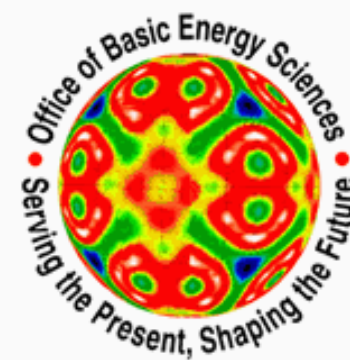


Photo Credit: I. Tsukerman, Seefeld, Austria, January,

2009

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# Solids in Superstrong and Ultrafast Optical Fields

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- **Introduction: strongly nonlinear phenomena in condensed matter in high fields: Wannier Stark localization and Zener tunneling**
- **Adiabatic states of the solid in strong field**
- **Reversible photocurrents in dielectrics**
- **Attosecond field control of dielectrics**
- **Conclusions**

# A Theory of the Electrical Breakdown of Solid Dielectrics

Clarence Zener

*Proc. R. Soc. Lond. A* 1934 **145**, 523-529

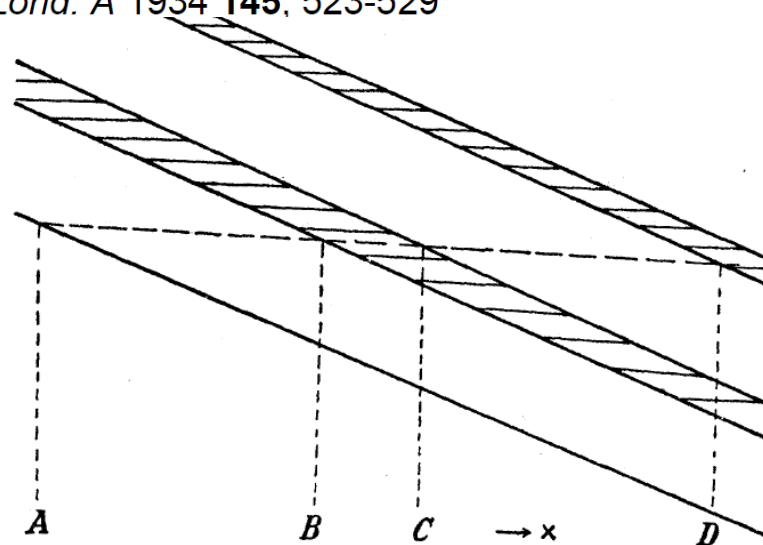


FIG. 1.—“ Potential barrier ” diagram. The shaded regions represent zones of forbidden energy in the presence of an electric field.

Stationary field, bulk solid, bands remain the same

## Strong-Field Phenomena in Condensed Matter

- L. Miaja-Avila, C. Lei, M. Aeschlimann, J. L. Gland, M. M. Murnane, H. C. Kapteyn, and G. Saathoff, *Laser-Assisted Photoelectric Effect from Surfaces*, *Phys. Rev. Lett.* **97**, 113604-1-4 (2006)
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For an insulator, optical field is adiabatic with respect to the bandgap  $\Delta_{vc}$  which greatly exceeds the optical frequency,  $\hbar\omega \ll \Delta_{vc}$ .

In an adiabatic field  $F$ , electron states in each band are localized in the field direction (Wannier-Stark localization). Their localization length  $L_{WS}$  is determined by the phase mismatch with respect to the lattice:

$$ka = \frac{p}{\hbar} a \sim 1, \quad \text{or}, \quad \frac{\sqrt{m^* e F L_{WS}}}{\hbar} a \sim 1, \quad \text{or} \quad L_{WS} \sim \frac{\hbar^2}{m^* a^2 e F}$$

The localization becomes strong when

$$L_{WS} \sim a, \quad \text{or} \quad F \sim \frac{\hbar^2}{m^* a^3 e} \sim 0.1 - 1 \frac{\text{V}}{\text{\AA}}$$

Wannier-Stark states of a given band are identical wave packets shifted by a lattice period. Correspondingly, their energies form an equidistant Wannier-Stark ladder separated by Bloch frequency  $\omega_B = \frac{eEa}{\hbar}$

## Metallization of Nanofilms in Strong Adiabatic Electric Fields

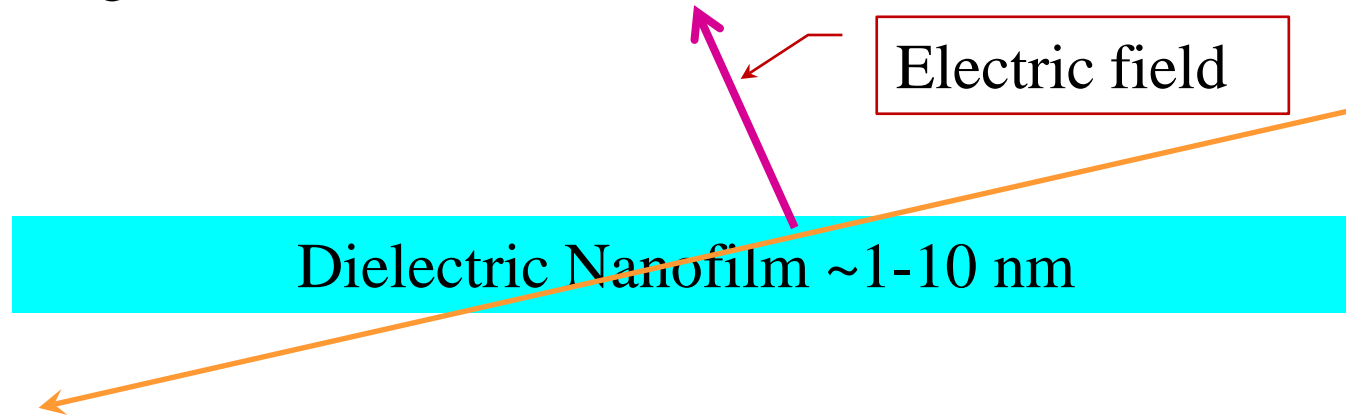
Maxim Durach,<sup>1</sup> Anastasia Rusina,<sup>1</sup> Matthias F. Kling,<sup>2</sup> and Mark I. Stockman<sup>1,2,\*</sup>

