Investigation of carrier dynamics in a laser-excited $Fe_1/(MgO)_3(001)$ heterostructure from real-time TDDFT

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Abstract

We explore the layer-resolved dynamics of optically excited carriers in a metal/insulator heterostructure in the framework of real-time time-dependent DFT.

In $Fe_1/(MgO)_3(001)$, we observe a marked anisotropy in the response to in- and out-of-plane polarized light, which changes its character for different frequencies [1,2]. The Fe layer is efficiently addressed by photon energies below the bulk MgO band gap and in-plane polarization, whereas the MgO part is excited for frequencies around and above the MgO band gap and out-of-plane polarization [1,2].

Moreover, we identified a concerted excitation mechanism which involves two simultaneous excitations via interface states [2]: From Fe-states in the vicinity of the Fermi level to the conduction band of MgO and, likewise, from the valence band of the insulator to metal states just above the Fermi level.

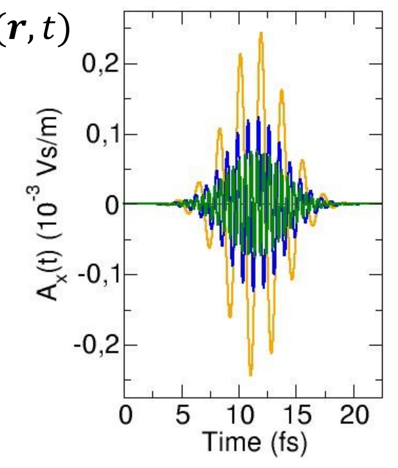
Computational details

- Evaluation of time-dependent Kohn-Sham equations with the all-electron full-potential linearised augmented-plane wave code ELK [3]

$$i\frac{\partial \psi_j(\mathbf{r},t)}{\partial t} = \left(\frac{1}{2}\left(-i\nabla + \frac{1}{c}\mathbf{A}_{ext}(t)\right)^2 + v_s(\mathbf{r},t) + \frac{1}{2c}\sigma\cdot\mathbf{B}_s(\mathbf{r},t) + \frac{1}{4c^2}\sigma\cdot\left(\nabla v_s(\mathbf{r},t)\times -i\nabla\right)\right)\psi_j(\mathbf{r},t)$$

$\mathbf{A}_{ext}(t)$ is the vector potential of applied laser field and the final term is the spin-orbit coupling (SOC) term [4,5]

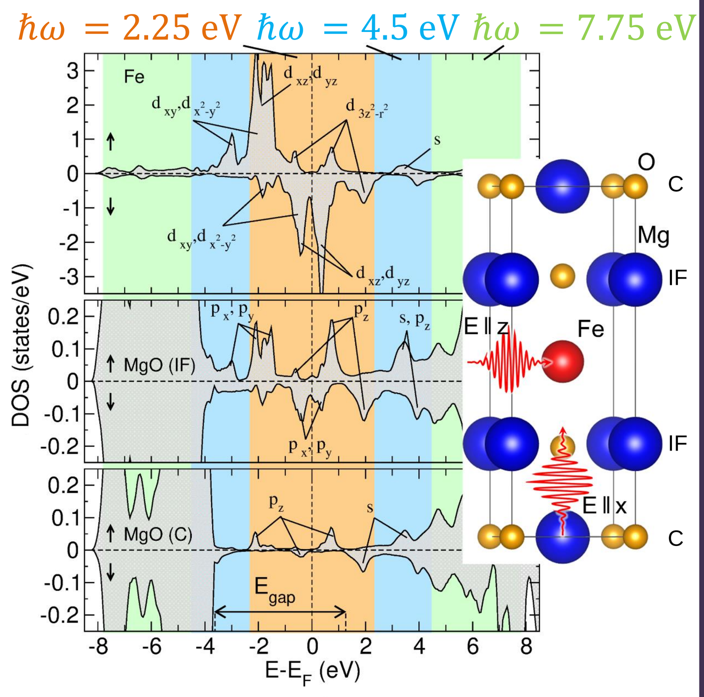
- Time propagation of the electronic subsystem only, ions are fixed
- Adiabatic LSDA: PW92 [6]
- Plane wave cut off parameter: RK_{max}=7
- K-points 8×8×3
- Time step length: Δt = 2.4 as



