Ultrafast dynamics and coherent excitations of 4f-orbitals derived electronic states in the Kondo semi metal CeSb

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Kondo (in a nutshell)

Magnetic impurity scattering

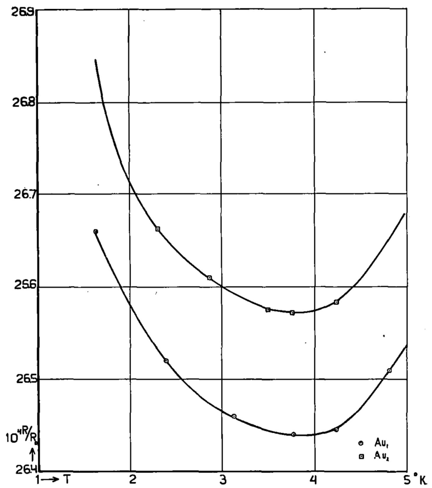
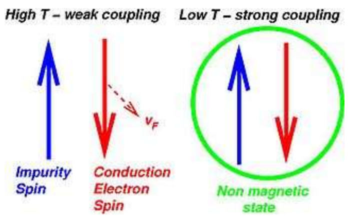


Fig. 1. Resistance of *Au* between 1°K. and 5°K.

W.J. de Haas, J. de Boer, G.J. van dën Berg, Physica **1**, 1115 (1934)
W. Meissner, B Voigt, Ann. Phys. **399**, 761 (1930)

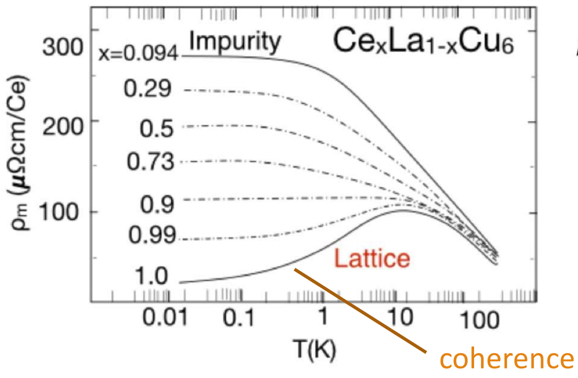
Kondo singlet (screening) at low *T* < T_K

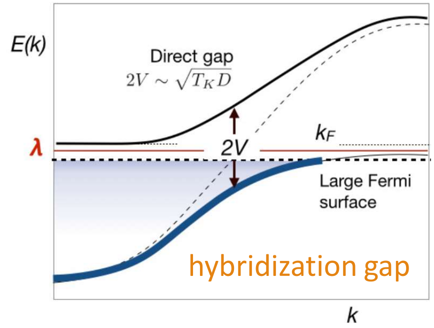


$$\rho_{\mathrm{spin}} = c\rho_M\{1 + (3zJ/\varepsilon_F)\log T\}$$

J. Kondo, Progress of Theoretical Physics **32**, 37 (1964)

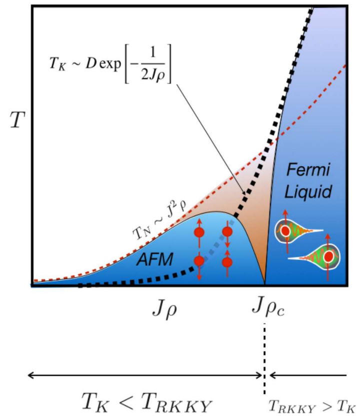
Dense Kondo systems - Kondo lattice

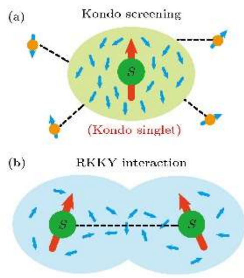




P. Coleman, *arXiv:1509.05769* (2015)

Competition: Kondo screening - RKKY



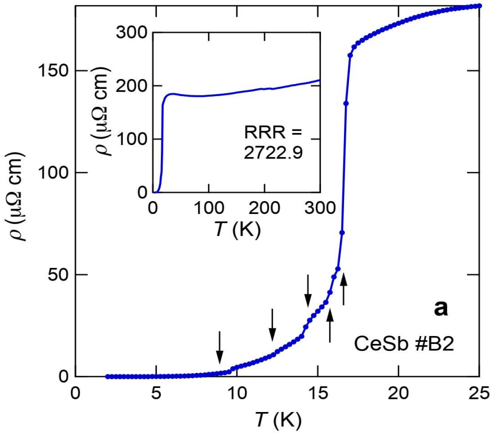


$T_K \sim De^{-1/(2J\rho)}$

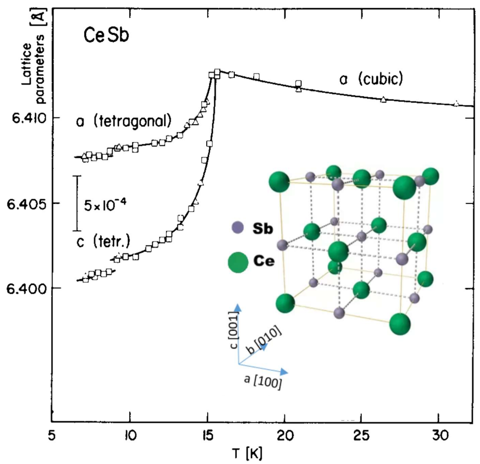
$T_{\mathrm{RKKY}} \sim J^2\rho$

S. Doniach, Physica **91B,** 231 (1977)
P. Coleman, *arXiv:1509.05769* (2015)
L. Yu et al., Acta Phys. Sin. **70**, 017402 (2021)

