

laser-induced phenomena: from atomic to condensed matter systems

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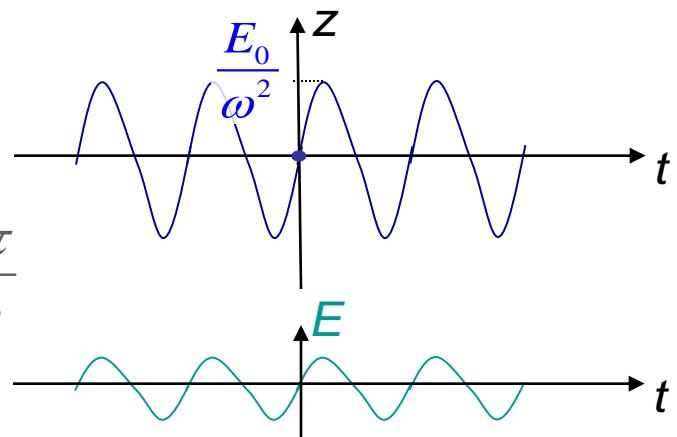
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overview

- charged particle dynamics in an oscillating electric field
- ultrafast change of optical properties of electronic systems
- emission time of photo-ejected electrons

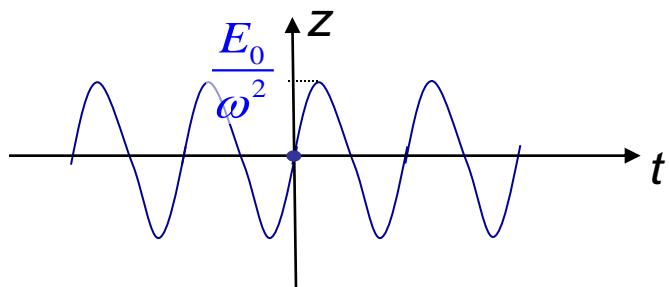
$$z(t=0) = 0, m = 1, \hbar = 1 = c, e = 1, T = \frac{2\pi}{\omega}$$



$$\ddot{z} = -E_0 \sin \omega t \Rightarrow \begin{cases} \dot{z} = \frac{E_0}{\omega} \cos \omega t \\ z = \frac{E_0}{\omega^2} \sin \omega t \end{cases}$$

$$\frac{1}{T} \int_0^T \dot{z}^2 dt = \frac{E_0^2}{4\omega^2} =: U_p$$

ponderomotive potential



$$\ddot{z} = -E_0 \sin \omega t \Rightarrow \begin{cases} \dot{z} = \frac{E_0}{\omega} \cos \omega t \\ z = \frac{E_0}{\omega^2} \sin \omega t \end{cases}$$

$$\frac{1}{T} \int_0^T \dot{z}^2 dt = \frac{E_0^2}{4\omega^2} =: U_p$$

Intensity I

$$I = \frac{\epsilon_0}{2} c E_0^2 \text{ (SI)}, \mapsto \frac{1}{8\pi} c E_0^2 \text{ (CGS)}$$

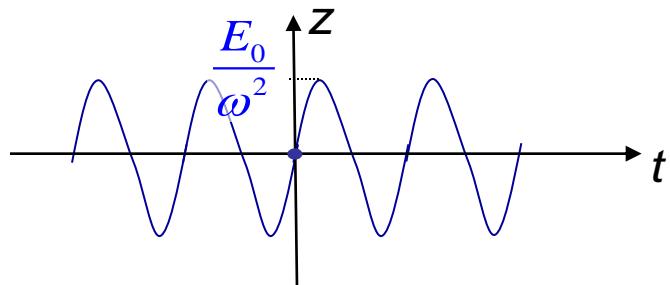
$$I \left[\frac{W}{cm^2} \right] \sim 1.3 \cdot 10^{-3} E_0^2$$

$$E_{a.u.} = \frac{e}{4\pi\epsilon_0 a_0^2} \sim 5 \times 10^9 \frac{V}{cm} = 1 \text{ a.u.}$$

$$1 \times I_{a.u.} \sim 3.5 \times 10^{16} \frac{W}{cm^2}$$

$$E_0^2 \sim \frac{N(\omega) \hbar \omega}{V} \Rightarrow I \sim \frac{N}{V}$$

$$\ddot{z} = -E_0 \sin \omega t \Rightarrow \begin{cases} \dot{z} = \frac{E_0}{\omega} \cos \omega t \\ z = \frac{E_0}{\omega^2} \sin \omega t \end{cases}$$



$$\frac{1}{T} \int_0^T \dot{z}^2 dt = \frac{E_0^2}{4\omega^2} =: U_p$$

$$\frac{E_0^2}{4\omega^2} = U_p \sim 10^2 \text{ keV}$$

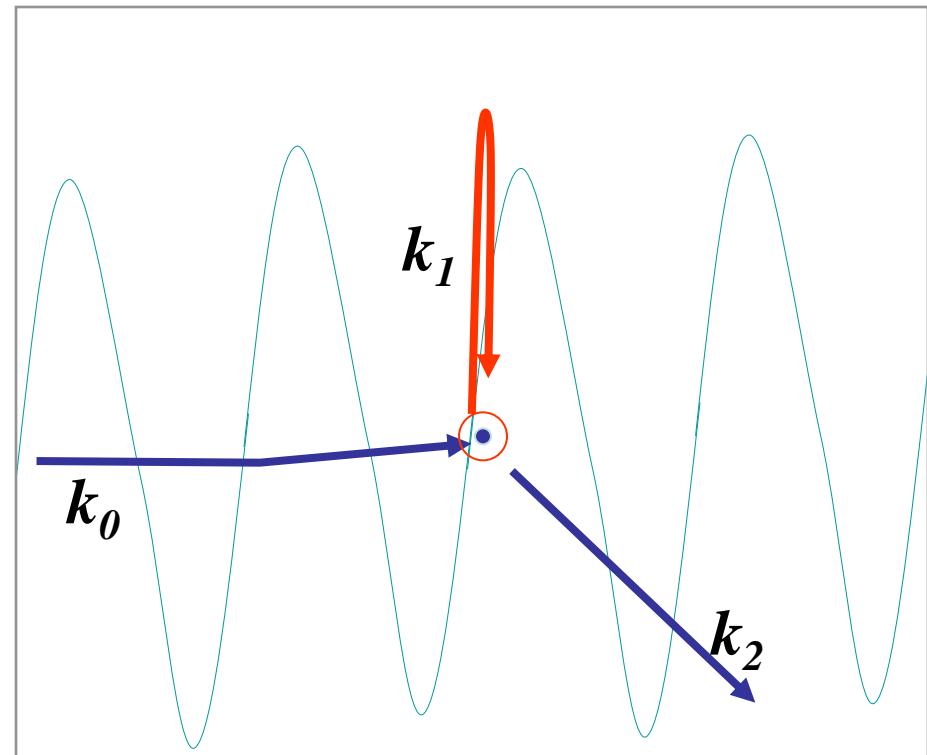
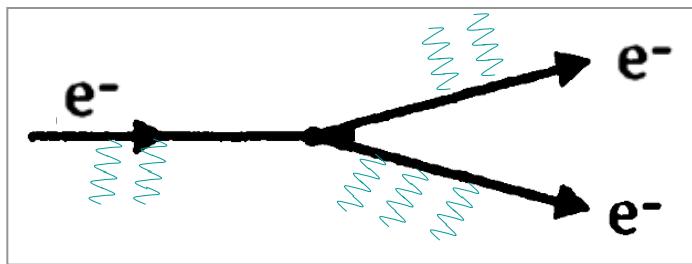
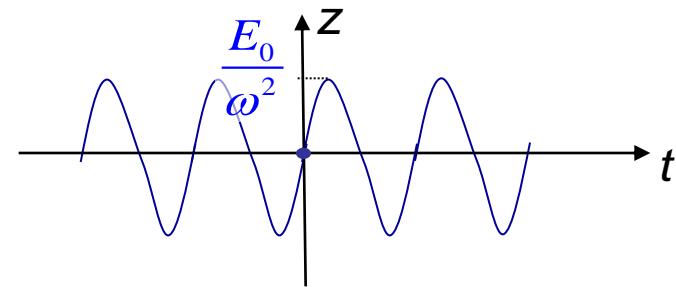
light sources

