



All-optical reconstruction of k-dependent transition dipole moment by solid harmonic spectra from ultrashort laser pulses

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Abstract: Band structure and transition dipole moment play important roles in high-order harmonic generation from solid materials. In this work we provide a new all-optical technique to reconstruct the momentum-dependent transition dipole moment using the harmonic spectrum from MgO crystal driven by an ultrashort mid-infrared laser pulse. Under the influence of the ultrashort laser pulse, the emitted photon energy and the crystal momentum form a one-to-one match, in the same way between the intensity of the harmonic above the minimum bandgap and the square of the amplitude of the transition dipole moment, resulting in a realization of directly probing the transition dipole moment. Our all-optical method paves a way to image the two-dimensional transition dipole moment of crystals with the inversion symmetry.

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1. Introduction

High-order harmonic generation (HHG) can be produced by the interaction of an intense laser pulse with crystalline solids [1–10]. Compared with HHG from gaseous media [11–22], due to the high electronic density and the periodic characteristic of crystals, it has been demonstrated that the more efficient harmonic emission can be achieved in experiments [1,2], which offers opportunities to obtain strong attosecond light sources and detect ultrafast photoelectrons in the condensed matter phase [23–25].

Physical mechanisms [1,26–32] including intraband and interband ultrafast electron dynamics are mostly used to explain solid HHG. The process can be described by the “three-step model” [27,33,34] which is similar to the case of gaseous media: first, an electron tunnels from the valence band to the conduction one forming an electron-hole pair; then the laser field accelerates the electron (hole) on the conduction (valence) band, which leads to the intraband current; finally, the electron recombines to the hole with a photon emitted. Information of band structure and transition dipole moment (TDM) are encoded in both intra-band and interband harmonic spectra. Therefore, all-optical techniques using intra- and inter-band HHG have been applied to reconstruct the band structure [5,35–38].

Based on the sensitivity of the momentum-dependent bandgap to the oscillation phase of even harmonics from a two-color driving field, Vampa *et al.* reconstructed the bandgap of ZnO along Γ -M direction [5]. Taking advantage of the harmonic power as a function of the driving laser's intensity, Lanin *et al.* retrieved the electron band structure by the intraband high-harmonic generation of ZnSe in sub-100-fs mid-infrared pulses [35]. You *et al.* illustrated that HHG from MgO is sensitive to the atomic-scale structure, which indicate the possibility for utilizing an

optical technique to map the interatomic potential and the valence electron density [36]. Making use of harmonic spectra in perpendicular polarization with respect to the driving field, Luu *et al.* retrieved the Berry curvature of α -quartz [37]. By utilizing the orientation dependent harmonic spectra, Yu *et al.* imaged the two-dimensional band information of *h*-BN crystal [38].

To date, all-optical technique about solid HHG mostly focused on reconstructing the band structure [5,35,39]. In the present work, taking into account harmonic spectra from the interaction of a solid and ultrashort mid-infrared laser pulses, we offer an all-optical method to map the *k*-dependent TDM of the solid. Under the influence of an intense laser pulse, the electron (hole) moving on a band can reach the edge of the first Brillouin zone, at the same time the electron excitation and the interband polarization between different bands are closely related to the *k*-dependent TDM [40,41]. In the present paper, the band structure and the *k*-dependent TDM of the sample are accurately calculated by ab-initio software. It is found that, by linking the harmonic photon energy and the pseudomomentum, where the electron recombines into hole, the *k*-dependent TDM can be reconstructed from the interband HHG spectrum, which is originated from a single quantum orbit. Furthermore, in a wide range of the carrier envelope phase (CEP) of the ultrashort laser pulse, the reconstruction of the *k*-dependent TDM can be directly realized by the harmonics from the interband polarization.

2. Theory and models

2.1. Semiconductor Bloch equations

The HHG process from MgO crystal in an ultrashort mid-infrared laser pulse is investigated by the solution of three-band semiconductor Bloch equations (SBEs) [38,42], which can be derived from time-dependent schrödinger equation [40,41]. The SBEs for describing the laser-crystal interaction with the laser polarization along the Γ -X direction of MgO read [40]. (Atomic units are used throughout this article unless stated otherwise):

$$\begin{aligned} \frac{\partial}{\partial t} p_{\mathbf{k}}^{c_1 v} = & -i \left(\varepsilon_{c_1}(\mathbf{k}) - \varepsilon_v(\mathbf{k}) - \frac{i}{T_2} \right) p_{\mathbf{k}}^{c_1 v} + i (f_{\mathbf{k}}^{c_1} - f_{\mathbf{k}}^v) \mathbf{d}_{c_1 v}(\mathbf{k}) E(t) \\ & + E(t) \nabla_{\mathbf{k}} p_{\mathbf{k}}^{c_1 v} + i E(t) (\mathbf{d}_{c_2 v}(\mathbf{k}) p_{\mathbf{k}}^{c_1 c_2} - \mathbf{d}_{c_1 c_2}(\mathbf{k}) p_{\mathbf{k}}^{c_2 v}) \end{aligned} \quad (1)$$