<sup>1</sup>*Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303, USA*

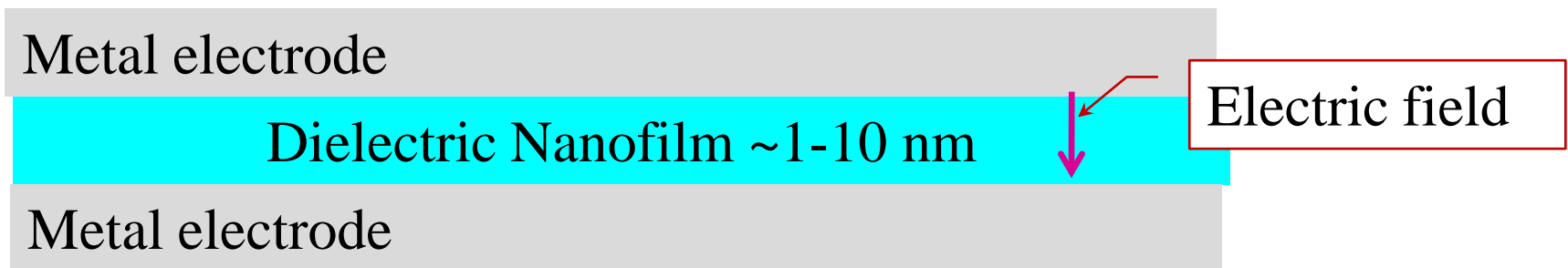
<sup>2</sup>*Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Straße 1, D-85748 Garching, Germany*  
(Received 28 April 2010; revised manuscript received 21 July 2010; published 18 August 2010)

We introduce an effect of metallization of dielectric nanofilms by strong, adiabatically varying electric fields. The metallization causes optical properties of a dielectric film to become similar to those of a plasmonic metal (strong absorption and negative permittivity at low optical frequencies). This is a quantum effect, which is exponentially size-dependent, occurring at fields on the order of  $0.1 \text{ V/\AA}$  and pulse durations ranging from  $\sim 1 \text{ fs}$  to  $\sim 10 \text{ ns}$  for a film thickness of  $3\text{--}10 \text{ nm}$ .

1. Grazing incidence: field almost normal to the nanofilm



2. Capacitor geometry (e.g., gate oxide of a MOSFET)



## Band structure of nanofilm in applied adiabatic (quasi-stationary) field

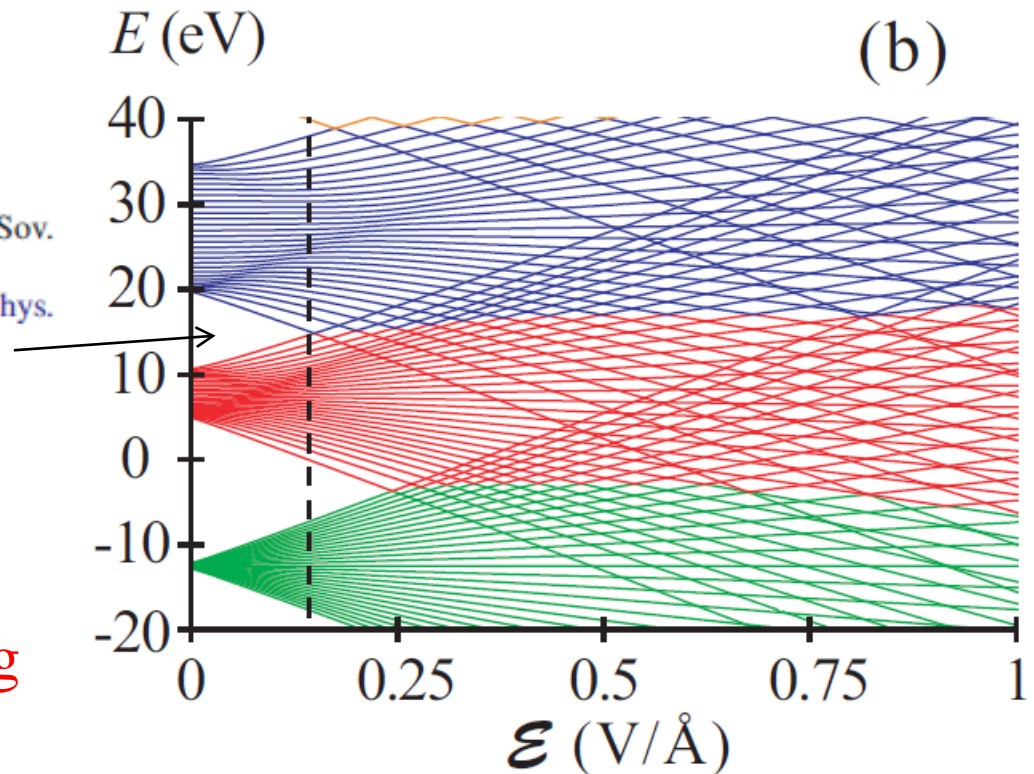
W. Franz, Z. Naturforsch. A 13, 484 (1958).  
 L. Keldysh, J. Exp. Theor. Phys. 34, 1138 (1958) [Sov. Phys. JETP 7, 788 (1958)].  
 D. A. B. Miller, D.S. Chemla, and S. Schmitt-Rink, Phys. Rev. B 33, 6976 (1986).

Electron states ( $m > 0$ ) shift against the field (along the force: energy decreases).

Hole states ( $m < 0$ ) shift along the field (against the force: energy increases).

