ICQM
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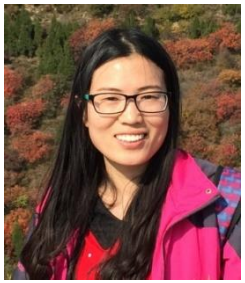
for Quantum Materials

Identify two CDW amplitude modes with extremely small energy scales in LaAgSb2 by ultrafast pump-probe measurement

Nan-Lin Wang



Collaborators：

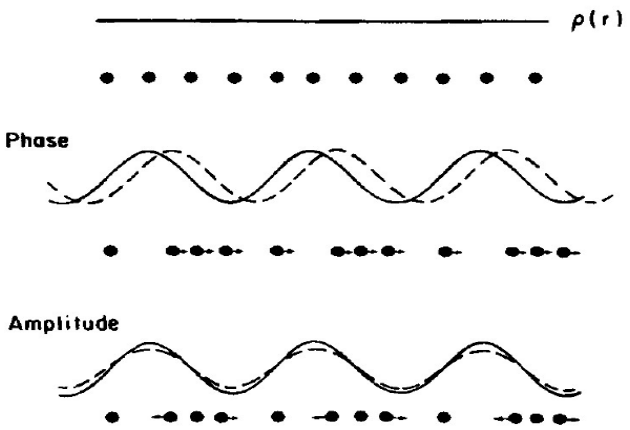
R. Y. Chen, S. J. Zhang, M. Y. Zhang, T. Dong

ICQM, School of Physics,
Peking University, Beijing, China

Density modulation

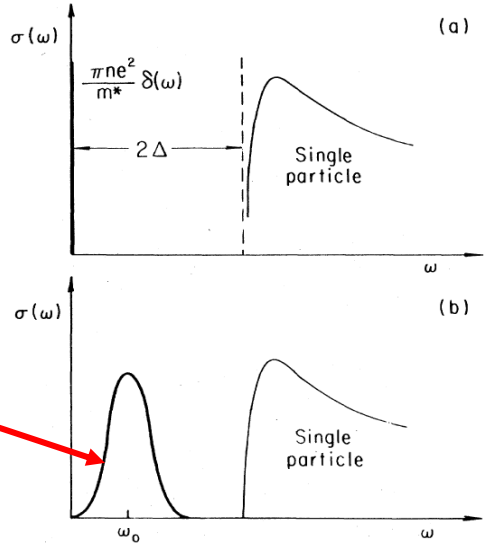
About CDW condensate:

Single particle excitation: an energy gap;
Collective excitations: amplitude and phase modes.



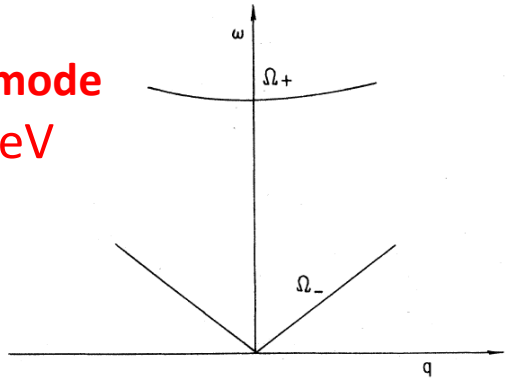
Phase excitation corresponds to the translational motion of the undistorted condensate

$\omega_\phi(q=0)=0.$ because no change in condensation energy



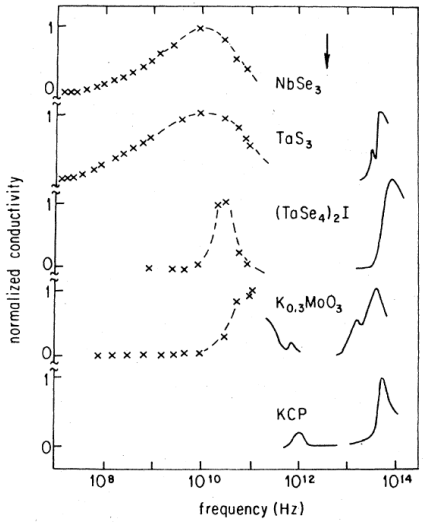
Pinning by impurities

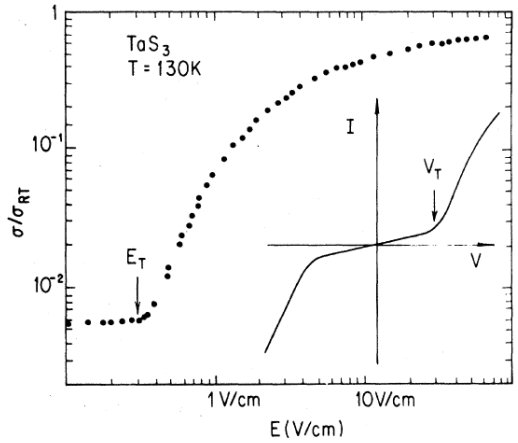
amplitude mode
gap ~10 meV



G. Gruner, RMP (1988)

The pinning/depinning of phase mode has dramatic effect on charge transport properties. By applying dc electric field, the phase mode can be driven into a current-carrying state, leading to nonlinear current-voltage characteristics.





G. Gruner, RMP (1988)

However, effect of amplitude mode (Raman active) on the physical properties was much less studied.

Here we show that a layered CDW compound LaAgSb2 has unusually small energy scales of the amplitude modes.

Outline

- Introduction about the material

  Two CDW orders in LaAgSb2 with very small $2\mathbf{K}_F$

- Optical spectroscopy data

  CDW energy gaps

- Ultrafast pump-probe measurement

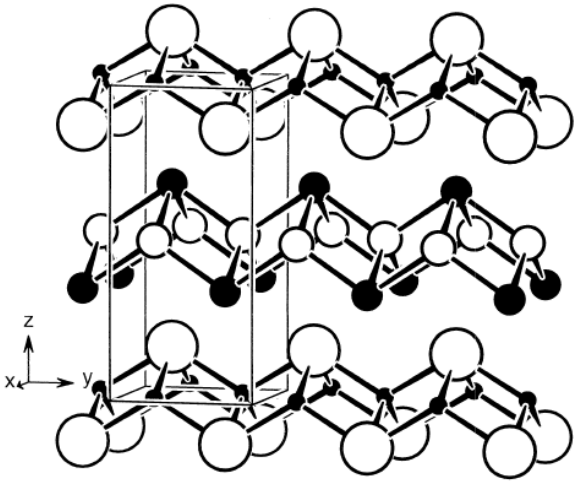
  Key result: Identify two CDW amplitude modes with extremely small energy scales

- Summary

R. Y. Chen et al., Phys. Rev. Lett. 118, 107402 (2017)

About the material

LaAgSb2 has ZrCuSi2 structure



- **First report of quaternary silicide arsenide ZrCuSiAs structure**
  V. Johnson and W. Jeitschko, J. Solid State Chem. 11, 161 (1974)
- **First ternary compound with similar structure ZrCuSi2**
  H. Sprenger, J. Less-Common Met. 34, 39 (1974)
- **Related compounds followed:**
  HfCuSi2 (1975), CaMnBi2 (1980), UCuAs2 (1987),

JOURNAL OF SOLID STATE CHEMISTRY **115**, 305–308 (1995)

Ternary Arsenides $ACuAs_2$ and Ternary Antimonides $AAgSb_2$ (A = Rare-Earth Elements and Uranium) with $HfCuSi_2$-Type Structure

