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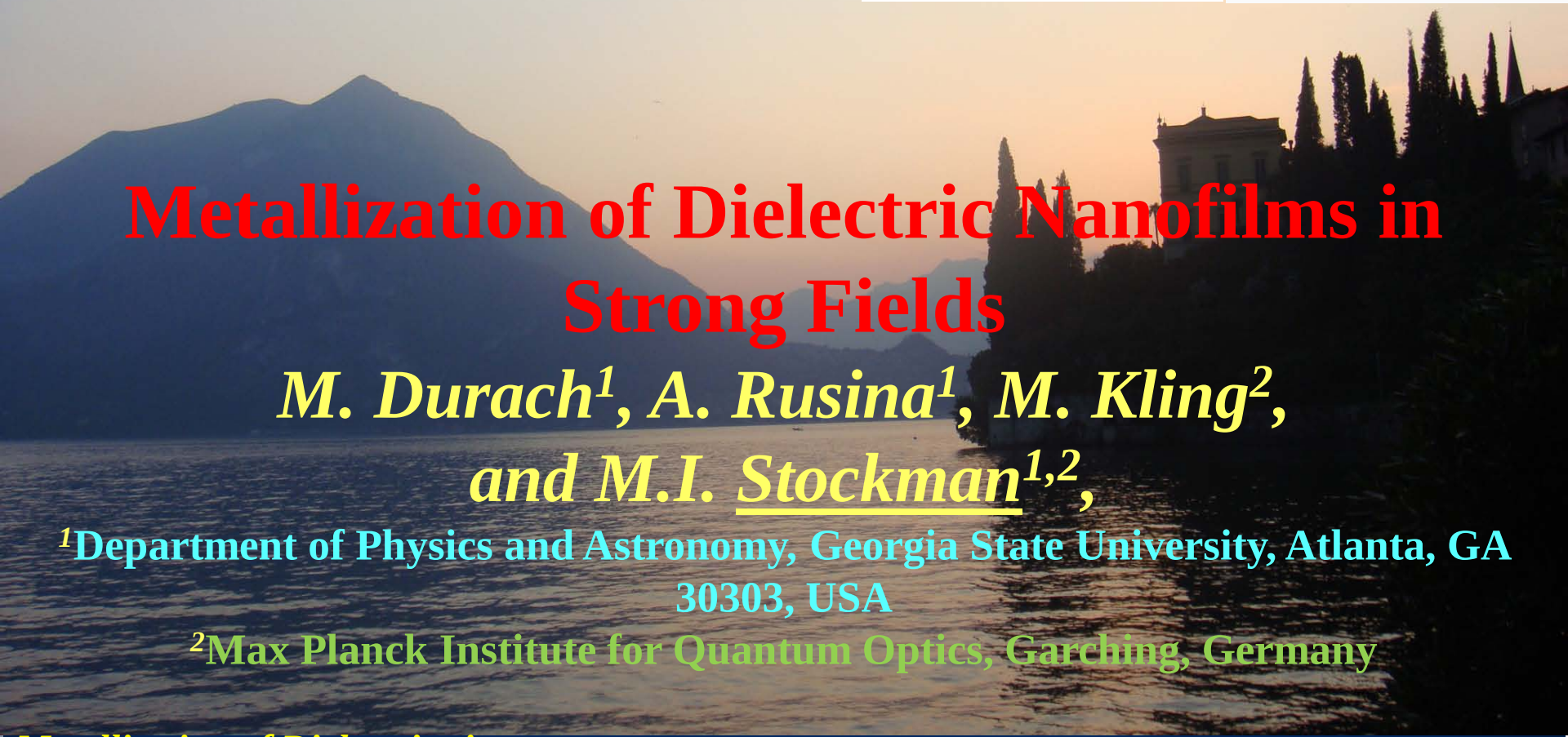

Metallization of Dielectric Nanofilms in Strong Fields

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High-field phenomena:

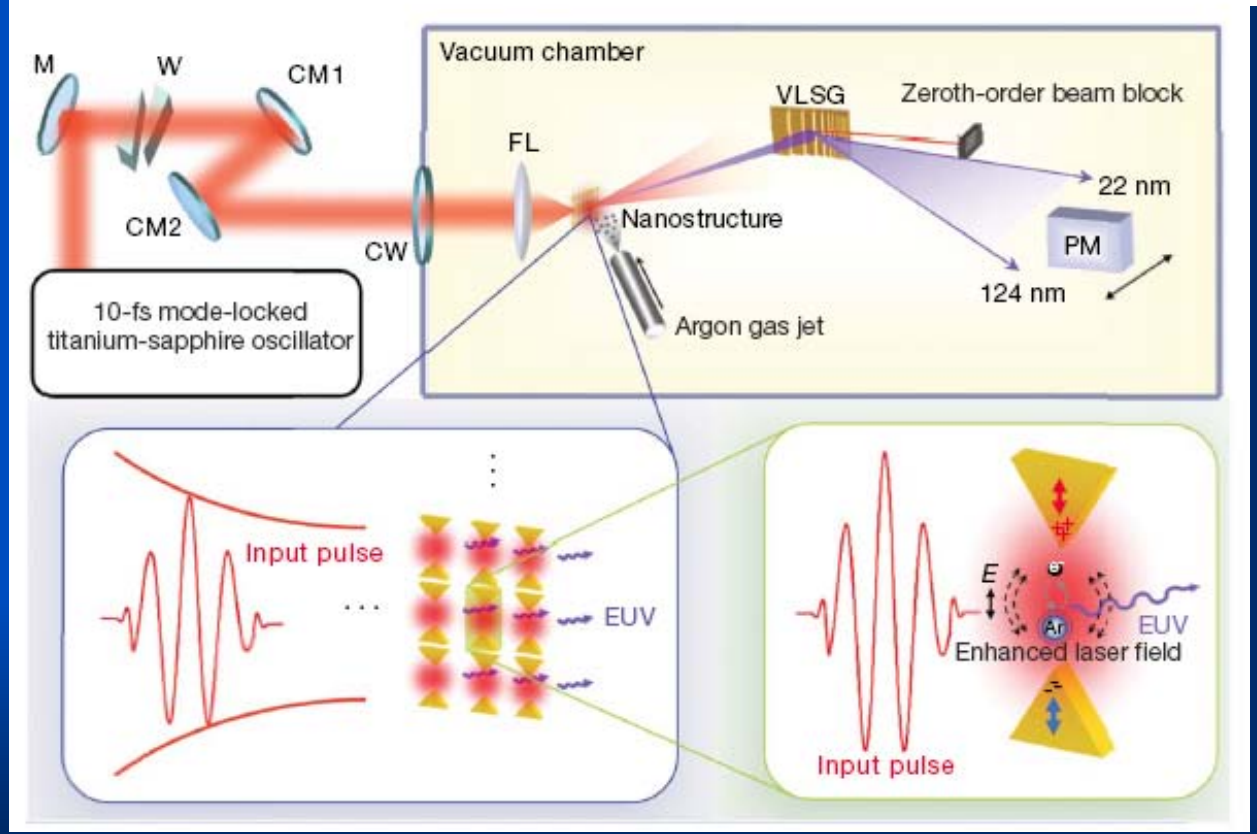
Optical fields are comparable to or stronger than atomic fields

Vol 453|5 June 2008|doi:10.1038/nature07012 nature

LETTERS

High-harmonic generation by resonant plasmon field enhancement

'oo Kim[1]



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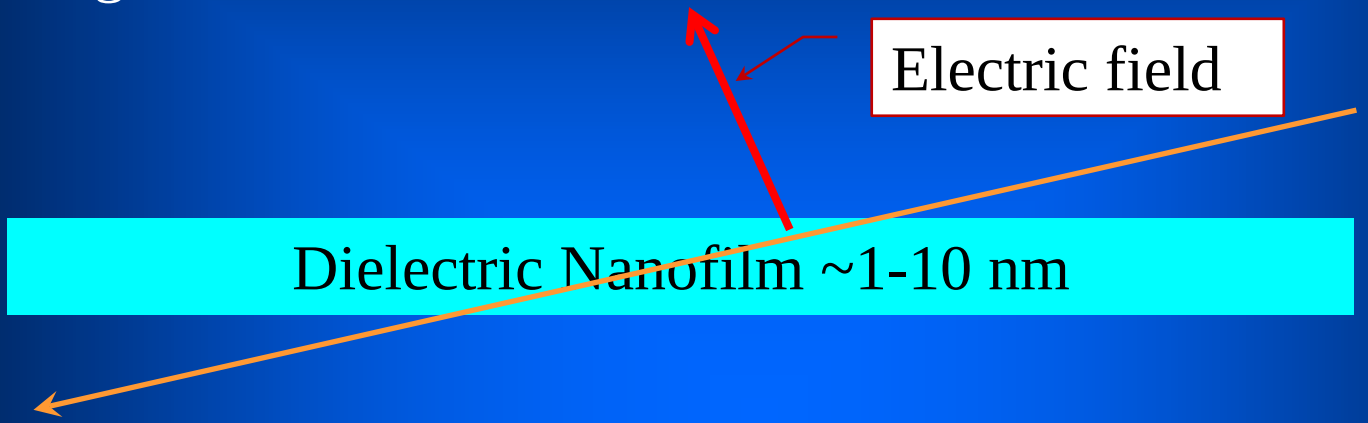
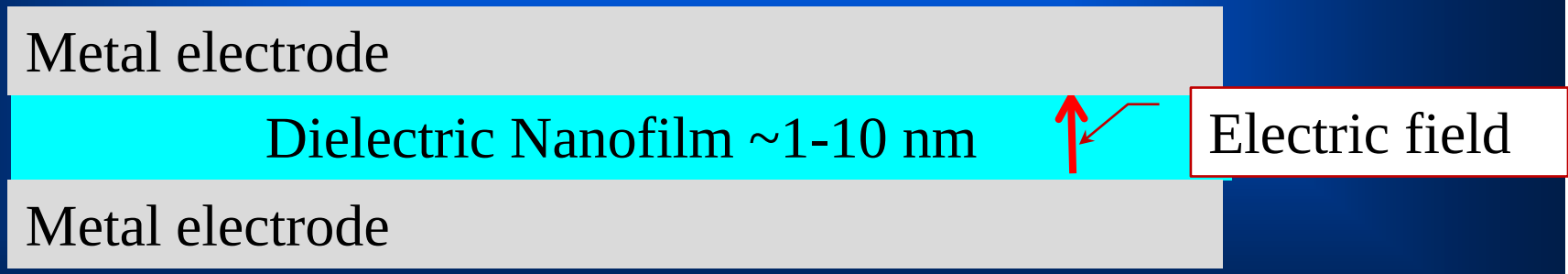
1. Grazing incidence: field almost normal to the nanofilm



