Dependence of dynamic Lorentz frequency shift on carrier-envelope phase and including local field effects

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Abstract: We investigate the local field effects in a ZnO dense medium. Our results show due to the local-field effects, the Lorentz shifts can be found in the reflected spectra driven by the few-cycle laser pulse. Moreover, the dynamic Lorentz shifts depend sensitively on the carrier-envelope phase (CEP) of the few-cycle laser pulse, which provides a useful means to obtain the CEP information by the frequency shifts.

OCIS codes: (190.7110) Ultrafast nonlinear optics; (320.7150) Ultrafast spectroscopy; (320.2250) Femtosecond phenomena; (190.5530) Pulse propagation and solitons

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1. Introduction

In recent years, due to the remarkable advancement of ultrashort laser pulses, it is feasible to generate light pulses with durations comparable to the carrier oscillation cycle [1]. For few-cycle laser pulses, many new phenomena have attracted extensive attention in resonant extreme nonlinear optics [2–9]. For example, the phenomenon of the carrier-wave Rabi flopping was firstly predicted in two-level system by Hughes [2], and it then was proved in experiment [3]. When few-cycle laser pulses propagate in a resonant two-level medium, a large red-shift in the reflected laser pulse is found [4].

It is well established that when laser pulse envelope contains only a few cycles, the time variation of the electric field in a pulse depends on the carrier-envelope phase (CEP). Hence

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the CEP will become an important parameter for studying nonlinear optics [10–12]. Especially, in few-cycle regime, many CEP-dependent effects have been successfully demonstrated both theoretically and experimentally under the condition of the resonance enhancement [5–7,13–17]. For instance, a signal comes from the different Rabi sidebands interference around twice the laser center frequency, which strongly depends on the CEP of the few-cycle laser pulse [7]. Recently, when considering an electrical bias, the CEP-dependent optical rectification means that the CEP information can be full extracted [14]. Moreover, in our previous works, the CEP dependence of the pulse duration and intensity may be utilized to measure the CEP of few-cycle laser pulses in polar molecule [15]. The CEP can effectively control the reflected spectral signals and the higher spectral components in the presence of a static electric field [16,17].

When the pulse propagates in a dense medium (i.e., media having many particles within a cubic resonance wavelength), it is necessary to take into account the local-field effects (LFE) [18–23]. The local-field can couple with the medium dipole moment in a simple cubic lattice, which is related to the macroscopic laser field E_x and volume polarization $\vec{P}$ in the dense medium [22,23]: $E_{loc} = E_x + \frac{\vec{P}}{3\varepsilon_0}$, where ε_0 is the electric permittivity in the vacuum.

Many works have considered the LFE when laser interacts with the medium [22–32]. The generalized Maxwell-Bloch equations were presented with the LFE [22]. The reflection and transmission of ultrashort light pulses have been investigated [23]. In particular, the LFE plays an important role on generation of the Lorentz shift [26–32]. The dynamic Lorentz shift in dense superradiant amplifier has been found [26]. A signature of near dipole-dipole interaction may provide a useful method for measuring the strength of the interaction [27]. Moreover, in an experiment, the excitation dependence of the Lorentz local-field shift can be measured [29]. However, almost of these works are based on the slowly-varying-envelope approximation (SVEA) and the rotating-wave approximation (RWA) to solve the Maxwell-Bloch equations. In few-cycle regime, the SVEA and the RWA will break down [2,4,8]. In our work, we investigate the new effects of the Lorentz shift driven by few-cycle laser pulses and solve numerically the full-wave Maxwell-Bloch equations without adopting the SVEA and the RWA. It is shown that the dynamic Lorentz shift can occur in the reflected spectra due to the LFE of the ZnO dense medium. Importantly, the Lorentz shift depends strongly on the CEP of the few-cycle laser pulse.

This paper is organized as follows. The interaction of few-cycle pulses with a dense medium when considering the LFE is described in Sec. 2. The characteristics of the dynamic Lorentz shift with adjusting the CEP are presented in Sec. 3. We summarize the results in Sec. 4.

