of the current-carrying valence $p$ orbitals in circularly polarized laser fields can be used to produce spin-polarized photoelectrons [32]. Using ultraviolet circularly polarized laser fields, it was predicted that the photoelectron spin polarization can be enhanced by momentum gating [33], which was verified in the experiment [34].

Most of the previous studies focused on the amplitude of the electron wave packet emitted from the current-carrying orbitals, while its phase distribution remained unexplored. The phase distribution of the electron wave packet is key to understanding the wavelike property of the photoelectron. In a single-color circularly or elliptically polarized laser field, the electrons emitted at different ionization times within an optical cycle are streaked to different emission angles [35–40], suppressing intracycle interferences of the electron wave packets in the PMD. As a result, the phase information of the electron wave packet is absent in a single-color circularly or elliptically polarized laser field. Recently, using circularly polarized two-color laser fields, fruitful interference structures have been observed in the PMDs of strong-field ionization of noble gas atoms [18,41,42]. However, those interference patterns were interpreted without taking the effect of the initial orbital into account.

In this work, we study the effect of the atomic $p_\pm$ orbitals on the phase distributions of the ionizing electron wave packets in circularly polarized two-color laser fields. A strong 400-nm right circularly polarized laser field rotates in the anticlockwise direction and a perturbative 800-nm circularly polarized laser field is added to generate subcycle photoelectron interference in the PMD. The two fields have either the same helicity (co-rotating) or opposite helicity (counter-rotating). To avoid deformation of the initial $p_\pm$ orbitals in the strong laser fields [31], the 800-nm field is chosen to be weak enough. Based on the numerical solution of the time-dependent Schrödinger equation (TDSE) and the strong-field approximation (SFA) model, we find that the interference fringes in the PMDs in co-rotating fields are nearly the same for the $p_\pm$ orbitals, while in counter-rotating fields the maxima and minima of the interference fringes for the $p_+$ and $p_-$ orbitals are roughly opposite. Using the SFA, we show that the photoelectron wave packets emitted from the $p_+$ and $p_-$ orbitals carry different phase distributions, which are imprinted into the interference fringes in the PMDs. We further show that the phase difference of the electron wave packets released within an optical cycle is approximately linear to the ionization time difference in the circularly polarized two-color laser field. Because the ionization time difference in the counter-rotating fields is larger than that in the co-rotating fields, the effect of the atomic $p_\pm$ orbitals on the phase distribution can be easily revealed in the counter-rotating fields.

2. Theoretical methods

2.1. TDSE calculation

We numerically solve the two-dimensional TDSE of a model neon atom ionized by circularly polarized two-color laser fields. With the single-active-electron approximation, the TDSE of a $2p$ valence electron of the model neon atom with $m = \pm 1$ in strong laser fields can be expressed as,

$$\frac{i}{\hbar} \frac{\partial \Psi_{\pm}(r; t)}{\partial t} = \left[-\frac{1}{2} \nabla^2 + V(r) + r \cdot E(t)\right] \Psi_{\pm}(r; t),$$

where $\Psi_{\pm}(r; t)$ is time-dependent wave function for the magnetic quantum numbers of $m = \pm 1$. $V(r)$ is the effective potential of the model neon atom, which is expressed as [27],

$$V(r) = -\frac{Z(r)}{\sqrt{r^2 + a}},$$

(2)
with \( Z(r) = 1 + 9 \exp(-r^2) \) and the soft-core parameter \( a = 2.88172 \) to match the ionization potential of the neon atom \( I_p = 0.793 \) a.u. [27,43]. \( r \) is the electron position. The circularly polarized two-color laser fields \( \mathbf{E}(t) = E_x(t)\hat{e}_x + E_y(t)\hat{e}_y \) is written as,

\[
E_x(t) = \frac{f(t)}{\sqrt{2}} \left[ E_{400} \cos(2\omega t) + E_{800} \cos(\omega t + \Delta \varphi) \right]
\]

\[
E_y(t) = \frac{f(t)}{\sqrt{2}} \left[ E_{400} \sin(2\omega t) \pm E_{800} \sin(\omega t + \Delta \varphi) \right],
\]

where \( f(t) \) is laser pulses envelope. \( E_{800} \) and \( E_{400} \) are the amplitudes of the 800-nm and 400-nm laser fields, respectively. \( \omega \) is the frequency of the 800-nm field. In the second term in the second row of Eq. (3), the sign is “+” (“-”) for co-rotating (counter-rotating) circularly polarized two-color laser fields. \( \Delta \varphi \) is the relative phase between the two-color components. Due to the rotational symmetry, we choose \( \Delta \varphi = 0 \) in this study. The initial electron wave function \( \Psi^0 \) for magnetic quantum numbers \( m = \pm 1 \) is given by \( \Psi^0 = \frac{1}{\sqrt{2}}(\Psi_x \pm i\Psi_y) \) with the normalized eigenfunctions \( \Psi_x \) and \( \Psi_y \) [43]. The degenerate states \( \Psi_{x,y} \) are obtained with imaginary time propagation method [44].