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



PROCEEDINGS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

A Theory of the Electrical Breakdown of Solid Dielectrics

Clarence Zener

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

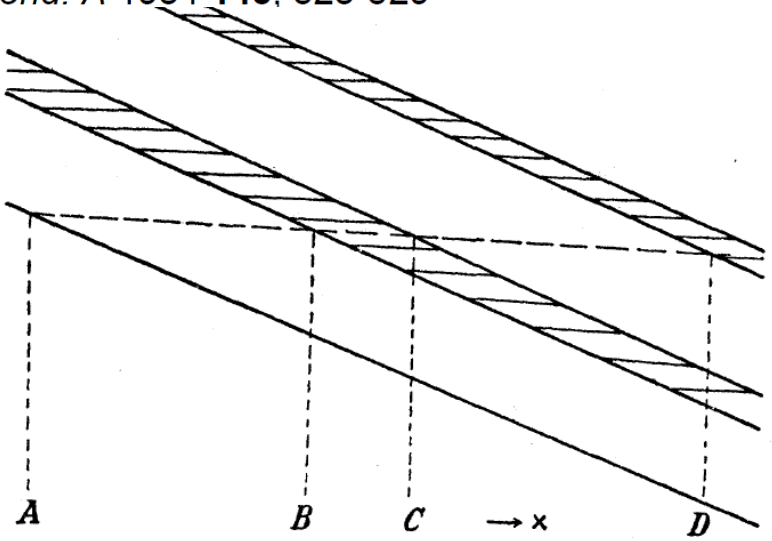


FIG. 1.—"Potential barrier" diagram. The shaded regions represent zones of forbidden energy in the presence of an electric field.

Field ~1-10 V/Å: Electron acquires the band-gap energy within the unit cell

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M. Durach, A. Rusina, M. F. Kling, and M. I. Stockman, *Metallization of Nanofilms in Strong Adiabatic Electric Fields*, Phys. Rev. Lett. **105**, 086803-1-4 (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). The is a quantum effect, which is exponentially size-dependent, occurring at fields on the order of 0.1 V/Å and pulse durations ranging from ~ 1 fs to ~ 10 ns for film thickness $3-10$ nm.

Kronig-Penney model with applied electric field:

$$V(\mathbf{r}) = \begin{cases} U(x)+U(y)+U(z)+e\mathcal{E}x & |x| < L/2 \\ \infty & |x| \geq L/2 \end{cases}$$

$$U(x) = -\alpha \sum_{n=-\infty}^{\infty} \delta(x-na)$$

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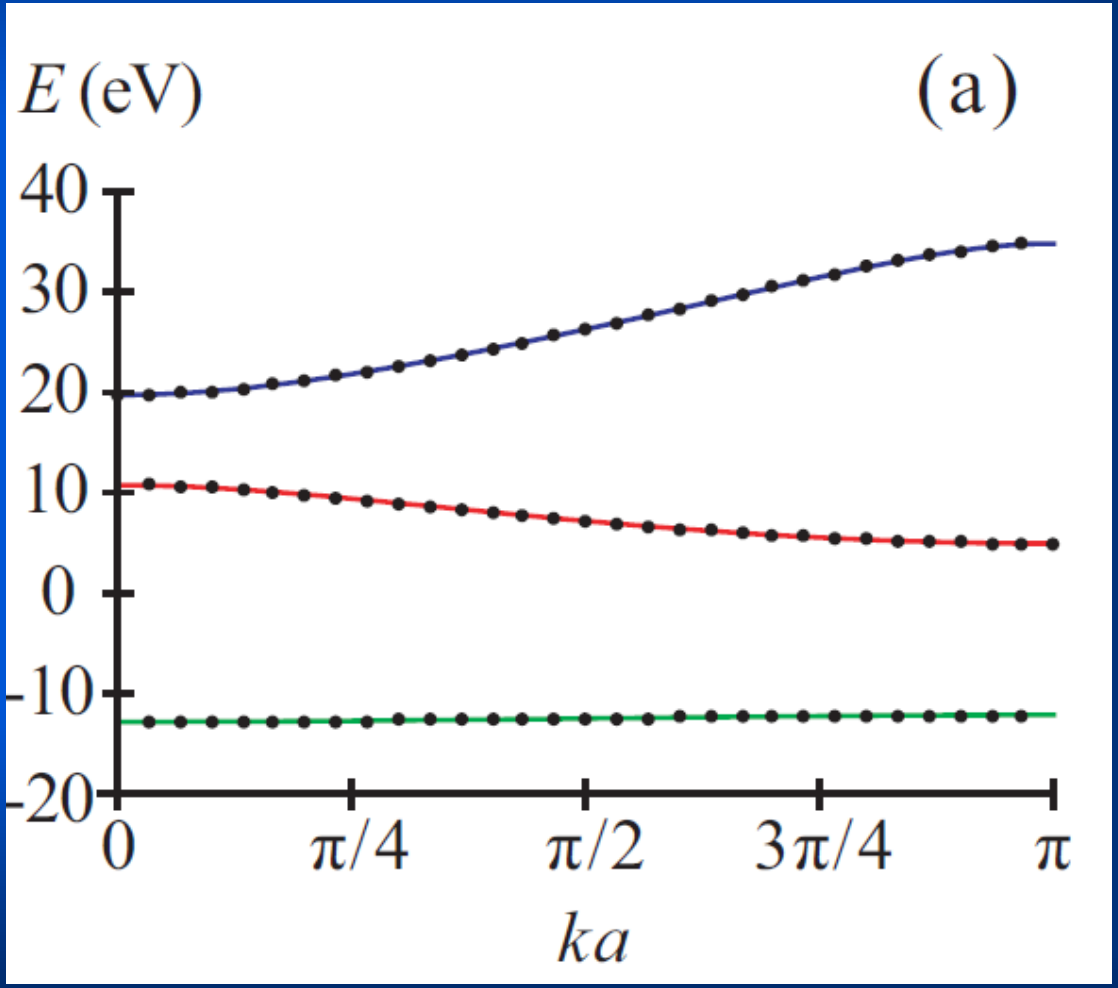
Band structure of the solid (lines) and nanofilm (dots)



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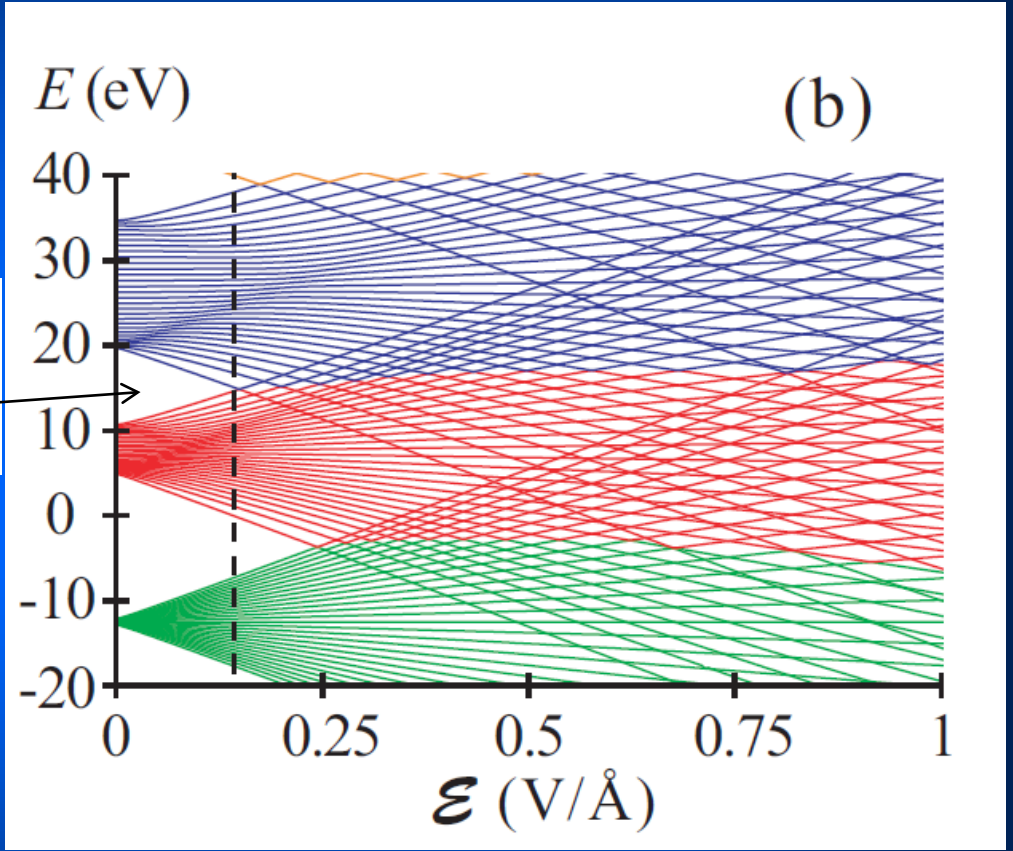
Band structure of nanofilm in applied adiabatic (quasi-stationary) field

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).