2. Theory

As done in Refs [2,4,30], we consider the propagation of the few-cycle pulse in a two-level medium. The pulse initially propagates in the free-space region, then it partially enters the ZnO dense medium at z = 12 μm, and partially reflects backward. The backward reflected pulse is detected at z = 3 μm. Taking the initial laser field propagating along z direction, and polarized along x direction, the Maxwell equations are the following form:

$$\frac{\partial H_y}{\partial t} = -\frac{1}{\mu_0}\frac{\partial E_x}{\partial z}, \quad \frac{\partial E_x}{\partial t} = -\frac{1}{\varepsilon_0\varepsilon_s}\frac{\partial H_y}{\partial z} - \frac{1}{\varepsilon_0\varepsilon_s}\frac{\partial P_x}{\partial t}, \tag{1}$$

where μ_0 is the magnetic permeability in the vacuum. $P_x = 2Nd\,\mathrm{Re}[\rho_{12}]$ is the macroscopic nonlinear polarization which connects with the off-diagonal element of the density matrix in the medium. The number density of medium is taken to be $N = 4.0\times10^{20}\,cm^{-3}$ and the dipole matrix element d = 0.19 e•nm. ε_s is the relevant dielectric constant of the medium and $\varepsilon_s = 4$

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[8], while within the vacuum, $P_x = 0$ and $\varepsilon_s = 1$. The Bloch equations including the LFE take the forms

$$\frac{\partial \rho_{12}}{\partial t} = -\omega_{12}\rho_{12} + i\frac{d}{\hbar}E_{loc}n - \frac{1}{\tau_2}\rho_{12}, \tag{2}$$

$$\frac{\partial n}{\partial t} = i\frac{2d}{\hbar}E_{loc}(\rho_{12} - \rho_{12}^{*}) - \frac{1}{\tau_1}n, \tag{3}$$

where ρ_{12} is the off-diagonal element of the density matrix. $n = \rho_{22} - \rho_{11}$ is the population difference between the excited and ground states. In the dense medium, the local-field is $E_{loc} = E_x + \frac{P}{3\varepsilon_0} = E_x + \frac{Nd}{3\varepsilon_0}\left(\rho_{12} + \rho_{12}^{*}\right)$ which shows the LFE is relevant to the macroscopic coherent polarization [22,23], while without the LFE, $E_{loc} = E_x$. The transition energy $\hbar\omega_{12}$ is close to the ZnO band gap ($E_s = 3.3$ eV). The excited-state lifetime and the dephasing time are set to $\tau_1 = \infty$ and $\tau_1 = 50$ fs, respectively [7,8]. The full-wave Maxwell-Bloch equations are solved by employing Yee's finite-difference time-domain (FDTD) discretization scheme [33–38]. Mur absorbing boundary conditions [39] are incorporated with FDTD discretization in order to avoid the influence of the finite-space computational domain. The temporal and spatial increments Δt and Δz are chosen to ensure $c\Delta t \le \Delta z$ [40], i.e., $\Delta t = 2.5\times10^{-18}$ s and $\Delta z = 1.5\times10^{-9}$ m.

The initial laser field propagating along z direction is

$$E_x(t=0,z) = E_0 \sec h[1.76(z/c - z_0/c)/\tau_0]\cos[\omega_0(z - z_0)/c + \varphi], \tag{4}$$

where E_0 is the peak field strength of the envelope. φ is the initial CEP, and τ_0 is the full width at half maximum (FWHM) of the pulse intensity envelope. The choice of z_0 ensures that the pulse penetrates negligibly into the medium at t = 0. The excitation pulse has a central frequency corresponding to a photon energy of $\hbar\omega_0$ = 1.4 eV and a FWHM intensity of $\tau_0 = 5$ fs. In our work, the Rabi frequency is comparable to the band gap frequency, so it is relevant to carrier-wave Rabi flopping. The envelope area of the input laser field we used is about 5π within the medium (the pulse area is defined as $A = \frac{d}{\varepsilon_s\hbar}\int E(t)dt$, where $E(t)$ is the envelope of the laser pulse), which is in the regime of the carrier-wave Rabi flopping (>4π) [2,8].