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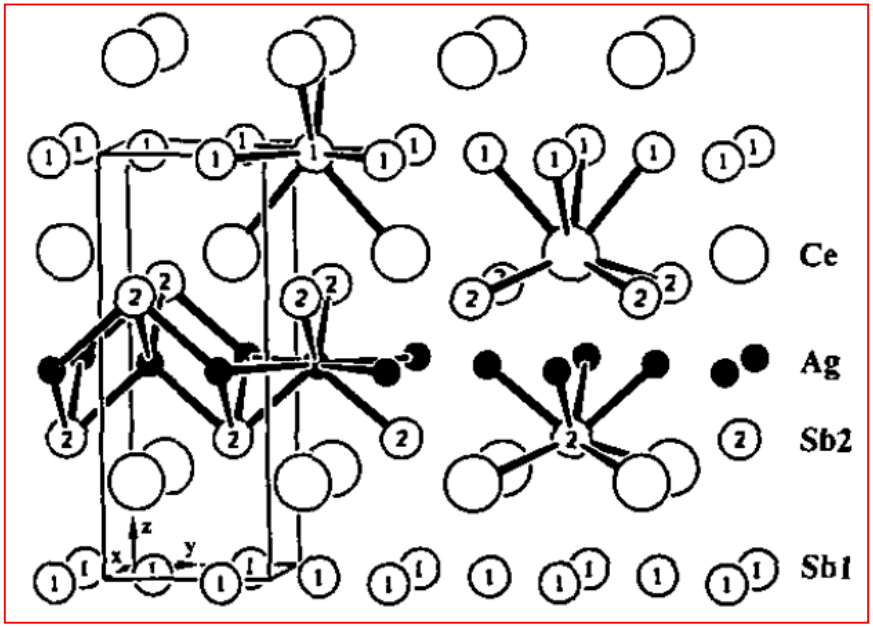
Received May 13, 1994; accepted July 15, 1994

Lattice Constants of Arsenides and Antimonides with the Tetragonal $HfCuSi_2$-Type Structure[a]

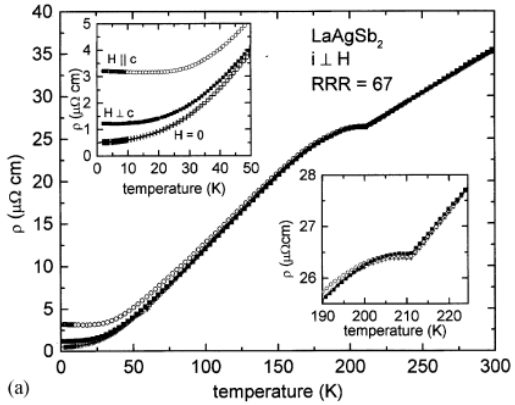
Compound	a (pm)	c (pm)	c/a	V (nm^3)
$YCuAs_2$	388.62(8)	987.10(3)	2.540	0.1491
$LaCuAs_2$	404.8(3)	1027(1)	2.537	0.1683
$CeCuAs_2$	401.81(7)	1010.4(2)	2.515	0.1631
$PrCuAs_2$	399.42(8)	1006.8(3)	2.521	0.1606
$NdCuAs_2$	396.7(1)	1005.5(5)	2.535	0.1582
$SmCuAs_2$	393.50(6)	997.2(2)	2.534	0.1544
$GdCuAs_2$	391.05(4)	992.9(2)	2.549	0.1518
$TbCuAs_2$	389.42(4)	987.9(2)	2.537	0.1498
$DyCuAs_2$	388.20(4)	984.8(2)	2.537	0.1484
$HoCuAs_2$	387.24(9)	982.2(3)	2.536	0.1473
$ErCuAs_2$	385.83(7)	978.9(3)	2.537	0.1457
$TmCuAs_2$	384.87(7)	976.9(3)	2.538	0.1447
$YbCuAs_2$	384.5(1)	974.5(5)	2.535	0.1441
$LuCuAs_2$	383.42(7)	974.2(3)	2.541	0.1432
$UCuAs_2$[b]	394.8(2)	953.3(5)	2.415	0.1486
$YAgSb_2$	427.65(3)	1048.8(1)	2.452	0.1918
$LaAgSb_2$	439.03(6)	1084.0(2)	2.469	0.2089
$CeAgSb_2$	436.3(1)	1069.9(4)	2.452	0.2037
$PrAgSb_2$	434.99(6)	1067.0(3)	2.453	0.2019
$NdAgSb_2$	433.53(3)	1063.0(2)	2.452	0.1998
$SmAgSb_2$	431.2(1)	1055.5(5)	2.448	0.1962
$GdAgSb_2$	429.52(7)	1050.6(2)	2.446	0.1938
$TbAgSb_2$	428.33(7)	1047.6(2)	2.446	0.1922
$DyAgSb_2$	427.43(5)	1044.2(3)	2.443	0.1908
$HoAgSb_2$	426.62(9)	1042.2(3)	2.443	0.1897
$ErAgSb_2$	425.65(8)	1039.0(3)	2.441	0.1883
$TmAgSb_2$	425.29(7)	1039.0(3)	2.443	0.1880
$UAgSb_2$	432.40(6)	1031.8(2)	2.386	0.1929

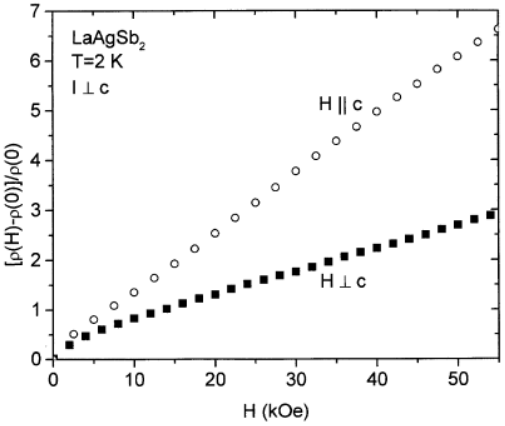


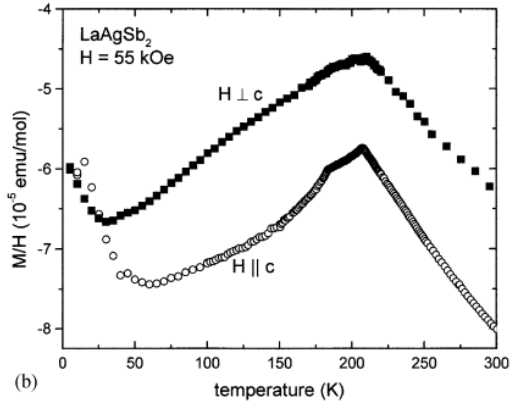
Basic properties

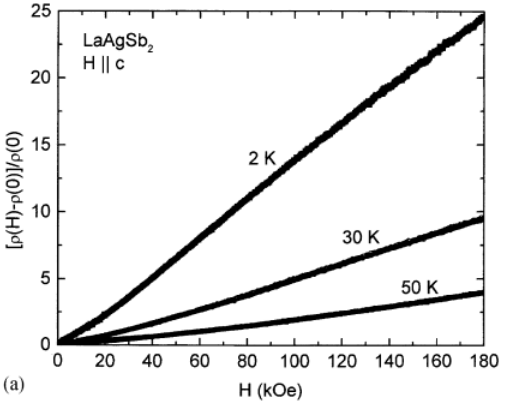
K.D. Myers et al. / Journal of Magnetism and Magnetic Materials 205 (1999) 27–52