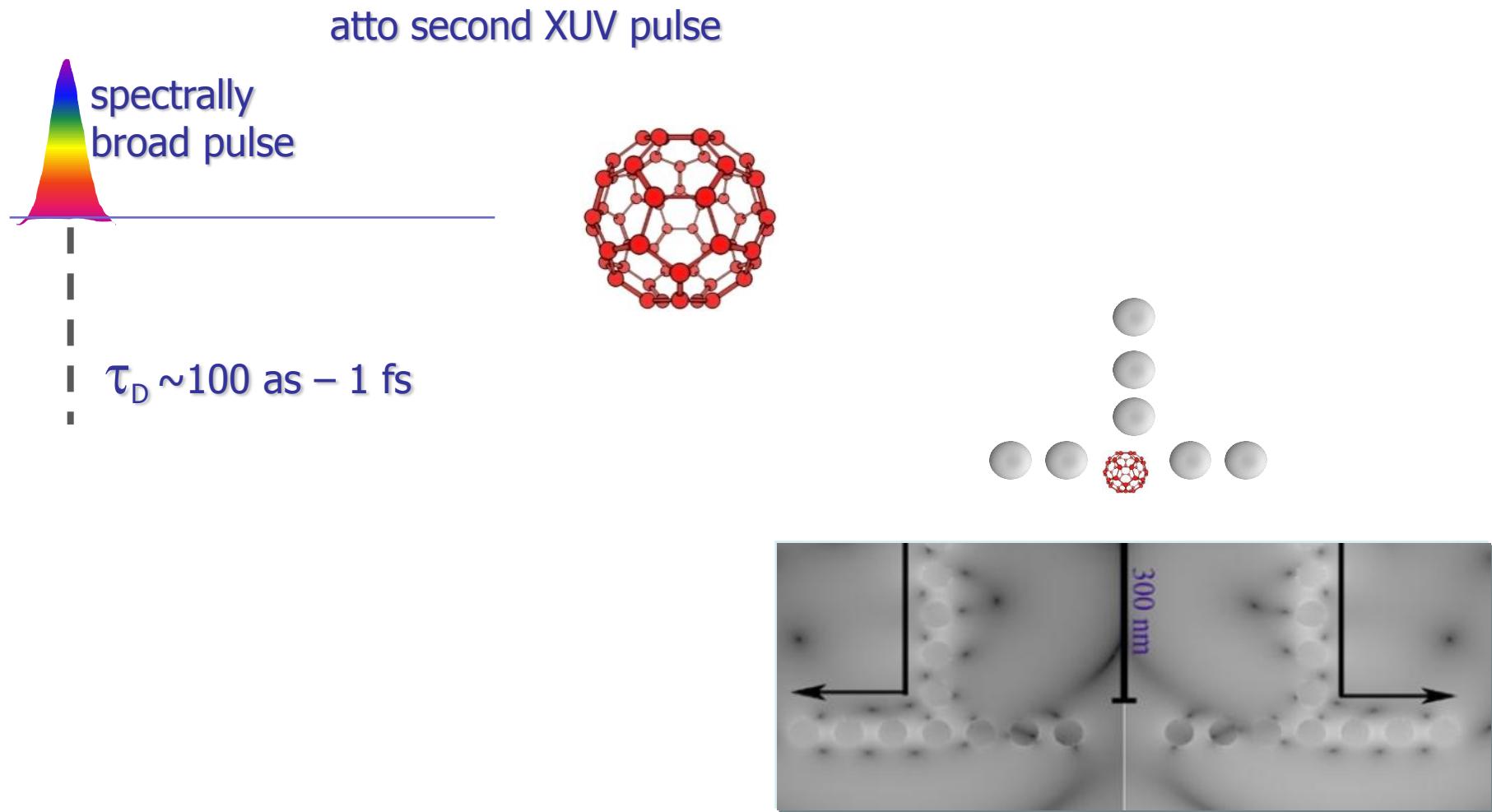
Source	λ [μm]	intensity [$W cm^{-2}$]	Photon density [cm^{-3}]
lamp	0.58	10^{-3}	10^5
sun	0.1-1	10^{-1}	10^7
cw CO_2 -Laser	10	10^{10}	10^{19}
Ti-Saphir-Laser	0.8	10^{18}	10^{26}
Petawatt Livermore	1.06	10^{21}	10^{29}

introduction

classical dynamics

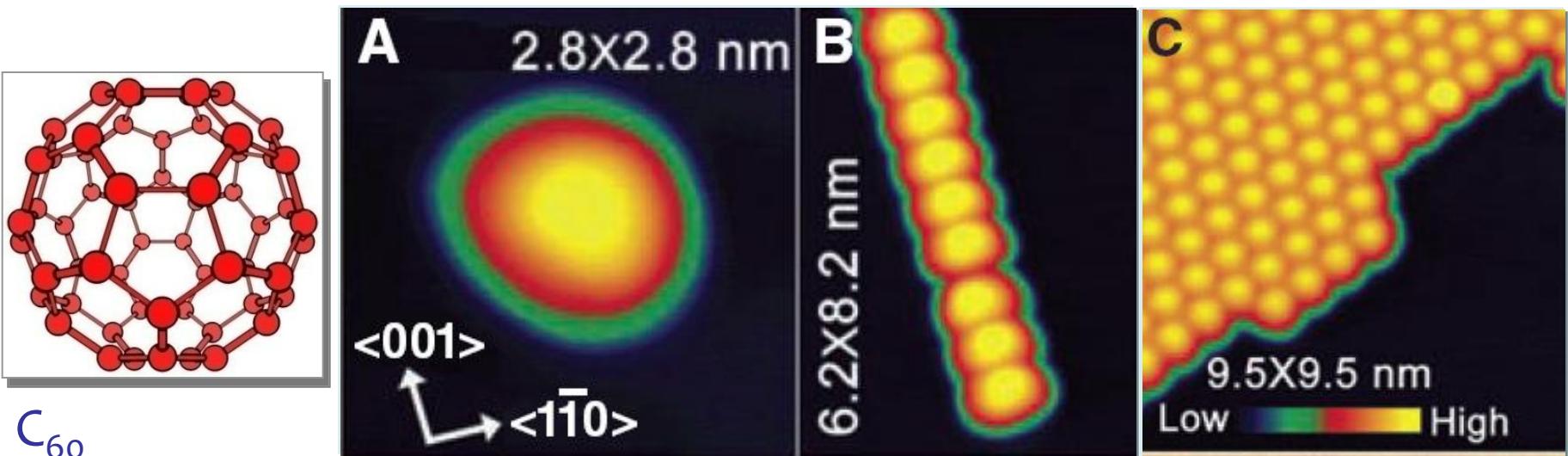
$$m\ddot{z} = -E_0 \sin \omega t \Rightarrow \begin{cases} \dot{z} = \frac{E_0}{m\omega} \cos \omega t \\ z = \frac{E_0}{m\omega^2} \sin \omega t \end{cases}$$





experiments

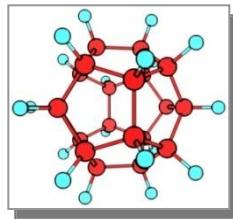
evidence of superatom states



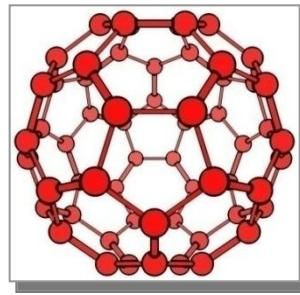
Feng *et al.*, *Science* **320**, 359 (2008); *ACS NANO* **3**, 853 (2009);
Nano Letters **10**, 4830 (2010).