$$\begin{aligned} \frac{\partial}{\partial t} p_{\mathbf{k}}^{c_2 v} = & -i \left(\varepsilon_{c_2}(\mathbf{k}) - \varepsilon_v(\mathbf{k}) - \frac{i}{T_2} \right) p_{\mathbf{k}}^{c_2 v} + i (f_{\mathbf{k}}^{c_2} - f_{\mathbf{k}}^v) \mathbf{d}_{c_2 v}(\mathbf{k}) E(t) \\ & + E(t) \nabla_{\mathbf{k}} p_{\mathbf{k}}^{c_2 v} + i E(t) (\mathbf{d}_{c_1 v}(\mathbf{k}) p_{\mathbf{k}}^{c_2 c_1} - \mathbf{d}_{c_2 c_1}(\mathbf{k}) p_{\mathbf{k}}^{c_1 v}) \end{aligned} \quad (2)$$

$$\begin{aligned} \frac{\partial}{\partial t} p_{\mathbf{k}}^{c_2 c_1} = & -i \left(\varepsilon_{c_2}(\mathbf{k}) - \varepsilon_{c_1}(\mathbf{k}) - \frac{i}{T_2} \right) p_{\mathbf{k}}^{c_2 c_1} + i (f_{\mathbf{k}}^{c_2} - f_{\mathbf{k}}^{c_1}) \mathbf{d}_{c_2 c_1}(\mathbf{k}) E(t) \\ & + E(t) \nabla_{\mathbf{k}} p_{\mathbf{k}}^{c_2 c_1} + i E(t) (\mathbf{d}_{v c_1}(\mathbf{k}) p_{\mathbf{k}}^{c_2 v} - \mathbf{d}_{c_2 v}(\mathbf{k}) p_{\mathbf{k}}^{v c_1}) \end{aligned} \quad (3)$$

$$\frac{\partial}{\partial t} f_{\mathbf{k}}^v = -2\text{Im} [\mathbf{d}_{c_1 v}(\mathbf{k}) E(t) p_{\mathbf{k}}^{v c_1} + \mathbf{d}_{c_2 v}(\mathbf{k}) E(t) p_{\mathbf{k}}^{v c_2}] + E(t) \nabla_{\mathbf{k}} f_{\mathbf{k}}^v \quad (4)$$

$$\frac{\partial}{\partial t} f_{\mathbf{k}}^{c_1} = 2\text{Im} [\mathbf{d}_{c_1 c_2}(\mathbf{k}) E(t) p_{\mathbf{k}}^{c_2 c_1} + \mathbf{d}_{c_1 v}(\mathbf{k}) E(t) p_{\mathbf{k}}^{v c_1}] + E(t) \nabla_{\mathbf{k}} f_{\mathbf{k}}^{c_1} \quad (5)$$

$$\frac{\partial}{\partial t} f_{\mathbf{k}}^{c_2} = 2\text{Im} [\mathbf{d}_{c_2 c_1}(\mathbf{k}) E(t) p_{\mathbf{k}}^{c_1 c_2} + \mathbf{d}_{c_2 v}(\mathbf{k}) E(t) p_{\mathbf{k}}^{v c_2}] + E(t) \nabla_{\mathbf{k}} f_{\mathbf{k}}^{c_2} \quad (6)$$

Here, $\varepsilon_{\lambda}(\mathbf{k})$ ($\lambda = v, c_1, c_2$) is the energy of the valence (conduction) band, $f_{\mathbf{k}}^{\lambda}$ ($\lambda = v, c_1, c_2$) is the electron density in the corresponding band, $p_{\mathbf{k}}^{\lambda_1 \lambda_2}$ ($\lambda_1 \lambda_2 = c_1 v, c_2 v, c_1 c_2$) and $\mathbf{d}_{\lambda_1 \lambda_2}(\mathbf{k})$ are the micropolarization and the transition dipole moment between two bands. T_2 is the dephasing time, which is set to a quarter-cycle of the incident laser pulse in this paper. The vector potential of the linear polarized laser field is $A(t) = \frac{e_0}{\omega} f(t) \cos(\omega t + \varphi)$, and the envelope function $f(t)$ is

the Gaussian form, where E_0 , ω and φ are the amplitude, the central frequency and the CEP of the electric field, respectively. Specially, amplitudes of initial and final instants for the vector potential are chosen as zero. The laser electric field can be produced by $E(t) = -\partial_t A(t)$. The intraband current $\mathbf{J}_{intra}(t)$ and interband current $\mathbf{J}_{inter}(t)$ can be derived from SBEs :

$$\mathbf{J}_{intra}(t) = \sum_{\lambda=c_1, c_2, v} \int_{BZ} \mathbf{v}_\lambda(\mathbf{k}) f_{\mathbf{k}}^\lambda d\mathbf{k} \quad (7)$$

$$\mathbf{J}_{inter}(t) = \sum_{\lambda_1 \lambda_2} \frac{\partial}{\partial t} \int_{BZ} \mathbf{d}_{\lambda_1 \lambda_2}(\mathbf{k}) p_{\mathbf{k}}^{\lambda_1 \lambda_2} d\mathbf{k} + c.c. \quad (8)$$

where $\mathbf{v}_\lambda(\mathbf{k}) = \nabla_{\mathbf{k}} \varepsilon_\lambda(\mathbf{k})$ is the group velocity. The harmonic spectrum is calculated from Fourier-transforming the time-dependent total current onto the polarization direction

$$S_{HHG} \propto \left| \int_{-\infty}^{\infty} [\mathbf{J}_{intra} + \mathbf{J}_{inter}] e^{i\omega t} dt \right|^2 \quad (9)$$