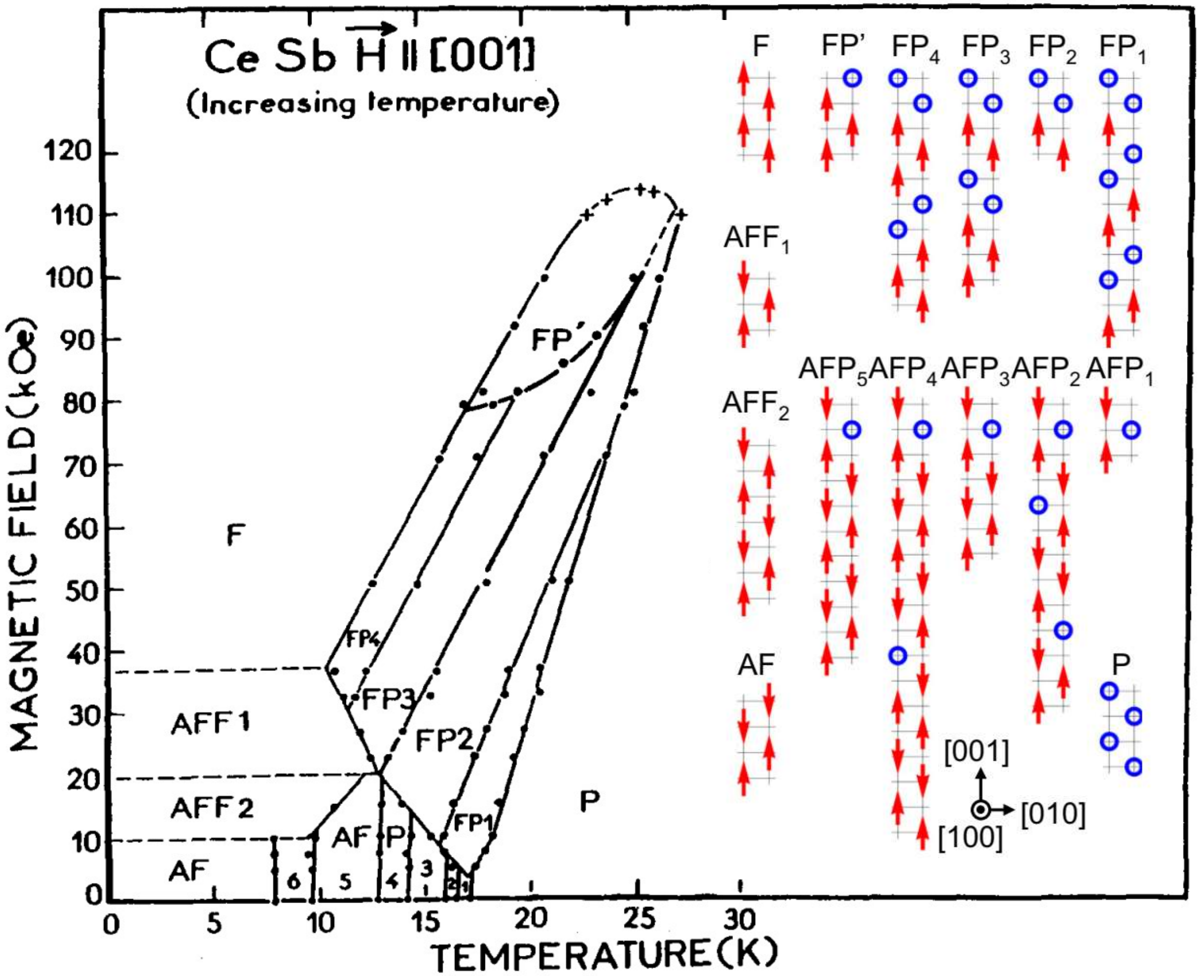
CeSb – low-*T* phase diagram



L. Ye et al., PRB 97, 081108(R) (2018)



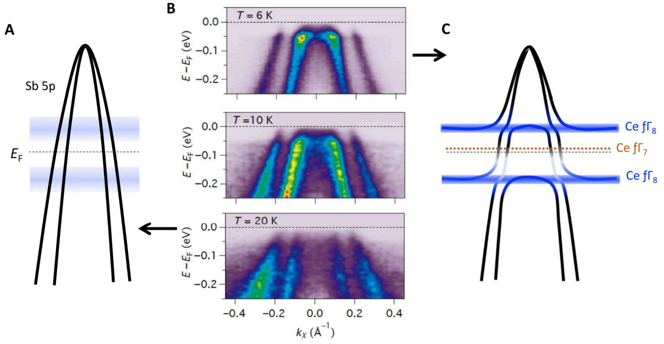
F. Hulliger et al., Journal of Low Temperature Physics **20**, 269 (1975)



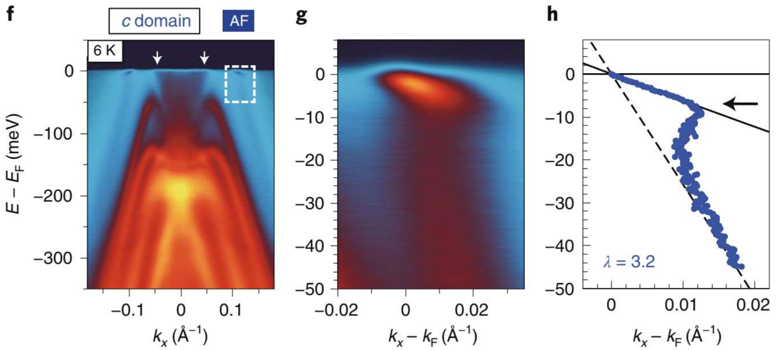
J. Rossat-Mignod et al., Physica 130B, 555 (1985)
L. Ye et al., PRB 97, 081108(R) (2018) (illustration)

Electronic structure in the magnetically ordered state

Γ-point (hybridization) (AF)

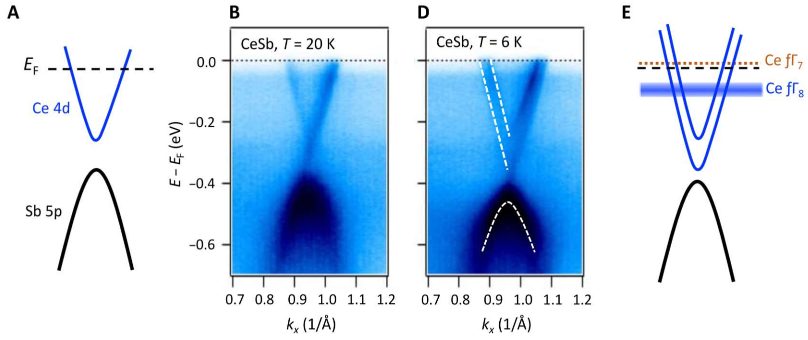


Jang et al., Sci. Adv. **5**, 7158 (2019)



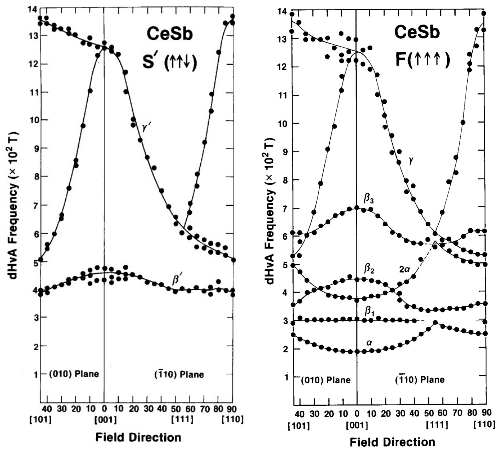
Arai et al., Nature Materials **21**, 410 (2022)

X-point (exchange splitting) (AF)



Jang et al., Sci. Adv. **5**, 7158 (2019)

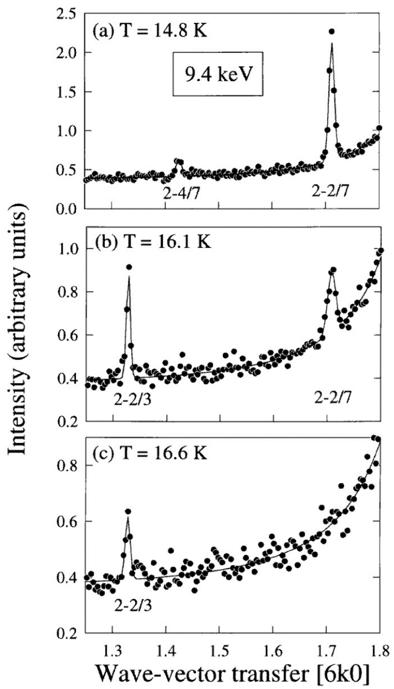
AFF & F states (de Haas-van Alphen)



H. Aoki eta al., Journal of Magnetism and Magnetic Materials **52**, 389 (1985)

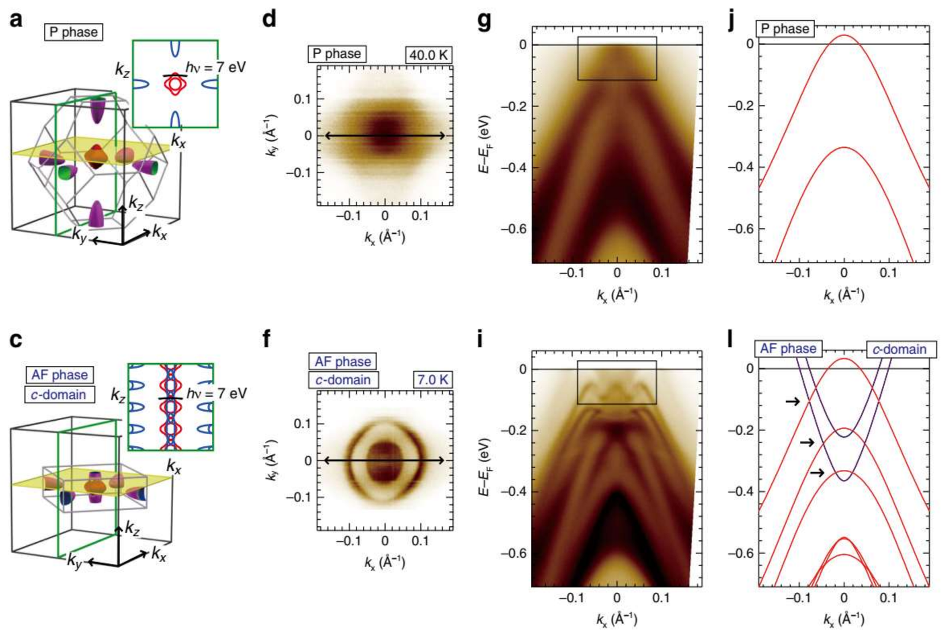
Periodic lattice modulation (BZ back folding)