LaAgSb2









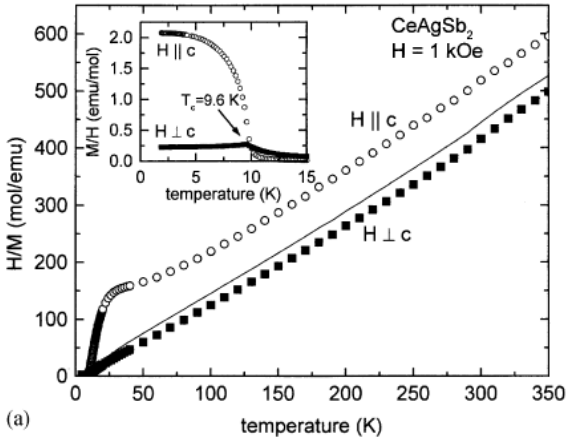
Two phase transitions

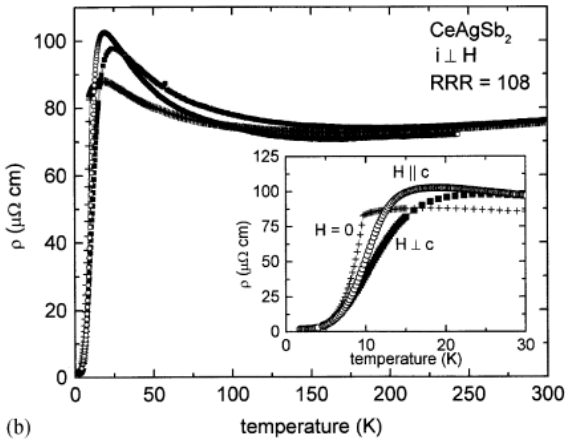
CDW or SDW speculated

Linear MR

Dirac fermion？

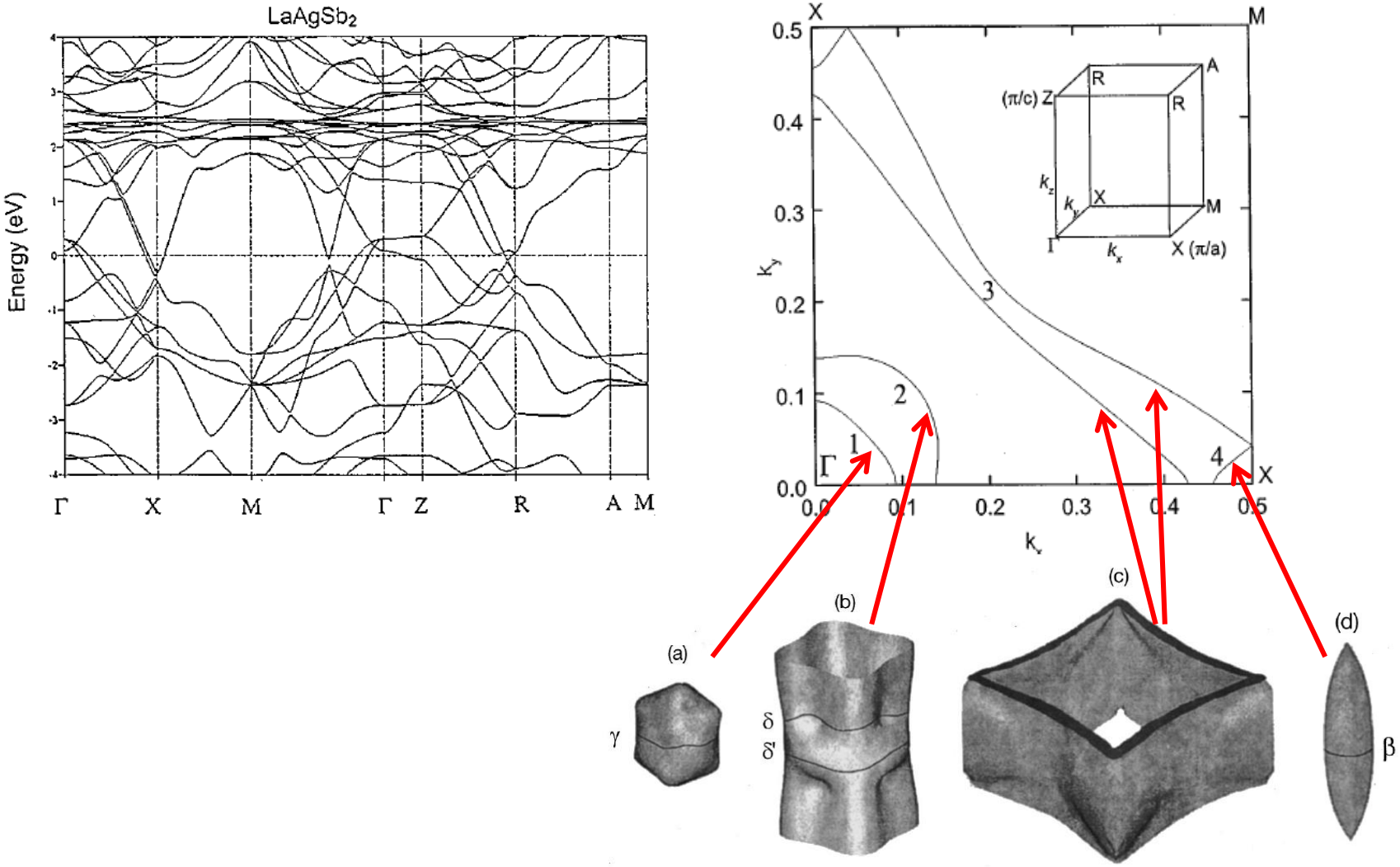
CeAgSb2





Heavy fermions

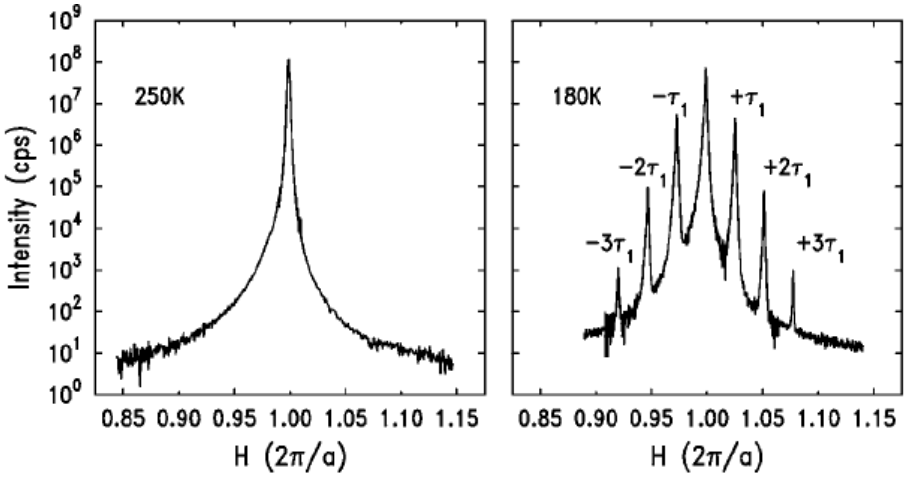
LDA band structures



K. D. Myers et al., PRB 60, 13731 (1999)

Synchrotron X-ray diffraction

C. Song et al. PRB 68, 035113 (2003)



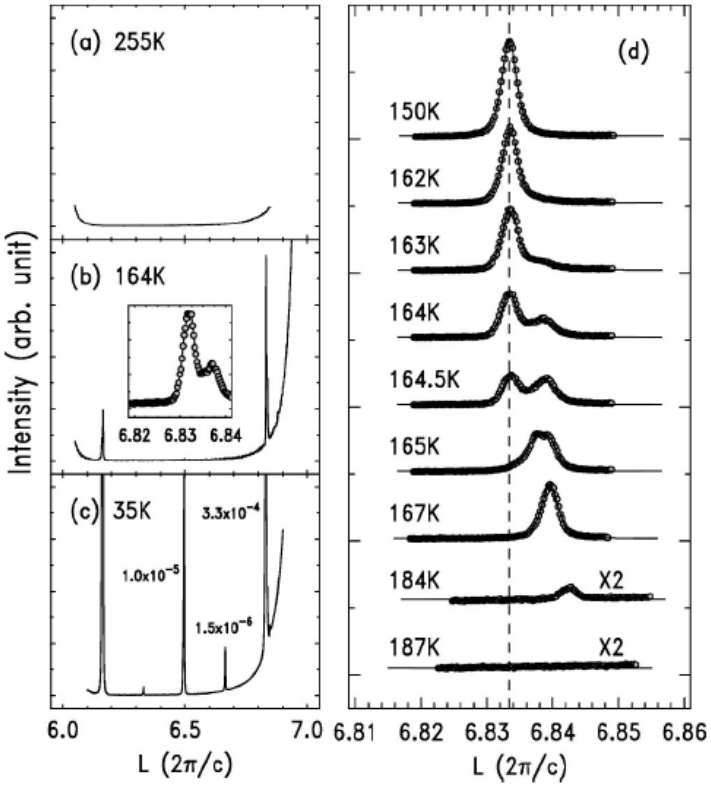
Along (0 0 1) direction

$$T_{1,\mathrm{CDW}} = 207\ \mathrm{K}.$$

$$\tau_1 \sim 0.026(2\pi/a)$$

$$2\pi/\tau_1 \sim 40a$$

a periodic charge and lattice modulation

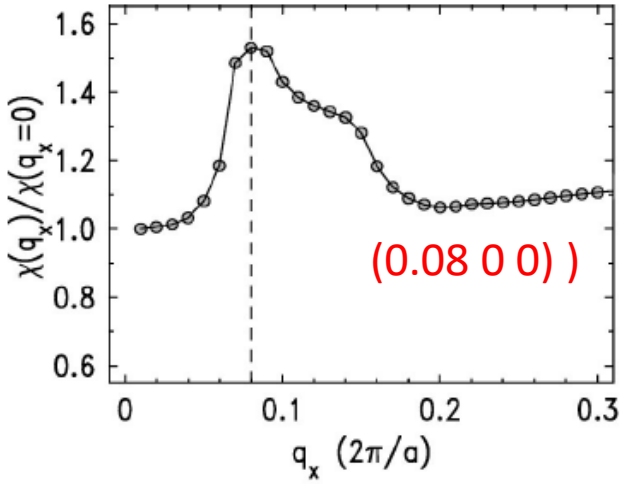


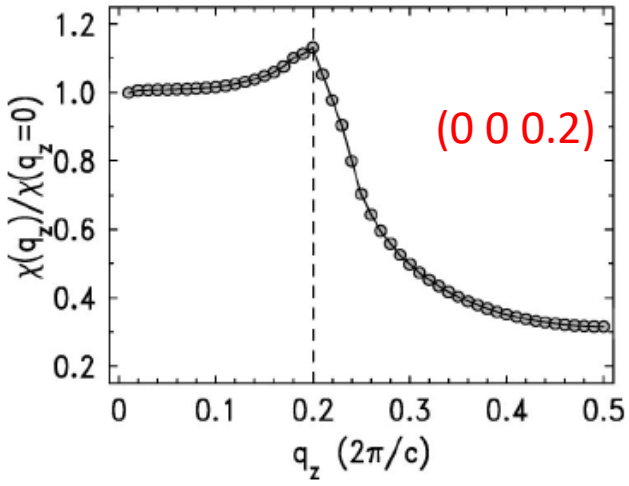
Along (0 0 1) direction around (1 0 7) peak