3. Results and discussions

We first study the reflected spectra of the few-cycle laser pulses when considering the LFE. Figure 1 shows that a peak can be obtained because of the resonant enhancement. The phenomenon is a special result for the few-cycle laser pulse, which accords with the previous works [7,8]. Interestingly, comparing with the case without the LFE ($E_{loc} = E_x$), the peak will generate a significant Lorentz shift near the resonant frequency due to the existence of the LFE (see dashed line). The origin of the phenomenon is the renormalization of the transition frequency, i.e., $\omega_{12}^{'} = \omega_{12} + \Delta_l n$, where $\Delta_l = \frac{Nd^2}{3\hbar\varepsilon_0}$. This shows that the Lorentz shift depends strongly upon the population difference n. When the system starts from its ground state n =

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−1, the LFE leads to a decrease of the resonant frequency by an amount of $-\frac{Nd^2}{3\hbar\varepsilon_0}$, which is stressed in the related Refs [23,26,29]. Moreover, the peak intensity of reflected spectrum reduces obvious because the effective Rabi frequency decreases (see solid line) [41].

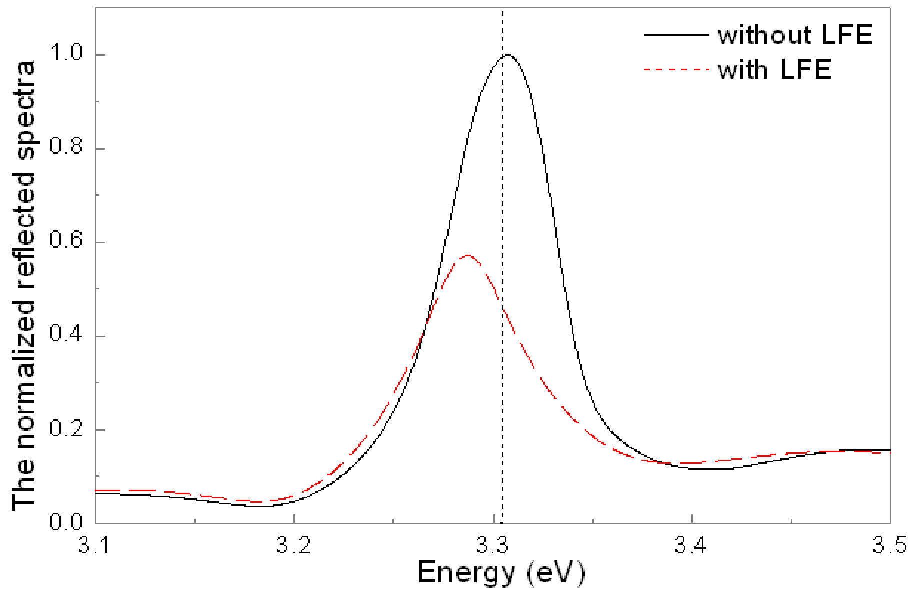

Fig. 1. (Color online) The Lorentz shift of the normalized reflected spectra.

Then we discuss the relation between the dynamic Lorentz shift and the CEP of the reflected few-cycle laser pulse with the LFE. As shown in Fig. 2, the dynamic Lorentz shifts of the reflected spectra become evident with adjusting the CEP. For $\phi = 0$, the dynamic Lorentz shift of the reflected spectrum is $\delta 1 = \text{-}19.85$ THZ (see Fig. 2, solid line), while for $\phi = 0.5\pi$, the dynamic Lorentz shift significantly increases ($\delta 2 = \text{-}29.24$ THZ). The reason can be explained by the population differences near the front face of the medium in Fig. 3(a). The transitions between the ground and excited levels represented by the population distributions can be reversed because of the two-photon absorption. As expressed in Eq. (2), the LFE will obviously affect the population distribution, which directly influence the Lorentz shifts near the resonant frequency. Interestingly, the population distributions change with the CEP of the few-cycle laser pulse. Importantly, the population differences are same for the cases of the CEPs $\varphi = 0$ and $\varphi = \pi$ [see Fig. 3(a)]. This means that the period of the CEP-dependent spectral signal is π. The view can be further proved through the reemitted field $E_{em}(t)$, i.e., single photon emission [see Fig. 3(b)]. When the resonant excitation is considered, the macroscopic coherent polarization $P_x(t)$ will build up gradually near the front face of the medium driven by the incident laser field, which acts as a source of a reemitted field $E_{em}(t)$, i.e., $E_{em}(\omega) \propto FFT\left(\frac{\partial P_x}{\partial t}\right)$ [15,42].