systems

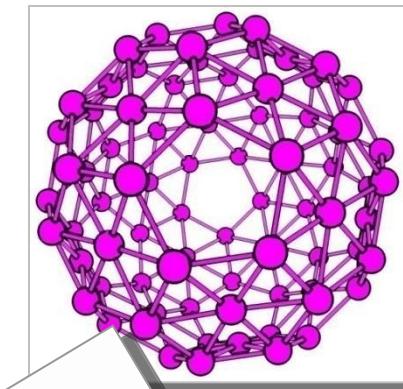
quasi spherical molecules



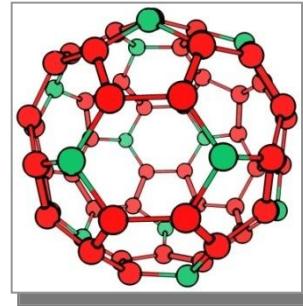
dodecahedrane
 $C_{20}H_{20}$



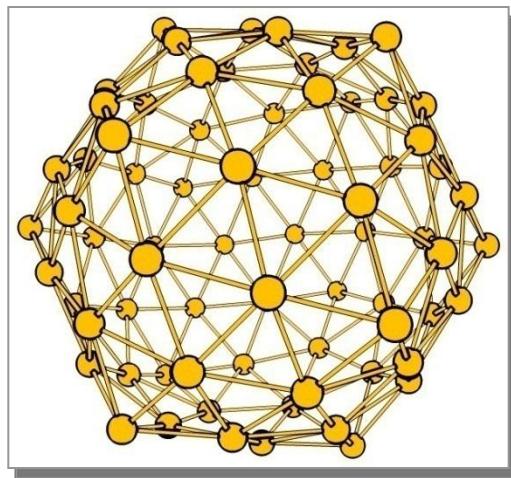
C_{60}



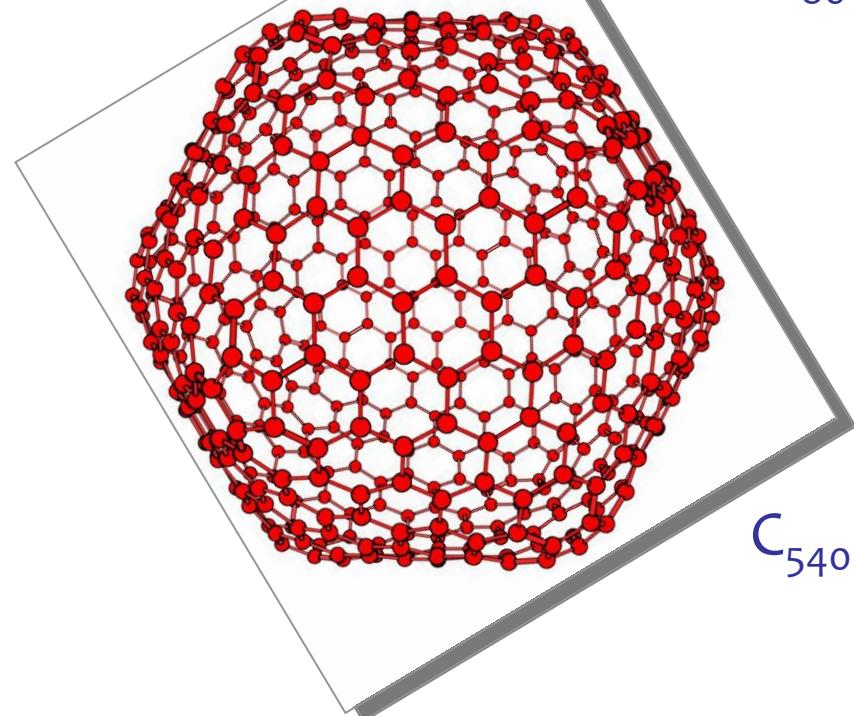
B_{80}



dodeca-aza[60]
fullerene $C_{48}N_{12}$



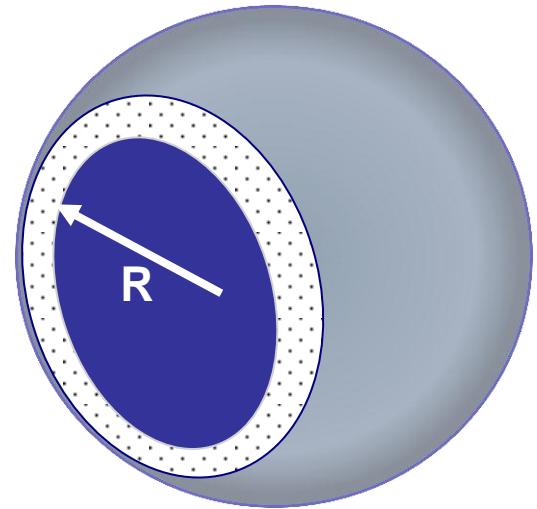
Au_{72}



C_{540}

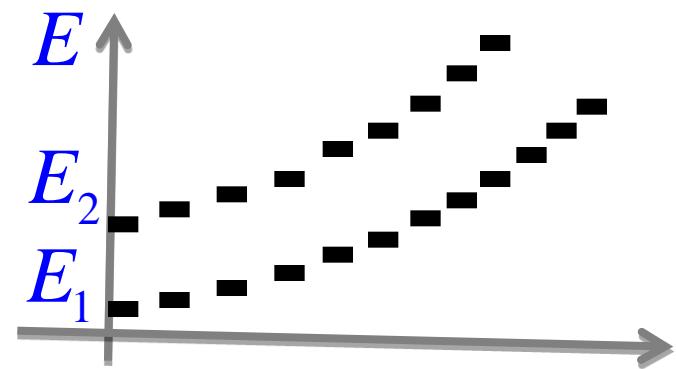
- ***single-particle states*** in quasi spherical systems
- ***inter-particle correlations*** → quasiparticles:
 - decay and energetic shift of single-particle states
 - transient dynamics of quasi particles
- ***collective response*** → coupled plasmons
- ***buildup of collective response*** → transient dynamics

$$H = h_r + \frac{L^2}{2mr^2} + V(r)$$



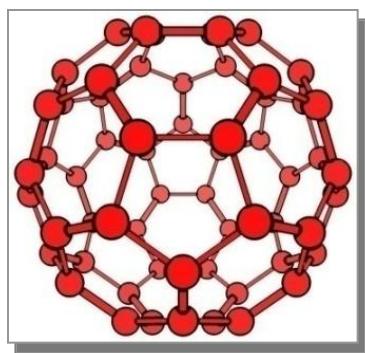
$$E = E_n + \frac{\hbar^2 \ell(\ell+1)}{2mR^2} = E_n + \frac{p_l^2}{2I}$$