2.2. Band structure and transition dipole moment

Band structure and TDM have an important effect on the harmonic dynamic in the interaction of the strong laser pulse with the crystal [1,25]. Here, k-dependent energy bands and TDM are achieved from the first-principles calculations using the VASP code. Geometry of MgO crystal with symmetry group $Fm3m$ is performed within the generalized gradient approximation (GGA) in the parametrization of Perdew, Burke, and Ernzerhof (PBE). The calculations use an energy cutoff with 400 eV, and a k-point Monkhorst pack mesh of $10 \times 10 \times 10$ in the Brillouin zone. The optimized lattice parameters of MgO are $a=b=c=4.213 \text{ \AA}$. Figure 1(a) shows a portion of the band structure of MgO along the Γ -X direction by using the HSE06 hybrid function with the parameter AEXX=0.43. It can be seen that, degenerate heavy-hole and light-hole bands above the split-off one are highest valence bands, and two lowest conduction bands intersect at the boundary of the Brillouin zone. This band dispersion agrees well with the experimental band structure. When MgO is irradiated by a linearly polarized weak laser pulse along the (001) crystal plane, only three bands marked by VB, CB1 and CB2 in Fig. 1(a) predominantly contribute to the crystal harmonic spectra, because non-zero transition matrix elements between the three bands exist in most k points. This feature was demonstrated in recent work by Ghimire *et al.*[43]. Furthermore, the minimum bandgap between CB1 and VB equals to 7.8 eV, which is consistent with the experimental data. Based on the accurate band structure, the k-dependent TDM can be obtained from the momentum matrix element $\xi_{\lambda_1 \lambda_2}(\mathbf{k})$ ($\lambda_1 \lambda_2 = c_1 v, c_2 v, c_1 c_2$)

$$\mathbf{d}_{\lambda_1 \lambda_2}(\mathbf{k}) = \frac{i \xi_{\lambda_1 \lambda_2}(\mathbf{k})}{\varepsilon_{\lambda_1}(\mathbf{k}) - \varepsilon_{\lambda_2}(\mathbf{k})} \quad (10)$$

$$\xi_{\lambda_1 \lambda_2}(\mathbf{k}) = i \int_{cell} u_{\lambda_1, \mathbf{k}}^*(\mathbf{r}) \nabla_{\mathbf{k}} u_{\lambda_2, \mathbf{k}}(\mathbf{r}) d\mathbf{r} \quad (11)$$

where $u_{\lambda, \mathbf{k}}(\mathbf{r})$ ($\lambda = c_1, c_2, v$) is the periodic part of the Bloch function for the conduction (valence) band with the crystal momentum $\mathbf{k}$. TDMs between each pair of bands are presented in Fig. 1(b). Because MgO crystal has inversion symmetry, the phases of TDMs can be set to be zero [40].

2.3. Classical trajectory derivation

According to the semiclassical recollision model for bulk solids [33,34], as the electron (hole) accelerates in a band, it gets the crystal momentum, which can be written in terms of the vector

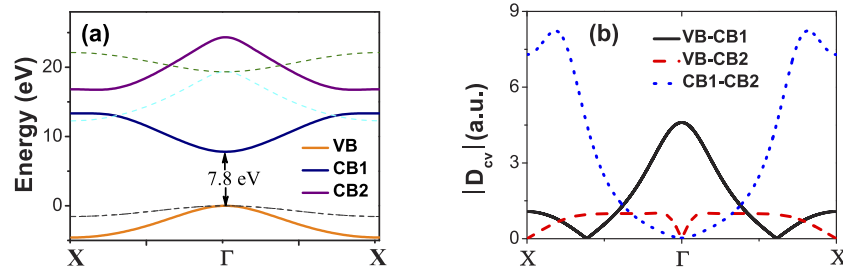


Fig. 1. (a) Band dispersion of MgO along the Γ -X direction; (b) k-dependent transition dipole moments from the first-principles calculations. The three bands (VB, CB1 and CB2) mainly contributing to HHG are marked by the solid lines.

potential $\mathbf{A}(t)$ of the laser field [27]:

$$\mathbf{k}(t) = \mathbf{A}(t) - \mathbf{A}(t') \quad (12)$$

Here, t' and t are ionization and recombination instants, respectively. For motions of the electron and the hole in real space, one can solve the above equation together with

$$x_e(t) = \int_{t'}^t \nabla \varepsilon_c[\mathbf{k}(\tau)] d\tau \quad (13)$$

$$x_h(t) = \int_{t'}^t \nabla \varepsilon_v[\mathbf{k}(\tau)] d\tau \quad (14)$$

When displacements of the electron and the hole are well matched, they can recombine and emit a harmonic photon with the energy equal to the bandgap between conduction and valence bands at the recollision time. Thereby, by using the recollision model, one can gain relations between the harmonic photon energy and times of creation or recollision, and trajectories of electrons (holes) in the $\mathbf{k}$ or real space [33,34]. Because velocities of electrons (holes) in a band m is determined by the formula $\mathbf{v}_m(\mathbf{k}) = \nabla_{\mathbf{k}} \varepsilon_m(\mathbf{k})$, trajectories of electrons (holes) depend on the band structure of the solid.