In the numerical simulations, the intensities of the fundamental field (800 nm) and second harmonic (400 nm) are \( 3 \times 10^{12} \) W/cm\(^2\) and \( 3 \times 10^{14} \) W/cm\(^2\), respectively. We use a trapezoidal pulse with a flat part of \( 4T_0 \) and \( 2T_0 \) turn-on and turn-off, in which \( T_0 \) is the optical cycle of the 800-nm field. The time step is set to be \( \Delta \tau = 0.1 \) a.u. The cartesian grid space ranges from -600 a.u. to 600 a.u. for both \( x \) and \( y \) directions with a spatial step of 0.1 a.u. After the laser pulse is finished, the wave function is further propagated for an additional four optical cycles to ensure that the ionized part is thoroughly separated from the bound part [45].

### 2.2. SFA simulation

In the SFA model, the transition amplitude of an electron from atomic ground state \(|\psi_0\rangle\) to continuum state \(|\psi_p\rangle\) with canonical momentum \( p \) is given by,

\[
M(p) = -i \int_{-\infty}^{\infty} \langle \psi_p | H(t) | \psi_0 \rangle dt,
\]

where \( H(t) = r \cdot \mathbf{E} \) is electron-laser interaction operator. The continuum state can be approximately denoted by the plane-wave ket vector \(|\mathbf{r} p \rangle = (2\pi)^{3/2} e^{i\mathbf{p} \cdot \mathbf{r}}. \) Thus, the transition amplitude is expressed as,

\[
M(p) = \int_{-\infty}^{\infty} \langle \psi_0 | [\mathbf{r} \cdot \mathbf{E}(t)] | \mathbf{r} p \rangle e^{-iS(p)} dt,
\]

where \( S(p) = -\int_0^T \mathbf{E}(t) dt \) is the laser vector potential. \( S(p) \) is the classical action. The initial orbital of the neon atom \( \psi_0 \) is given by the asymptotic expression \( \psi_0(r) = (\theta \phi | \psi_0 \rangle = A r^{-l} \exp(-\kappa r) Y_{lm}(\theta, \phi) \) [46]. Here \( Y_{lm}(\theta, \phi) \) is spherical harmonics function, \( l \) and \( m \) are orbital and magnetic quantum numbers, respectively. \( \kappa = \sqrt{2} T_p \) and \( \nu = \frac{Z}{\kappa} \) (\( Z = 1 \) for atoms) are related to the ionization potential.

The integral in Eq. (5) can be solved by the saddle-point method. Since the term \( S(p) \) varies faster than \( \langle \mathbf{p} + A(t) | \mathbf{r} \cdot \mathbf{E}(t) | \psi_0 \rangle \) in time, only the integral around the saddle point has a large contribution. The saddle point is determined by the equation \( [\mathbf{p} + A(t)]^2 / 2 + I_p = 0 \). Thus, Eq. (5) can be rewritten as [46–48],

\[
M_{2p_c}(p) = C_s \sum_{s} \sqrt{\frac{3}{8\pi\kappa}} \left( \frac{q_s}{i\kappa} \right)^{l+1} \left( \frac{2\pi}{S_c} \right)^{(\nu+1)/2} \nu s e^{-iS_s},
\]

where \( C_s \) is a constant independent of the magnetic quantum number. \( \mathbf{q}_s = \mathbf{p} + A(t_s) \) satisfies the saddle point equation \( \mathbf{q}_s = -\kappa^2 \). Here \( q_s \rightarrow i\kappa \) according to Ref. [48]. \( Y_s(\mathbf{q}_s) = \)}
\[ [\mathbf{p}_x + \mathbf{A}_x(t_s)] \pm i[\mathbf{p}_y + \mathbf{A}_y(t_s)], \] where “±” represents the magnetic quantum numbers \( m = \pm 1 \).

\[ S''_s = -\mathbf{E}(t_s) \cdot [\mathbf{p} + \mathbf{A}(t_s)], \] \( t_s = t_0 + it_i \) is the complex saddle point time. The real part of the saddle point time \( t_0 \) corresponds to the release time of the electron. One can find that the preexponential factor in Eq. (6) is related to the magnetic quantum number, which will affect the phase and amplitude distributions of the electron wave packets released from \( p_\pm \) orbitals.