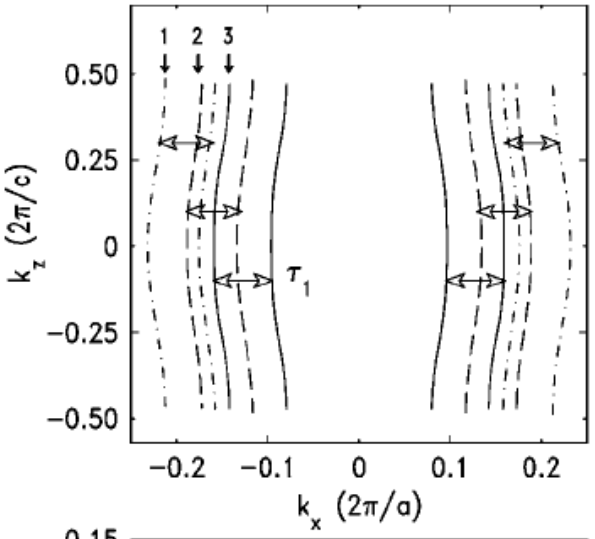
$$T_{2,\mathrm{CDW}} = 186\ \mathrm{K}$$

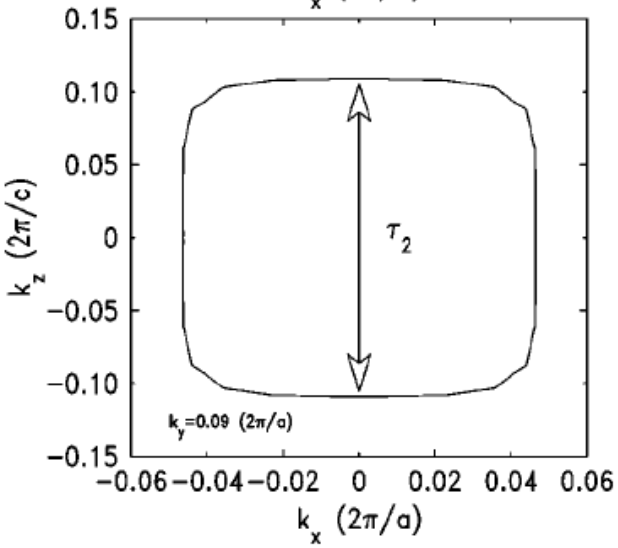
$$\tau_2 \sim 0.16(2\pi/c)$$

(0.026 0 0) of τ_1-CDW
(0 0 0.166) of τ_2-CDW.









Generalized susceptibility

C. Song et al. PRB 68, 035113 (2003)

PHYSICAL REVIEW B **86**, 155213 (2012)

Multiband effects and possible Dirac states in $LaAgSb_2$

Kefeng Wang (王克锋) and C. Petrovic
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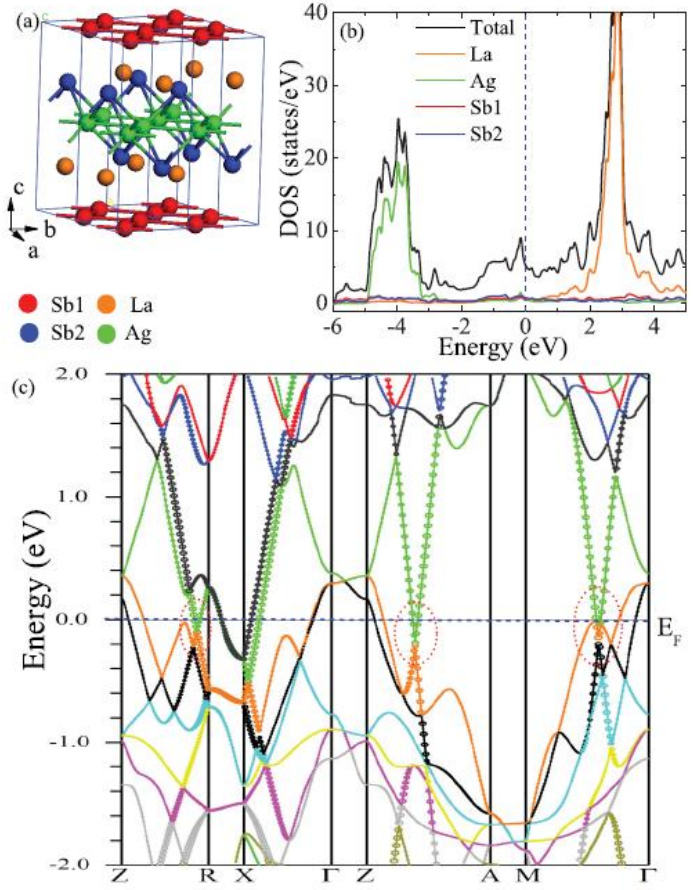
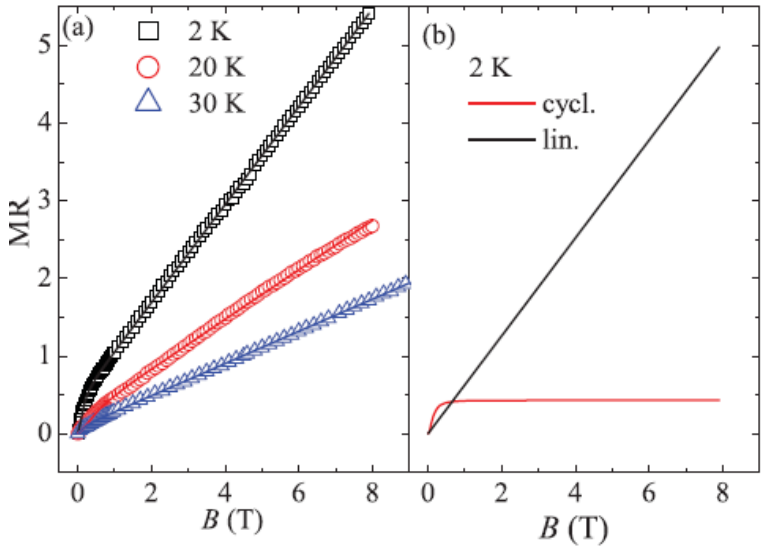
(Received 13 July 2012; revised manuscript received 4 September 2012; published 25 October 2012)





$$\mathrm{MR} = \frac{1}{2\pi}\left(\frac{e^2}{\varepsilon_\infty \hbar v_F}\right)^2 \frac{N_i}{en^2} B \ln(\varepsilon_\infty)$$