3. Reconstruction of k-dependent TDM

Based on the real TDM from the first-principles calculations, the black solid and red short-dashed lines in Fig. 2(a) present the harmonic spectra of MgO along the Γ -X direction, which are obtained from two-band and three-band SBEs, respectively. Here the laser parameters are 5 fs of full width at half maximum (FWHM), 2400 nm in wavelength, 0 for carrier envelope phase and $1.0 \times 10^{13} \text{ W/cm}^2$ in peak intensity, and the electric field of the laser pulse is plotted in Fig. 4(a). It is worth to mention that sub-optical-cycle and carrier-envelope-phase-stable light pulses can be produced by the coherent waveform synthesis from ultrabroadband optical parametric chirped-pulse amplifiers [44]. The harmonic spectrum from the three-band calculation exhibits a two-plateau structure, and the first plateau with the cutoff at 18 eV is approximately one order of magnitude in harmonic intensity higher than the second one, which is similar with the results from the recent works about the solid HHG from the multiband structure [10,45,46]. For the first harmonic plateau, because the direct coupling of the second conduction band with the valence band is small, the harmonic spectrum from the two-band model is almost same with that from the three-band calculation. In this work, we try to explore the relation between the harmonics in the first plateau and the crystal reciprocal structure. Hence, we focus on the HHG spectra from the two-band model in the following. The green dash-dotted and the blue dashed curves in

Fig. 2(b) show the harmonic spectra from the intraband current and the interband polarization, respectively. For comparison, the total harmonic spectrum is also presented here. It is clear that the harmonics with the energies above the minimum bandgap between CB1 and VB are dominated by the interband current, which agrees with other theoretical and experimental results for ZnO, MgO, and GaAs exposed to mid-infrared laser pulses [1,26,33,43,47–49]. Furthermore, harmonics with energies from the bandgap to the cutoff are almost continuous and smooth.

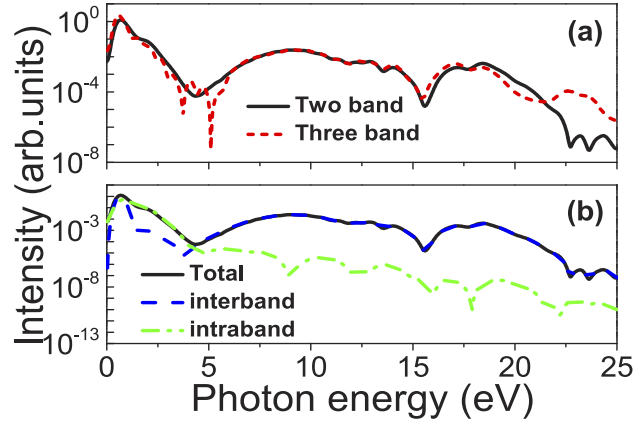


Fig. 2. (a) Harmonic spectra of MgO from two-band (black solid curve) and three-band models (red short-dashed curve); (b) harmonic spectra produced by interband (blue dashed curve) and intraband currents (green dash-dotted curve) from the two-band calculation.

In order to understand the continuous characteristic of the harmonic spectrum from the MgO crystal, emission times of continuous harmonics are investigated by the time-frequency analysis method. Figure 3 exhibits the time-frequency distribution of the HHG spectrum corresponding to the black solid curve in Fig. 2(a). One can notice that above-threshold harmonics are primarily generated by one quantum path. This is confirmed by the dependence of the harmonic photon energy with the emission instant calculated by the semiclassical recollision model, as shown in the purple solid curve from Fig. 3. Therefore, it is possible that the k-dependent TDM can be retrieved by using the harmonic spectrum from one quantum path.

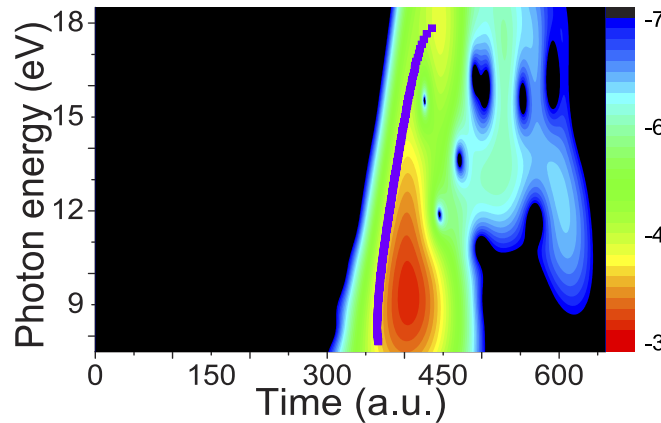


Fig. 3. Time-frequency distribution of the HHG corresponding to the black solid curve in Fig. 2(a). The purple solid curve is the photon energy vs the emission time from the semiclassical recollision model.