$$\psi_E(R, \theta, \phi) = \chi_{E_n}(R) Y_{\ell,m}(\Omega)$$

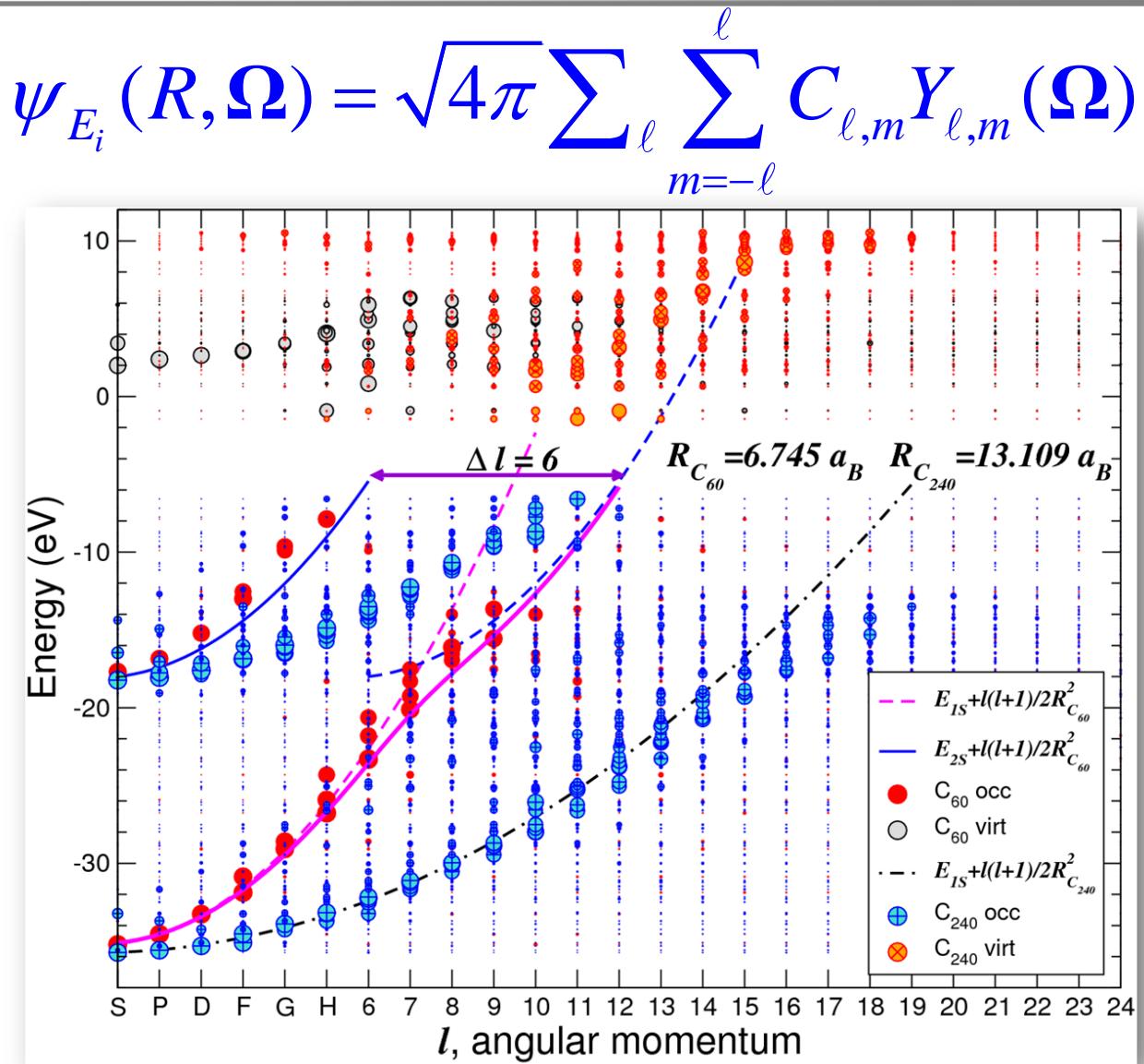


spherical molecules

single particle states

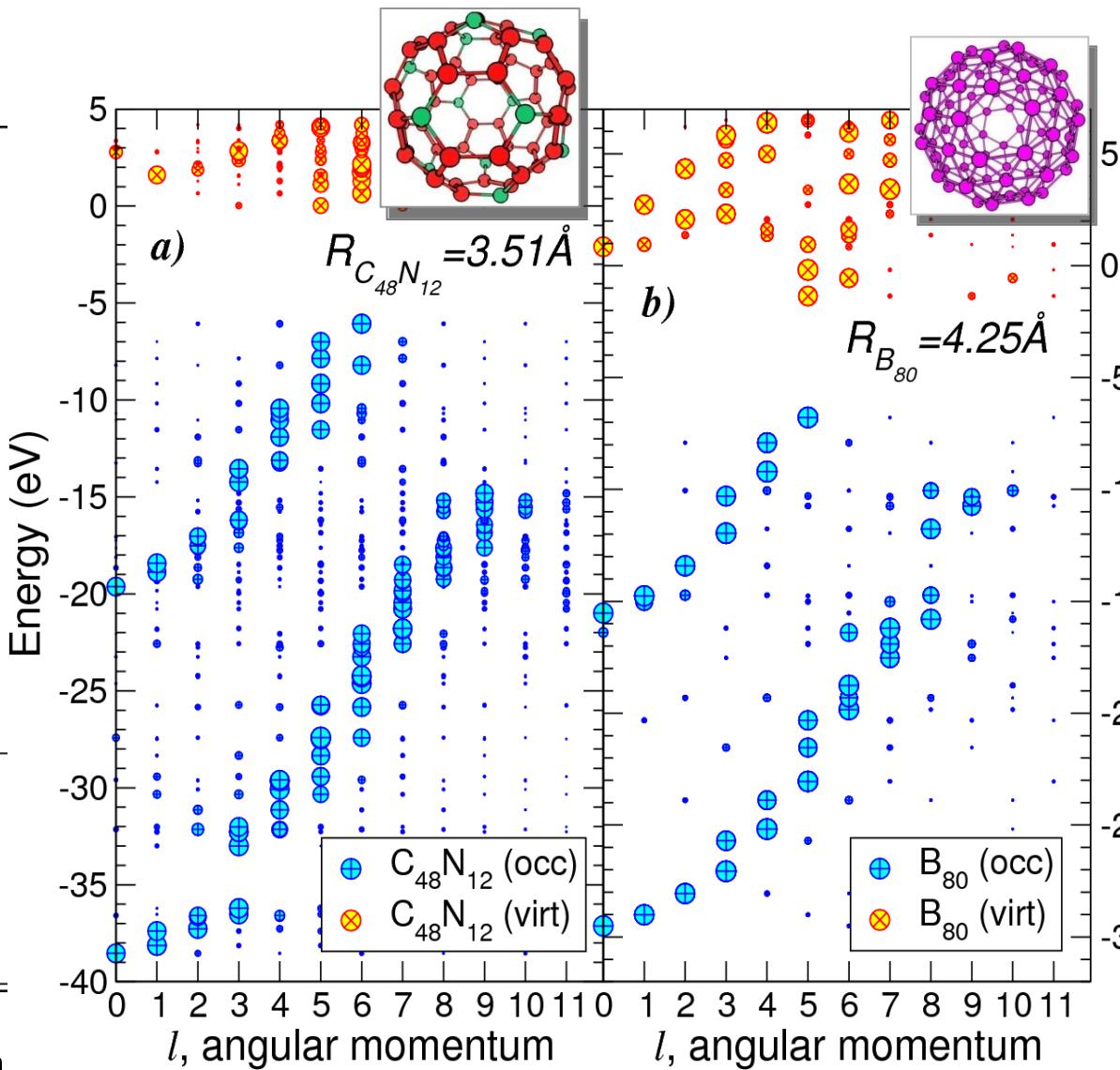
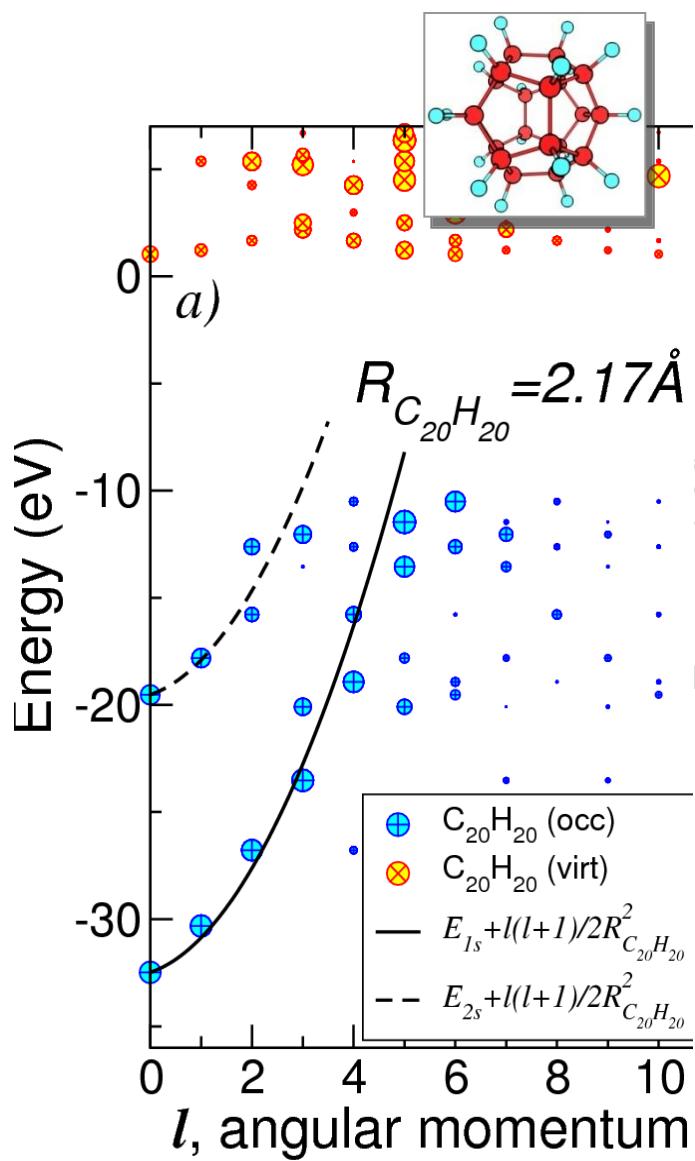


C_{60}



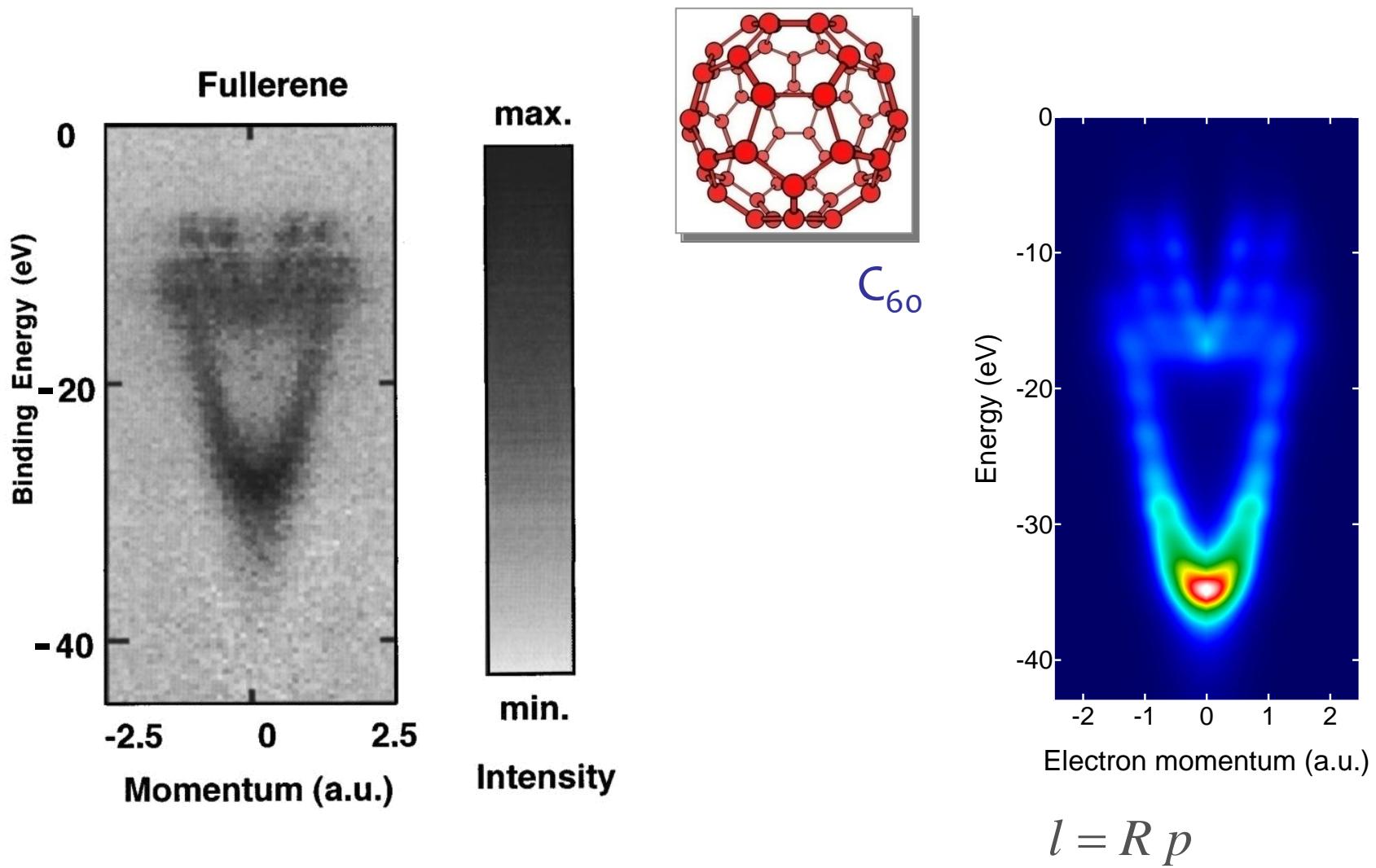
spherical molecules

single particle states



spherical molecules

exp. vs. theory



quasiparticle dynamics

life time

many-body effects on single-particle states

$$G_0(\mathbf{r}_1, \mathbf{r}_2; \omega) = \sum_{\alpha} \frac{\phi_{\alpha}(\mathbf{r}_1)\phi_{\alpha}(\mathbf{r}_2)}{\omega - [\varepsilon_{\alpha} - i \operatorname{sgn}(\varepsilon_{\alpha} - \varepsilon_F) \delta]},$$

single-particle Green's function

$$W = \frac{\nu}{1 - \Pi_0 \nu}$$

screened Coulomb interaction

$$\Sigma(\mathbf{r}_1, \mathbf{r}_2, \omega) = \frac{i}{2\pi} \int d\omega' e^{-i\delta\omega} G_0(\mathbf{r}_1, \mathbf{r}_2, \omega + \omega') W(\mathbf{r}_1, \mathbf{r}_2, \omega') \text{ self energy}$$

$$G = G_0 + G_0 \Sigma G$$

interacting Green's function

Pavlyukh, Berakdar, Hübner, Phys. Rev. Lett. **100**, 116103 (2008)

Pavlyukh, Berakdar, Chem. Phys. **135**, 201103 (2011).