3. Results and discussions

We first show in Figs. 1(a) and 1(b) the PMDs in a single-color 400-nm right circularly polarized field with a peak intensity of \( 3 \times 10^{14} \) W/cm\(^2\) by solving the TDSE for \( p_+ \) and \( p_- \) orbitals, respectively. The PMDs are shown in polar coordinate with \( p_r = \sqrt{p_x^2 + p_y^2} \) being the momentum drift and \( \theta \) being the electron emission angle in the laser polarization plane. The maxima in the PMDs have been normalized to be unity. One can see that a series of above threshold ionization (ATI) peaks appear in the PMDs for both \( p_+ \) and \( p_- \) orbitals, which are separated by an energy interval of \( \sim 3.1 \) eV. Those ATI peaks result from the interference of electron wave packets released periodically in time at different laser cycles, i.e., intercycle interference [49]. Compared with the case of \( p_- \) orbital, the electron released from \( p_+ \) orbital achieves a larger momentum drift \( p_r \) in the laser field. This agrees with previous studies [24,26,31]. One can clearly see in Fig. 1 that the photoelectron momentum spectra are independent of the emission angle for both \( p_+ \) and \( p_- \) orbitals, which comes from the fact that the intracycle interference is absent in a single-color circularly polarized field. Thus the phase information of the released electron wave packets can not be revealed in a single-color circularly polarized field.

To generate intracycle interference of the electron wave packets, we add a weak 800-nm circularly polarized laser field co-rotating or counter-rotating with respect to the strong 400-nm field. In Figs. 2(a) and 2(b), we show the PMDs in the co-rotating circularly polarized two-color laser fields from the TDSE calculation for the \( p_+ \) and \( p_- \) orbitals, respectively. One can see that the sidebands appear between adjacent 400-nm ATI peaks in the PMDs for both \( p_+ \) and \( p_- \) orbitals. The intensity of the sidebands and main ATI peaks oscillate roughly \( \pi \) out of phase with the emission angle, which is consistent with recent experimental results [18,20,41]. Comparing Figs. 2(a) with 2(b), one can see that the electron released from \( p_+ \) orbital achieves a larger momentum drift in the laser field, which is the same as in Fig. 1. Moreover, the oscillation of each sideband or main ATI peak is nearly the same for \( p_+ \) and \( p_- \) orbitals. The corresponding SFA simulation with the same laser pulses are shown in Figs. 2(c) and 2(d), in which the interference fringes in the PMDs are nearly the same as the TDSE results.
Fig. 2. The PMDs of Ne in co-rotating circularly polarized two-color laser fields for atomic $p_\pm$ orbitals calculated by the TDSE [(a)(b)] and the SFA [(c)(d)]. The left and right columns correspond to $p_+$ and $p_-$ orbitals, respectively.

In Figs. 3(a) and 3(b), we also show the PMDs in the counter-rotating circularly polarized two-color laser fields from the TDSE calculation for $p_+$ and $p_-$ orbitals, respectively. Similar to Fig. 2, the sidebands also appear between adjacent 400-nm ATI peaks in the PMDs. The intensity of both main ATI and sideband peaks oscillates with respect to the emission angle. Compared with Fig. 2, though only the rotating direction of the weak 800-nm field is reversed, those oscillations in the PMDs are dramatically changed. Interestingly, those oscillations from

Fig. 3. The same as Fig. 2 but in counter-rotating circularly polarized two-color laser fields.
\(p_+\) and \(p_-\) orbitals are very different. For the electron final momentum \(p_r \lesssim 1\) a.u., the maxima and minima within each ATI peak for \(p_+\) and \(p_-\) orbitals are roughly opposite along the electron emission angle direction. In Figs. 3(c) and 3(d), the corresponding SFA results are shown, which also agree with the TDSE results.

In order to clearly see the difference of the interference fringes in the PMDs between \(p_+\) and \(p_-\) orbitals, in Figs. 4(a) and 4(b), we show the lineouts taken at \(p_r = 0.85\) a.u. from the TDSE calculation in the co-rotating and counter-rotating circularly polarized two-color laser fields for \(p_+\) and \(p_-\) orbitals, respectively. All lineouts reveal three interference maxima along the emission angle direction. In Fig. 4(a), the maxima and minima of the lineouts are almost the same for \(p_+\) and \(p_-\) orbitals in the co-rotating fields. However, it is obvious in Fig. 4(b) that the lineouts for \(p_+\) and \(p_-\) orbitals oscillate out of phase in the counter-rotating fields. We also show the same lineouts from the SFA simulation in Figs. 4(c) and 4(d), which are consistent with the TDSE results.