To more clearly understand time profiles of harmonics, we also plot the time evolution of the k -dependent electron density on the conduction band, as presented in Fig. 4(b). It can be observed that, electrons in the crystal are excited from the valence band to the conduction band at the peak of the ultrashort laser field, and then are oscillated by the driving laser field. When the electric field decays to zero, some of excited electrons can recombine to holes in the valence band. The time evolution of the excited electron can also be predicted by classical trajectories calculated from the semiclassical recollision method. Figure 5 presents two typical trajectories of electrons and holes in the real coordinate space. When the electronic trajectory (solid lines) intersects with the hole's one, then the photon is emitted. It can be seen from Fig. 5, the electron reencounters with the hole only once in the ultrashort laser pulse, which leads to the continuum harmonic spectrum.

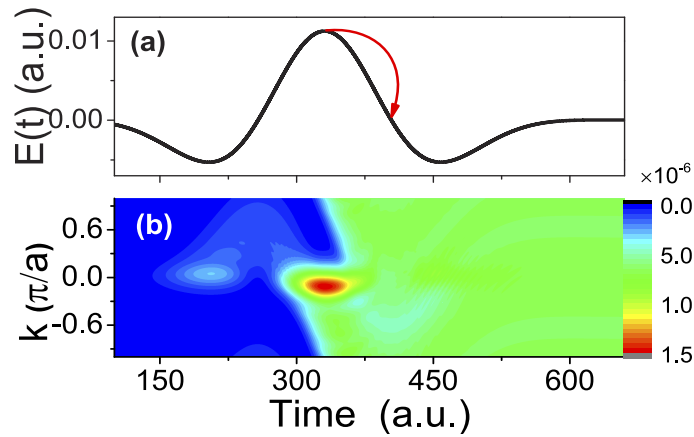


Fig. 4. (a) Electric field of the ultrashort laser pulse. The arrow indicates a typical electron orbit: the electron is excited into the conduction band at the starting point of the arrow and recombines to the valence band at the ending point of the arrow with a 15 eV photon emitted. (b) Time evolution of the electronic density distribution at different crystal momentums in the conduction band.

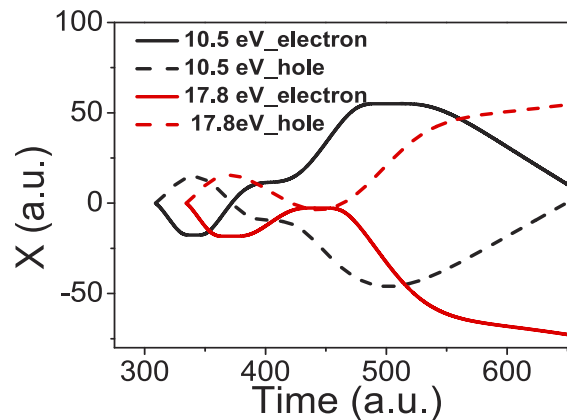


Fig. 5. Real space trajectories of electrons (solid lines) and holes (dashed lines) corresponding to the emissions near the bandgap and the cut-off energy.

In order to better explain the crystal harmonic generation from the interband current, *Keldysh's* approach used for explaining HHG can be extended from atom to solid. In *Keldysh's* scheme [26], the interband current due to a single quantum orbit is proportional to

$$\mathbf{d}_{c1v}^*(\mathbf{k}_r) \mathbf{d}_{c1v}(\mathbf{k}_i') \mathbf{F}(t_i') e^{-i\omega t_r - i\mathbf{S}(\mathbf{k}_r, t_r, t_i') - (t_r - t_i')/T_2} \quad (15)$$

where ω is the frequency of the harmonic photon, $\mathbf{S}$ is the action, t_i' and t_r are ionization and emission times, $\mathbf{k}_i'$ and $\mathbf{k}_r$ are corresponding crystal momentums, respectively. From Fig. 4, one can see that, electrons are mostly excited near the amplitude peak of the electric field and crystal momentums at ionization times are mostly same. Moreover, only one quantum trajectory mainly contributes to the harmonics shown by the black solid curve in Fig. 2. According to Eq. (15), the yield of HHG from the interband current is approximately proportional to

$$P(\mathbf{k}_r) \propto \left| d_{c1v}(\mathbf{k}_r) e^{-(t_r - t_i')/T_2} \right|^2 \quad (16)$$

In addition, when the CEP of the ultrashort laser pulse equals to zero, the difference between t_r and t_i' as a function of the photon energy approximately exhibits a smooth upward trend line from 9 eV to 16 eV. However, the time difference shows a sharp rise in the energy range from 16 to 18 eV, as presented from Fig. 6. After reasonably ignoring (photon energy from 8 to 16 eV) the exponential factor about $t_r - t_i'$, then the harmonic yield is directly related to the square of absolute value of the transition dipole moment at the emission instant. As a result, it is reasonable to extract TDMs at $\mathbf{k}$ points where photons emitted.