quasiparticle dynamics

life time

quasiparticle states of C₆₀

State	ℓ	E _{HF} (eV)	$\Re E_{GW}$ (eV)	$\Im E_{GW}$ (meV)
HOMO-3	3	-12.61	-11.39	136
HOMO-2	4	-9.93	-9.07	41
HOMO-1	4	-9.69	-9.03	30
HOMO	5	-7.89	-7.48	15
LUMO	5	-0.95	-1.15	-10
LUMO+1	6	0.77	0.22	-29
LUMO+2	5	2.03	1.48	-36
LUMO+3	0	2.10	1.71	-13

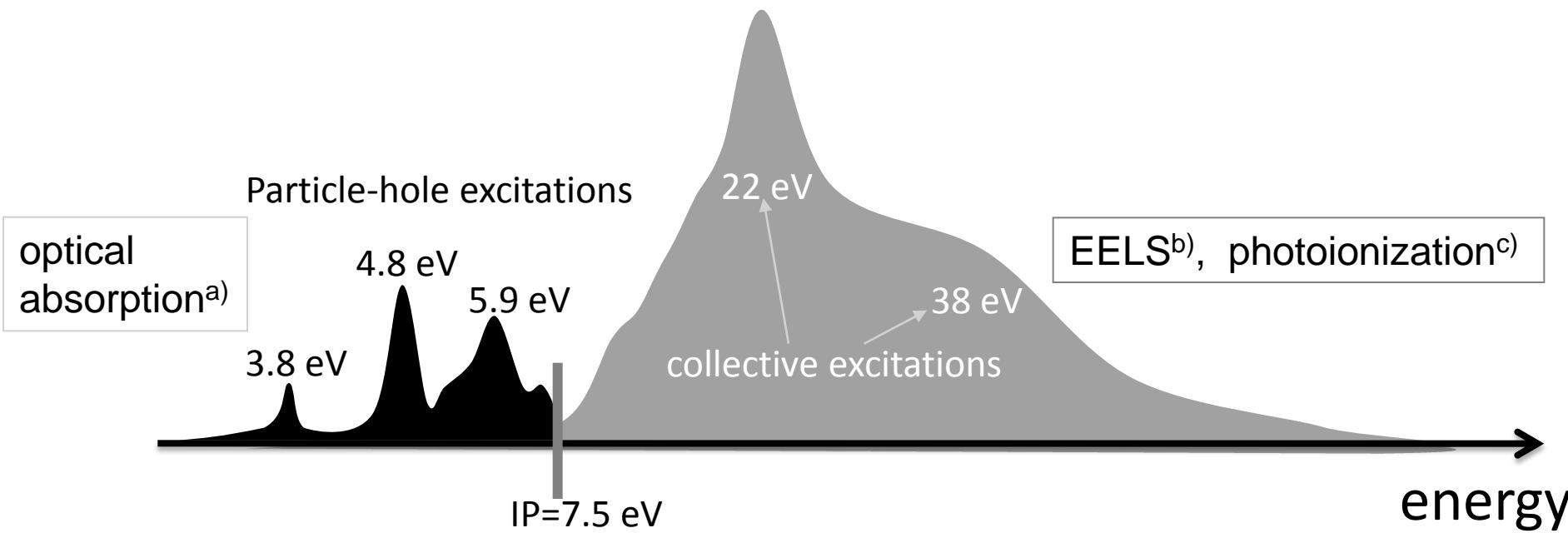
IP_{HF}=7.9 eV, IP_{GW}=7.5 eV, IP_{exp}=7.5 eV exp: Hertel *et al.*, Phys. Rev. Lett. **68**, 784 (1992)

transient dynamics of quasi-particles → Pavlyukh, Berakdar, J. Chem. Phys. 135, 201103 (2011).

excitations

absorption

excitation spectrum of C₆₀



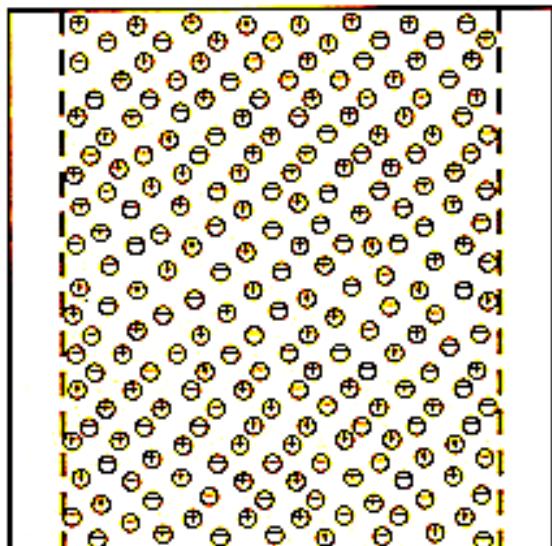
a) • Bauernschmitt *et al.*, J. Am. Chem. Soc. **120**, 5052 (1998)

b) • Hansen *et al.*, Chem. Phys. Lett. **181**, 367 (1991)

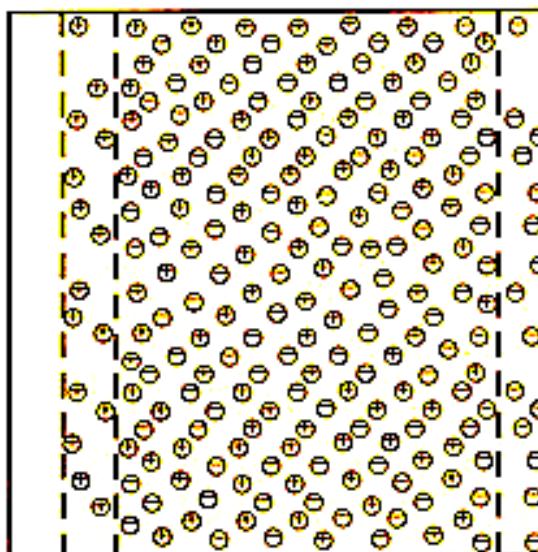
c) • Scully *et al.*, Phys. Rev. Lett. **94**, 065503 (2005)
• Hertel *et al.*, Phys. Rev. Lett. **68**, 784 (1992)