X-ray diffraction



D.F. McMorrow et al., J. Phys.: Condens. Matter **9,** 1133 (1997)

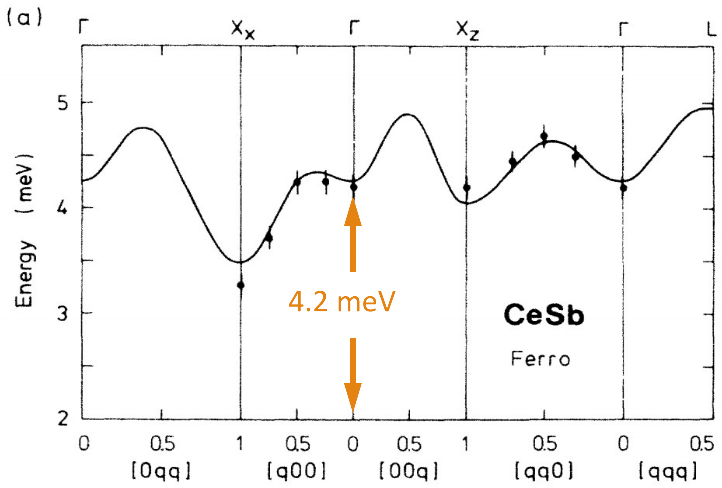
BZ back folding



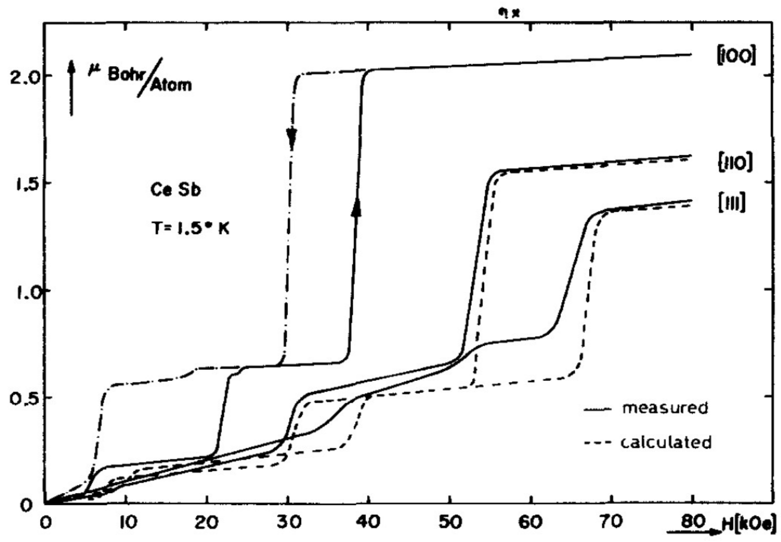
K. Kuroda et al., Nat Commun **11**, 2888 (2020)

Large anisotropy (Ising-like behavior)

Magnetic inelastic neutron scattering



B Hälg, A Furer, PRB **34**, 6258 (1986)

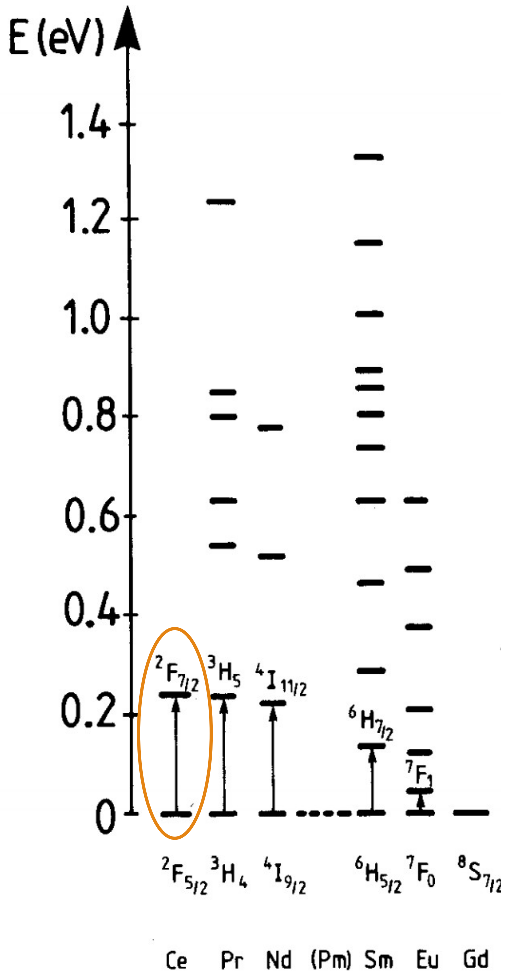


G. Busch, O. Vogt, Physics Letters **25A**, 449, (1967)

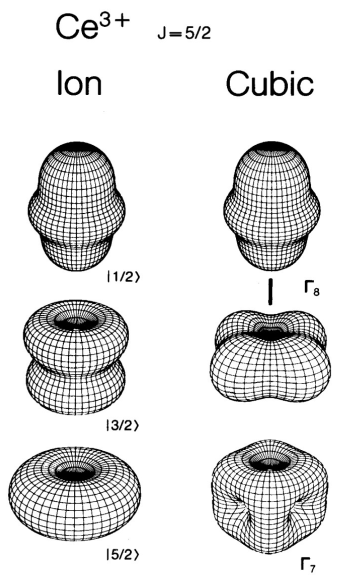
Magnetic moments are pinned along the modulation wave vector, *q*, along the [001] (tetragonal) direction. In the case of *H* along <110> or <111> (cubic) directions one observes just a projection due to the domains with the moments along <100> (cubic) directions.