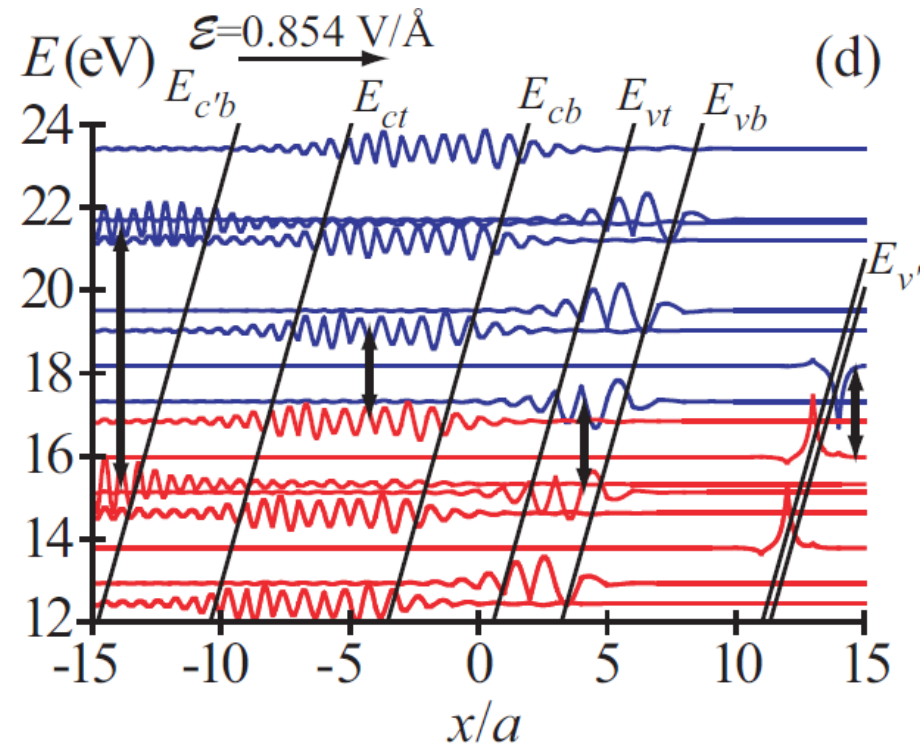
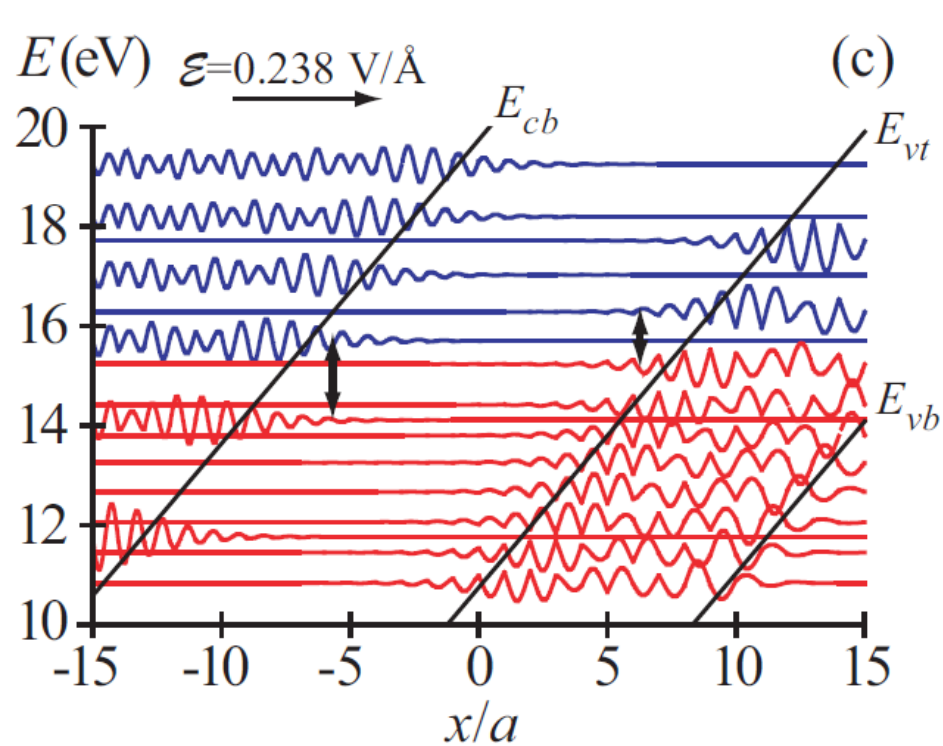
In high fields, the bandgap collapses linearly in the field

$$\mathcal{E}_m = \frac{E_g}{eL}, \quad E_{b,t}(\mathcal{E}) = E_{b,t} \mp e\mathcal{E} \frac{L}{2}, \quad E_g(\mathcal{E}) = E_g - e\mathcal{E}L.$$