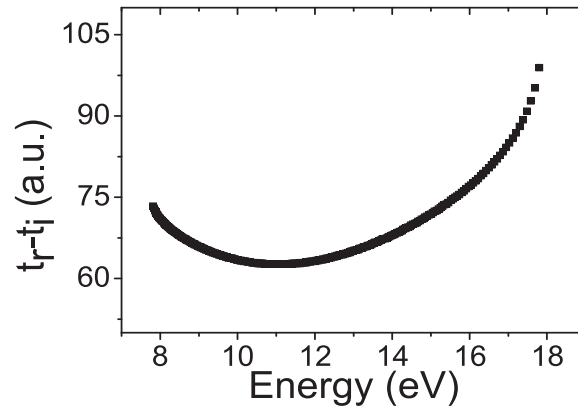


Fig. 6. The dependence of the difference between the recombination and the ionization instants with the photon energy.

Next, we explore the dependence of the amplitude of TDM from the first-principles calculations with the bandgap, as shown by the blue dotted curve in Fig. 7. Here, the harmonic spectrum shown by the black solid curve in Fig. 2(b) is also presented. Clearly, the shape of TDM almost coincides with that of the harmonic spectrum from the minimum bandgap to the cutoff. Meanwhile, it has been confirmed that, the energy of the harmonic photon equals to the bandgap energy, which is determined by the crystal momentum when the electron recombines with the hole. Consequently, the harmonic photon energy and the crystal momentum have a one-to-one match at the emission instant.

In terms of above analyses, it is possible to image the TDM between conduction and valence bands by adopting harmonic spectra from the single quantum trajectory. Here, the strength of a

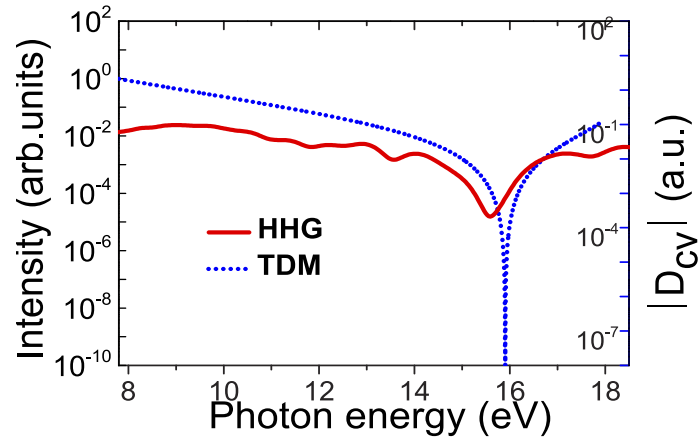


Fig. 7. The amplitude of TDM from the first-principles calculations versus the bandgap (the blue dotted line), and the corresponding harmonic spectrum shown by the black solid line in Fig. 2(a).

harmonic photon with energy n eV is defined by integrating the harmonic signal from $n-0.1$ eV to $n+0.1$ eV, and the intensity of the harmonic is normalized to the most intense harmonic above the minimum bandgap. Then, the harmonic with energy n eV in the plateau is chosen, and the corresponding harmonic energy is marked in the bandgap, as shown by the blue circles in Fig. 8. Due to the one-to-one correspondence between the photon energy and the crystal momentum at the emission instant, if the difference between and is calculated from the semiclassical recollision method, the relation between the harmonic intensity and the k -dependent transition dipole moment can be achieved from the formula 16. Naturally, one can reconstruct the k -dependent transition dipole moment by the harmonic spectrum from the ultrashort laser pulse. Figure 8(a) shows the reconstruction of the k -dependent bandgap and TDM by directly using the harmonic spectrum from the black solid curve in Fig. 2. By considering the exponential factor about the difference between emission and ionization instants, the reconstruction by indirectly adopting

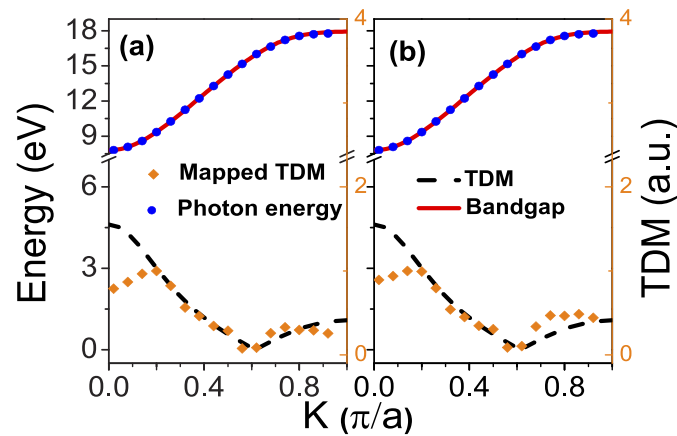


Fig. 8. Reconstruction of the TDM (orange diamonds) by directly (a) or indirectly (b) using the HHG spectrum. For comparison, the TDM (black dash-dotted line) and bandgap (red solid line) from the first-principles calculations are also presented here. The harmonic photon energy used to map the TDM is marked in the bandgap (blue circles).

the harmonic spectrum is presented in Fig. 8(b). For comparison, the TDM (black dash-dotted line) and bandgap (red solid line) from the first-principles calculations are also exhibited here. In both cases, the mapped k-dependent TDM (orange diamonds in Fig. 8) in most k points is closely consistent with that from the real TDM. Especially, the TDM's reconstruction from taking the exponential factor is no much difference from that from directly using the harmonic spectrum. The above results clearly illustrate that the TDM can be mapped by harmonic spectra from ultrashort laser pulses.