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

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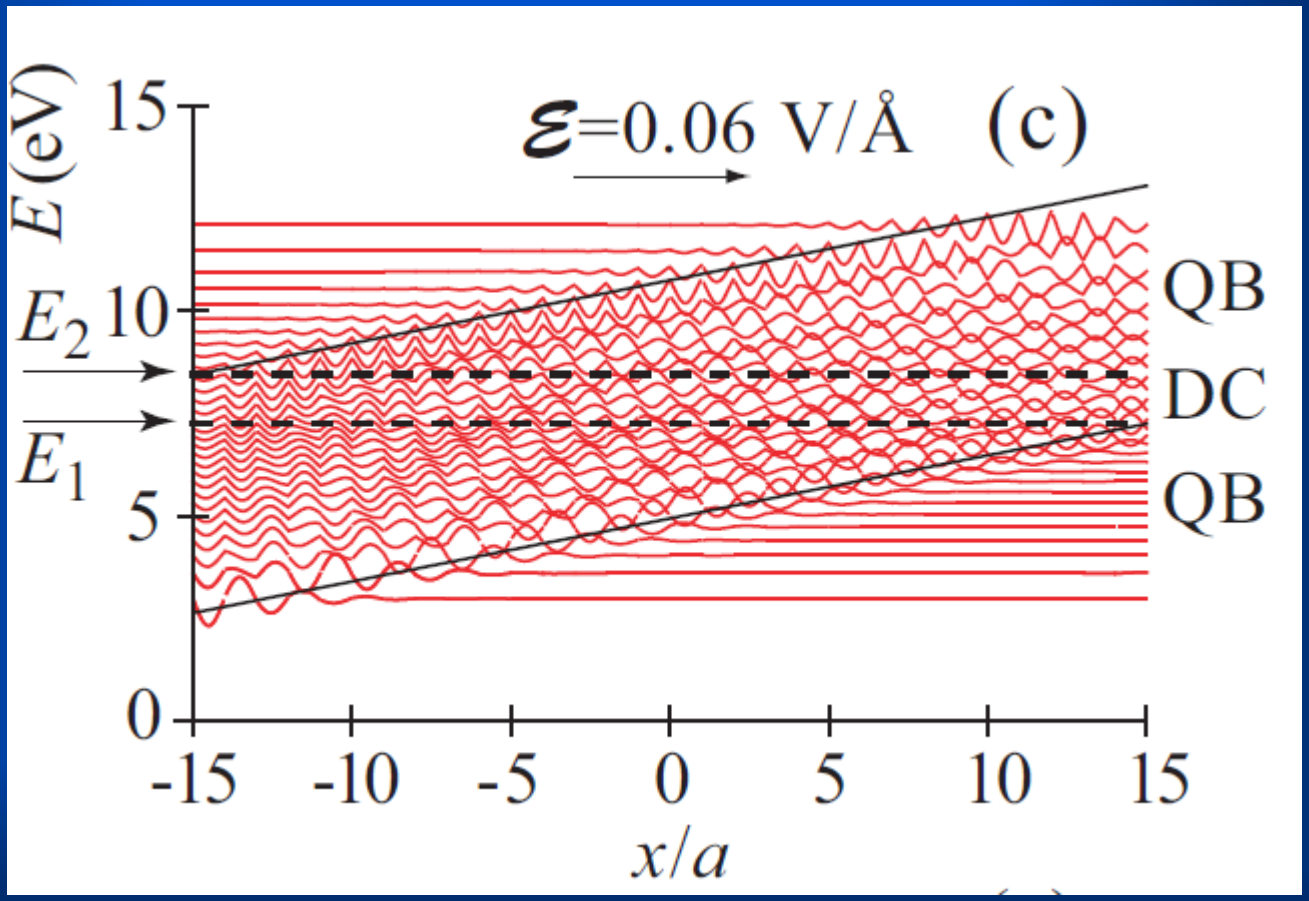
Valence band states in a moderate field



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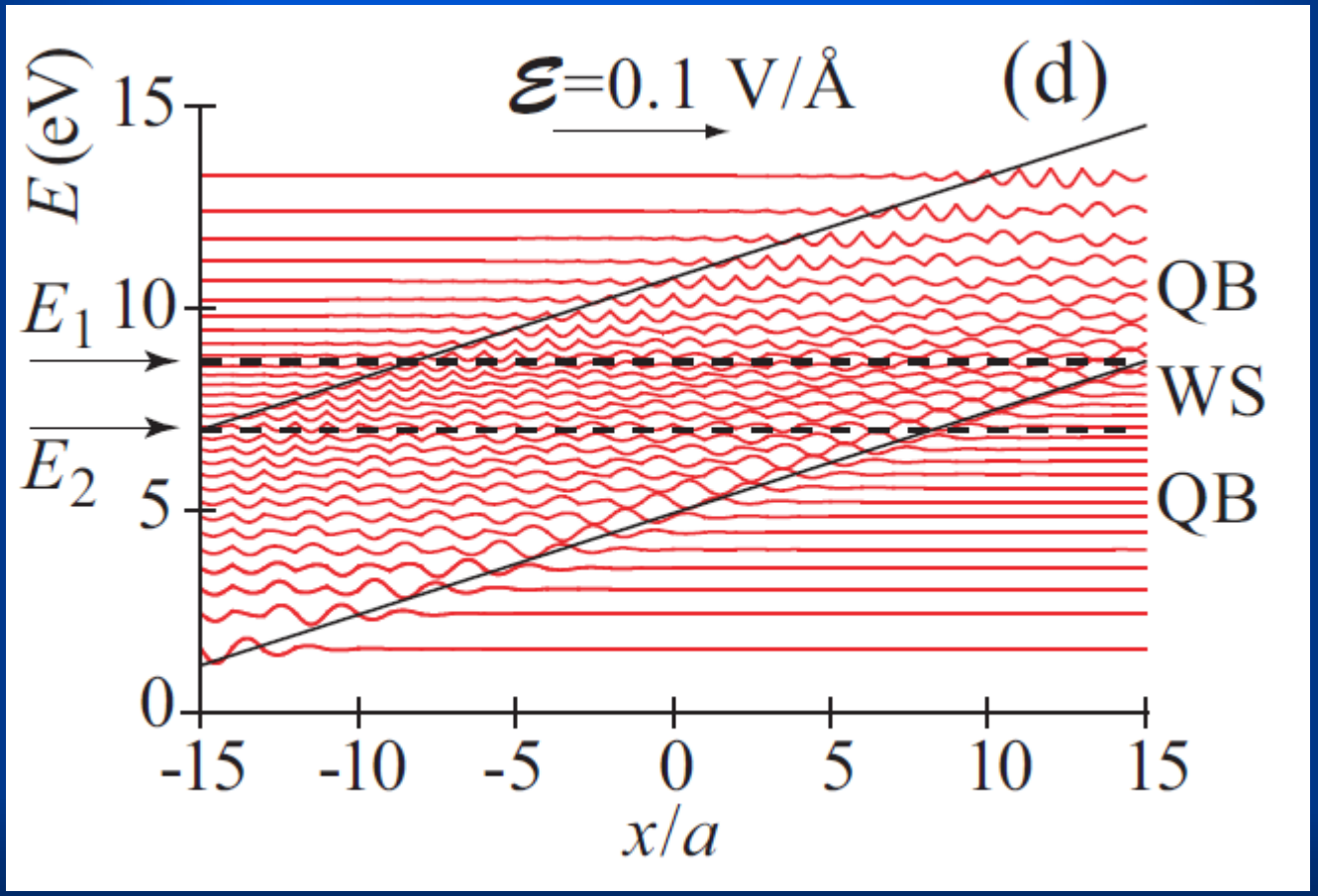
Valence band states in a strong field: all states are localized