plasma oscillations

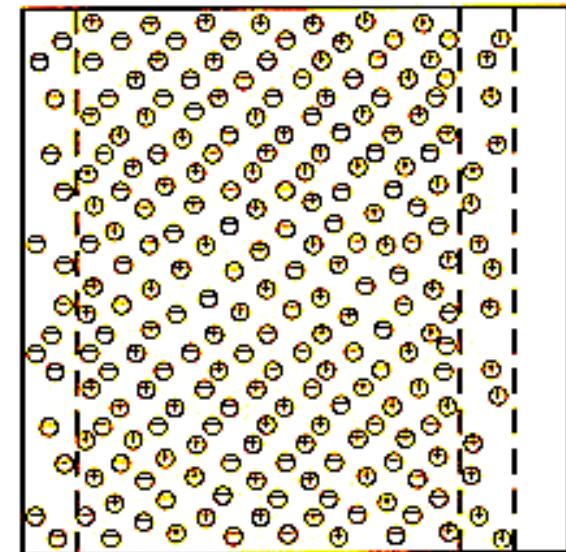
$t = 0$



$t = \frac{\pi}{2} \omega_{pe}^{-1}$



$t = \frac{3\pi}{2} \omega_{pe}^{-1}$



$$\omega_{pe}^2 = \frac{n_e e^2}{m_e \epsilon_0}$$

build up of plasmons

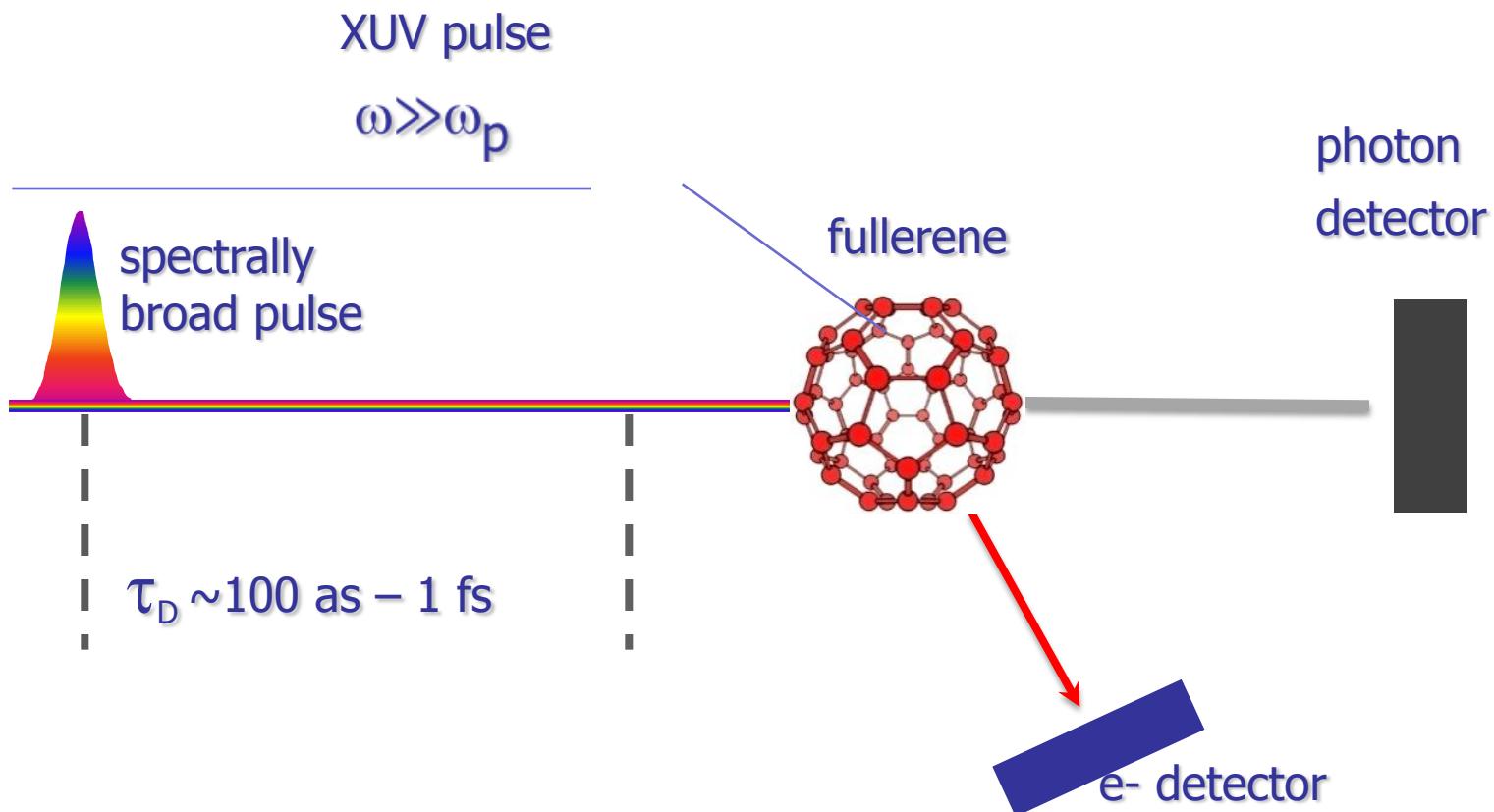
experimental proposal

$$\frac{\partial \hat{\rho}}{\partial t} + \frac{i}{\hbar} [\hat{E} + \hat{V}^{\text{ind}} + \hat{V}^{\text{ext}}, \hat{\rho}] = \frac{\hat{\rho}^{\text{l.e.}} - \hat{\rho}}{\tau}$$

build up of plasmons

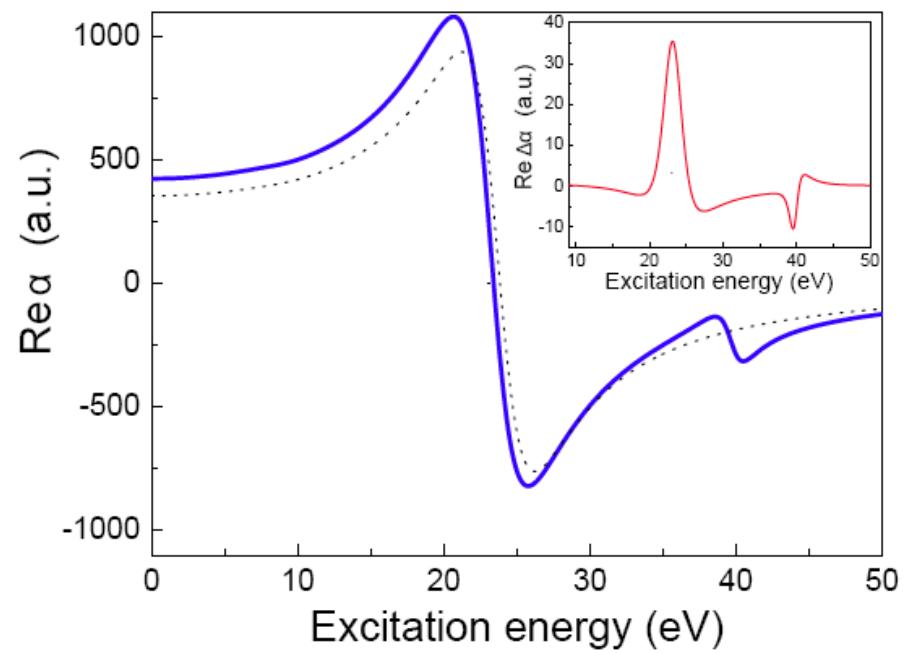
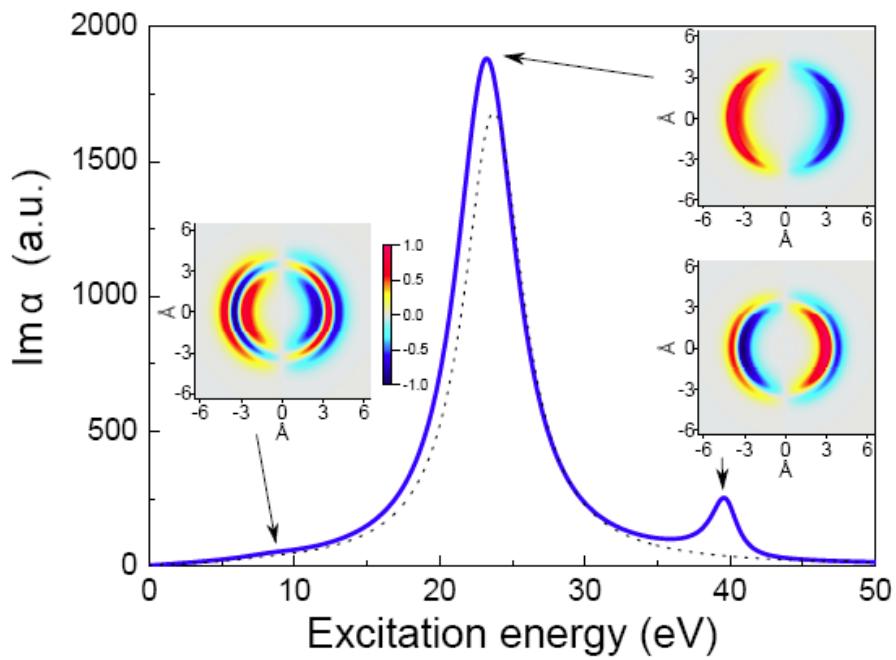
experimental proposal

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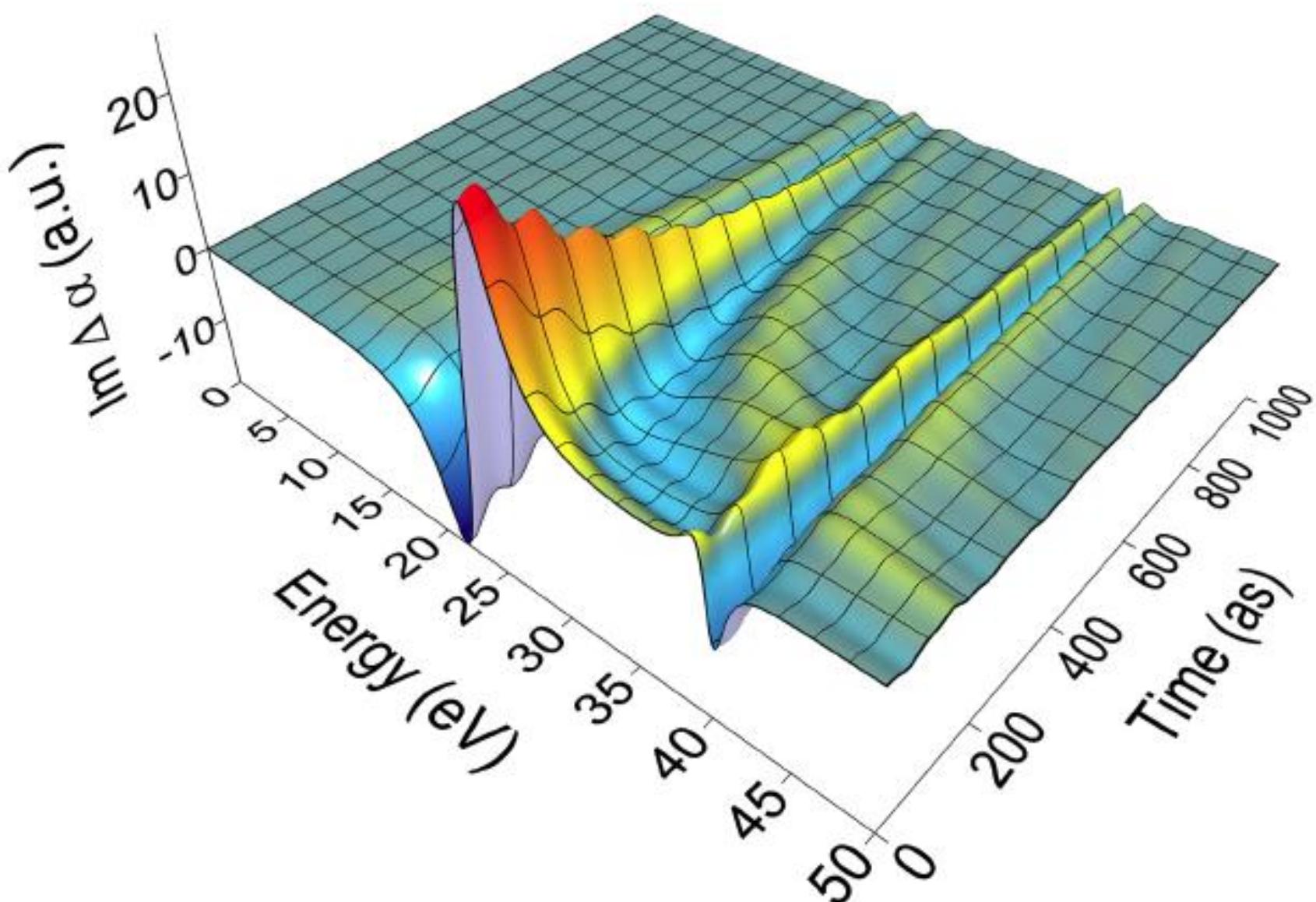