Vector potential of laser pulses

Layer-resolved density of states

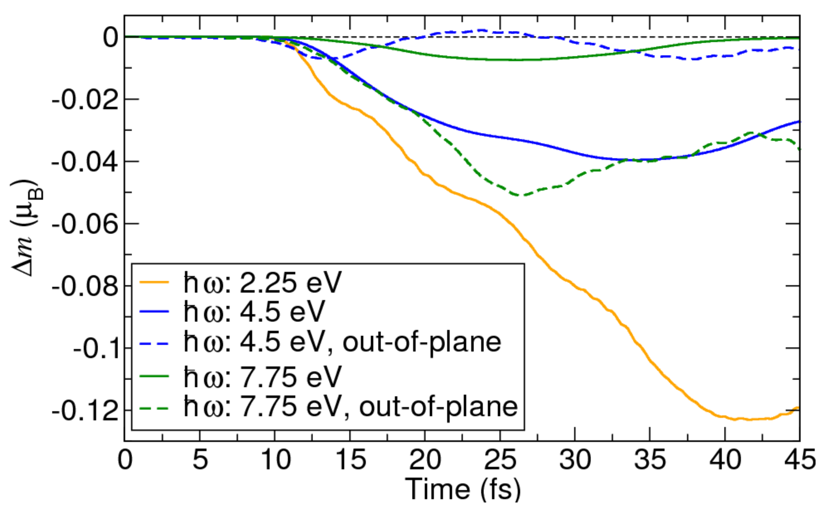
- Correspondence of peaks in the Fe and MgO(IF) partial DOS between -3 and +3 eV: Considerable hybridization between 3*d* states of Fe and 2*p* states of apical oxygen.
- Exponential damping of IF states towards central layer



Colored areas: Expected energy range of direct excitations.

Magnetic moment

Magnetization change in the $Fe_1/(MgO)_3(001)$ heterostructure for different frequencies & directions of polarization:

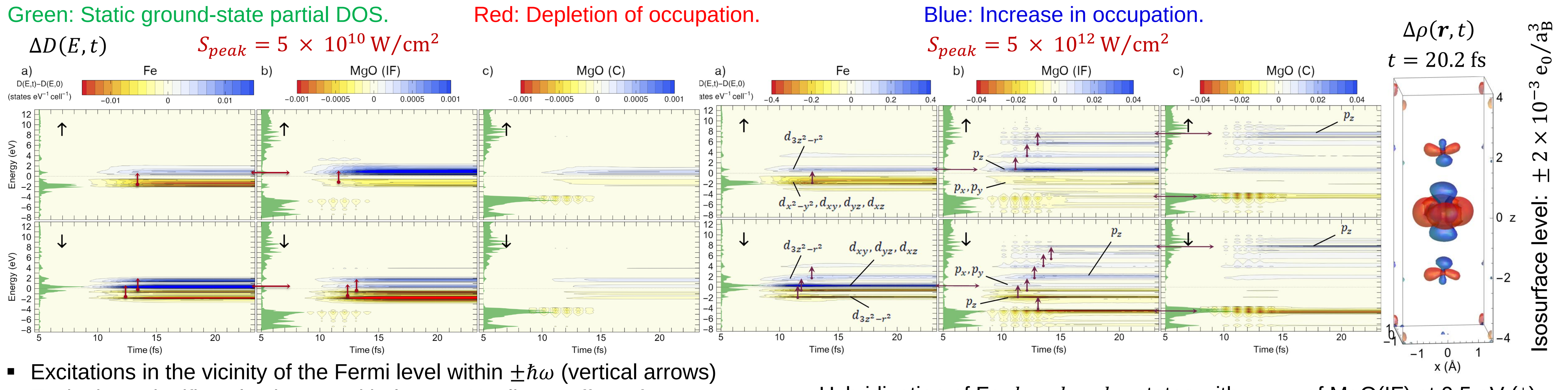


- Moderate time dependent change of magnetization in the Fe-layer induced by SOC
- Strongest changes for in-plane polarization and low frequencies