Next we use the SFA to understand the physical origin of the different interference patterns for the \(p_+\) and \(p_-\) orbitals. In fact, both intercycle and intracycle interferences of the electron wave packet will contribute to the PMDs. The intercycle interference of the electron wave packets leads to the ATI-like peaks along the \(p_r\) direction with no dependence on the emission angle \(\theta\), as shown in Fig. 1. Thus the oscillation of the main ATI and sideband peaks with respect to the emission angle comes from the intracycle interferences of the electron wave packets. We show in Figs. 5(a) and 5(b) the intracycle interference patterns calculated by the SFA for \(p_+\) and \(p_-\) orbitals in the co-rotating circularly polarized two-color laser fields. The corresponding results in the counter-rotating circularly polarized two-color laser fields are shown in Figs. 5(c) and 5(d). To rule out the effect of the amplitude of the ionizing electron wave packet on the intracycle interference patterns, we have normalized the probability to the maximum at each \(p_r\).

One can see that the intracycle interference in the co-rotating fields reveals a wavelike pattern. Comparing Figs. 5(a) with 5(b), we find that the wavelike interference patterns are nearly the same for \(p_+\) and \(p_-\) orbitals. In the counter-rotating fields, there are several node structures along the emission angle direction for \(p_r \lesssim 1\) a.u., which means that the phase difference of the electron wave packets contributing to the intracycle interference changes rapidly with \(\theta\). Moreover, the maxima and minima of the intracycle interferences in the \(\theta\) direction for \(p_+\) and \(p_-\) orbitals are
roughly opposite, as guided by the black dashed lines in Figs. 5(c) and 5(d). Therefore, the phase distributions for the electrons released from $p_+$ and $p_-$ orbitals are different, leading to different interference fringes in the PMDs.

![Fig. 5. The intracycle interference patterns in the co-rotating [(a)(b)] and counter-rotating [(c)(d)] circularly polarized two-color laser fields calculated by the SFA. The left and right columns correspond to $p_+$ and $p_-$ orbitals, respectively. The maximum for each $p_r$ has been normalized to be unity.](image)

To understand different roles of the atomic orbitals in co-rotating and counter-rotating circularly polarized two-color laser fields, we next study the subcycle electron dynamics. We show in Fig. 6 the electric field $E(t) = \sqrt{E_x^2 + E_y^2}$ of the two-color laser fields with almost one optical cycle. The electric field has one peak (three peaks) in one optical cycle for the co-rotating (counter-rotating) circularly polarized two-color laser fields. There are two electron wave packets released within one laser cycle with the same final momentum, which are indicated by WP1 and WP2 in Fig. 6. The interference of WP1 and WP2 corresponds to the intracycle interference in Fig. 5. In Fig. 6, we show the ionization times of WP1 and WP2 by the dashed lines for the emission angles of $-0.75\pi$ (red) and $-0.15\pi$ (black) with the final momentum of $p_r = 0.75$ a.u. As shown in Fig. 6(a), the ionization time difference $\Delta t_0$ between WP2 and WP1 is $\sim 0.51T_0$ and $\sim 0.49T_0$ for the emission angles of $-0.75\pi$ and $-0.15\pi$, respectively. Thus, $\Delta t_0$ is only slightly changed with the emission angle in the co-rotating fields. In the counter-rotating fields, as shown in Fig. 6(b), the ionization times of WP1 and WP2 have a large shift as compared with the case of the co-rotating fields. WP1 is released earlier while WP2 released later for the emission angle of $-0.15\pi$ as compared with Fig. 6(a). The case for the emission angle of $-0.75\pi$ is reversed. As a result, $\Delta t_0$ becomes $\sim 0.54T_0$ and $\sim 0.46T_0$ in the counter-rotating fields for the emission angles of $-0.75\pi$ and $-0.15\pi$, respectively. Thus the ionization time difference $\Delta t_0$ has a comparably large change with the emission angle in the counter-rotating fields. As we will show below, the difference in $\Delta t_0$ is the main reason for the different intracycle interference patterns of $p_+$ and $p_-$ orbitals in the co-rotating and counter-rotating circularly polarized two-color laser fields.