The CEP-dependent Lorentz shift will be more significant when the inversion symmetry of the system is broken, such as the presence of a static electric field. When a static electric field is added ($E_s = 2\% E_0$), the period of the CEP-dependent spectral signal becomes 2π as shown in Fig. 4. Moreover, due to the LFE, the dynamic Lorentz shifts δ depend crucially on the

CEP of few-cycle laser pulses. By analysis, the underlying physical mechanism for the phenomenon can be explained with the field strength. As presented in Ref [43], the

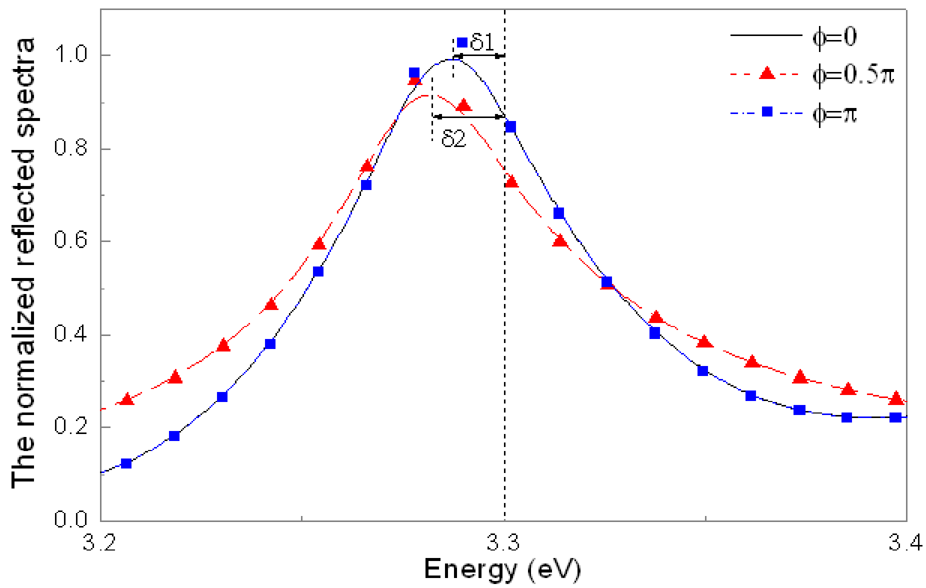

Fig. 2. (Color online) The dynamic Lorentz shift of the normalized reflected spectra of the few-cycle laser pulse with the LFE.

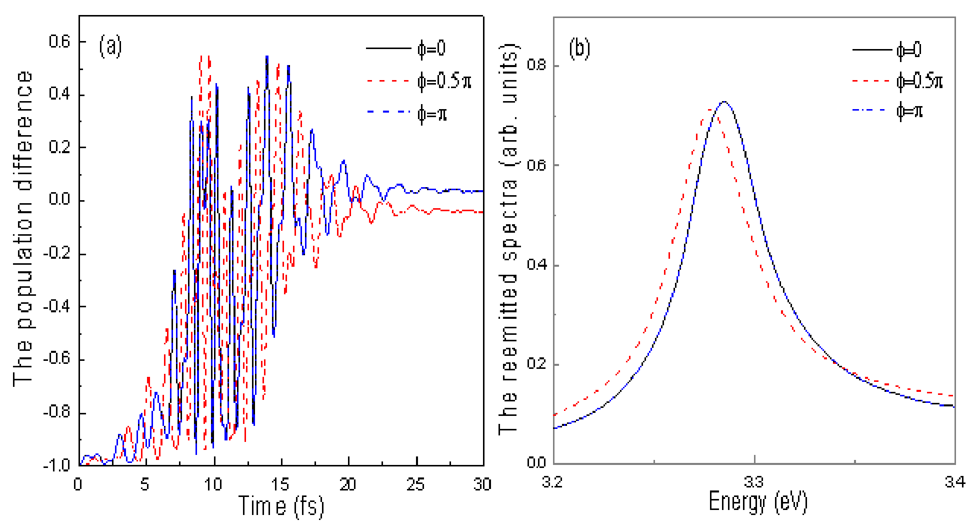

Fig. 3. (Color online) (a) The population difference n near the front face with the LFE. (b) The reemitted spectra [$E_{em}(\omega)$] near the front face of the medium.