Ce^{3+} crystal field splitting

SO splitting (free ion)



CF orbitals



$$|\Gamma_7\rangle = a|\pm 5/2\rangle - b|\mp 3/2\rangle,$$

$$|\Gamma_8\rangle = \begin{cases} |\pm\tfrac{1}{2}\rangle \\ b|\pm 5/2\rangle + a|\mp 3/2\rangle, \end{cases}$$

$$a = (1/6)^{1/2} \text{ and } b = (5/6)^{1/2}$$

P. Fulde, M. Loewenhaupt, Advances In Physics **34**, 589 (1985)

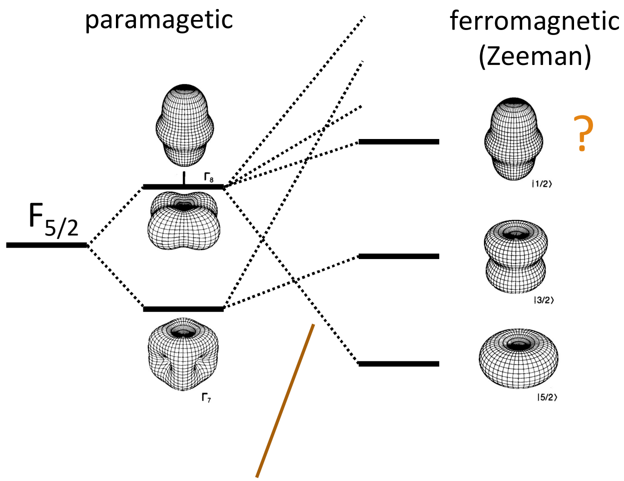
CF splitting in CeSb

paramagnetic (state, planes)

the ground state Γ_7

ordered planes

$\mu_{Ce^{3+}} \sim 2\mu_B$, the ground state close to |5/2>, Γ_8-like

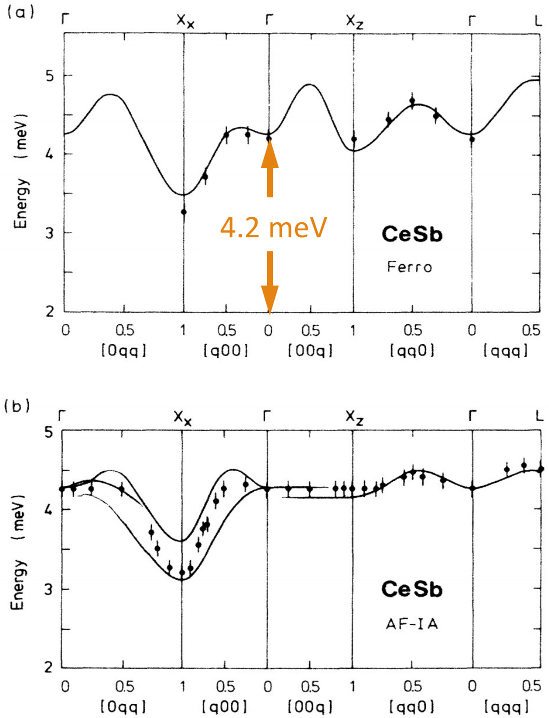


p-f mixing.

H. Takahashi and T. Kasuya, J. Phys. C: Solid State Phys. 18, 2721 (1985)

Magnetic (CF) excitations

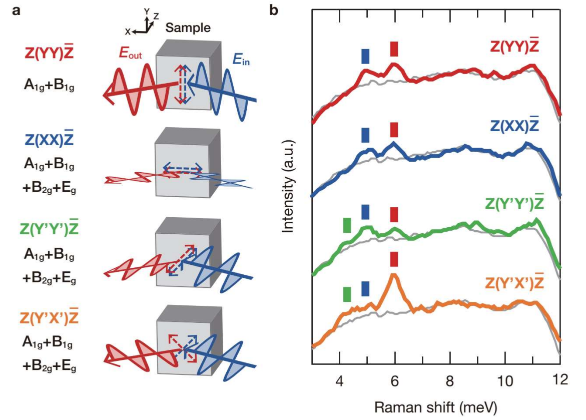
Inelastic neutron scattering (AF & F)

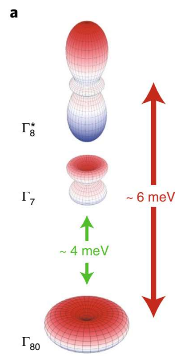


B Hälg, A Furer, PRB **34**, 6258 (1986)

- Two level model for low *T*.
  Excitation of the |5/2> -> |3/2> type (transverse magnon-like.
- CF effects negligible in the ordered state, Zeeman like splitting?

Raman scattering (AF)

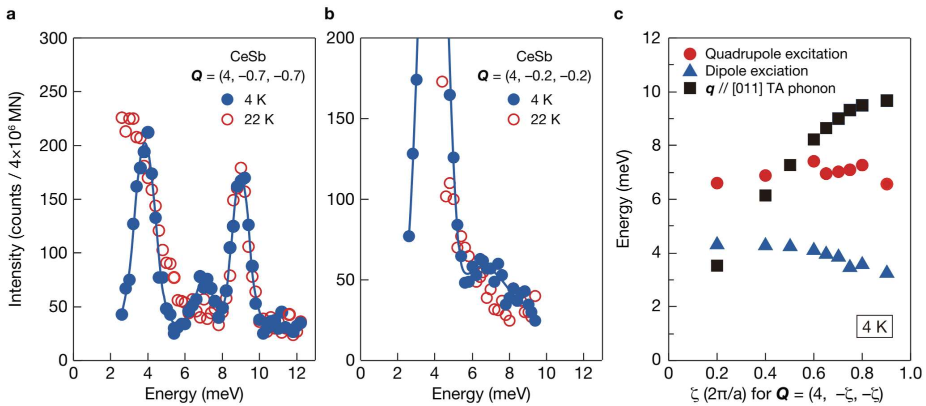




Arai et al., Nature Materials **21**, 410 (2022)

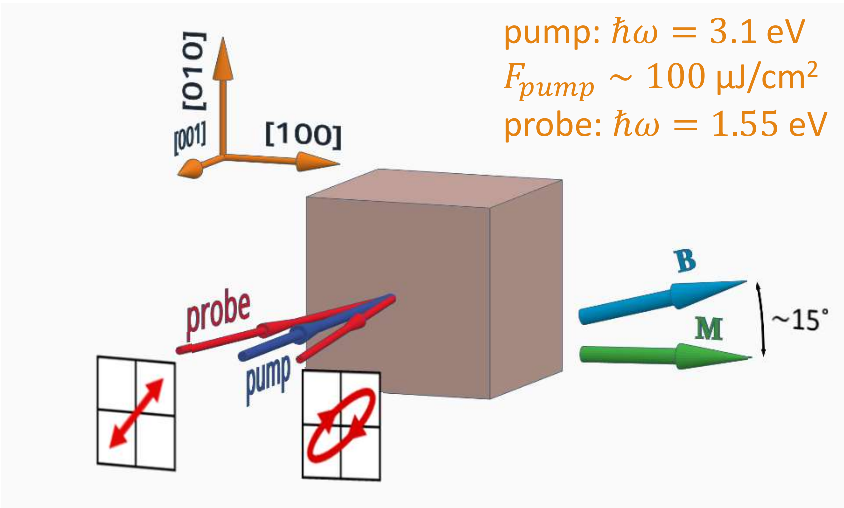
Splitting not Zeeman?

Inelastic neutron scattering (AF)



Arai et al., Nature Materials **21**, 410 (2022)

Experimental



MLB & MLD (assuming the cubic symmetry & $M_0 || [100]$)

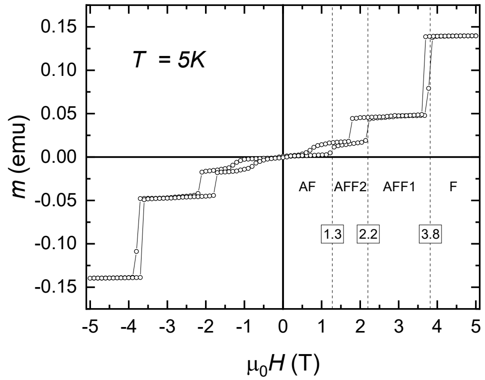
$$G = \begin{bmatrix} c_{11}M_x^2 + c_{21}\left(M_y^2 + M_z^2\right) & c_{55}M_xM_y & c_{55}M_xM_z \\ c_{55}M_xM_y & c_{11}M_y^2 + c_{21}\left(M_x^2 + M_z^2\right) & c_{55}M_yM_z \\ c_{55}M_xM_z & c_{55}M_yM_z & c_{11}M_z^2 + c_{21}\left(M_x^2 + M_y^2\right) \end{bmatrix}$$