$$B^* = \frac{1}{2e\hbar v_F^2}(E_F + k_B T)^2$$

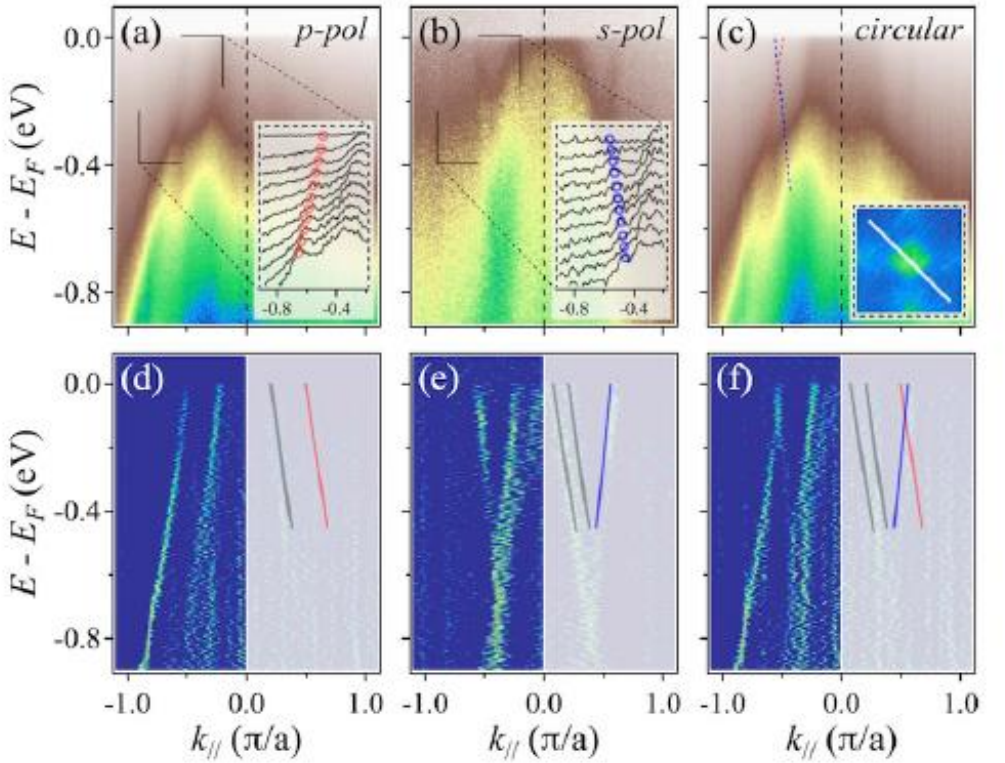
$$v_F \sim 1.46 \times 10^5 \ \mathrm{ms}^{-1}$$

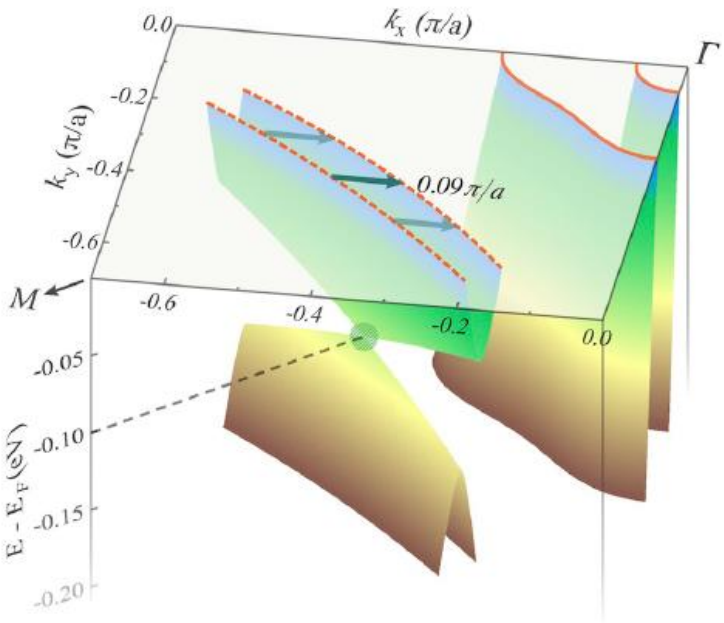
Linear MR could arise due to the quantum limit of Dirac fermions in the high magnetic field.

PHYSICAL REVIEW B **93**, 081105(R) (2016)

Observation of Dirac-like band dispersion in $LaAgSb_2$

X. Shi,[1] P. Richard,[1,2,*] Kefeng Wang,[3,†] M. Liu,[1] C. E. Matt,[4,5] N. Xu,[4,6] R. S. Dhaka,[4,6,7] Z. Ristic,[4,6,8] T. Qian,[1] Y.-F. Yang,[1,2] C. Petrovic,[3] M. Shi,[4] and H. Ding[1,2,‡]





Dirac point locates 100 meV below E_F

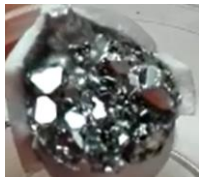
$V_{F1}=5.7\times10^5$ m/s $V_{F2}=8\times10^5$ m/s

Nesting wave vector 009 π/a

The relationship between Dirac fermions and CDW?

Single crystal growth and characterizations

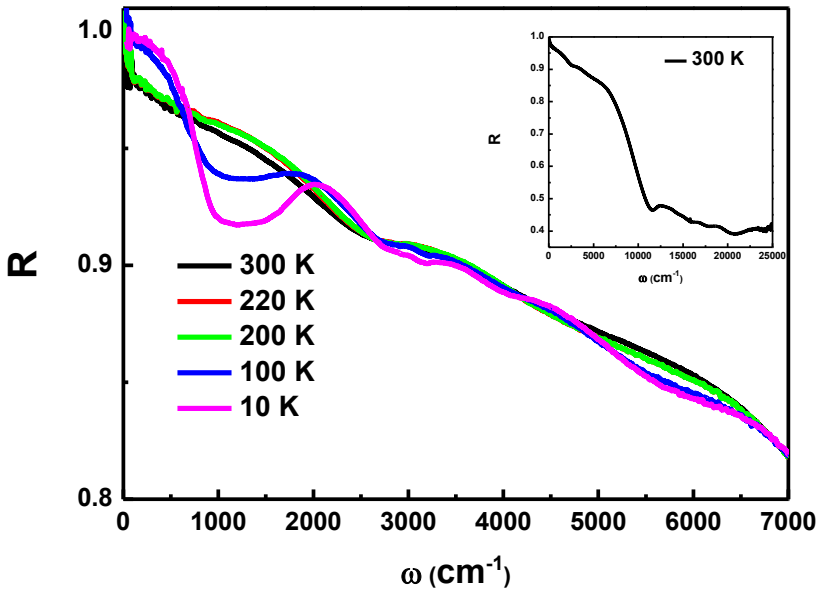
- We grew LaAsSb2 single crystal by Sb flux method.

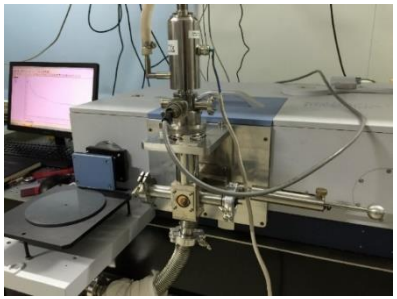


- The crystals were characterized by XRD, EDX, $\rho(T)$, $\chi(T)$.
- Optical spectroscopy
- Pump-probe measurement