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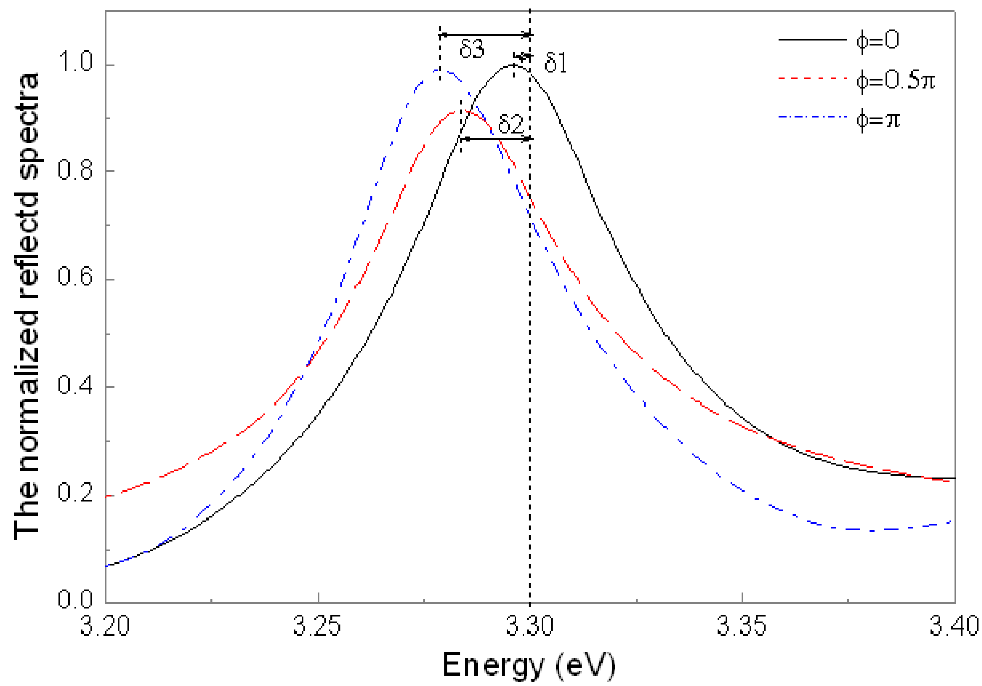

Fig. 4. (Color online) Same as in Fig. 2 but for the presence of a static electric field.

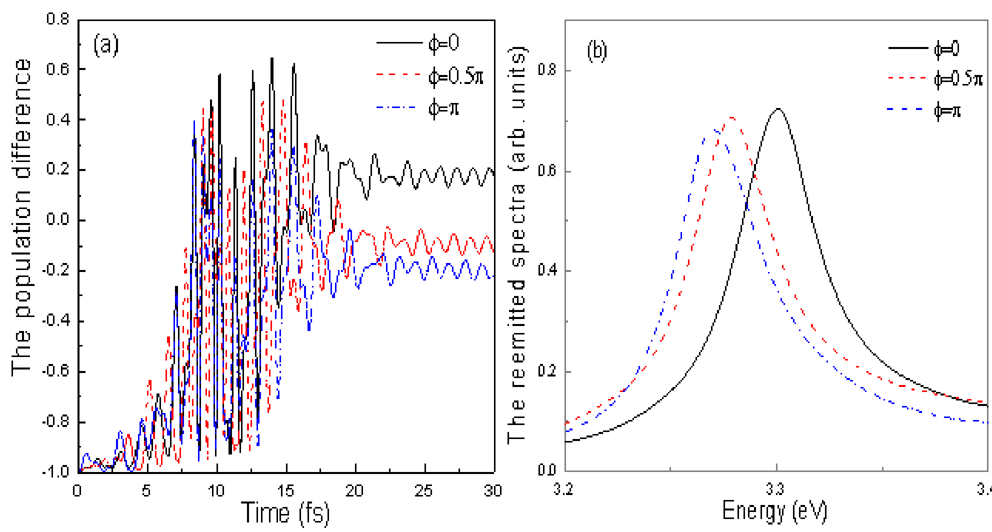

Fig. 5. (Color online) Same as in Fig. 3 but the presence of a static electric field.