Dependence of the excitation pattern on laser intensity $\hbar\omega = 2.25$ eV, $\vec{E} \parallel x$

Layer and time-resolved changes in the ΔTDDOS, $\Delta D(E,t) = D(E,t) - D(E,0)$ and change in the charge distribution relative to the initial state, $\Delta\rho(\mathbf{r},t)$:

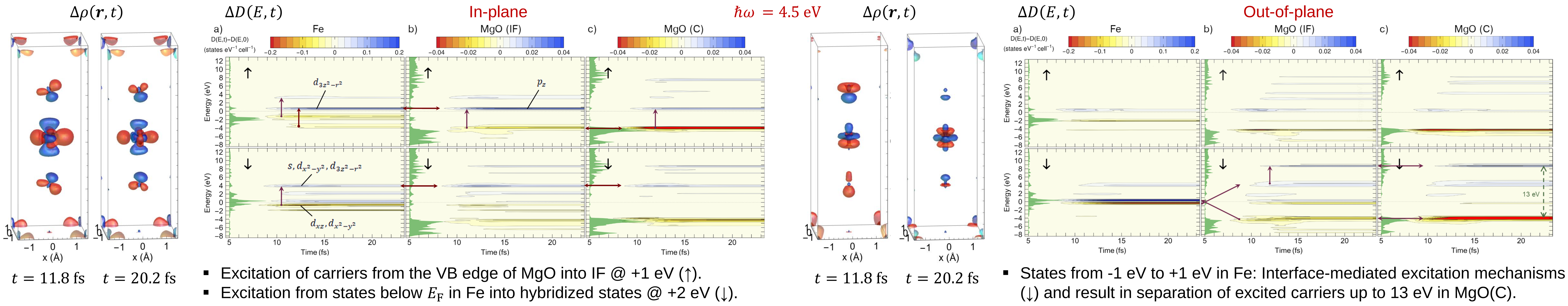
Green: Static ground-state partial DOS. Red: Depletion of occupation. Blue: Increase in occupation.



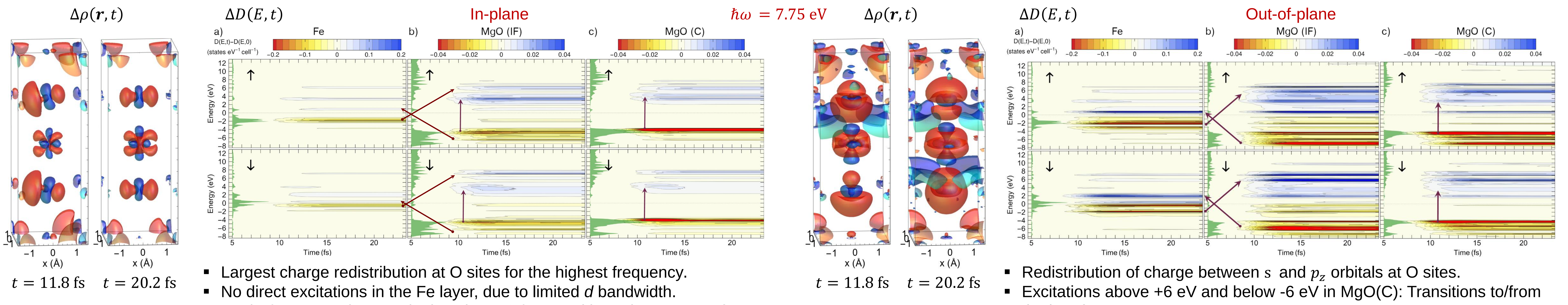
- Excitations in the vicinity of the Fermi level within $\pm\hbar\omega$ (vertical arrows)
- Excitations significantly above and below E_F: Nonlinear effects for $S_{peak} \geq 5 \times 10^{12}$ W/cm²
- Hybridization of Fe $d_{3z^2-r^2}$ states with p_z orbitals of MgO(IF) at +0.8 eV (↑)
- Hybridization of Fe d_{xy}, d_{xz}, d_{yz} states with p_x, p_y of MgO(IF) at 0.5 eV (↓)
- charge density redistribution @Fe: Depletion from in-plane (d_{xy} and $d_{x^2-y^2}$), accumulation of charge in out-of-plane orbitals (d_{xz}, d_{yz} and $d_{3z^2-r^2}$)