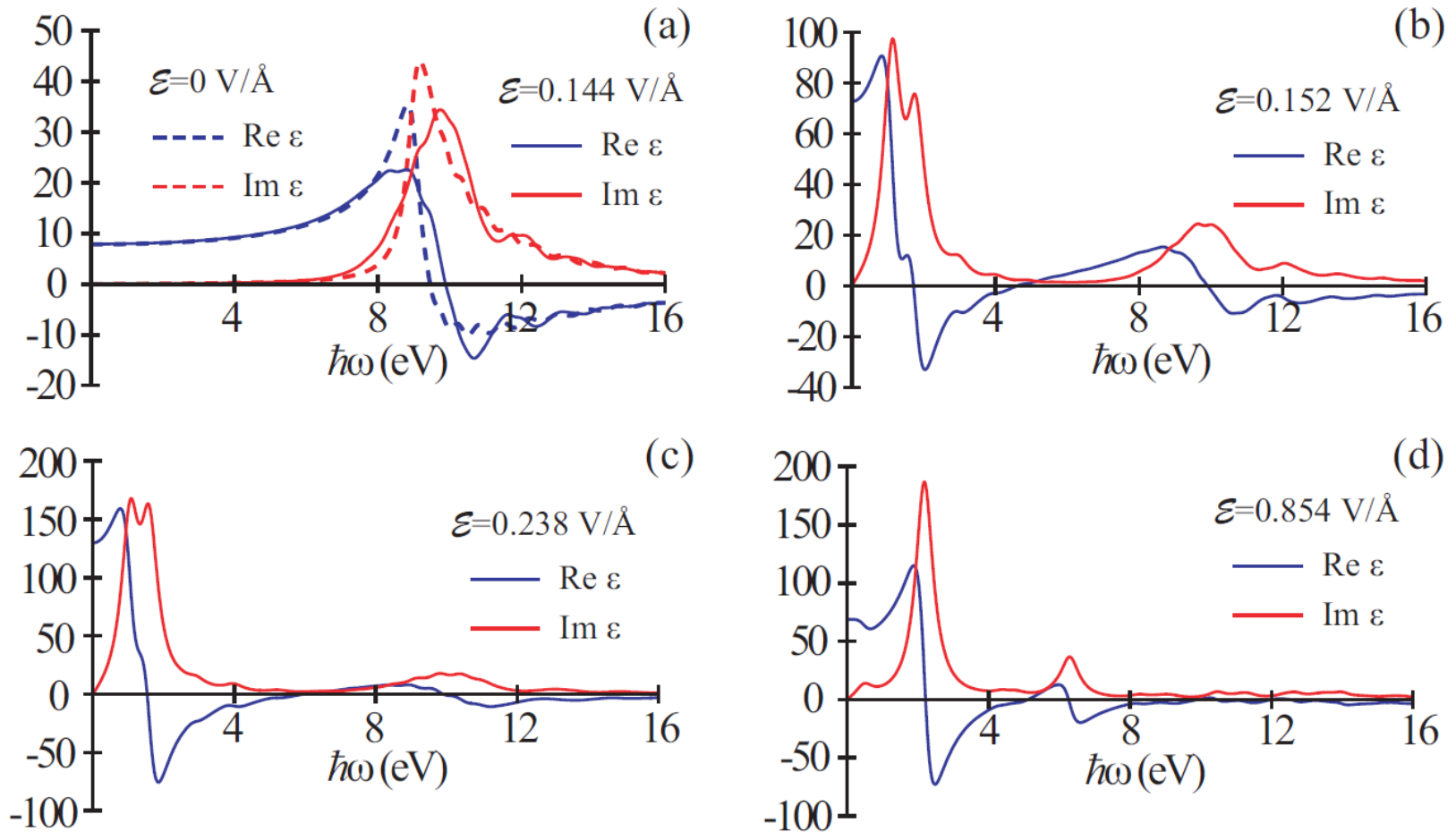




Developed metallization: From quantum bouncers to Wannier-Stark ladder of localized states, transition between which are the Bloch oscillations



## Optical spectra of nanofilm in moderately strong fields: Metallization at $0.151 \text{ V/\AA}$



# LETTER

doi:10.1038/nature11567

## Optical-field-induced current in dielectrics

Agustin Schiffrin<sup>1†</sup>, Tim Paasch-Colberg<sup>1</sup>, Nicholas Karpowicz<sup>1</sup>, Vadym Apalkov<sup>2</sup>, Daniel Gerster<sup>3</sup>, Sascha Mühlbrandt<sup>1,3</sup>, Michael Korbman<sup>1</sup>, Joachim Reichert<sup>3</sup>, Martin Schultze<sup>1,4</sup>, Simon Holzner<sup>1</sup>, Johannes V. Barth<sup>3</sup>, Reinhard Kienberger<sup>1,3</sup>, Ralph Ernstorfer<sup>1,3,5</sup>, Vladislav S. Yakovlev<sup>1,4</sup>, Mark I. Stockman<sup>2</sup> & Ferenc Krausz<sup>1,4</sup>

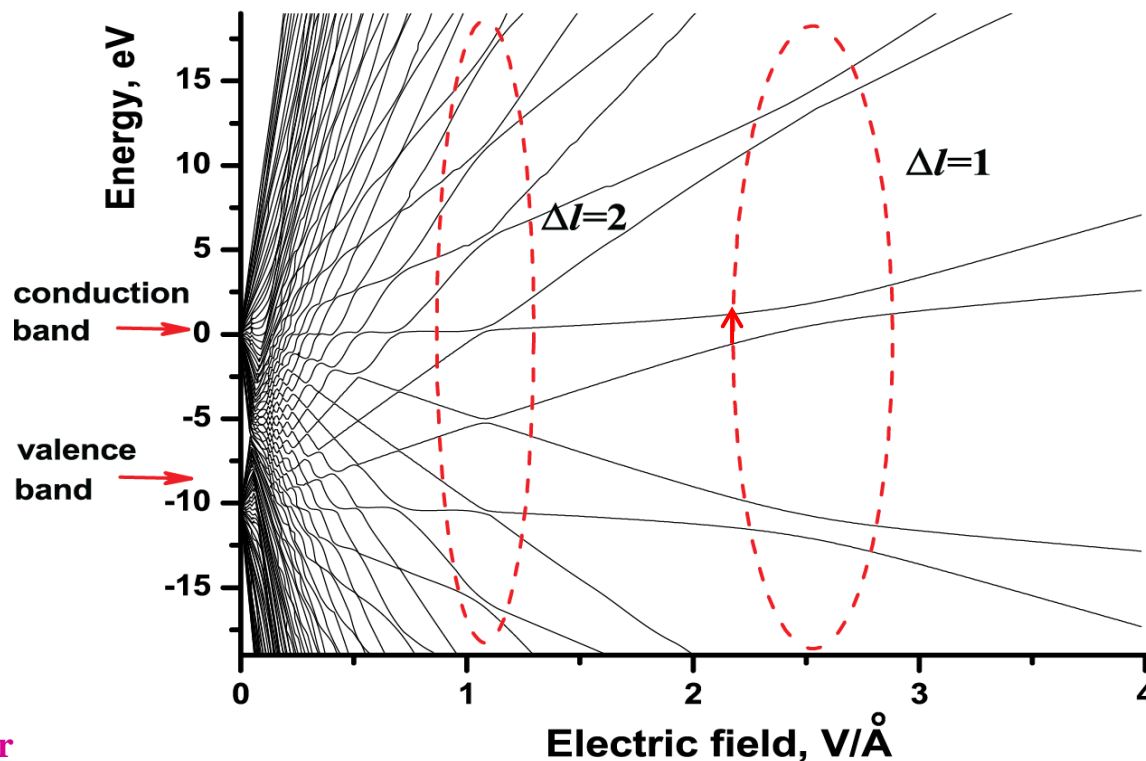
<sup>1</sup>Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, D-85748 Garching, Germany. <sup>2</sup>Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30340, USA. <sup>3</sup>Physik-Department, Technische Universität München, James-Frank-Strasse, D-85748 Garching, Germany. <sup>4</sup>Fakultät für Physik, Ludwig-Maximilians-Universität, Am Coulombwall 1, D-85748 Garching, Germany. <sup>5</sup>Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4–6, 14195 Berlin, Germany. †Present addresses: Department of Physics and Astronomy, University of British Columbia, Vancouver, British Columbia, V6T 1Z1 Canada; Quantum Matter Institute, University of British Columbia, Vancouver, British Columbia, V6T 1Z4 Canada.