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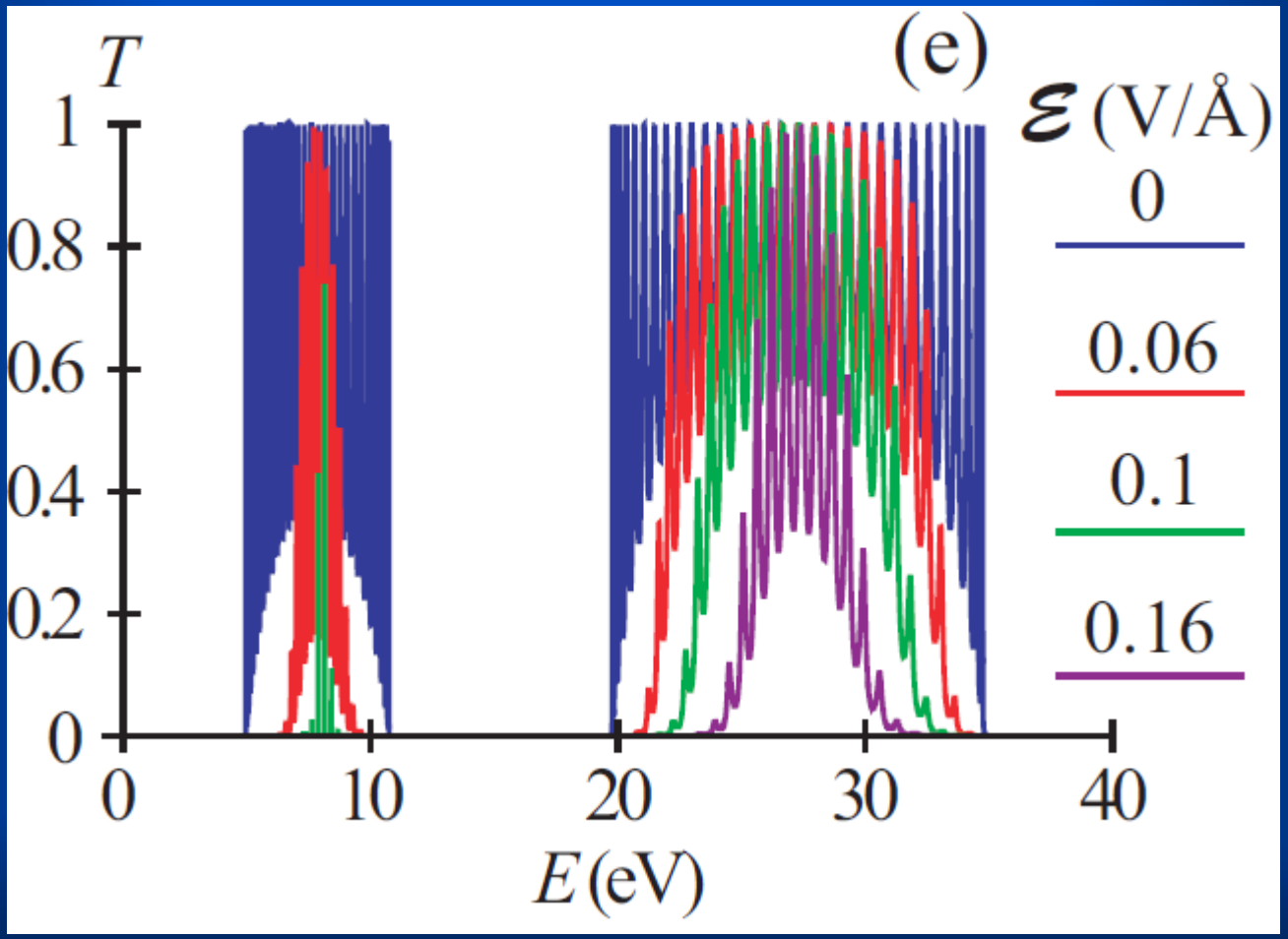
Band transparency collapses in moderate fields



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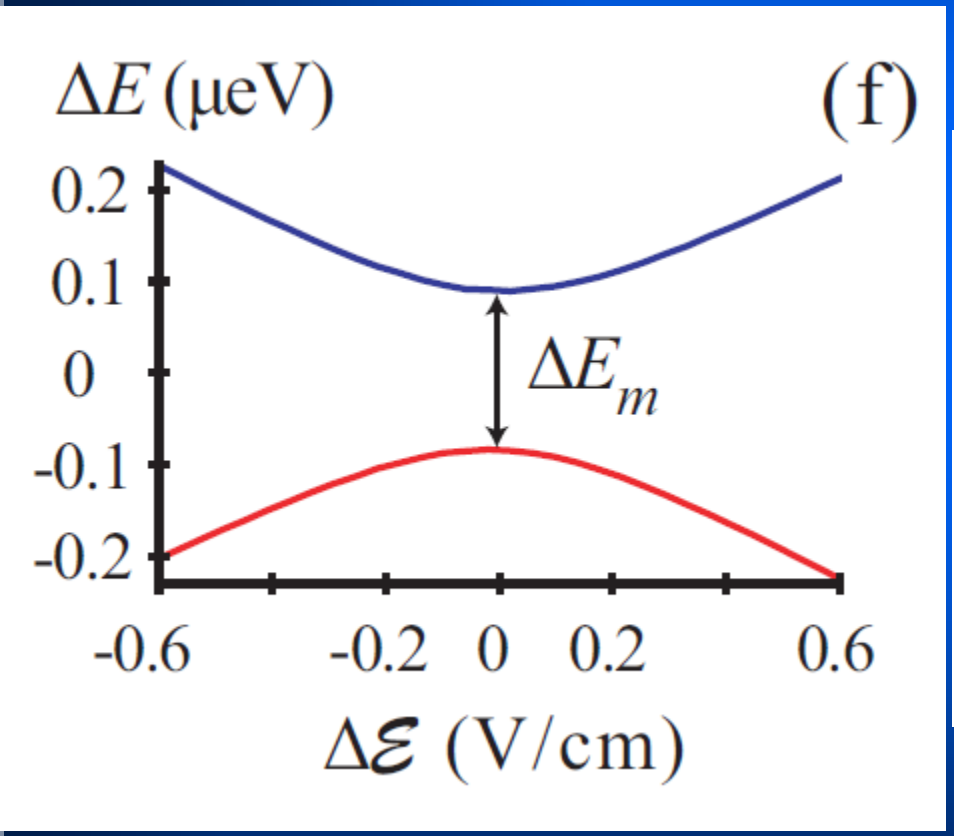
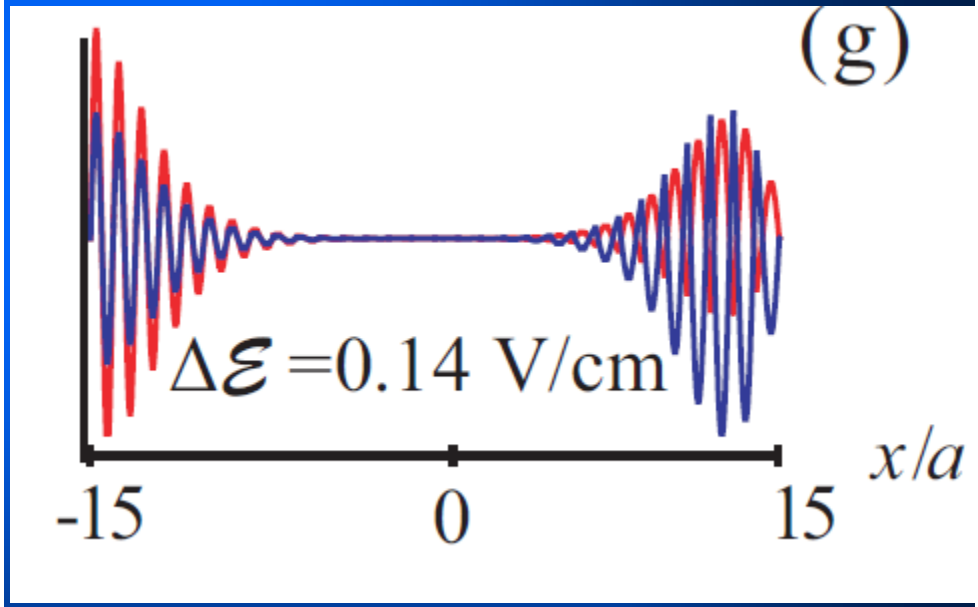
Anticrossing of the valence and conduction band





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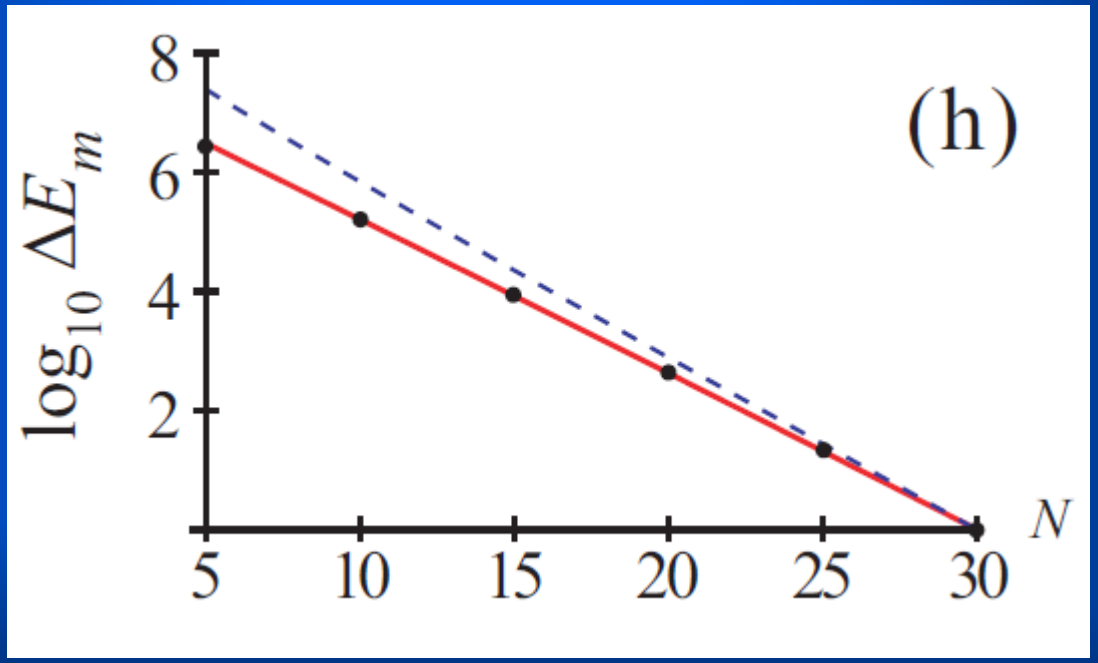
Splitting of bands as a function of the film thickness (number of unit cells) has an exponential dependence