populations of the states can be strongly phase dependent if the field strength is increased. As a result, a 2% variation in the field amplitude can affect the population distributions near the front face of the medium as shown in Fig. 5(a), which directly control the Lorentz shift decreasing or increasing. In order to demonstrate the validity of our analysis, the spectra of the reemitted field for different CEPs are performed to help interpret the CEP-dependent phenomena [see Fig. 5(b)].

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One thing that should be highlighted is that such alternative phenomena are not limited to the certain value but are valid for a range of the number density of medium *N*. As shown in Fig. 6, due to the LFE, the characteristics of the dynamic Lorentz shift depending on the CEP of few-cycle laser pulses also occur. Our results may provide a potential method to gain the information about the CEP of few-cycle laser pulses by measuring the dynamic Lorentz shifts.

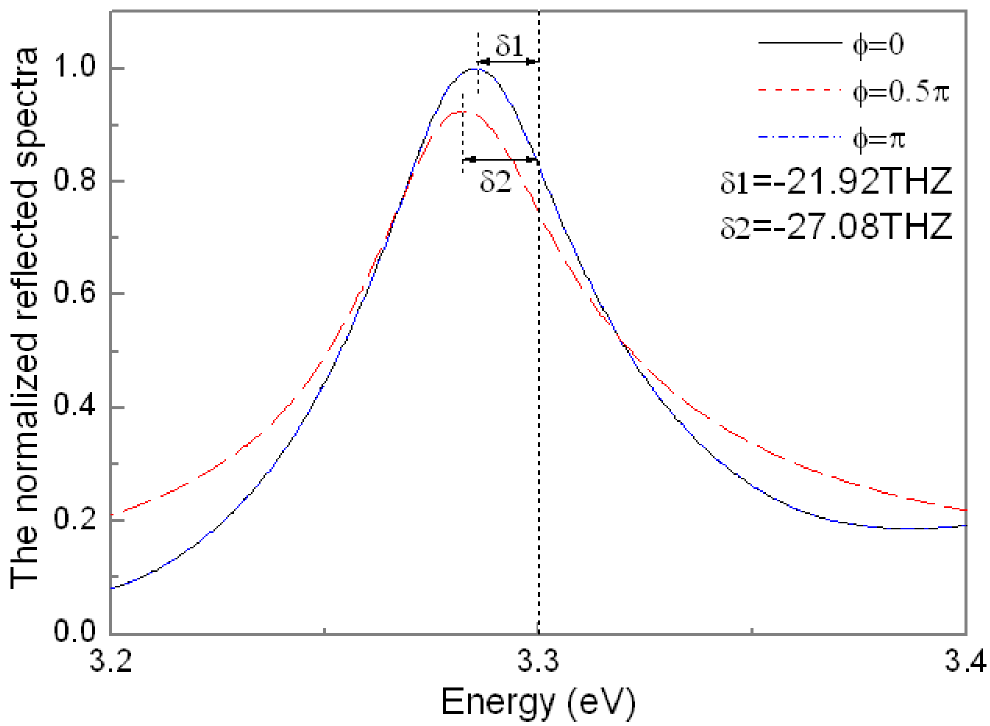

Fig. 6. (Color online) Same as in Fig. 2 but for the number density of medium $N = 3.0\times10^{20}\,cm^{-3}$.

4. Conclusions

In conclusion, we have studied the LFE on the reflected spectra of few-cycle laser pulses in a dense medium. It has been found that when considering the LFE, the Lorentz shift can be obtained in the reflected spectrum. Interestingly, the dynamic Lorentz shifts vary significantly with adjusting the CEP of the few-cycle laser pulse. Moreover, when the inversion symmetry of the system is broken, the CEP-dependent spectra effects will be further enhanced. Our results suggest that the CEP-dependent effect can provides a useful route to obtain the CEP information.

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