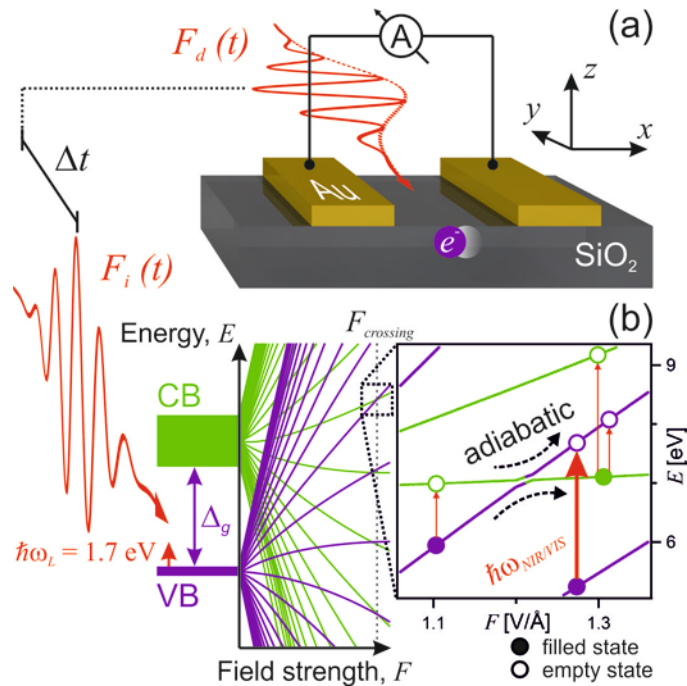
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In a strong ultrafast optical field, an  $n$ -anticrossing (Zener-type transition) with a tunneling across  $n$  lattice periods and transition into the empty conduction band occurs at a field

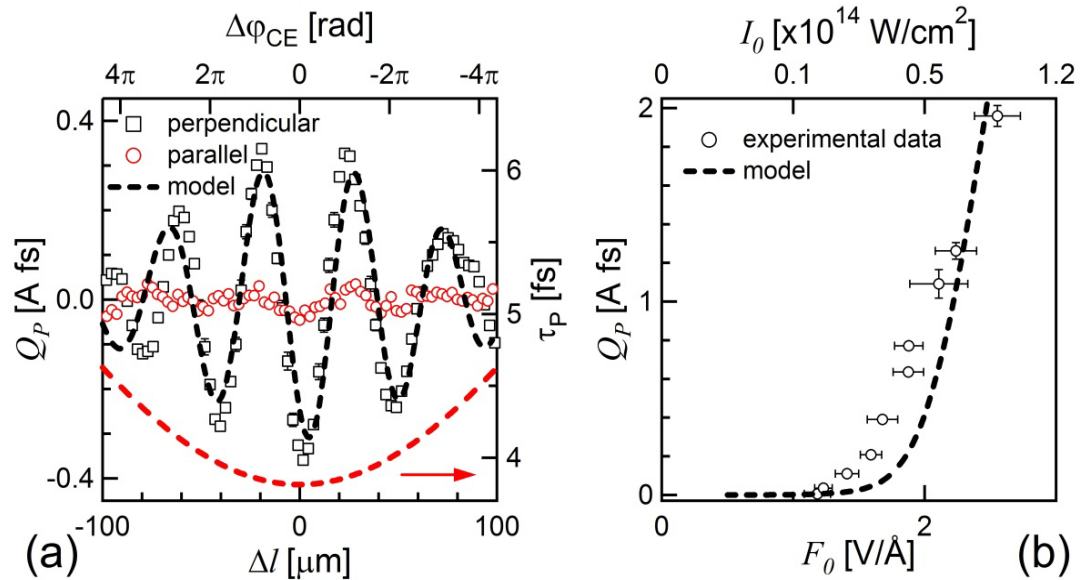
$$E \sim \frac{\Delta_g}{nea} \sim \frac{2}{n} \frac{V}{\text{\AA}}, \text{ where } \Delta_g \text{ is bandgap and } a \text{ is lattice period}$$

Wannier-Stark levels in adiabatic strong field for valence and conduction bands of silica



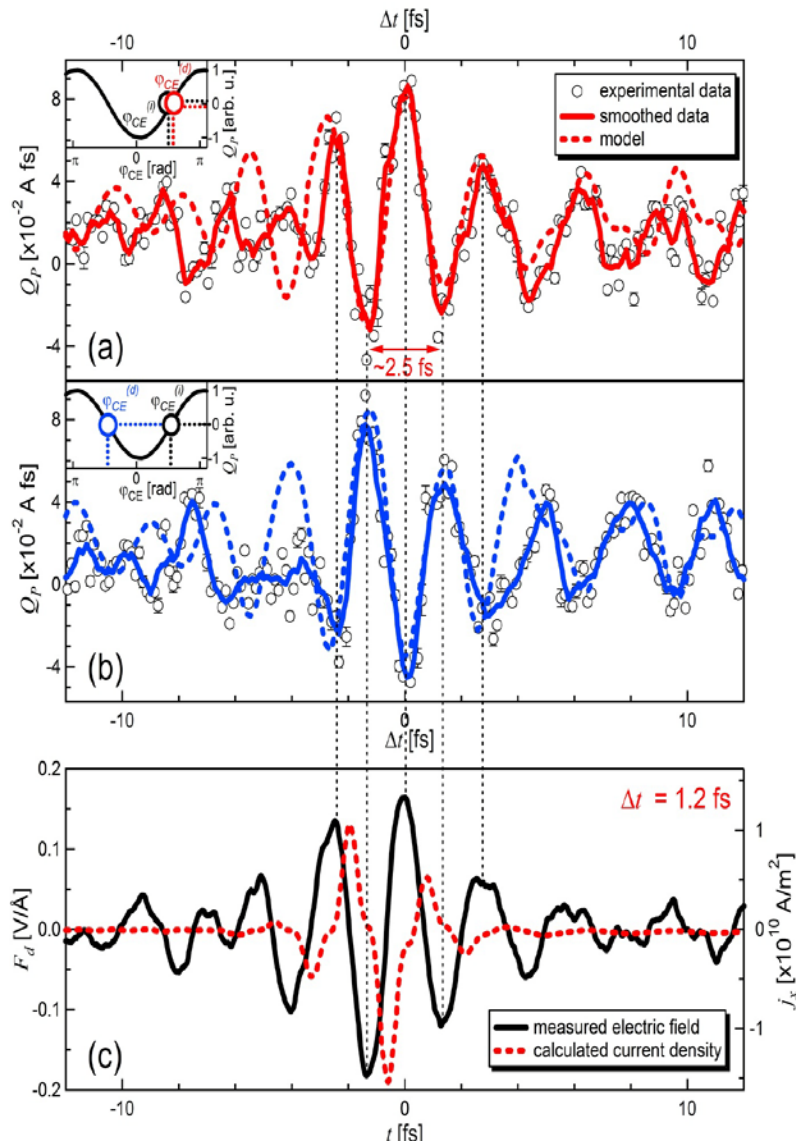


## Single pulse experiment



Carrier-envelope-phase control and intensity dependence of optical-field-generated electric current in SiO<sub>2</sub>.  
Field: normal or **parallel to electrodes**

Optical-field-induced conductivity and current control in a dielectric



## Double pulse experiment

Sub-femtosecond control of electric current with the electric field of light.