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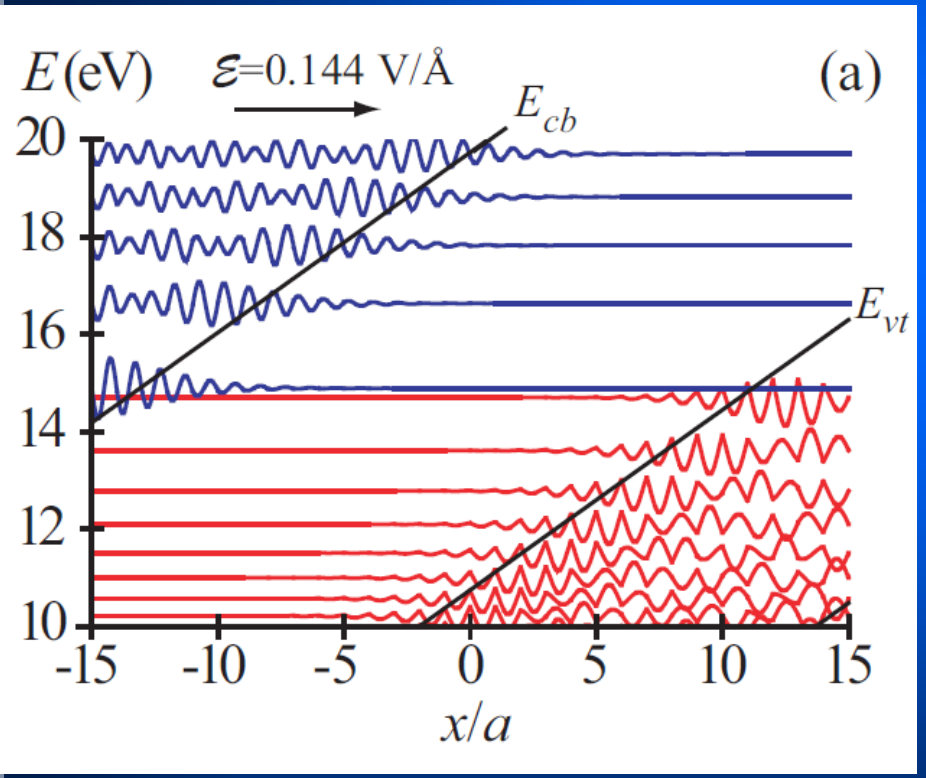
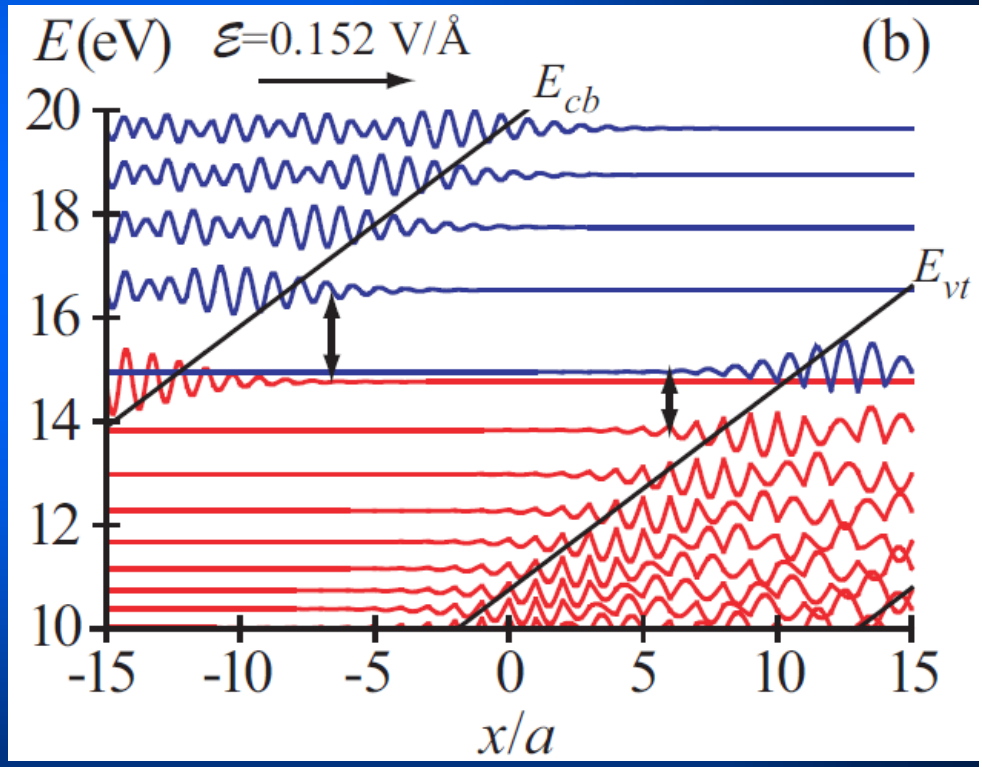
Adiabatic metallization: electrons remain in the ground state and are **transferred in space from one boundary to another** . **Now interband transitions are allowed due to the wave function overlap**





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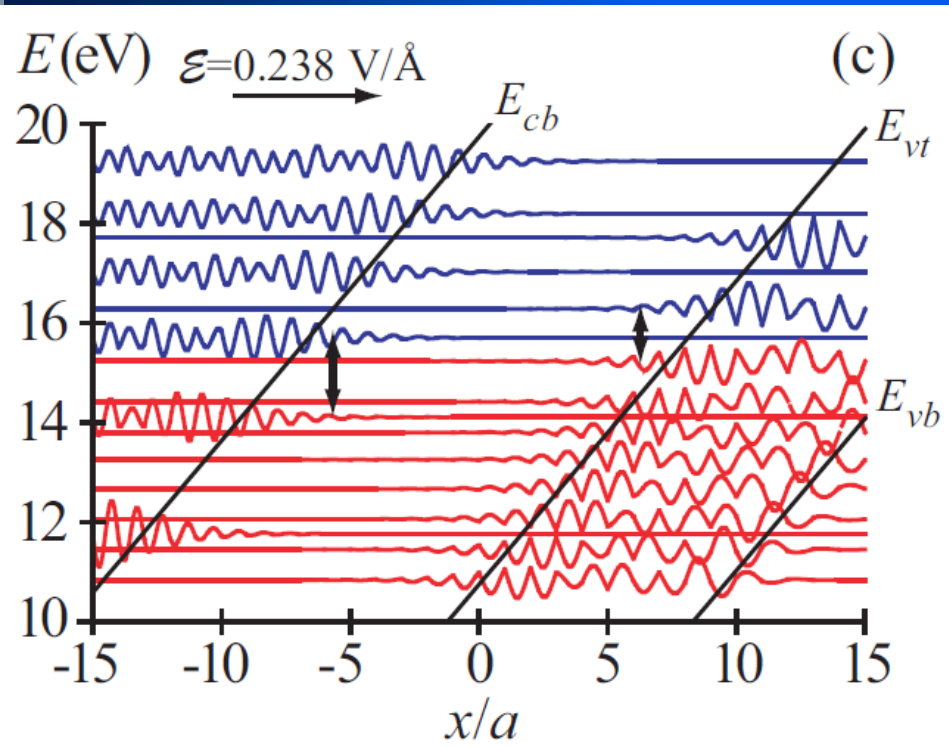
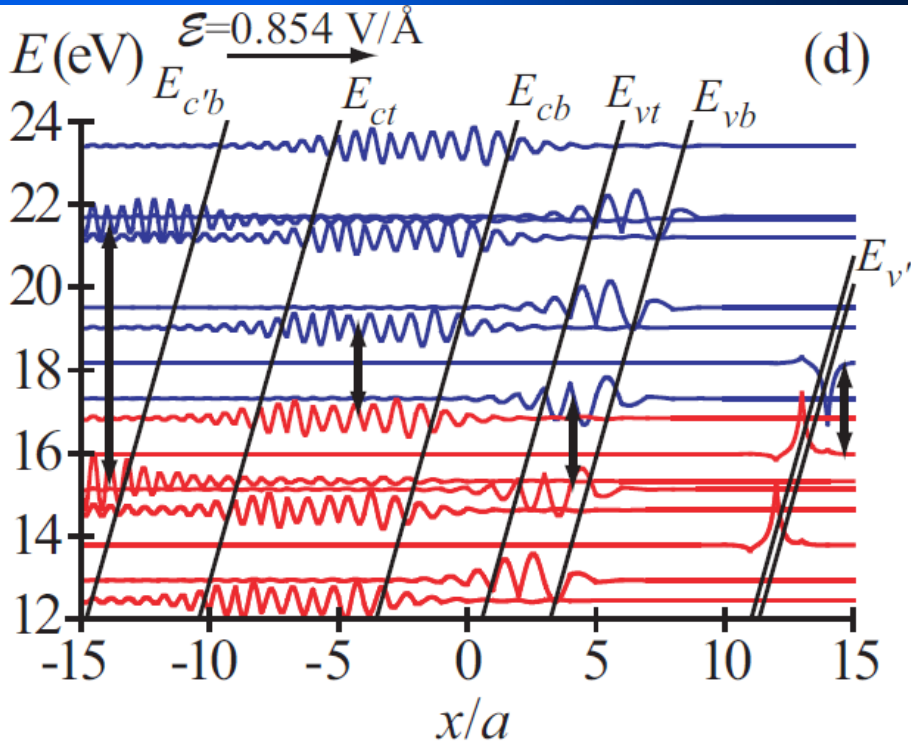
Developed metallization: From quantum bouncers to Wannier-Stark ladder of localized states, transition between which are the Bloch oscillations