build up of plasmons

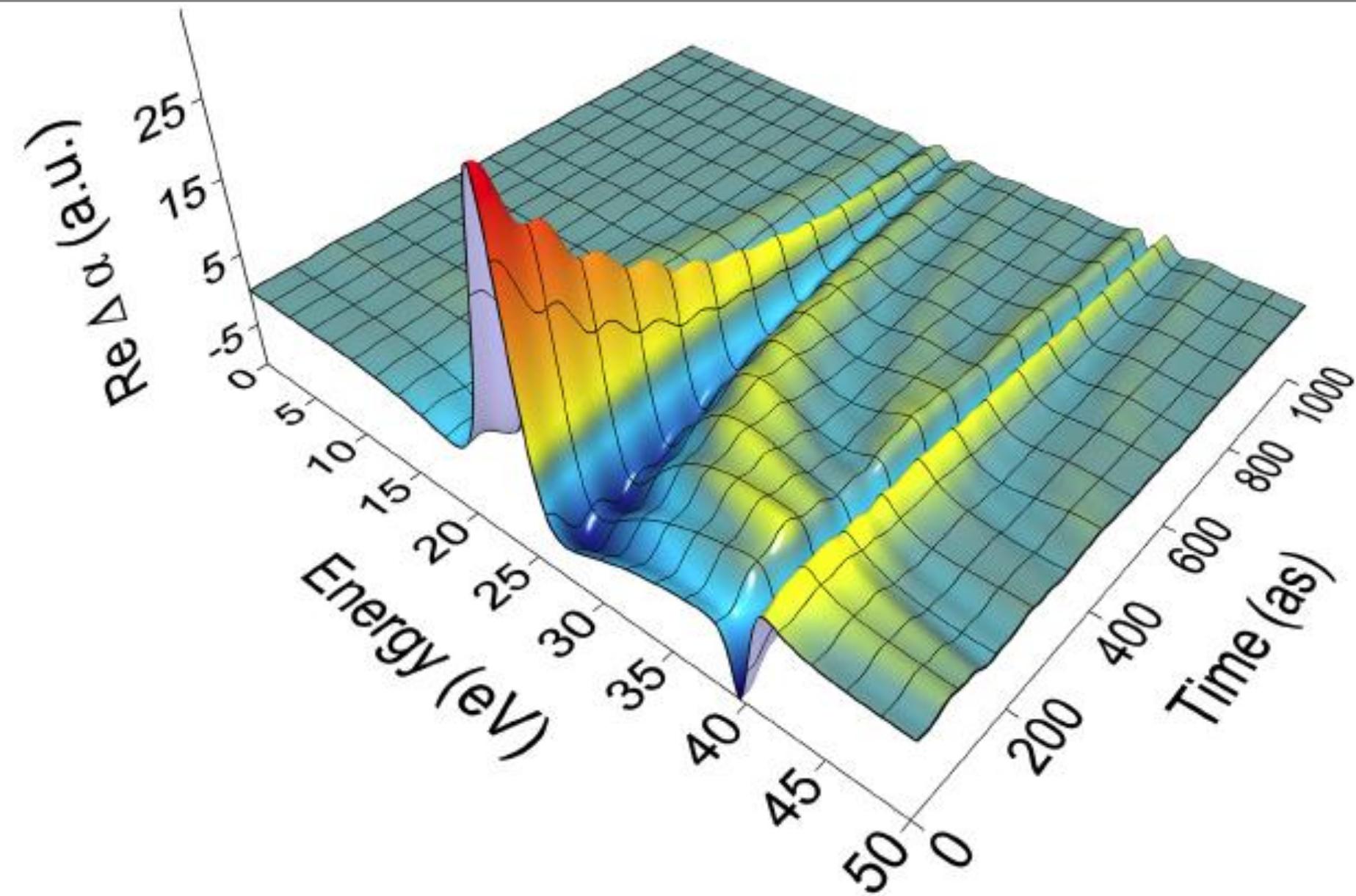
time asymptotic



time-dependent absorption and refraction

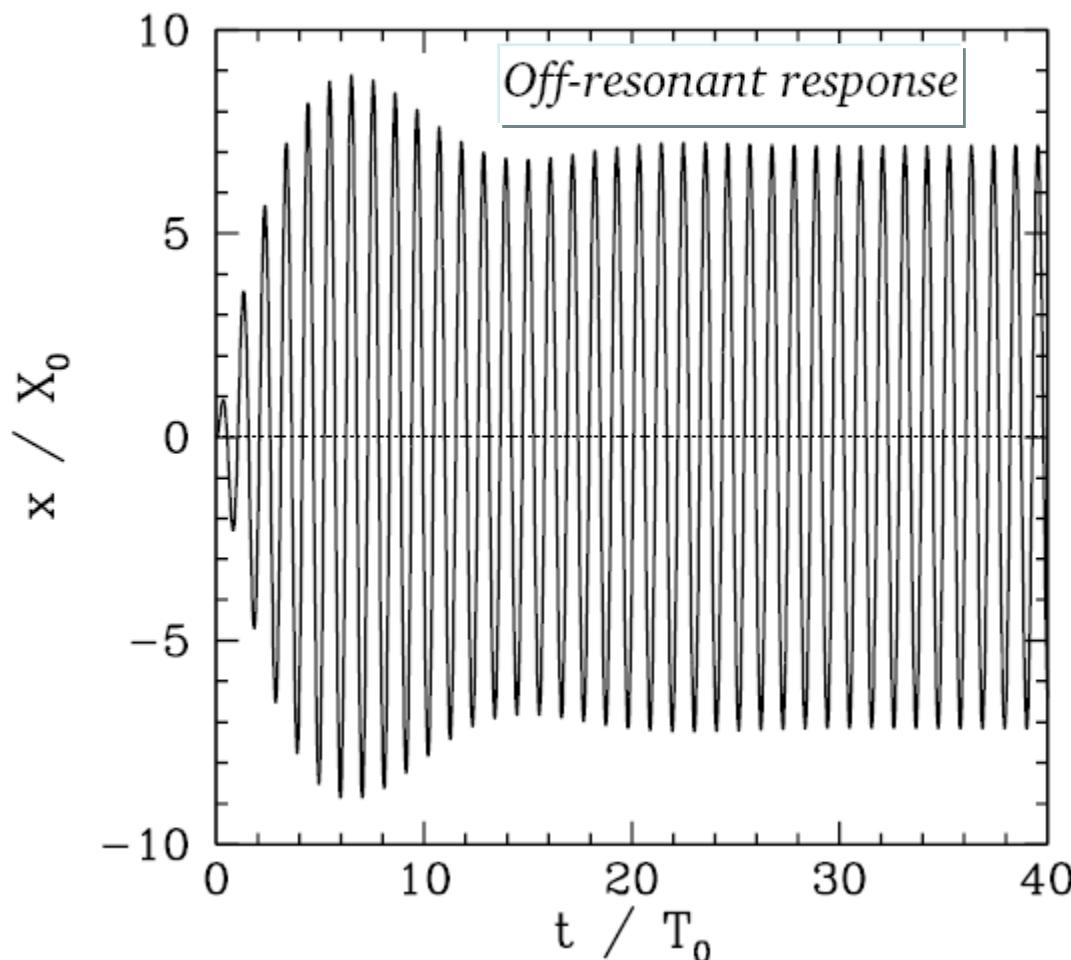


time-dependent absorption and refraction

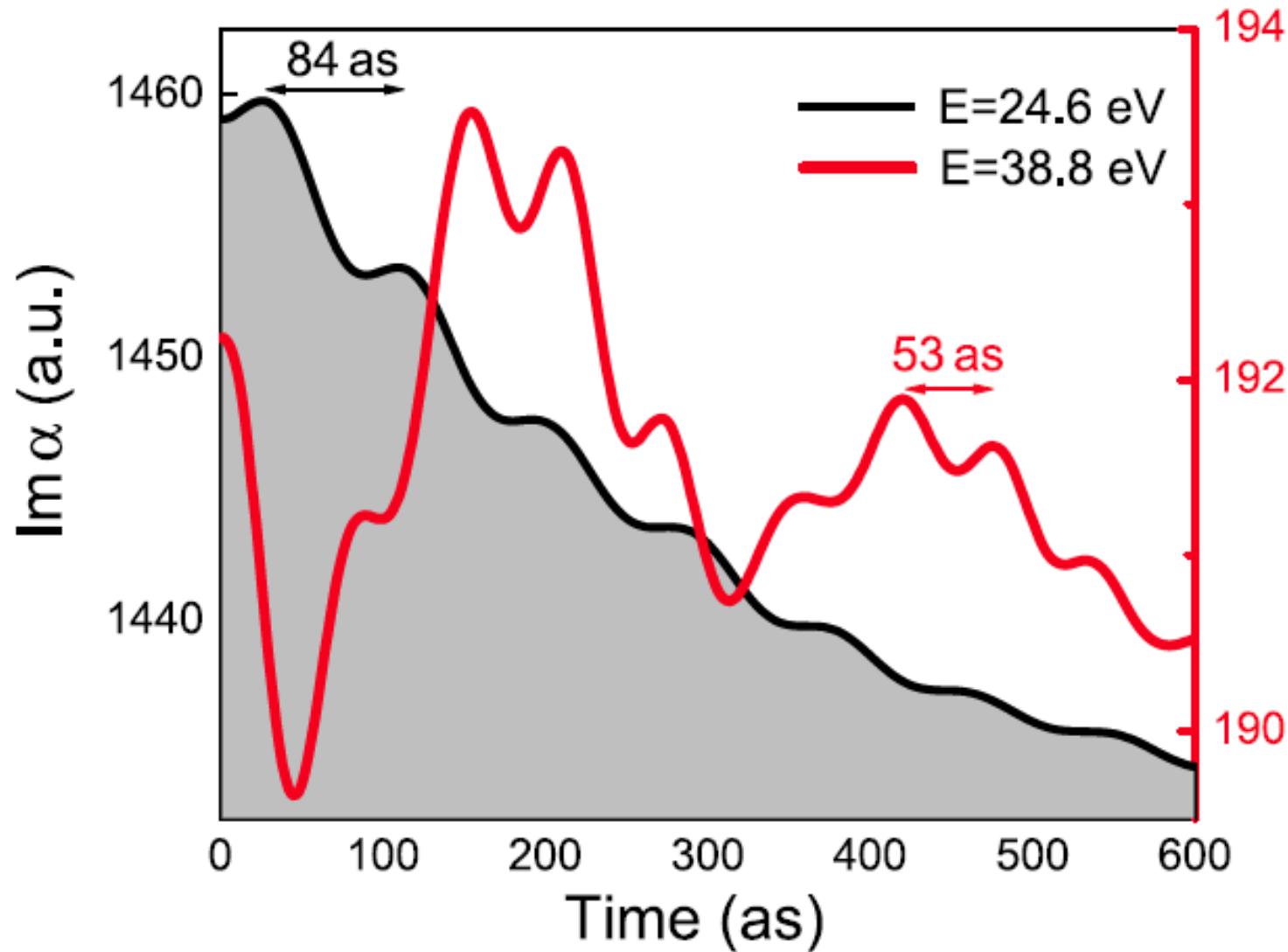


$$\ddot{x} + \gamma \dot{x} + \omega_0^2 x = \omega_0^2 X_0 \cos(\omega t).$$

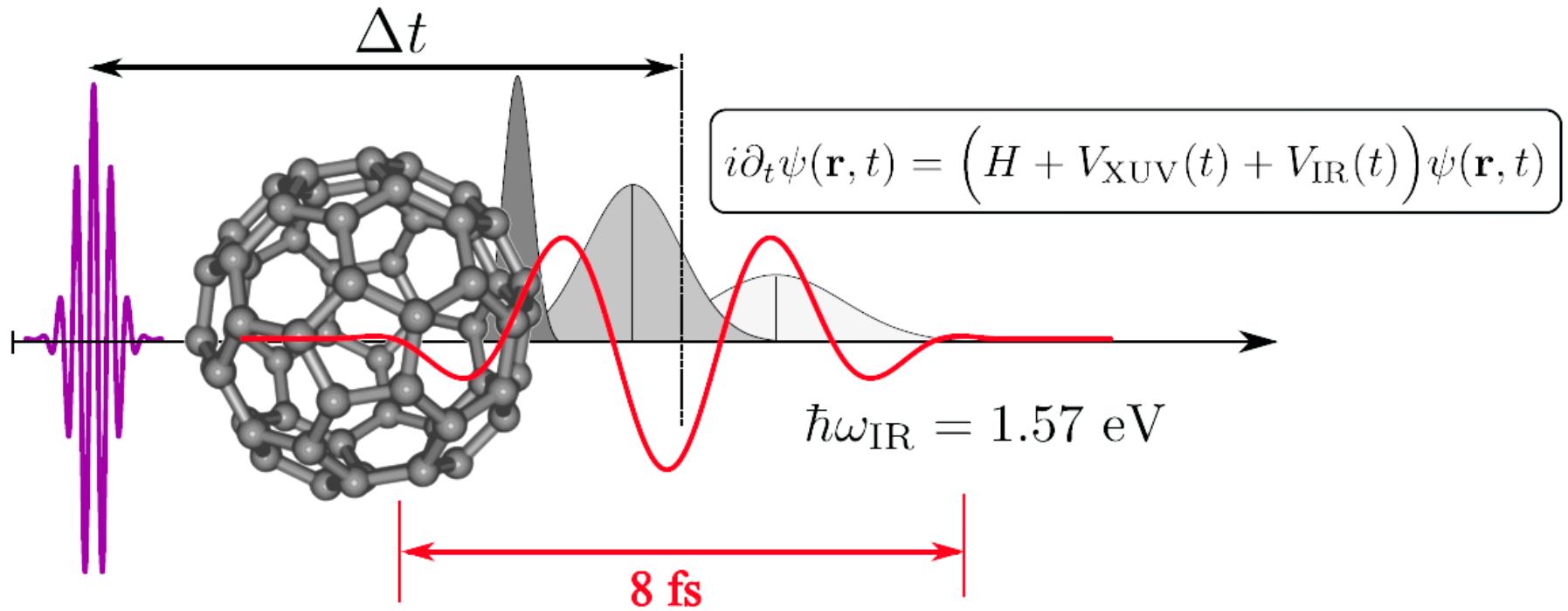
$$x(t) \simeq X_0 \left[\frac{2\omega_0(\omega_0 - \omega)}{4(\omega_0 - \omega)^2 + \gamma^2} \right] \left[\cos(\omega t) - e^{-\gamma t/2} \cos(\omega_0 t) \right] \\ + X_0 \left[\frac{\omega_0 \gamma}{4(\omega_0 - \omega)^2 + \gamma^2} \right] \left[\sin(\omega t) - e^{-\gamma t/2} \sin(\omega_0 t) \right].$$