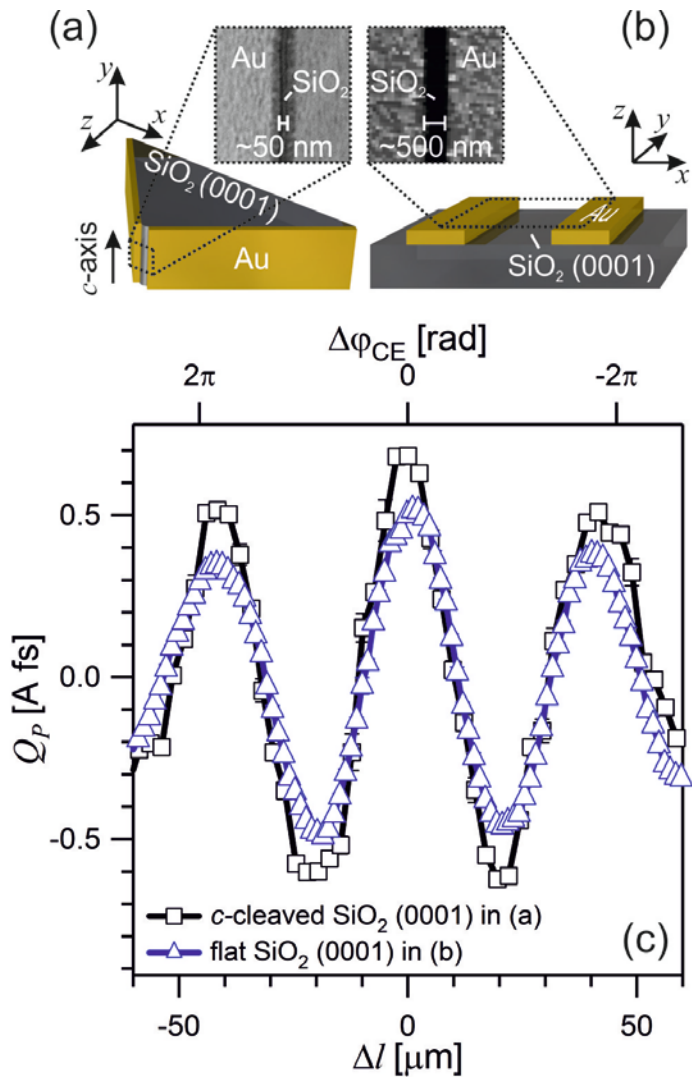
(a)-(b) Transferred charge versus delay between the injection and drive pulses.

(c) Real-time optical electric field of the VIS/NIR pulses retrieved from attosecond streaking. The red dashed curve displays the time-dependent current density as calculated from quantum mechanical model

**Effect does not depend on the gap size – it is non-plasmonic – and does not depend on crystallinity**

Effect of the dielectric crystallinity and of the gap size on the optical-field-induced electric current.

- (a) Schematic of a metal-dielectric-metal nanojunction obtained by cleaving the (0001) surface of monocrystalline SiO<sub>2</sub> (0001) and coating the adjacent surfaces with ~50 nm of evaporated gold
- (b) Schematic of a ~500 nm metal-dielectric-metal nanogap obtained via electron-beam lithography and evaporated gold deposition on monocrystalline SiO<sub>2</sub> (0001)
- (c) Phase-dependent component of the charge per pulse as a function of propagation length through a pair of fused silica wedges for polarization perpendicular (i.e. along the x coordinate) to the metal-dielectric interface . The observed effect is not sensitive to the crystallinity of the dielectric and its thickness.