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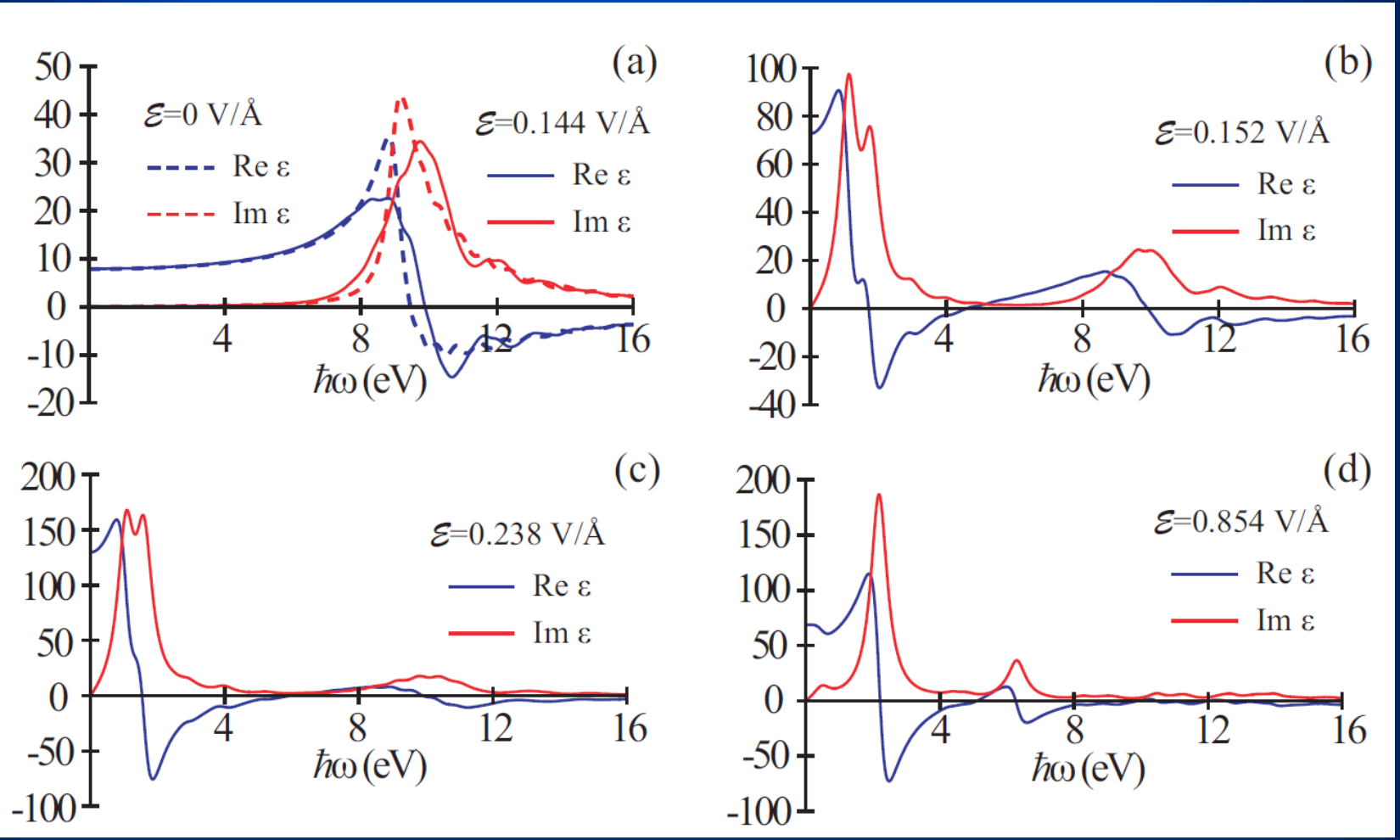
Spectra of nanofilm in moderately strong fields: metallization



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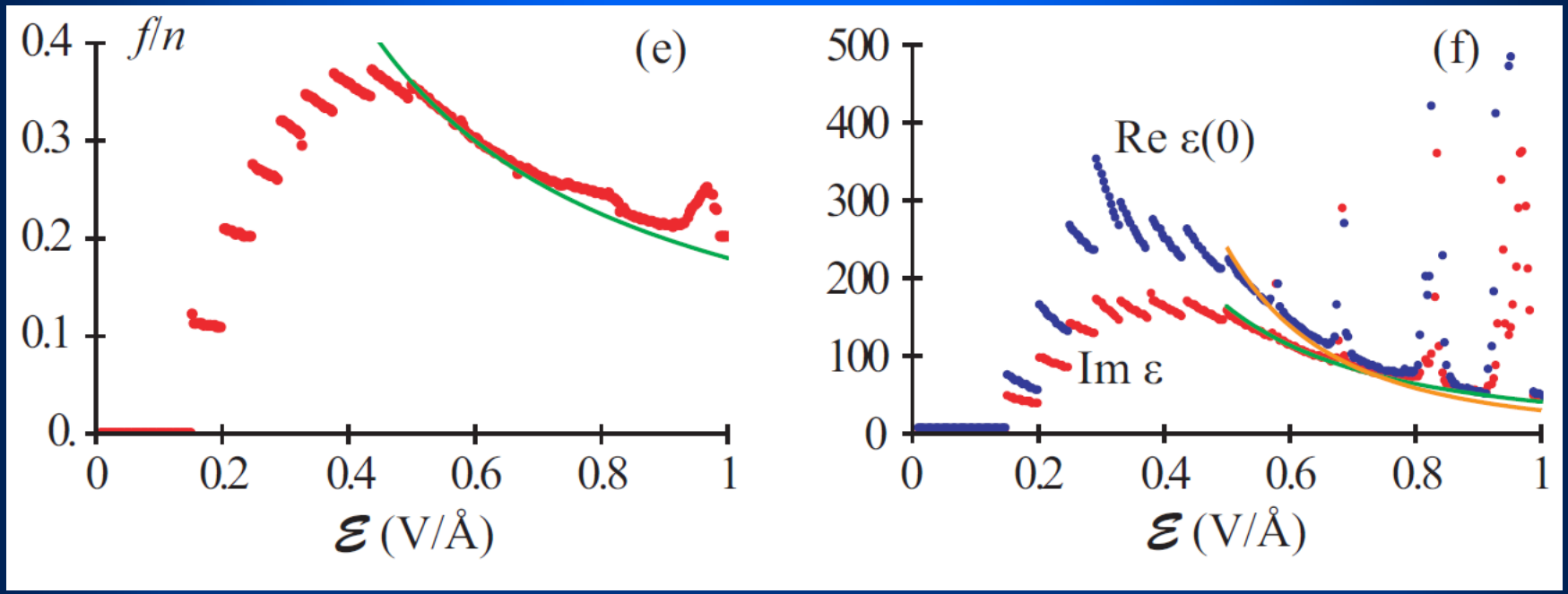
Oscillator strength and low-frequency dielectric permittivity of nanofilm as functions of the normal electric field



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Concluding (see also Sec. VIII of Supplemental Material[11]), we have predicted an effect of metallization in dielectric nanofilms induced by an adiabatically increasing applied field. The localized states crossing the Fermi surface cause optical absorption extending from very low (THz) frequencies over all optical region. In the near-ir and red spectral region, $\mathrm{Re}\,\varepsilon < 0$ is predicted. This property is characteristic of metals and allow for a multitude of nanoplasmonic effects. The metallization cardinally differs from the Zener breakdown in bulk crystals, which is clear from much lower fields required ($\mathcal{E}_m \sim 0.1$ V/Å for $L = 10$ nm). In fact, the metallization is defined not by the field $\mathcal{E}$ per se but by the total potential difference $\Delta U = \mathcal{E}_m L = E_g/e$. Due to the requirement of adiabatic passage to the metallized state, the rise time t_p of the applied electric field *exponentially* increases with $L\sqrt{\mu E_g}/\hbar$. For instance, for the considered case ($L = 10$ nm, $E_g = 9$ eV), $t_p \gtrsim 10$ ns, while for $L = 3$ nm the passage is much faster: $t_p \gtrsim 1$ fs.

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arXiv:1104.1642v1 [cond-mat.mes-hall] 8 Apr 2011

Ultrafast Dynamic Metallization of Dielectric Nanofilms by Strong Single-Cycle Optical Fields

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[2] *Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Straße 1, D-85748 Garching, Germany*
[3] *Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303, USA*
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(Dated: April 12, 2011)

We predict a dynamic metallization effect where an ultrafast (single-cycle) optical pulse with a $\lesssim 1$ V/Å field causes plasmonic metal-like behavior of a dielectric film with a few-nm thickness. This manifests itself in plasmonic oscillations of polarization and a significant population of the conduction band evolving on a ~ 1 fs time scale. These phenomena are due a combination of both adiabatic (reversible) and diabatic (for practical purposes irreversible) pathways.

PACS numbers: 73.20.Mf 42.65.Re 72.20.Ht 77.22.Jp

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Recent Results: Dynamic Metallization

Numerical solution of the density matrix equations (optical Bloch equations) for 5 monolayers (approx. 2 nm) nanofilm with bandgap approx. 5 eV. This simulates diamond or GaN (a wideband semiconductor).