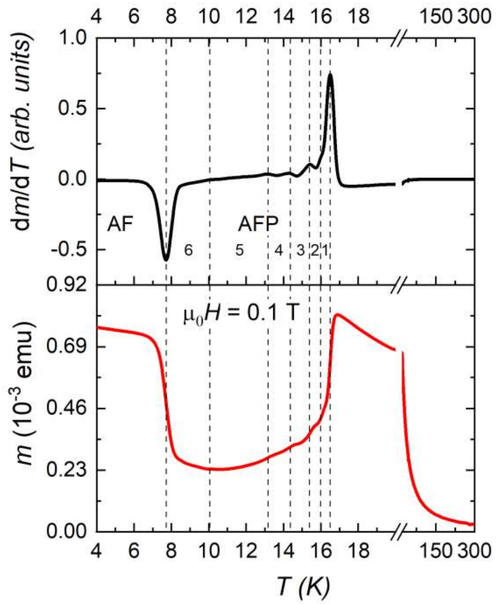
$$\Delta G \approx \begin{bmatrix} 2c_{11}M_0\Delta M_x & c_{55}M_0\Delta M_y & c_{55}M_0\Delta M_z \\ c_{55}M_0\Delta M_y & 2c_{21}M_0\Delta M_x & 0 \\ c_{55}M_0\Delta M_z & 0 & 2c_{21}M_0\Delta M_x \end{bmatrix}$$

MOKE (polar)

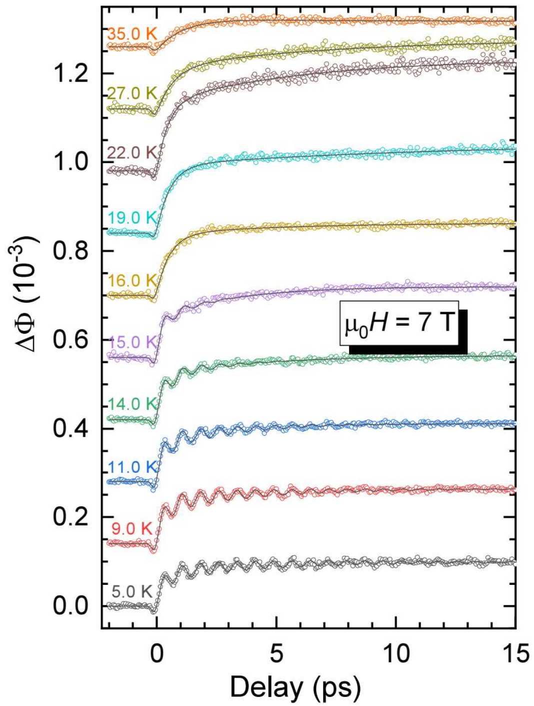
$$\Delta\Phi \propto \Delta M_z$$

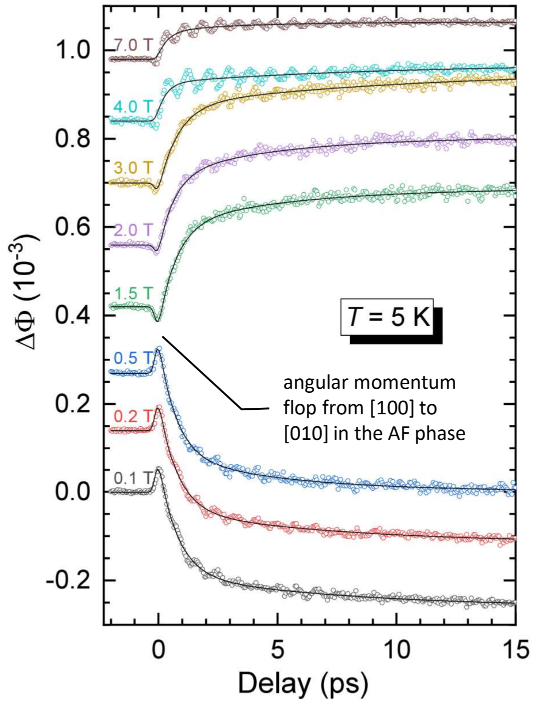
Sample characterization



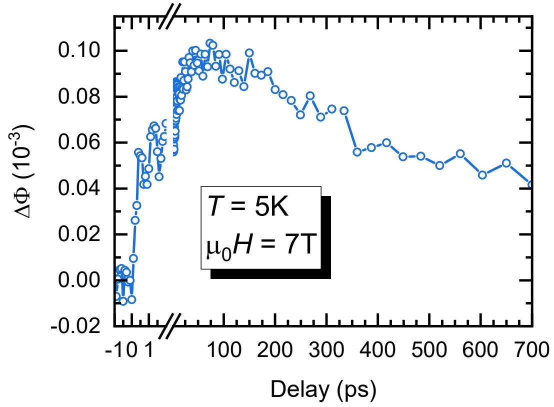


Data at glance

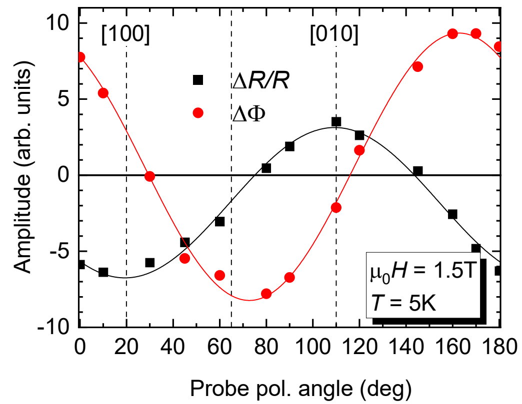


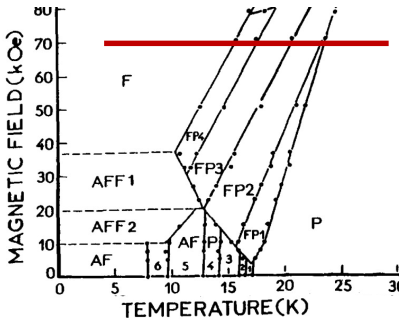


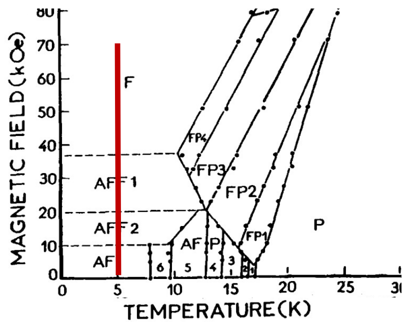
Slow dynamics



Angular dependence (probe)



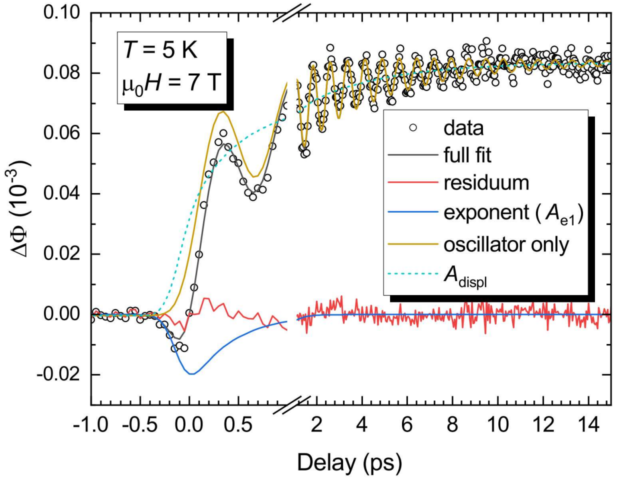




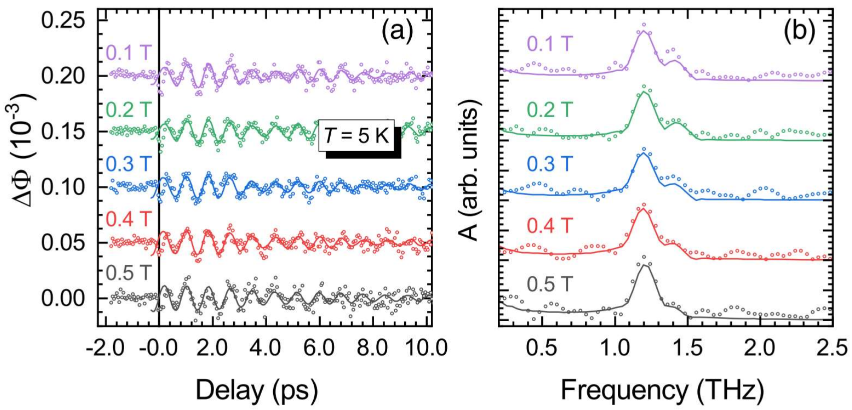
No dependence on the pump polarization angle.
No MOKE in the response.