Polarization dependence of the excitation pattern $S_{peak} = 5 \times 10^{12}$ W/cm²

Changes in the charge distribution, $\rho(\vec{r},t) - \rho(\vec{r},0)$ at t = 20.2 fs and layer-resolved changes in the ΔTDDOS, $\Delta D(E,t) = D(E,t) - D(E,0)$, for laser pulses with $\hbar\omega$ = 7.75 eV and different directions of polarization:

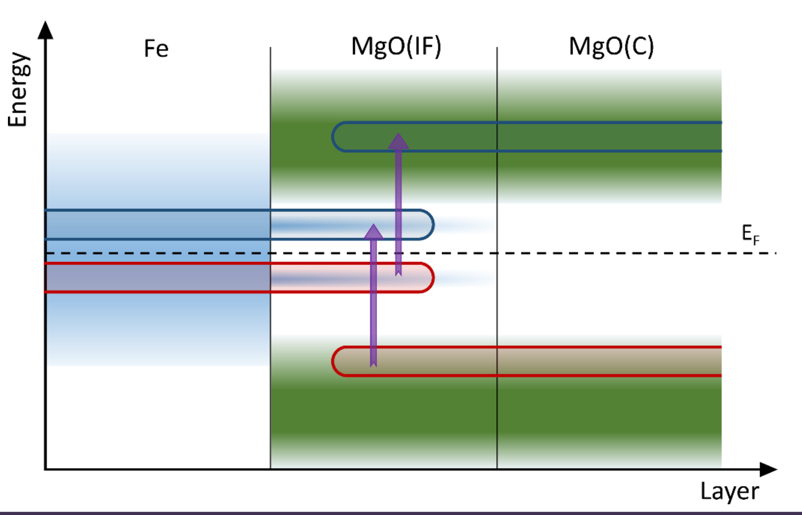


- Excitation of carriers from the VB edge of MgO into IF @ +1 eV (↑).
- Excitation from states below E_F in Fe into hybridized states @ +2 eV (↓).
- States from -1 eV to +1 eV in Fe: Interface-mediated excitation mechanisms (↓) and result in separation of excited carriers up to 13 eV in MgO(C).



- Largest charge redistribution at O sites for the highest frequency.
- No direct excitations in the Fe layer, due to limited *d* bandwidth.
- Vertical arrows: Direct excitations in MgO layers without the support of IF states.
- Redistribution of charge between *s* and p_z orbitals at O sites.
- Excitations above +6 eV and below -6 eV in MgO(C): Transitions to/from the interface states.

Concerted excitation mechanism



Two independent simultaneous excitations mediated by IF [2]:

- From valence band of insulator into an interface state above E_F
- From interface states below E_F into conduction band of MgO
- Bulk states serve as reservoir for excitation at IF

Conclusion

- Response of the system strongly depends on the photon frequency:
  $\hbar\omega$ = 2.25, 4.5 eV: Excitations take place predominantly in the Fe layers
  $\hbar\omega$ = 7.75 eV: Direct excitation within MgO layers
- Nonlinear response for the pulse with $S_{peak} = 5 \times 10^{12}$ W/cm² and $\hbar\omega$ = 2.25 eV
- Strong orbital dependence:
  Depletion of charge from in-plane orbitals of Fe and accumulation in out-of-plane orbitals for $\hbar\omega$ = 2.25, 4.5 eV, $\vec{E} \parallel x$
  Transfer of charge from d_{xz}, d_{yz} to e_g orbitals of Fe for 7.75 eV, $\vec{E} \parallel x$
- Anisotropic response for $\hbar\omega$ = 7.75 eV: Larger charge redistribution for out-of-plane polarization compared to the in-plane.
- Concerted excitation between VB/CB states of MgO and *d*-states of Fe via IF states allows simultaneous transfer of carriers between the subsystems in both directions: Accumulation of hot carriers in MgO

References

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We acknowledge funding through the DFG within SFB 1242, project C02 and MagnitUDE high performance computer at the University of Duisburg-Essen