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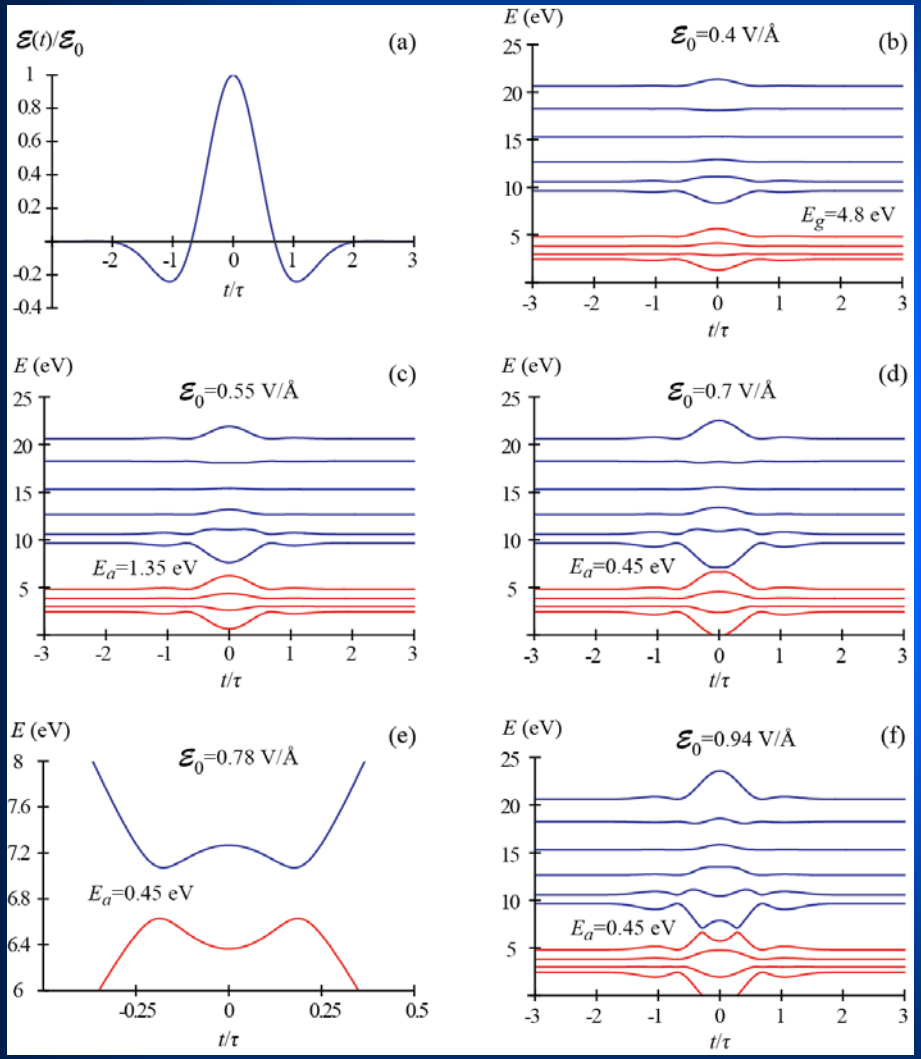
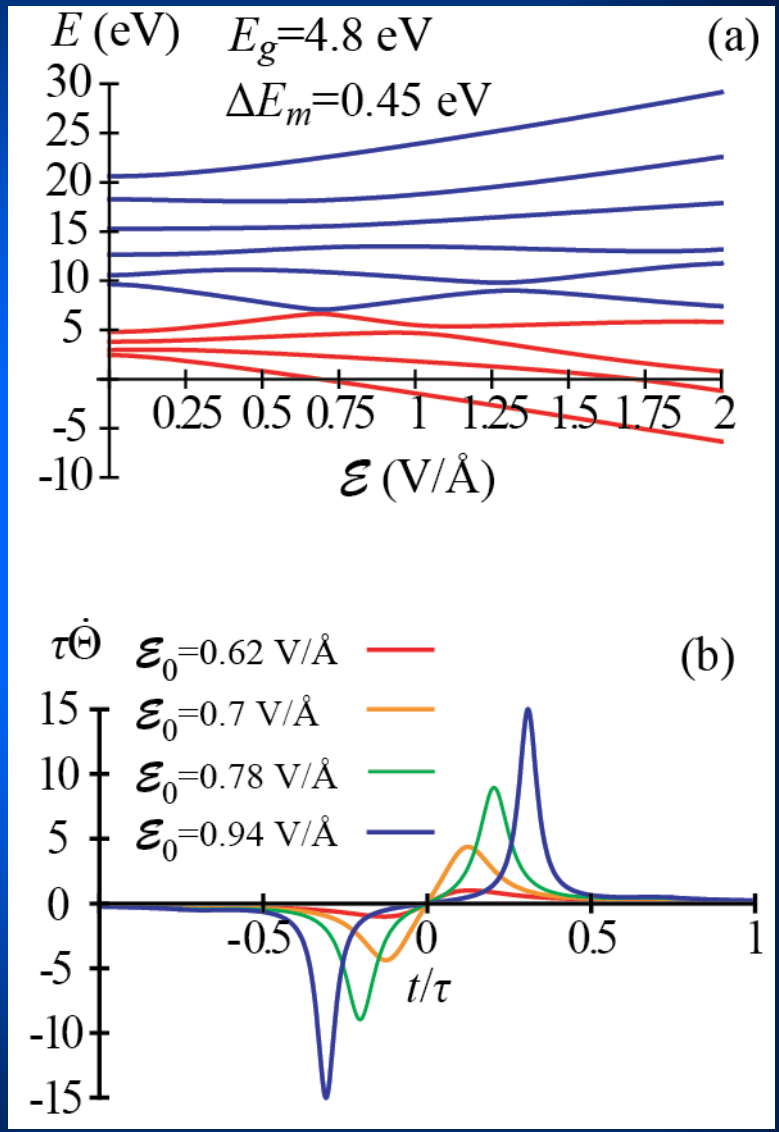
Adiabatic excitation: reproducing previous results





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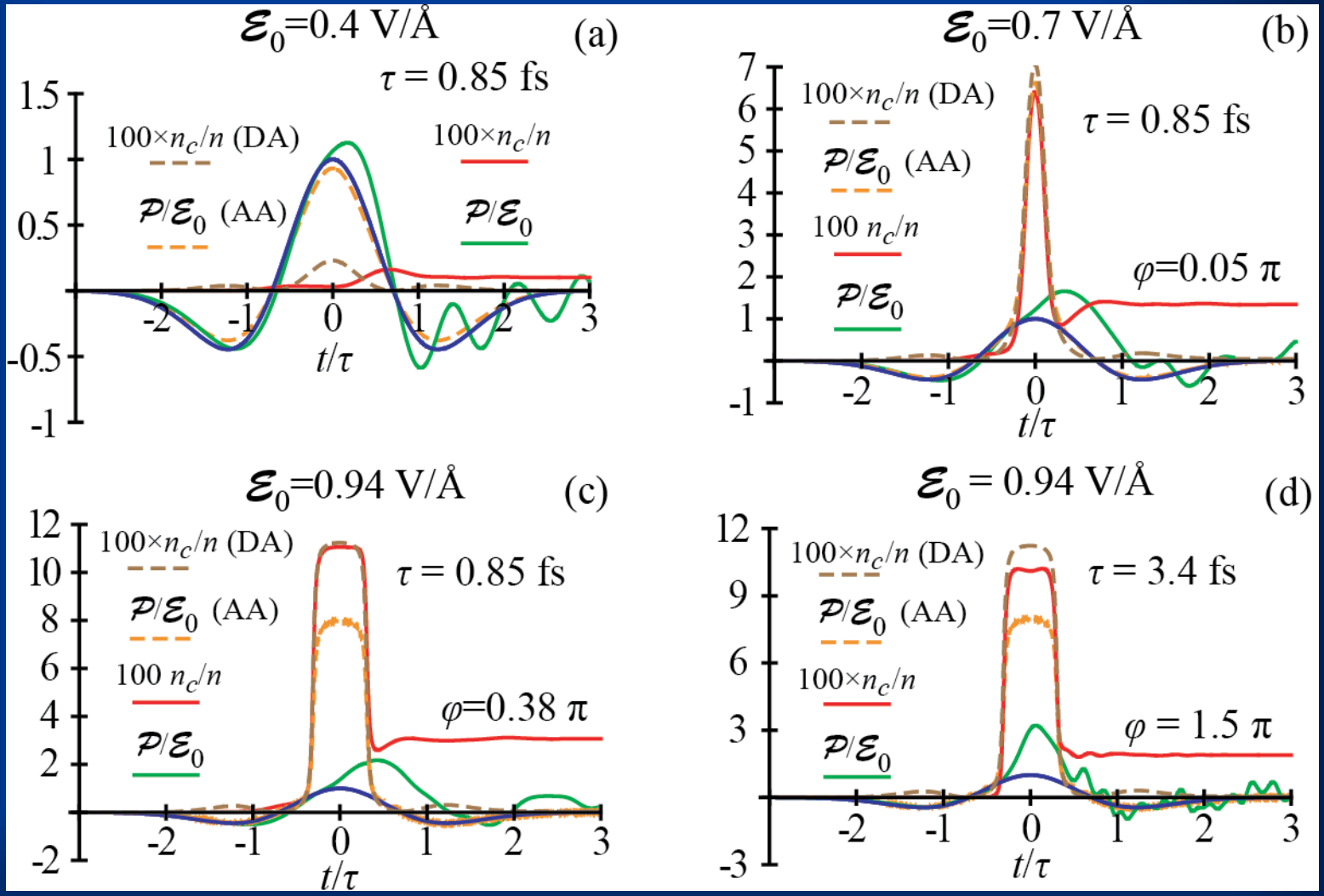
From non-adiabatic to adiabatic excitation