The intracycle interference pattern is determined by the phase difference between the wave packets WP2 and WP1. Using the SFA model, we can obtain the phase of the photoelectron wave packets released from different atomic orbitals. The transition amplitude in Eq. (6) consists of an exponential term and a preexponential factor, both of which contribute to the phase of the emitted electron wave packet. The exponential term corresponds to the classical action, whose real part $\Re[S_\gamma(p, t)]$ is the accumulated phase of the electron trajectory in the laser field. The
Thus, $\Delta \Phi$ plays an important role in forming the interference fringes in PMDs for
atomic orbital. As a result, the phase difference $\Delta \Phi$ has a large contribution to the interference fringes, resulting in different interference patterns for $p_+$ and $p_-$ orbitals, as shown in Fig. 3. Thus, the ionization time difference $\Delta \Phi$ plays an important role in forming the interference fringes in PMDs for $p_+$ and $p_-$ orbitals.

In Fig. 7, we show the distribution of $\Delta \Phi$ for $p_+$ orbital and $\delta t_0$ in the co-rotating and counter-rotating circularly polarized two-color laser fields. One can clearly see the relation between $\delta t_0$ and $\Delta \Phi$. Comparing Figs. 7(a) and 7(b) with Figs. 7(c) and 7(d), we find that the distributions of $\Delta \Phi$ and $\delta t_0$ with respect to the electron momentum and emission angle are

Fig. 6. The electric field $E(t) = \sqrt{E_{10}^2 + E_{20}^2}$ of the co-rotating (a) and counter-rotating (b) circularly polarized two-color laser pulses. The black and red arrows indicate the time difference of the electron wave packets released within the same laser cycle for the emission angle of $-0.75\pi$ (red arrows) and $-0.15\pi$ (black arrows), respectively, with the final momentum of $p_T = 0.75$ a.u. WP1 and WP2 indicate the first and second electron wave packets released within the same laser cycle, respectively.
very similar. In the co-rotating fields, the values of both $\delta t_0$ and $\Delta \Phi_0$ increase slowly with the emission angle. In the counter-rotating fields, the distributions of $\Delta \Phi_0$ and $\delta t_0$ reveal nearly the same oscillations with respect to the emission angle. The absolute values of $\delta t_0$ and $\Delta \Phi_0$ in the counter-rotating fields are much larger than those in the co-rotating fields. Thus the effect of the atomic orbital on the phase distribution of an ionizing electron wave packet can be easily revealed in the counter-rotating circularly polarized two-color laser fields. Due to the linear relation between $\Delta \Phi_0$ and $\Delta t_0$, the intracycle interference patterns have recorded the ionization time difference of the electron wave packets released within an optical cycle.

![Fig. 7.](image)

**Fig. 7.** The phase difference $\Delta \Phi_0$ for $p_+$ orbital [(a)(b)] and $\delta t_0$ [(c)(d)] between WP2 and WP1 with respect to the final momentum $p_r$ and the emission angle $\theta$ calculated by the SFA. The left (right) column corresponds to the co-rotating (counter-rotating) circularly polarized two-color laser pulse. $\Delta \Phi_0$ for $p_-$ orbital is the negative of that for $p_+$ orbital.

### 4. Conclusion

In summary, we have studied the effect of atomic $p_\pm$ orbitals on the phase distributions of the ionizing electron wave packets in circularly polarized two-color laser fields. We find that the photoelectron interference fringes in the PMDs are similar in the co-rotating fields for $p_+$ and $p_-$ orbitals while in the counter-rotating fields the interference fringes oscillate out of phase with respect to the emission angle for $p_+$ and $p_-$ orbitals. Based on the SFA model, we show that the electron wave packets released from $p_\pm$ orbitals carry different phase distributions. The atomic orbitals with different magnetic quantum numbers $m$ affect the preexponential factor of the transition amplitude, and the phase difference from the preexponential factor is approximately linear to the ionization time difference of the electron wave packets released within the same laser cycle. In co-rotating fields, the ionization time difference and the phase difference are small. As a result, the initial orbital has a minor effect on the interference structure, leading to similar interference patterns for $p_+$ and $p_-$ orbitals. On the contrary, in counter-rotating fields, the ionization time difference and the phase difference are large, which results in different interference fringes in the PMDs for $p_+$ and $p_-$ orbitals. Thus the effect of atomic orbitals on the phase distribution of the electron wave packet can be easily revealed in the counter-rotating circularly polarized two-color laser fields. Our study is useful for understanding the significant role of the atomic orbital in the photoelectron interference, which was usually neglected in previous studies [18,41]. The sensitivity of the photoelectron interference to the atomic $p_\pm$ orbitals may pave the way to produce high degree spin-polarized electron sources with considering the spin-orbit coupling.
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Disclosures
The authors declare no conflicts of interest.

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