Coherent oscillations

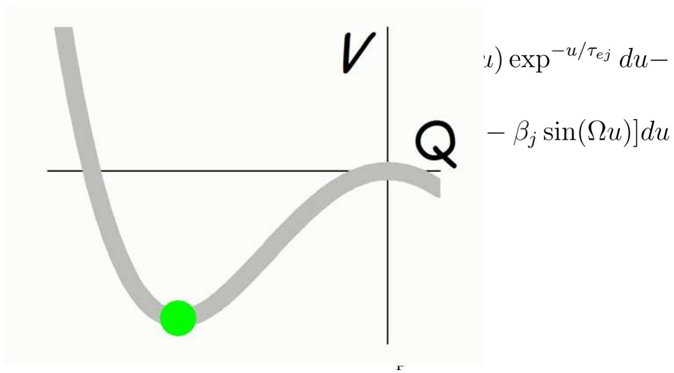
F phase (full DECP fit)

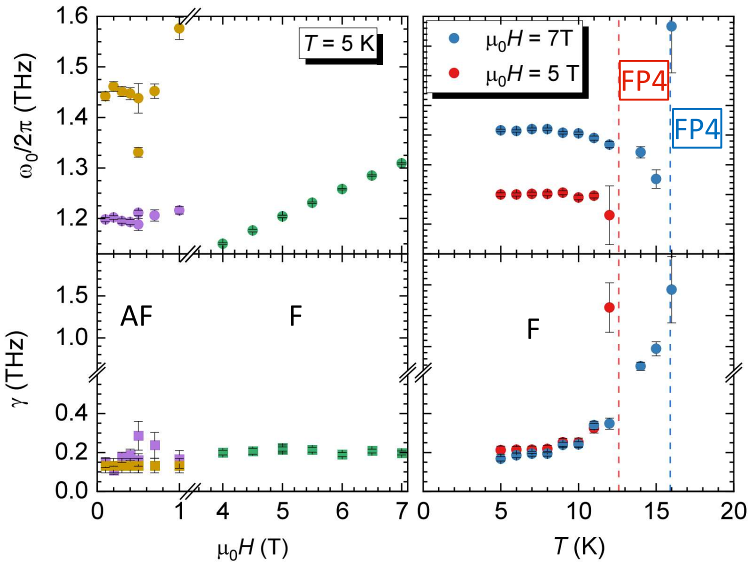


AF phase (residuum fit)



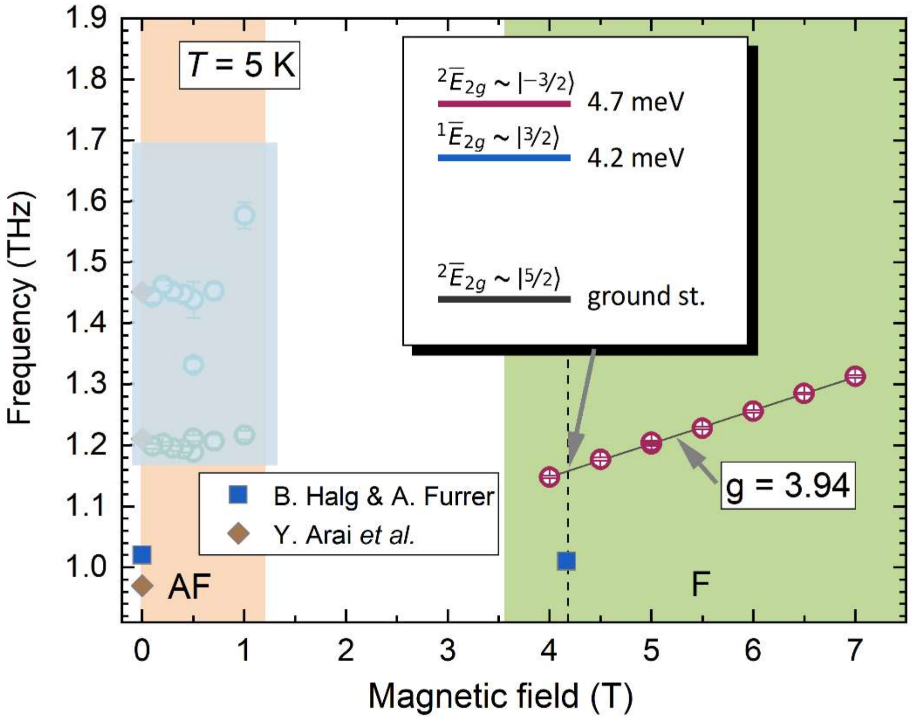
DECO model

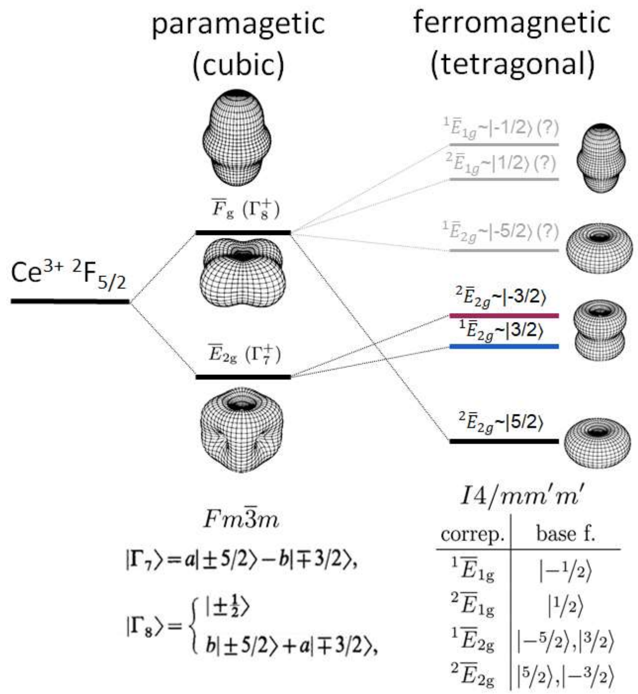




Zeiger et al., Phys. Rev. B 45, 768 (1992)

Origin of the coherent oscillations (F phase)

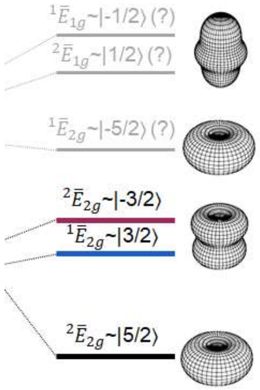




- The observed mode in the F phase cannot be |5/2> -> |3/2> spin-wave-like transition observed by INS since demagnetization effects can shift the frequency no more than: $\Delta\omega/\omega \sim M_0/2(H + H_A) \ = 1.6\%$
- Polarization dependence ($\Delta M \,||\, M_0$) suggests a longitudinal excitation between |5/2> -> |-3/2>.
- CF effects are not negligible, splitting not close to Zeeman as suggested by Halg et al..