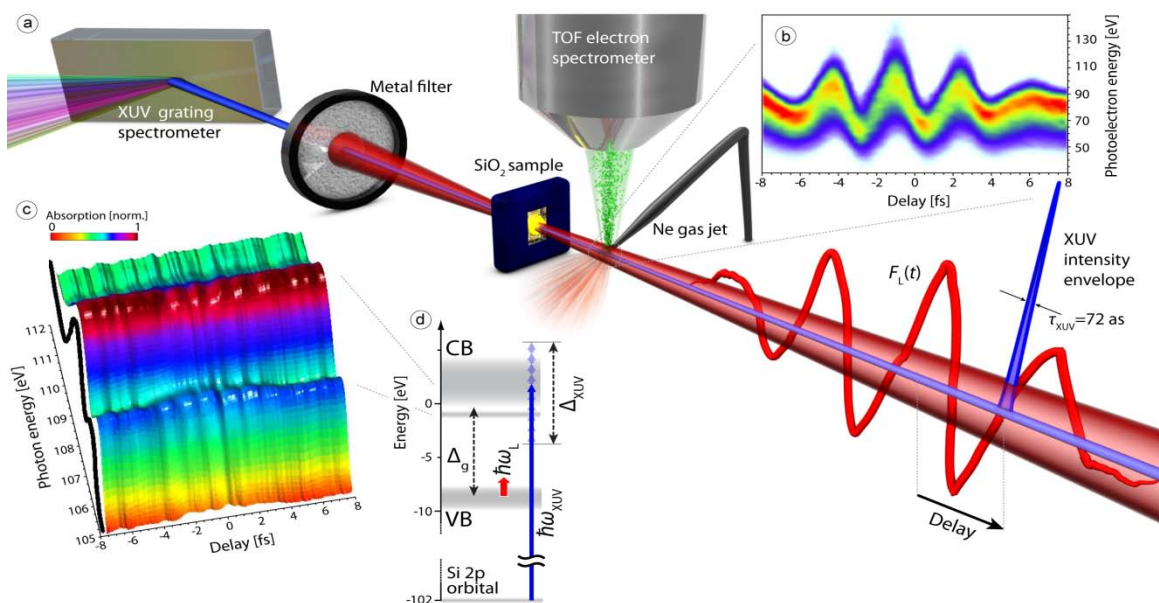


# Controlling dielectrics with the electric field of light

Martin Schultze<sup>1,2</sup>, Elisabeth M. Bothschafter<sup>1,3</sup>, Annkatrin Sommer<sup>1</sup>, Simon Holzner<sup>1</sup>, Wolfgang Schweinberger<sup>1</sup>, Markus Fiess<sup>1</sup>, Michael Hofstetter<sup>2</sup>, Reinhard Kienberger<sup>1,3</sup>, Vadym Apalkov<sup>4</sup>, Vladislav S. Yakovlev<sup>2</sup>, Mark I. Stockman<sup>4</sup> & Ferenc Krausz<sup>1,2</sup>

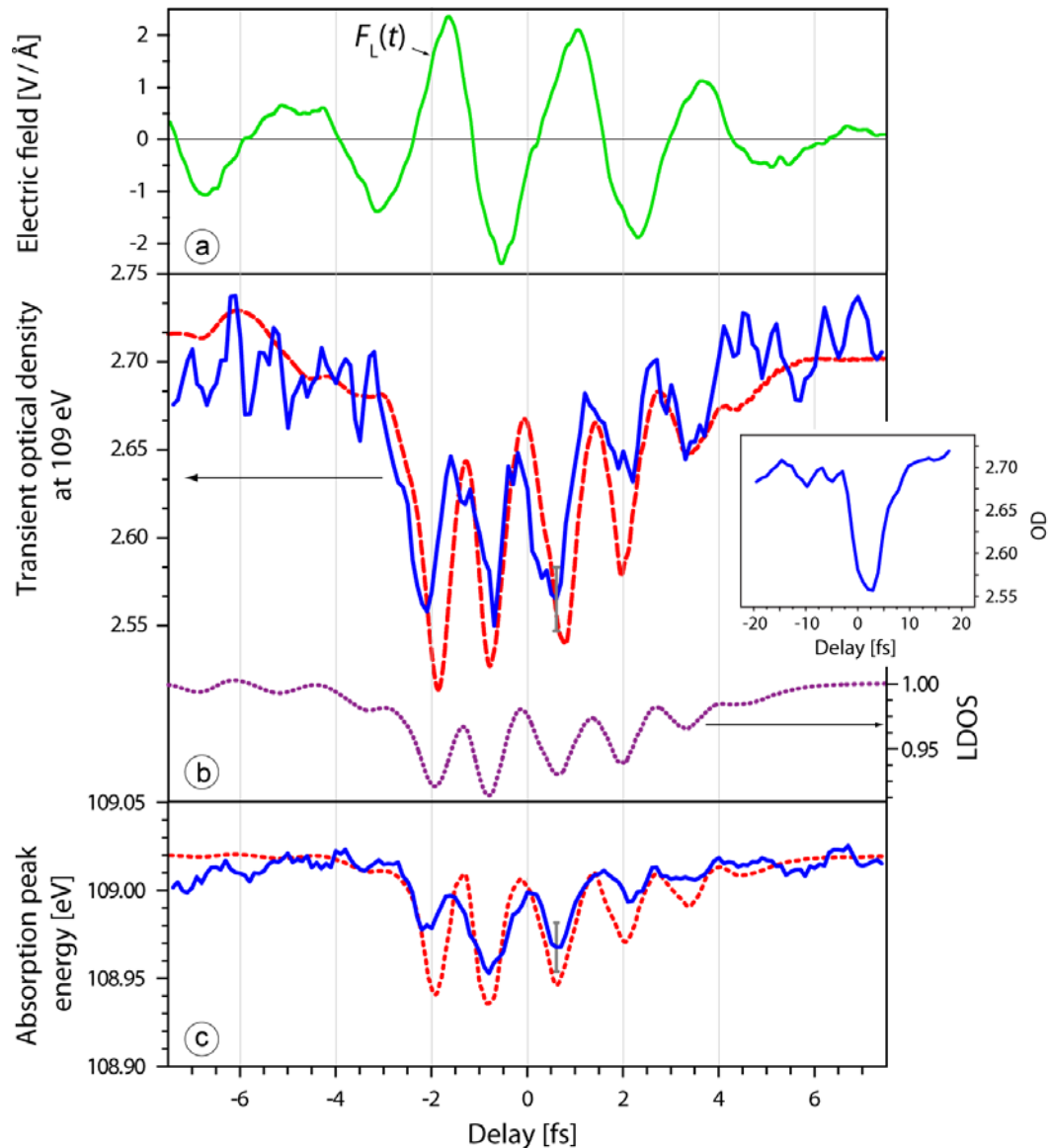
<sup>1</sup>Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, D-85748 Garching, Germany. <sup>2</sup>Fakultät für Physik, Ludwig-Maximilians-Universität, Geschwister-Scholl-Platz 1, D-80539 München, Germany. <sup>3</sup>Physik-Department, Technische Universität München, James-Frank-Strasse, D-85748 Garching, Germany. <sup>4</sup>Department of Physics, Georgia State University, Atlanta, Georgia 30340, USA.