For the ultrashort laser pulse, the CEP of which apparently changes the shape of the electric field and the characteristic of the harmonic spectrum. In the following, we examine the CEP effect on the reconstruction of the TDM. It can be seen from Fig. 8 and Fig. 9, when the CEP is altered from 0 to 0.3π , due to one trajectory dominating harmonics with energies above the bandgap, we can still map the TDM well by directly using harmonic spectra from ultrashort laser pulses. Thus, the reconstruction achieved in a wide range of CEP become feasible in practical experimental implementation.

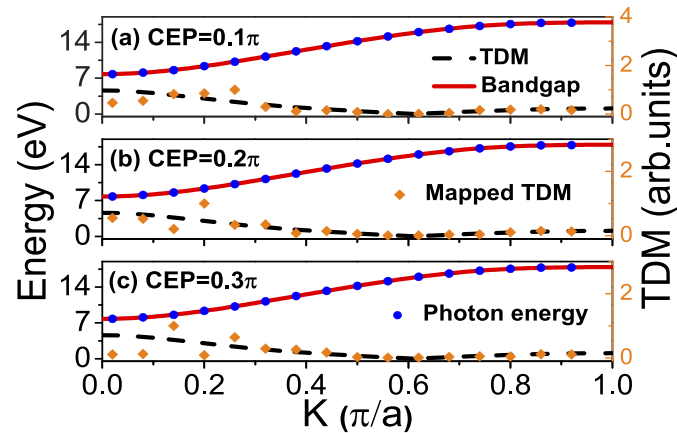


Fig. 9. Reconstruction of the TDM (orange diamonds) in the different CEP of the ultrashort laser pulse. The harmonic photon energy used to map the TDM is marked in the bandgap (blue circles).

It should be emphasized that the harmonic spectrum originated from the single quantum path is the successful key for our all-optical reconstruction method. According to our simulations, when the 2400 nm laser pulse's FWHM is extended to 7fs, we can still better map the TDM by the harmonic spectrum from the ultrashort laser pulse, as shown by Fig. 10(d). For the 800 nm or 1600 nm laser pulse frequently used in HHG experiments, if we keep the corresponding pulse's FWHM with 4 fs or 5 fs, the TDM also can be mostly reconstructed in a wide k range from 0.3 to 1.0, as presented by Fig. 11(d) and Fig. 12(d), respectively. In addition, the pump-probe scheme was extensively practiced for HHG from gaseous media [50,51], which also can be applied to control quantum trajectories of electrons in the conduction band for the solid. An ultraviolet pulse with a few hundred attosecond duration is used to pump electrons in the valence band to the conduction band. A weak multicycle mid-infrared pulse can be adopted to drive electrons in the conduction band. By controlling the time delay between pump and probe laser pulses, the interband excitation is mainly confined to the central half cycle of the probe pulse, thereby the interband harmonic from a single quantum trajectory can be achieved. In recent future, we will investigate all-optical reconstruction of the k-dependent TDM by this pump-probe method.

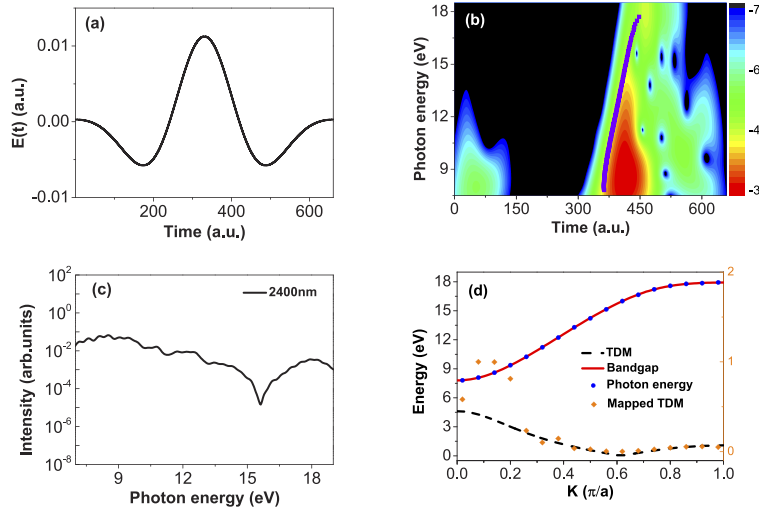


Fig. 10. (a) The electric field of the incident laser pulse. (b) Time profile of the harmonic spectrum, the purple solid curve is the dependence of the harmonic photon with the emission time from the classical calculation. (c) HHG spectrum from the ultrashort laser pulse. (d) Reconstruction of the TDM (orange diamonds) by the HHG spectrum. The laser parameters are 2400 nm in wavelength, 7 fs of FWHM, 0 for CEP, and $1.0 \times 10^{13} \text{ W/cm}^2$ in intensity.