Exciton "displacive" excitation mechanism

ferromagnetic
(tetragonal)

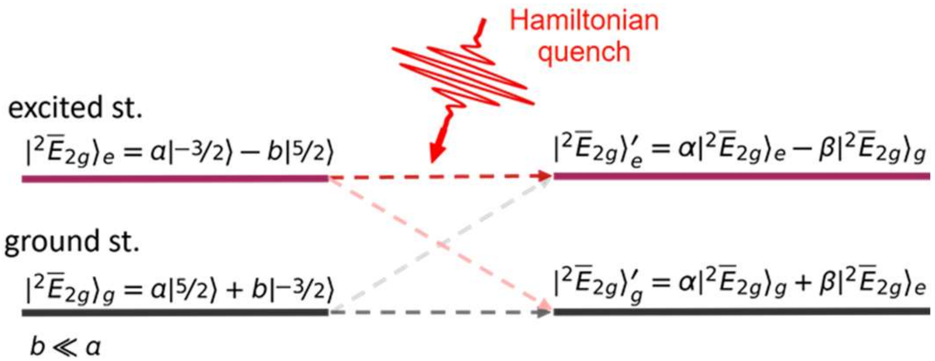


$I4/mm'm'$

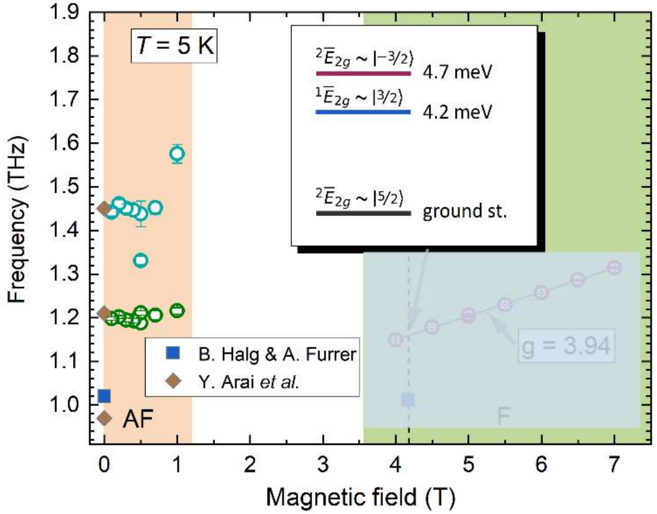
corep.	base f.
$^1\bar{E}_{1g}$	$\lvert -1/2\rangle$
$^2\bar{E}_{1g}$	$\lvert 1/2\rangle$
$^1\bar{E}_{2g}$	$\lvert -5/2\rangle, \lvert 3/2\rangle$
$^2\bar{E}_{2g}$	$\lvert 5/2\rangle, \lvert -3/2\rangle$

p-f mixing

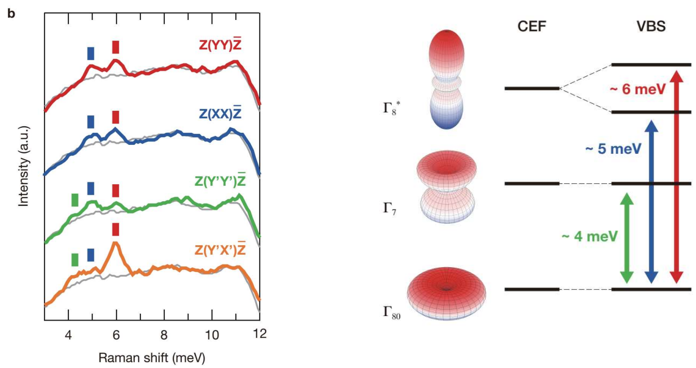
H. Takahashi and T. Kasuya, J. Phys. C: Solid State Phys. 18, 2721 (1985)



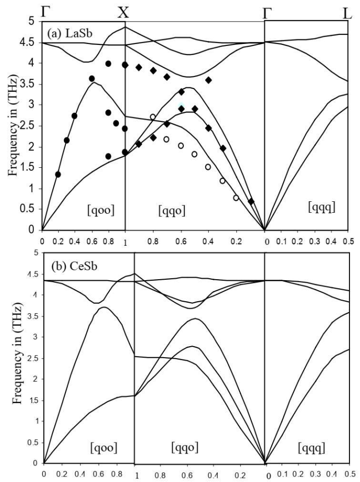
Origin of the coherent oscillations (AF phase)



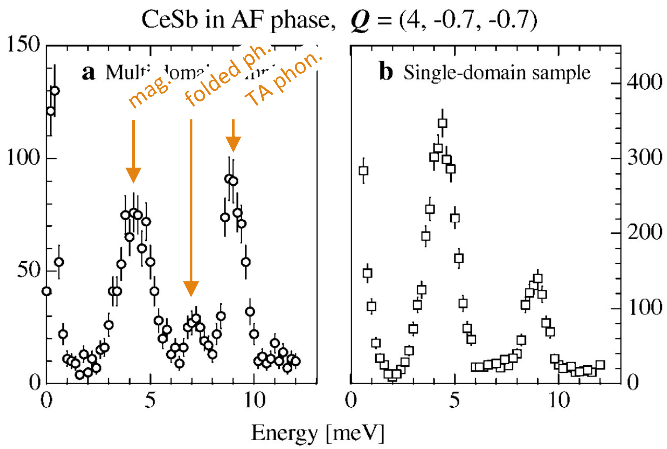
Raman scattering (AF)



Arai et al., Nature Materials **21**, 410 (2022).



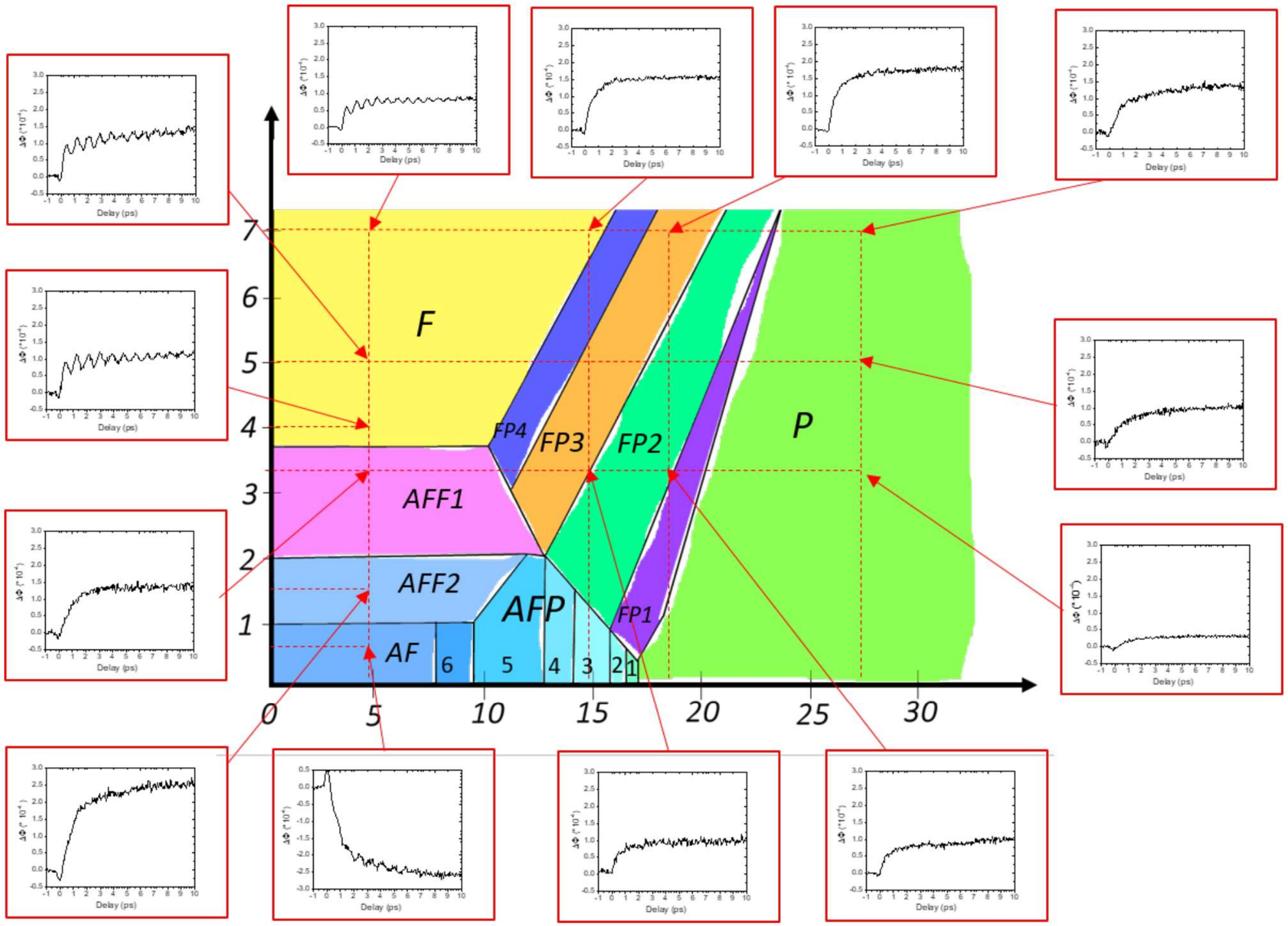
B. Rakshit et al., Optoelectron Adv Mat **2**, 37 (2008)



