Universality of ultrafast semi-metallization in dielectrics in PHz domain

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Abstract: The ultrafast semimetalization by light field of various materials have been studied. Despite of their different physical properties, similar semimetallization behavior has been observed, which can be well explained by Wannier Stark localization with Zener type tunneling, taking interband and intraband transition into account.

1. Introduction

Light-matter interaction has been long-standing topic of intensive research since 1930s, when modern physics was founded and thriving. For instance, controlling properties of a material using light has also drawn interest. The transition of fused silica into conductive state by optical field is lately demonstrated by Schiffrin, et. al., suggesting a feasibility of current control at a high rate of petahertz domain [1]. Motivated by this work, we performed comparative study among different species of dielectric crystals to answer the following question: will the semimetallization take place for other dielectrics? If so, what will be the aspect like? Three different crystals, quartz, sapphire and calcium fluoride, are subjected to intense laser field and optical-field-induced current is measured.

2. Method

~4fs (sub-2-cycle) VIS-NIR pulses with adjustable carrier-envelope-phase (CEP) are focused onto dielectric substrates on which two gold electrodes are fabricated in order to collect charges driven by optical field. (Fig. 1)

CEP of laser pulse train is manipulated in a way that CEP change of adjacent pulses is fixed to \( \pi \) radian, namely, CEP of every other pulse become equal. By doing so, CEP is modulated at a rate of a half repetition rate. A lock-in amplifier referenced with CEP modulation frequency is employed to extract CEP-dependent component out of overall laser-induced current.

3. Measurements

Charge transfer per pulse \( Q \) with respect to CEP of incident pulse \( \phi \) at a given intensity is measured for three materials (left panel of Fig. 2). \( Q \) versus \( \phi \) curves exhibit oscillatory feature with the same period, which coincides with the period of the irradiated laser field. This implies that charge transfer is driven by the optical field. If normalized, all curves are overlapped, showing similarity despite dissimilarities among the three media. Maximum charge transfer per pulse versus criticality parameter \( \delta \) is plotted in the right panel of Fig. 2. Criticality parameter \( \delta \) is defined as the ratio of laser field strength inside the medium to the critical field [2]. All three curves scale nearly
exponentially. If a proper constant is multiplied, three curves overlap, signifying that the three different materials undergo similar phenomena.

Fig. 2 (left) normalized transferred charge per pulse against CEP of incident pulse. (right) maximum transferred charge per pulse as a function of criticality parameter. All curves are scaled to ensure the best overlap.

4. Theoretical estimation

According to Zener, the probability of transition between two energy levels under the influence of electric field is given as

\[ \gamma = \frac{eF\hbar}{2ma\Delta} \exp\left( -\frac{ma\Delta^2}{4\hbar^2eF} \right) \]  

where \( e \) is the unit charge, \( a \) the lattice constant, \( F \) the field strength, \( \hbar \) the reduced Planck constant, \( m \) the electron mass, \( \Delta \) the band gap[3]. First, we tried to fit the experimental data using eq. (1) as shown in dashed lines in Fig. 3. Zener’s formula tends to fail for strong field case, i.e., \( \delta > 0.7 \). This is because only transition between top valence band and lowest conduction band is considered in Zener’s formalism.

Our new calculation [1,2] is performed taking more transition channels into account. We considered two top valence sub-bands and two lowest conduction sub-bands and assumed that transition is probable between two adjacent sub-bands. Consequently, two more transition paths, within the conduction and valence bands, are allowed. Based on this, charge density is calculated. By multiplying a constant, simulated result is superimposed onto experimental data, showing a good agreement. This indicates that intraband transition becomes significant at strong field while interband transition is dominant for low field.

Fig. 3 Experimental data (squares with error bars), fitted by Zener’s formula (dashed lines) and result of calculation considering interactions between four sub-bands nearby the Fermi level (solid lines).

5. References