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Field-Free Molecular Orientation Induced by a Single-Cycle THz Laser Pulse Train

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Abstract. We investigate theoretically the field-free orientation of CO molecules induced by a single-cycle THz laser pulse train. It is shown that the molecular orientation can be obviously enhanced by applying the pulse train. The laser intensity and pulse number have some effects on the molecular orientation. The high degree of field-free molecular orientation $|\langle\cos\theta\rangle|_{\max}=0.9246$ is obtained at temperature $T=0$ K. The variations of the maximum orientation degree with the experimentally available pulse number and peak intensity are given. Temperature T has a considerable influence on the field-free molecular orientation. The maximal field-free molecular orientation at $T=0$, 10, 20 and 30 K for $N=14$ and $E_0=1.8$ MV/cm are $|\langle\cos\theta\rangle|_{\max}=0.9188$, 0.7338, 0.6055 and 0.5154 in order, and the corresponding effective duration times of molecular orientation are $\Delta t=0.759$, 0.432, 0.297 and 0.117 ps.

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Key words: Single-cycle THz laser pulse train, pulse number, molecular orientation.

1 Introduction

Molecular alignment and orientation have drawn substantial attention of both physicists and chemists in the past few decades due to their extensive applications in relevant research fields [1, 2], such as high-order harmonic generation [3–6], chemical reaction dynamics [7–11], strong-field ionization [12], ultrafast molecular imaging [13–15] and attosecond science [16]. Alignment implies that the symmetry axis of molecule is localized along a laboratory-fixed axis (the polarization direction of the laser field), and orientation additionally requires the aligned molecules point along a particular direction. The

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molecular alignment has been realized experimentally [17–21]. However, the realization of molecular orientation is still a challenging issue.

Several ways have been proposed to manipulate molecular orientation. Originally, a strong dc-field, such as a hexapole field [22] or brute forced field [23], based on its interaction with the permanent dipole moment, was employed to achieve molecular orientation. However, this way can only obtain relatively low orientation degree, and strong dc-field often induces the stark effects influencing the experimental measurement. Alternatively, Friedrich *et al*. proposed a scheme for enhancing the molecular orientation by an intense laser field combined with a weak dc-field. The effectiveness of this scheme depends on the combined effects of the permanent dipole interaction and the anisotropic polarizability interaction [24, 25]. The maximal degree of molecular orientation achieved by using this scheme was greatly improved. However, the presence of an electric field may perturb the quantum states of the target molecule and limit applications of the oriented samples [26]. Additionally, an asymmetric laser field, such as a half-cycle pulse (HCP) [27–29], a few-cycle THz laser field [30, 31], a two-color laser field [32–36] and a multi-color laser field [37], was employed to steer molecular orientation via their interaction with molecular permanent dipole moment, anisotropic polarizability and hyperpolarizability. These ways can be used to realize effectively field-free molecular orientation. In experiment, however, it is difficult to generate intense HCPs at present. Moreover, the intense two-color or multi-color ultrafast laser fields potentially cause unwanted molecular excitations leading to molecular ionization or dissociation. Resulting from the recent breakthrough in THz technology, the intense single-cycle THz pulses have become available in the laboratory [38]. Kitano *et al*. obtained theoretically a high degree of field-free orientation of $\langle\cos\theta\rangle$=0.84 in the HBr molecule by the combination of a femtosecond laser field and a time-delayed single-cycle THz laser pulse [26]. In their scheme, a nonresonant femtosecond laser pulse was used to pre-excite molecules and a time-delayed THz pulse was used to orient molecules. After the nonresonant femtosecond laser pulse is over, the even rotational states are populated and the molecule sample is aligned. The transition frequency becomes resonant with the subsequent THz pulse after pre-excitation, the molecules can be consequently oriented. Fleischer *et al*. measured the field-free molecular orientation induced by an intense single-cycle THz laser pulse in the OCS gas sample [39]. Theoretically, single-cycle THz pulses was utilized to realize orientation of the HCN molecule [40].

In the present work, we investigate theoretically the efficient field-free molecular orientation steered by a single-cycle THz laser pulse train. We study the influence of the laser intensity and pulse number on the maximum orientation degree. We find that the nearly equal populations on neighboring rotational states can lead to a large orientation degree. The optimal pulse number in the range of peak intensity of E_0=[0.1,10.1] MV/cm are given. The influences of temperature on molecular orientation degree and duration time are discussed.