K. Iwasa et al., Appl. Phys. **A 74**, S1779 (2002).

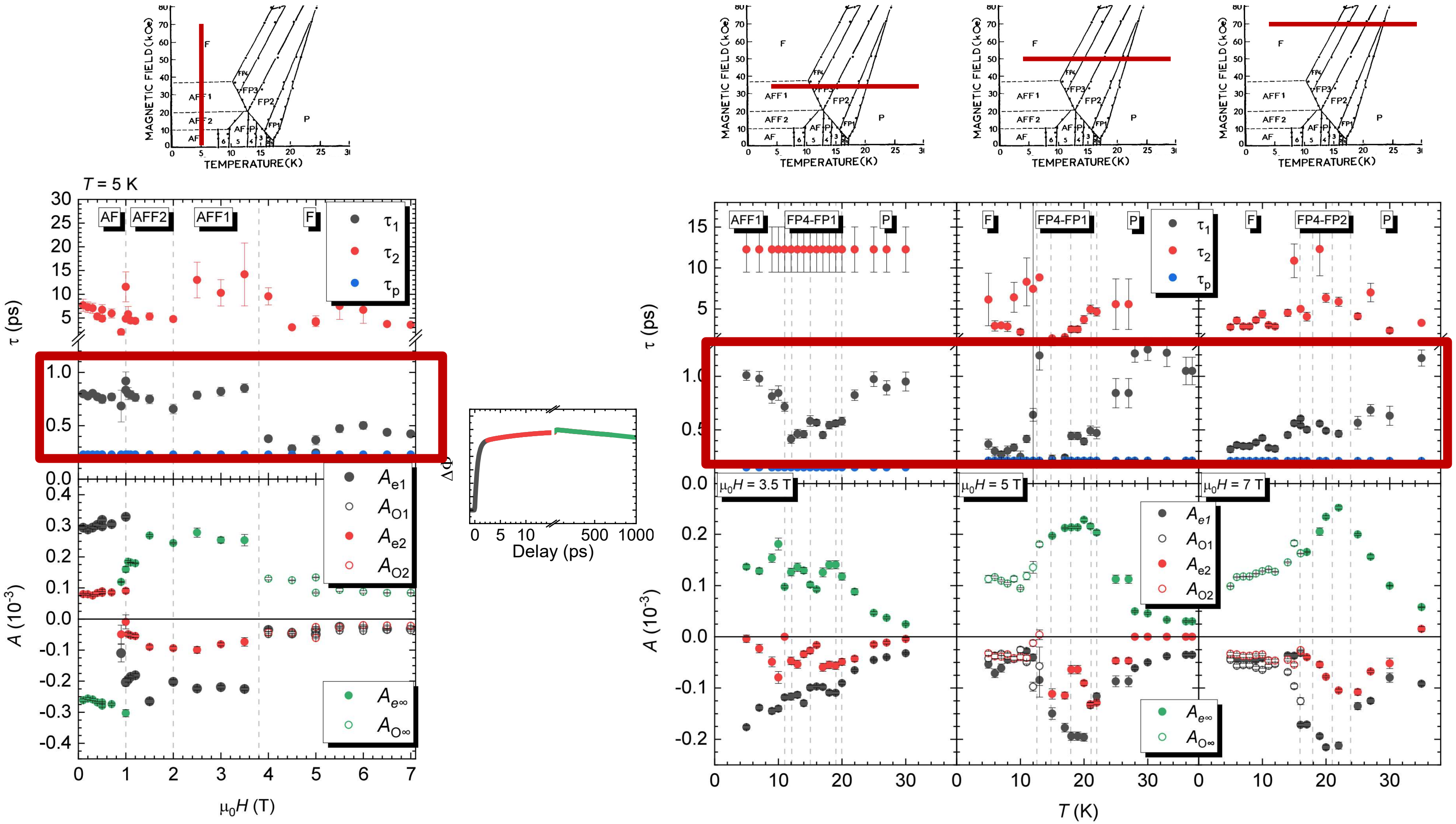
- Backfolding: 12 doubly degenerate CF-derived states. ($2\bar{E}_{1g} \oplus 4\bar{E}_{2g} \oplus 4\bar{E}_{1u} \oplus 2\bar{E}_{2u}$)
- No magnetic field dependence, but could be magnetic since $H \perp M_{sl}$ due to the angular momentum flop.
- Folded phonons (vibronic) also possible.

Data across the phase diagram



Exponential components

H - scan *T* - scans



The F and FP1-FP4 phases show faster sub-picosecond dynamics (faster quench) suggesting similar electronic structure different from the AF, AFF1 and AFF2 phases.

Credits

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arrs SLOVENIAN RESEARCH AGENCY

Research core funding : Grant No. P1-0040
Young researcher funding: Grant. No.PR-08428

cost EUROPEAN COOPERATION IN SCIENCE & TECHNOLOGY

CA17123 - Ultrafast opto-magneto-electronics for non-dissipative information technology (MAGNETOFON)

Summary & Conclusions

- We measured ultrafast magnetooptical response in most of the CeSb phase diagram.
- Coherent oscillations observed in the F and AF phases.
- Additional collective magnetic excitation observed in the F phase. CF effects strong also in the F phase.
- Two modes in the AF phase consistent with recent Raman results. Unclear origin, could be magnetic excitations or folded lattice modes or magnetic-lattice hybridized excitations.
- Non-oscillatory relaxation qualitatively similar in all phases. The sub-picosecond dynamics faster in the F(P) phases. Related to different electronic structures in the AF and AFF_n phases.

M. Naseska, N. D. Zhigadlo, Z. Jagličić, E. Goreshnik, and T. Mertelj, *Ultrafast Optical Polarimetry in Magnetic Phases of the Kondo Semimetal CeSb*, Phys. Rev. B **109**, 125140 (2024).