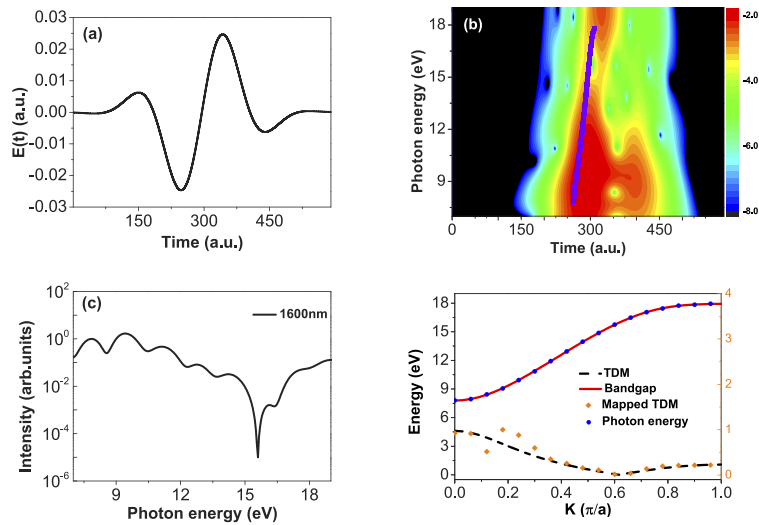


Fig. 11. (a) The electric field of the incident laser pulse. (b) Time profile of the harmonic spectrum, the purple solid curve is the dependence of the harmonic photon with the emission time from the classical calculation. (c) HHG spectrum from the ultrashort laser pulse. (d) Reconstruction of the TDM (orange diamonds) by the HHG spectrum. The laser parameters are 1600 nm in wavelength, 5 fs of FWHM, 0 for CEP, and $3.0 \times 10^{13} \text{ W/cm}^2$ in intensity.

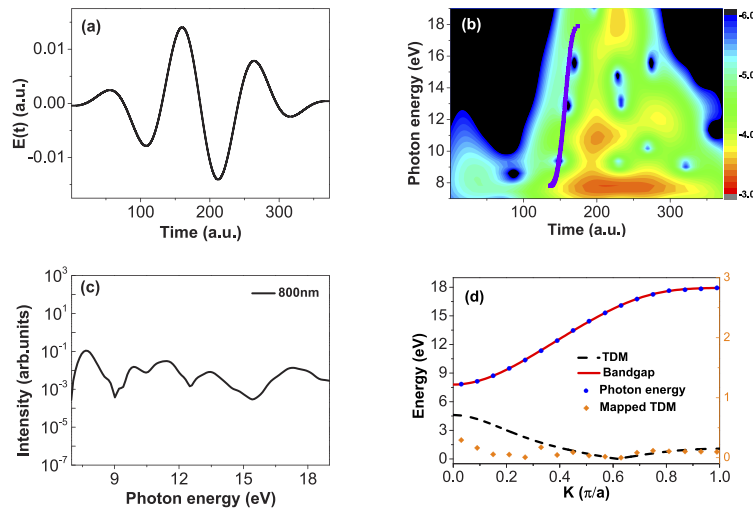


Fig. 12. (a) The electric field of the incident laser pulse. (b) Time profile of the harmonic spectrum, the purple solid curve is the dependence of the harmonic photon with the emission time from the classical calculation. (c) HHG spectrum from the ultrashort laser pulse. (d) Reconstruction of the TDM (orange diamonds) by the HHG spectrum. The laser parameters are 800 nm in wavelength, 4 fs of FWHM, 0 for CEP, and $8.0 \times 10^{12} \text{ W/cm}^2$ in intensity.

4. Summary

In this work, we proposed a new all-optical method to image the k -dependent transition dipole moment by the crystal harmonic spectrum in the ultrashort laser pulse. Based on the accurate band dispersion and the transition dipole moment between the conduction band and the valence one from the first-principles calculations, the interaction of the ultrashort mid-infrared laser pulse with MgO crystal is analyzed by solving SBEs. It is found that, due to the one-to-one correspondence between the photon energy and the crystal momentum at the emission instant, the intensity of the harmonic from a single quantum trajectory is proportional to the square of the TDM's value, then the k -dependent TDM between two bands can be directly retrieved by utilizing harmonics with energies above the minimum bandgap. Furthermore, by examining the orientation-dependent harmonic spectra, our all-optical measurement scheme can be expected to explore 2D k -dependent TDM in condensed matter with an inversion symmetry. Especially, the success of the all-optical reconstruction for crystals with the inversion symmetry would pave the way for extending the method to solids without symmetry, but the transition dipole phase should be considered.

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Disclosures

The authors declare no conflicts of interest.

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