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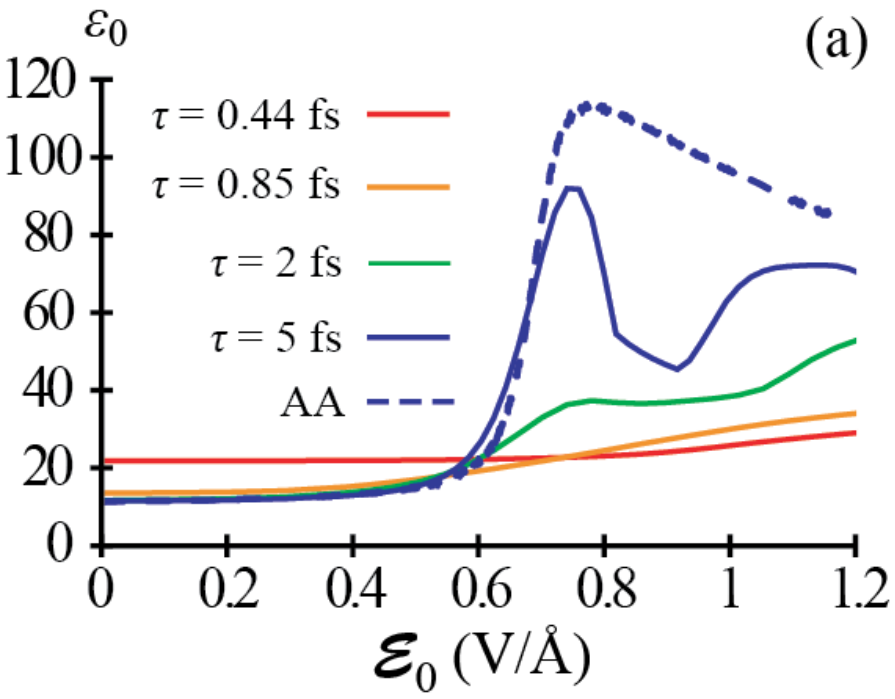
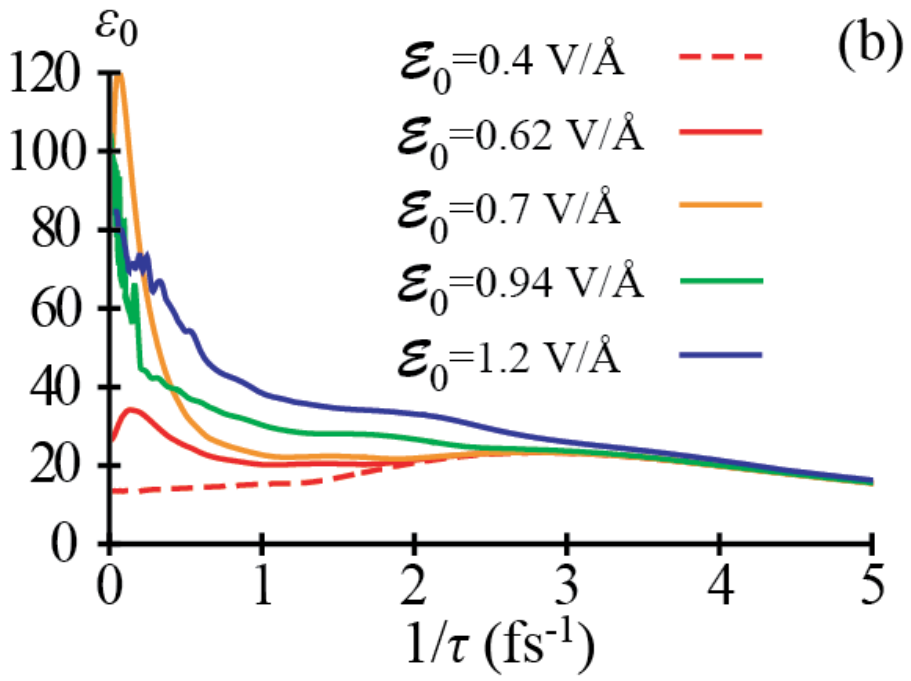
Effective permittivity of a dielectric subjected to a strong-field femtosecond pulse





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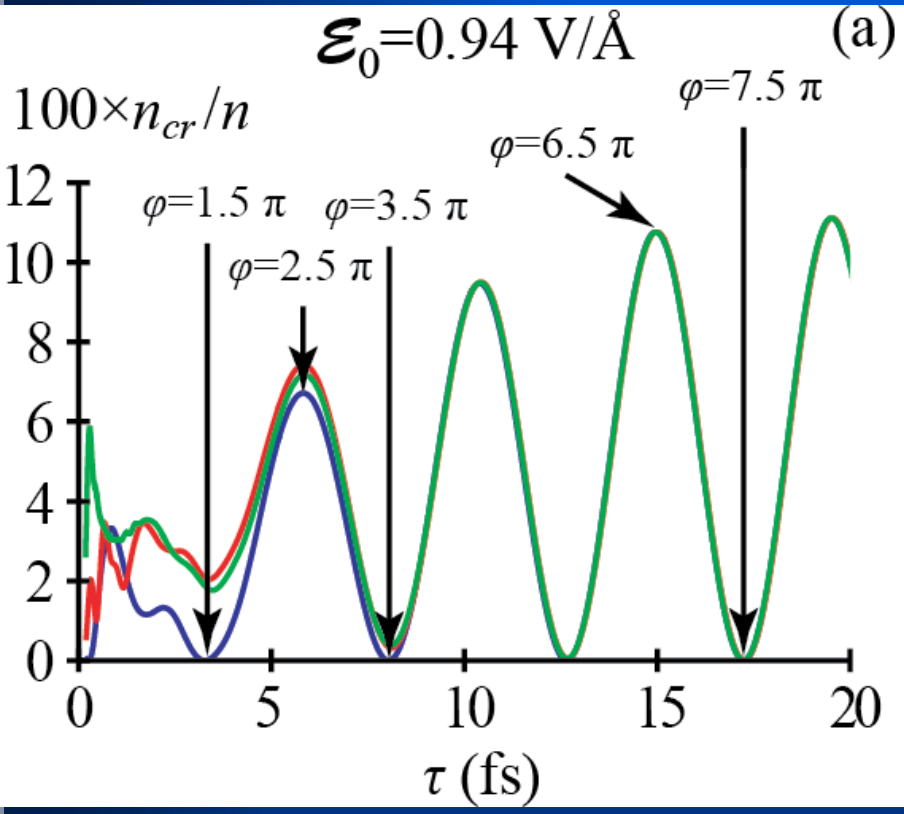
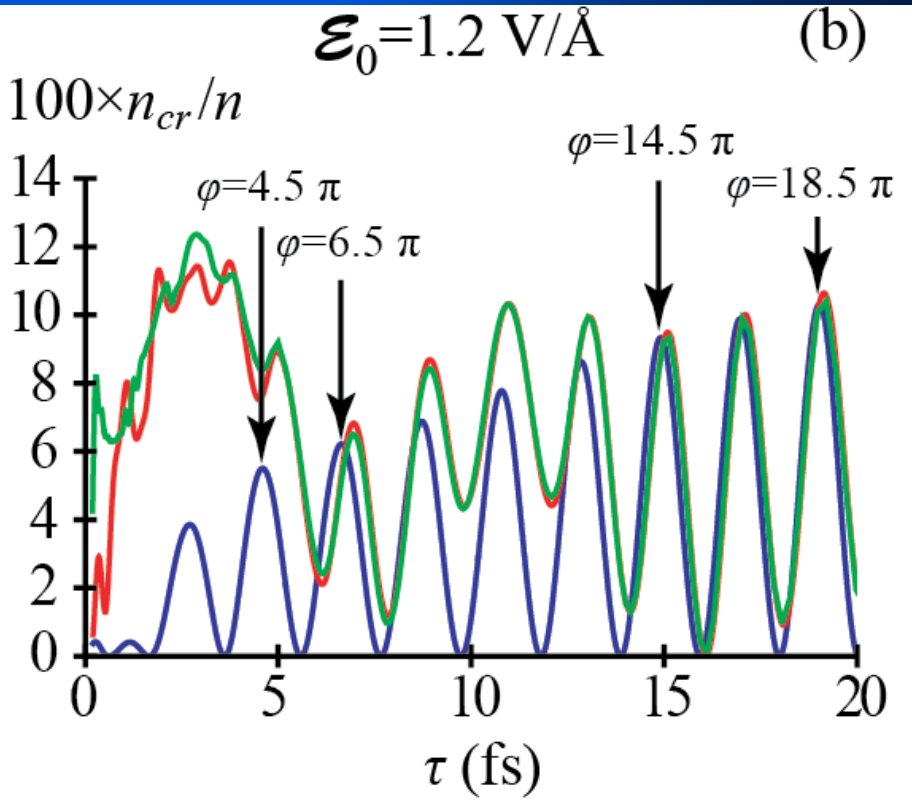
Residual Electron Population of the Conduction Band: Ramsey Fringes as an Effect of the Adiabatic Phase





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Conclusions on Dynamic Metallization

- For long pulses or non-metallizing (weak) fields, the dynamics is adiabatic, following the pulse field.
- The results of adiabatic metallization are fully reproduced
- For short pulses and/or high amplitudes, there is a severe breakdown of adiabaticity: the system is left with a large electron population in the conduction band and a large polarization oscillating after the pulse end


THE END