Optical spectroscopy measurement

We measured R(ω) (30-25000 cm^{-1}) using Bruker 80 v and 113 v spectrometers

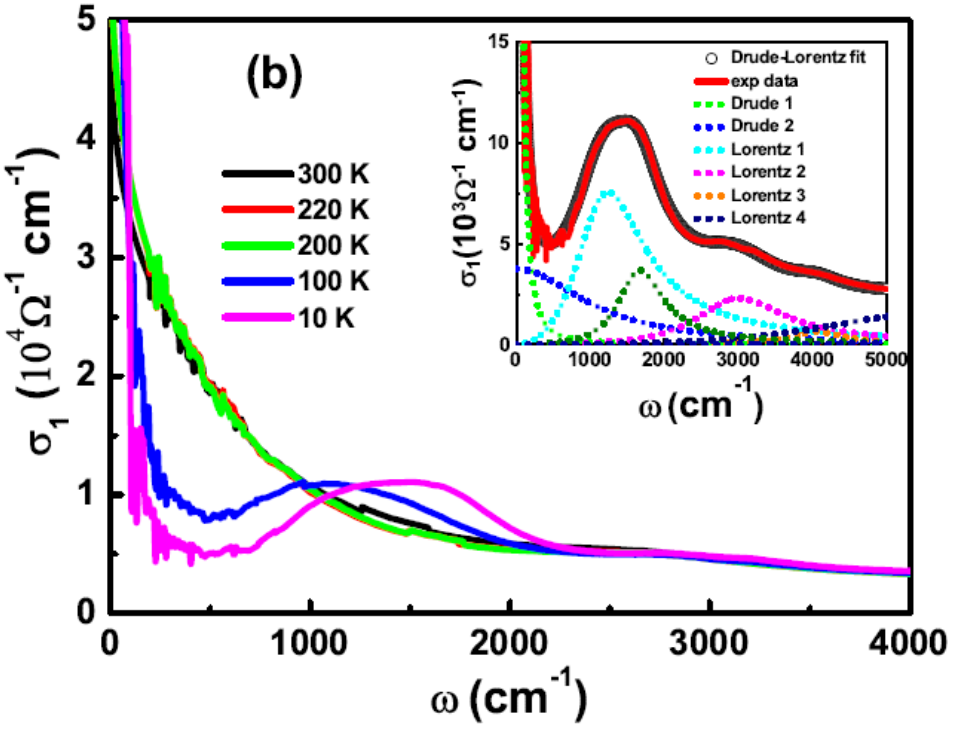




In-situ gold- or aluminum-deposit technique

$$R(\omega) \overset{KK}{\Rightarrow} \theta(\omega) \rightarrow \begin{cases} n(\omega), \kappa(\omega) \\ \varepsilon_1(\omega), \varepsilon_2(\omega) \\ \sigma_1(\omega), \sigma_2(\omega) \end{cases}$$

$$\theta(\omega) = -\frac{\omega}{\pi}\int_0^{\infty} \frac{\ln R(\omega') - \ln R(\omega)}{\omega'^2 - \omega^2} d\omega'$$



- ✓ **Observation of partial CDW gaps: *$\Delta_1 \approx 207$ meV; $\Delta_2 \approx 154$ meV***
- ✓ **Most part of the free carrier spectral weight was removed due to the opening of CDW gaps. ω_p=40400 cm^{-1} at 300 K to 25500 cm^{-1} at 10 K.**

the Drude-Lorentz model to fit the optical conductivity,

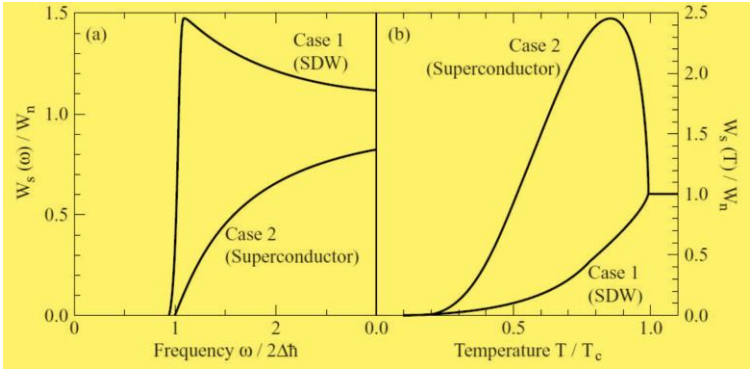
$$\epsilon(\omega) = \epsilon_\infty - \sum_s \frac{\omega_{ps}^2}{\omega^2 + i\omega/\tau_{Ds}} + \sum_j \frac{S_j^2}{\omega_j^2 - \omega^2 - i\omega/\tau_j}$$

Energy gaps for symmetry broken states：Coherence factors

$$\alpha_s = \int |M|^2 F(\Delta, E, E+\hbar\omega) N_s(E) N_s(E+\hbar\omega)[f(E) - f(E+\hbar\omega)]dE$$

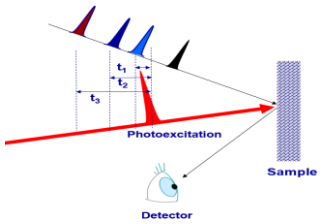
$$F(\Delta, E, E') = \frac{1}{2}(1 + \eta \frac{\Delta^2}{EE'})$$

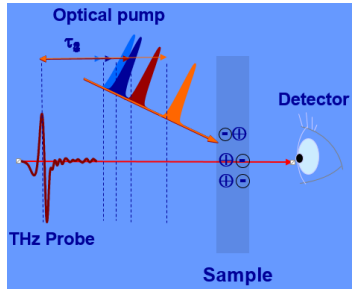
$$\eta = \begin{cases} -1 & caseI \\ +1 & caseII \end{cases}$$



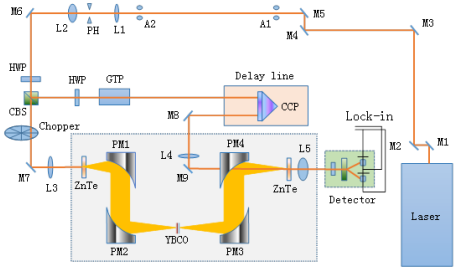
Efforts on time-resolved optical systems

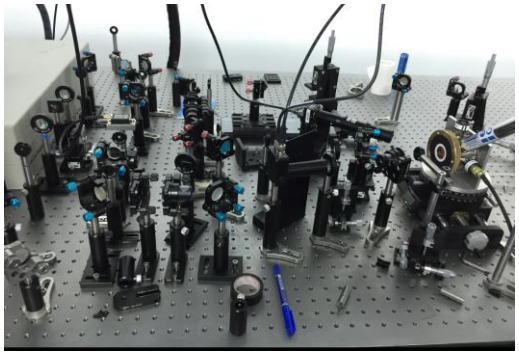
- Pump-probe with 80 MHz repetition rate
- Pump-probe with 1 KHz repetition rate

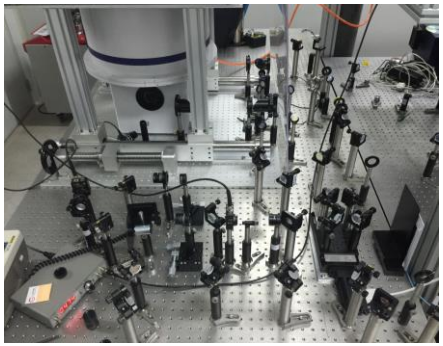


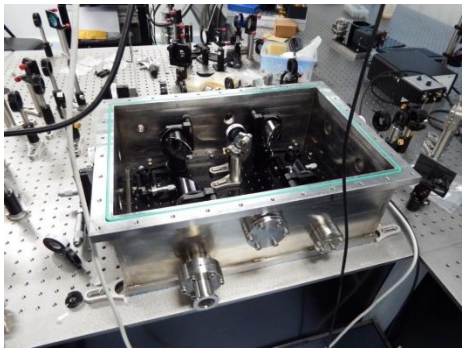


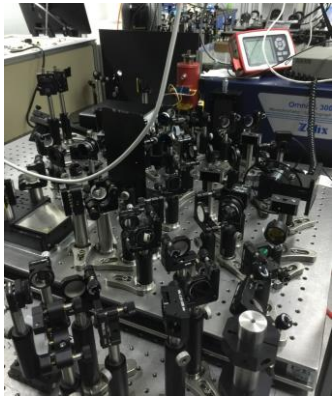
- Time domain terahertz spectroscopy
- Optical pump- THz probe
- THz pump- THz probe
- Mid-infrared pump –THz probe