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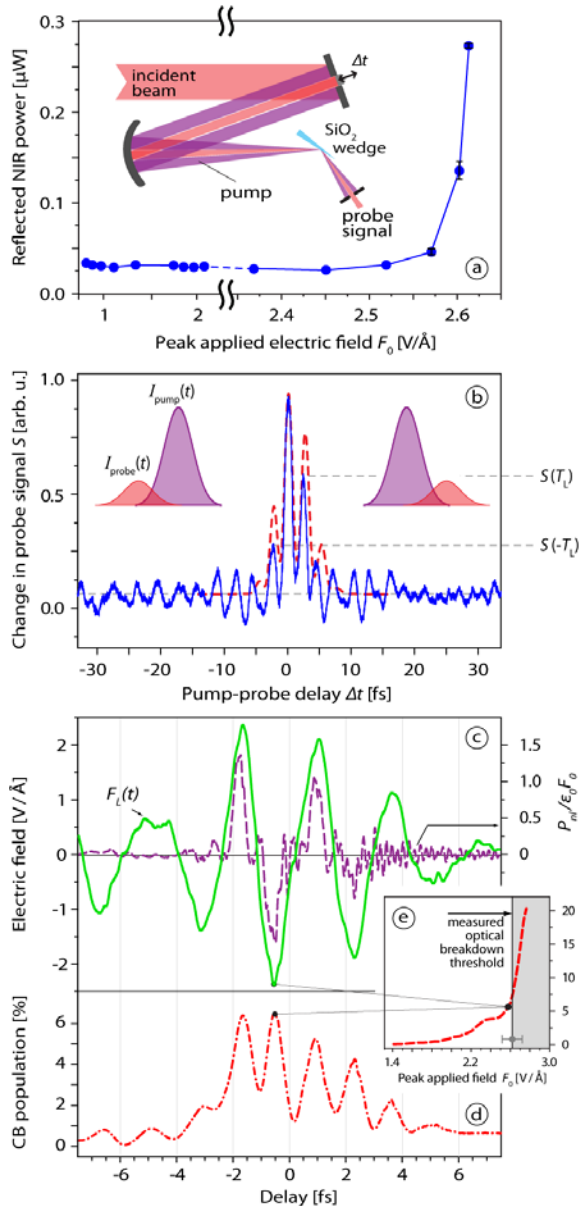
**Attosecond time-resolved strong-field-induced effects in SiO<sub>2</sub>.** Solid lines, experimental results; dashed lines, predictions of theoretical modeling.



(a) Electric field of the few-cycle NIR laser pulse impinging on the SiO<sub>2</sub> sample,  $F_L(t)$ , as extracted from attosecond streaking (see Fig. 1b).

(b) (Transient change of the OD integrated over a 1-eV bandwidth, as a function of the between the 72-as XUV probe and the NIR laser pulse (blue solid line), along with the prediction of quantum mechanical model (red dashed line). The inset shows the OD evolution in a more extended delay range, recorded with larger delay steps ( 0.5 fs).. Dashed violet line: Calculated local density of states (LDOS) at the position of a Si atom (integrated over the energy range accessed by the XUV pulse, for more details, see SI) versus delay of the XUV probe.

(c) Energy of the absorption peak at 109 eV subject to an optical-field-induced (ac-Stark) shift (measurement: blue solid line, calculation: red dashed line)



## Wave-cycle-resolved NIR femtosecond probing of strong-field-induced nonlinear reflectivity of SiO<sub>2</sub>.

- (a) Reflected power of p-polarized sub-4-fs NIR laser pulses incident at Brewster's angle on the thin wedged fused silica sample as a function of peak electric field (penetrating into the sample). The process is fully reversible for several thousand laser shots before irreversible damage occurs due to self-focusing rather inside the sample than on the surface
- (b) Reflected power of the sub-4-fs probe pulse (represented by the red beam) as a function of the delay with respect to the strong sub-4-fs pump pulse (illustrated by the violet beam). Dashed red line represent our model computations.
- (c) Electric field of the few-cycle NIR laser pulse impinging on the SiO<sub>2</sub> sample,  $F_L(t)$ , as extracted from attosecond streaking [see Fig. 1(b), green solid line], and the resulting internal net optical electric field  $F(t)$  predicted by our model (purple dashed line). (d) Computed transient evolution of the population of (unperturbed) CB states timed to the laser field. (e) Peak transient CB population versus peak applied field strength. The grey solid bar represents the experimental value of the optical breakdown threshold as determined from the reflectance measurement yielding the data of panel (a).

## CONCLUSIONS

- We have predicted and observed the fastest phenomena possible in optics,  $<1$  fs limited by the bandwidth of the valence and conduction bands
- A strong field  $\sim 1-2$  V/Å closes the bandgap adiabatically and makes the insulator highly-polarizable without damaging the solid
- Optical polarization induced in the presence of strong field  $E < 2.5$  V/Å causes currents  $\sim 1$  A in dielectrics for  $\sim 1$  fs without damage
- The charge transfer induced by the strong field is controlled by its carrier-envelope phase (it is in the direction of the maximal field)
- The charge transfer by the weak (probe) field follows its instantaneous value at the moment of the strong field maximum within  $<1$  fs time
- The attosecond absorption experiment shows that the electron population of the conduction band follows the strong field and, within the experimental precision  $\sim 1\%$  disappears within  $\sim 1$  fs after the strong field end
- Reversibly with  $\sim 1$  fs response time silica acquires semimetallic properties with conduction increased by 18 orders of magnitude

A dramatic sunset scene over a large body of water. The sky is filled with dark, heavy clouds, illuminated from below by the setting sun, creating a deep orange and red glow. In the foreground, the dark silhouettes of buildings are visible, with a few small lights glowing from windows. The water in the middle ground is dark, and numerous small sailboats are scattered across its surface, their sails catching the low light. The overall mood is serene and contemplative.

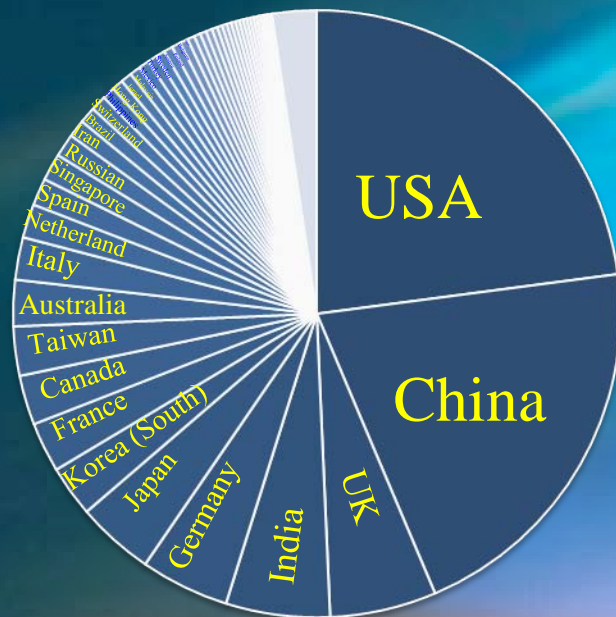
Thank you!

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