build up of plasmons



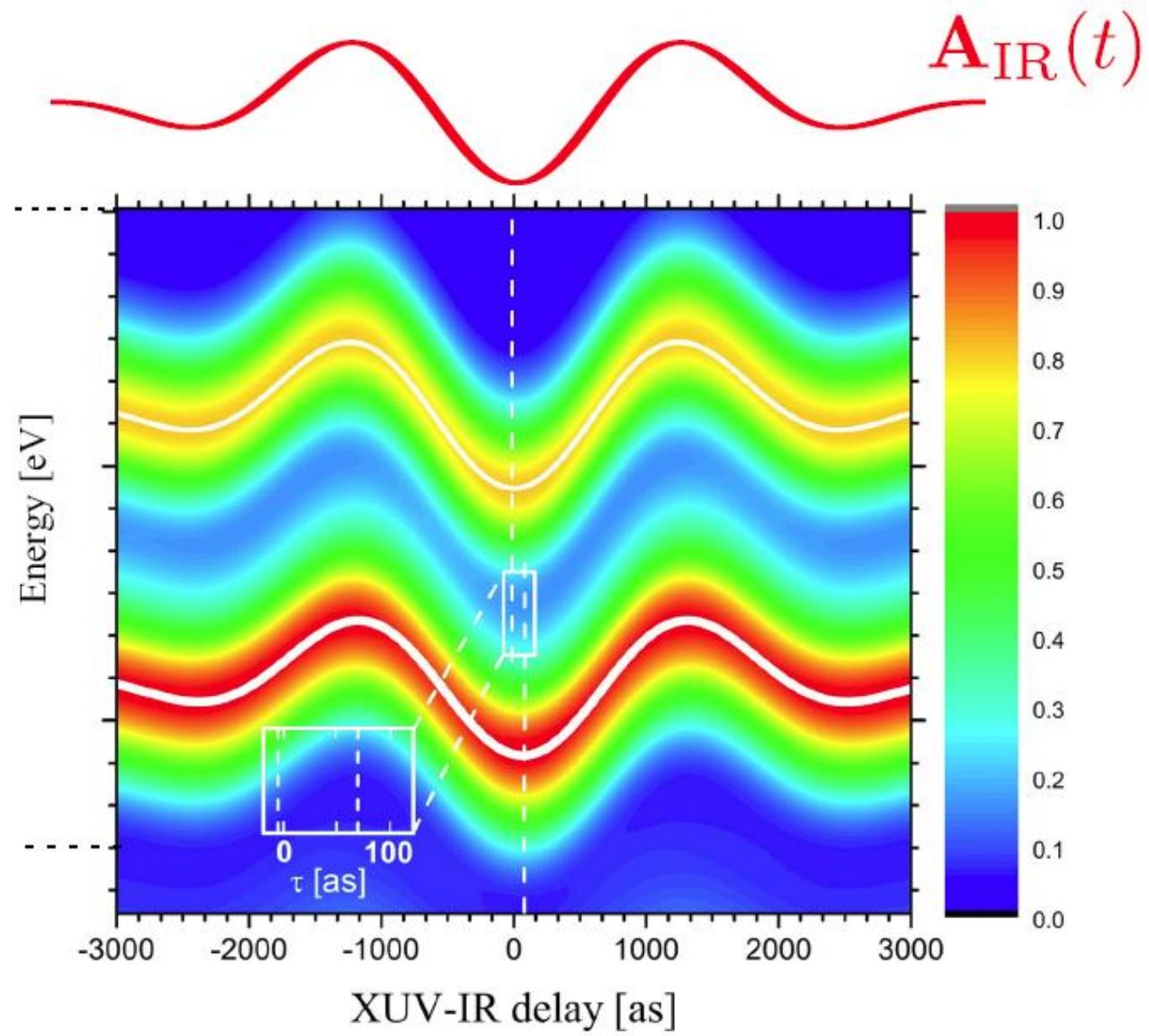
photoionisation in the time domain



streaking histogram → relative time of emission of the photoelectrons

photoionisation in the time domain

streaking histogram → relative time of emission of the photoelectrons from C₆₀



summary

-symmetry, topology → single-particle states

-inter-particle correlations →

1. decay and energetic shift of single-particle states
2. emergence of collective response

- quantum kinetics → build-up of inter-particle correlations
- Ultrafast XUV plasmonics behaviour is theoretically possible