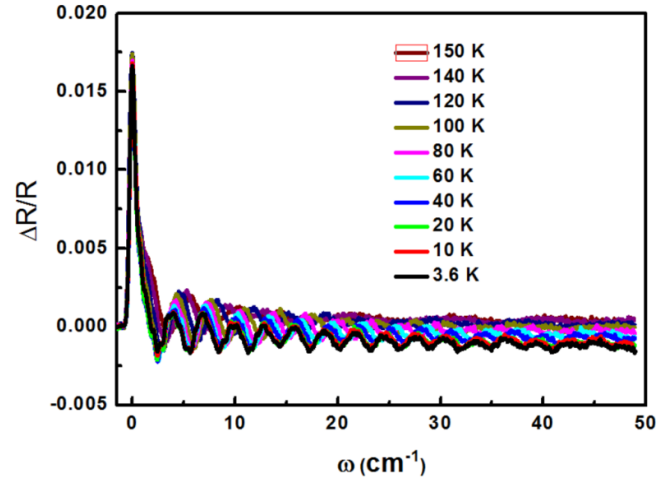


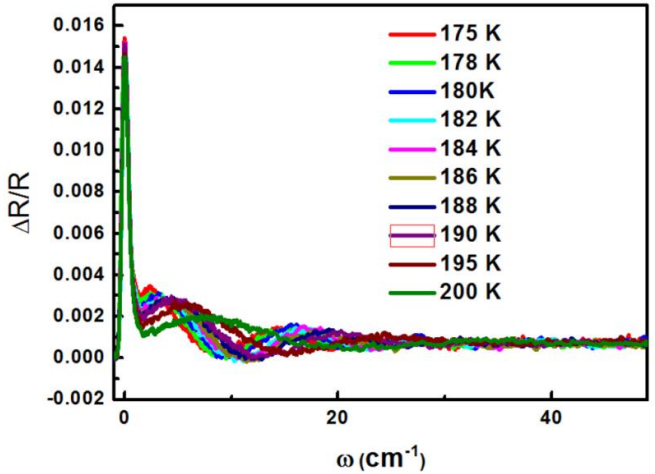


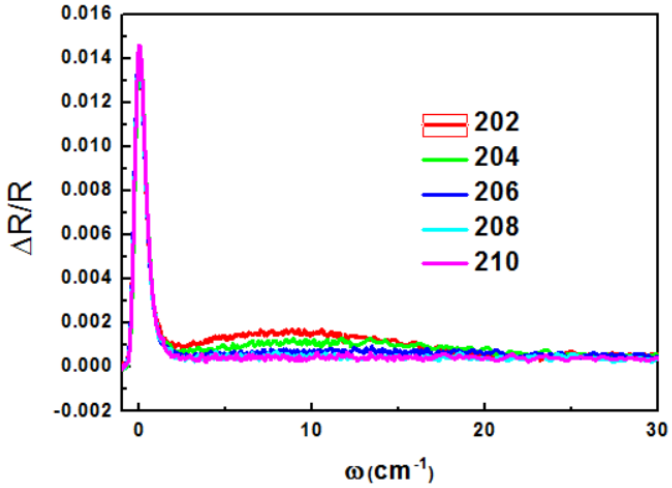


The transient photo-induced reflectivity ΔR/R as a function of time delay at different temperatures.

Ti:sapphire oscillator: 800 nm, pulse width 100 fs, repetition rate 80 MHz
Pump fluence is 1~ 2 μJ/cm^2







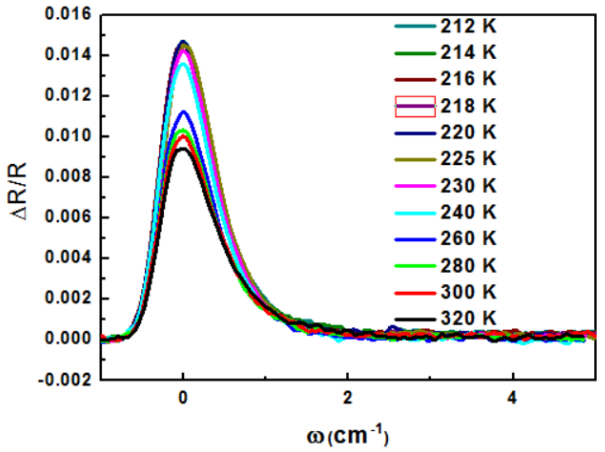
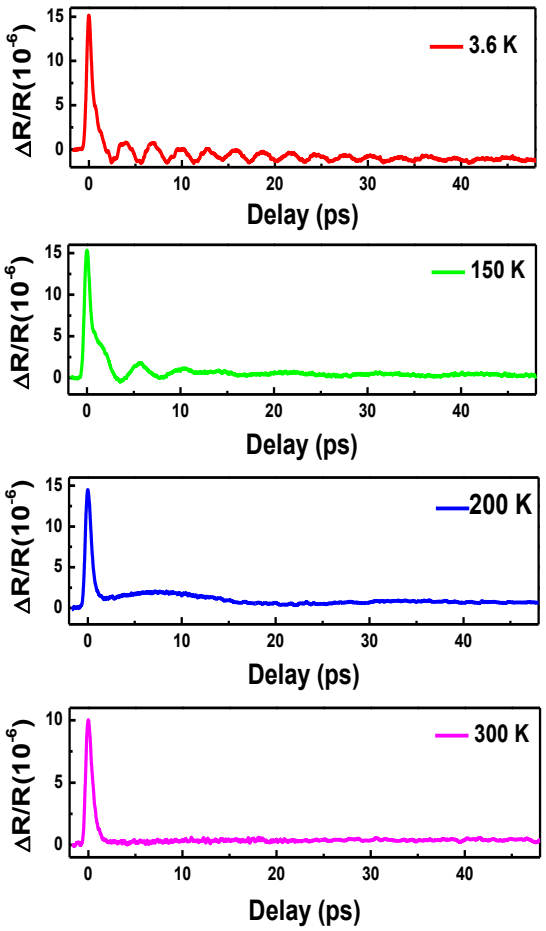
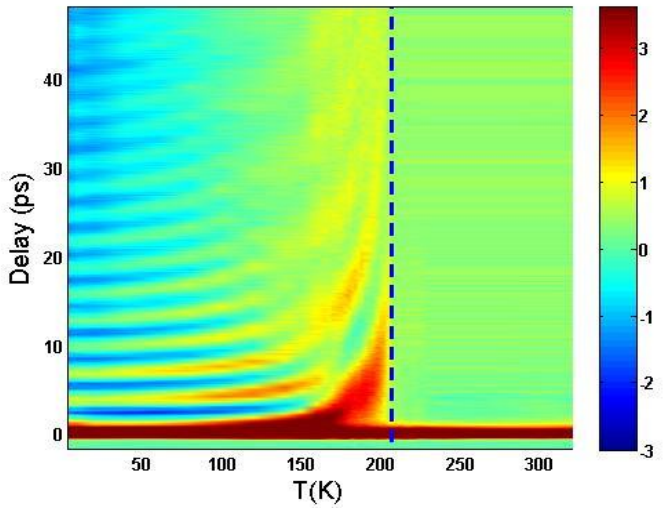
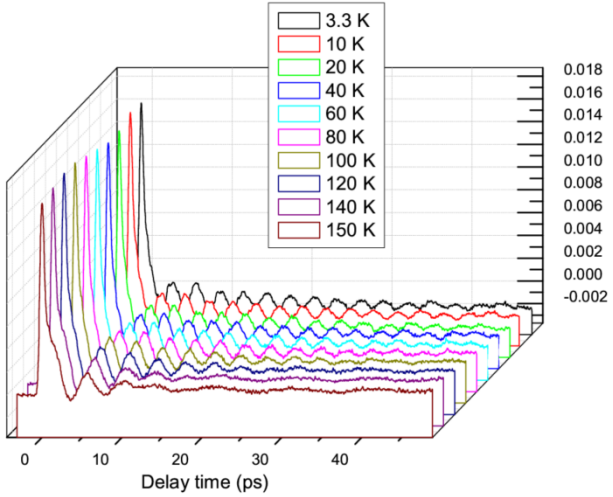


Photo-induced reflectivity



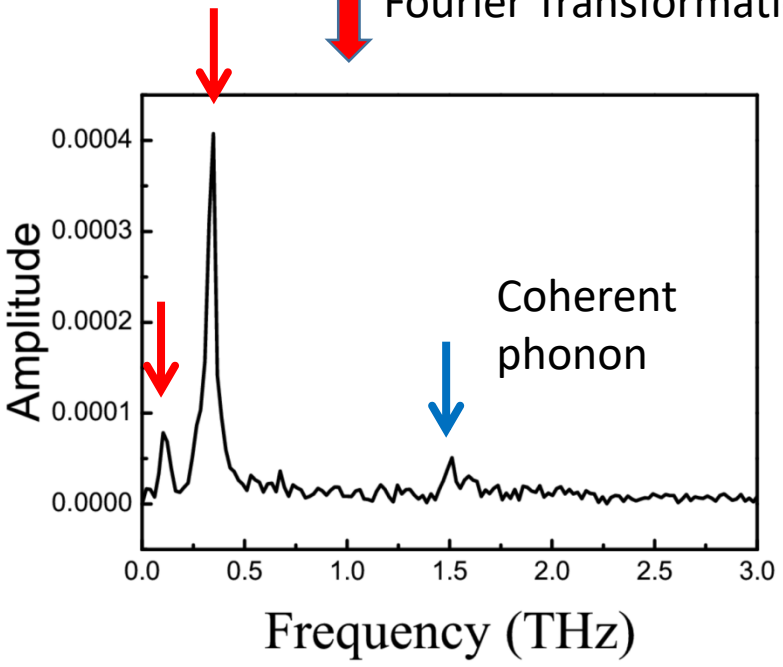


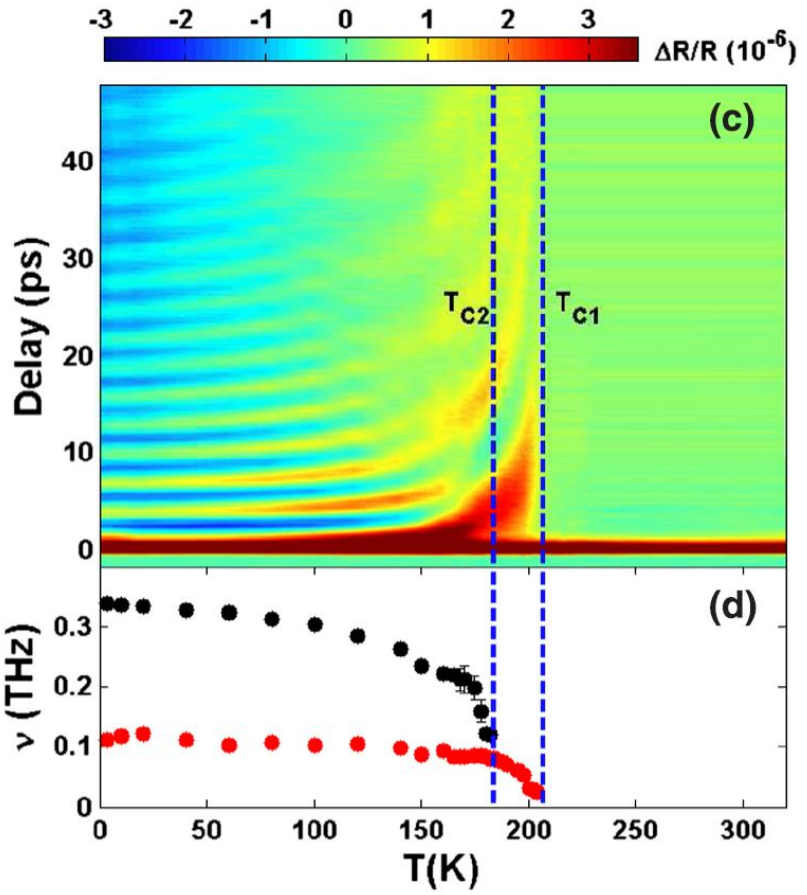
- ΔR/R = A exp (-t/τ) + C at high temperatures.
- Emergence of strong oscillations upon entering the CDW states.
- The amplitude A and relaxation time τ are almost constant across the two CDW transition temperatures.



amplitude modes

Fourier Transformation

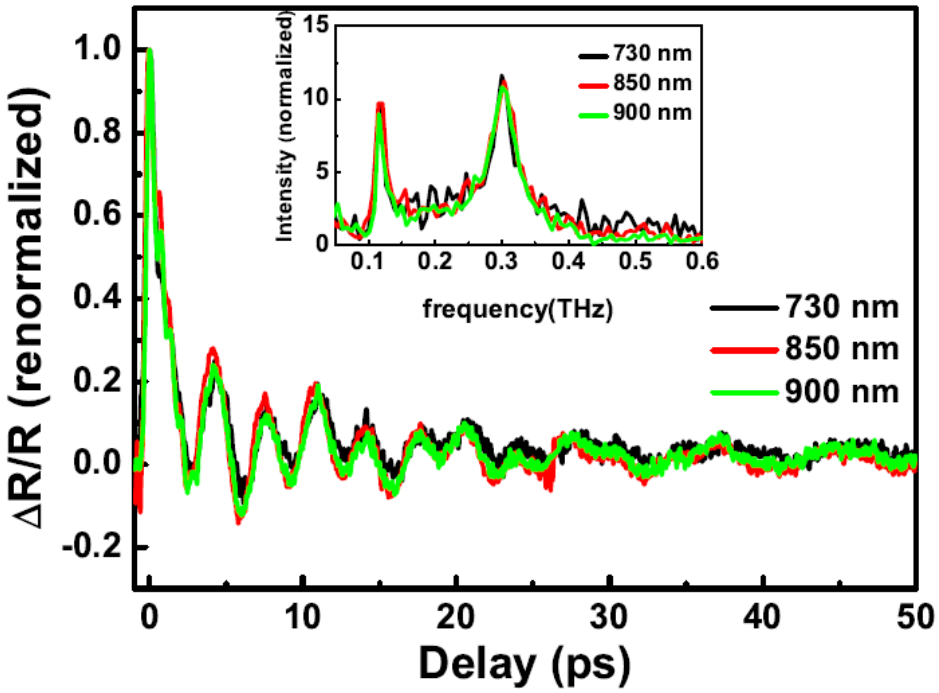


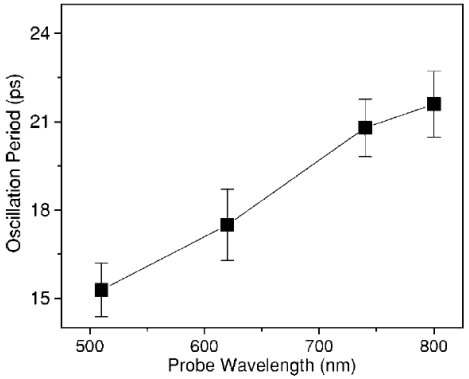


Extremely low energy scale of amplitude modes

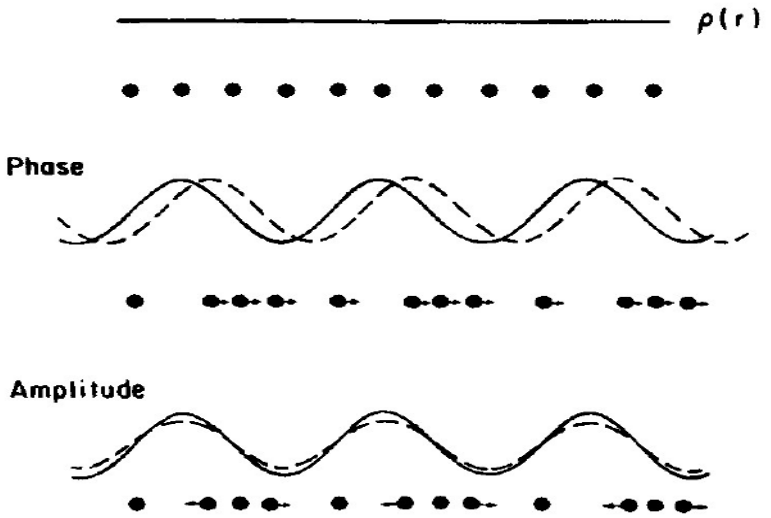
f1=0.12 THz (4 cm-1), corresponds to the higher CDW phase transition.
f2=0.34 THz (11 cm-1), corresponds to the lower CDW phase transition.

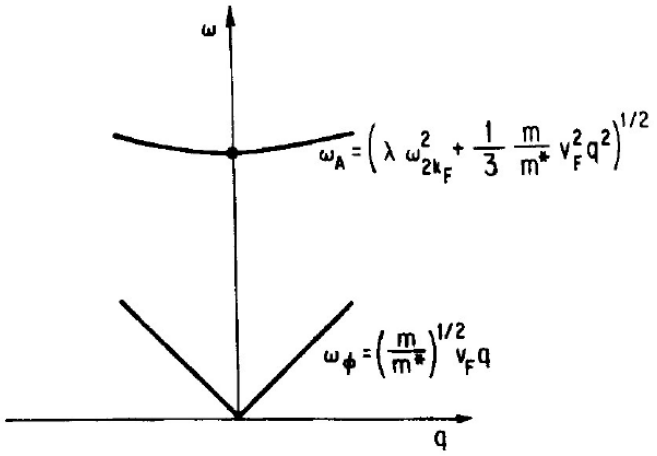
Coherent acoustic phonons can be excluded, because no energy shift was observed when the wave length of probe beam is changed.





Acoustic phonon for LSMO

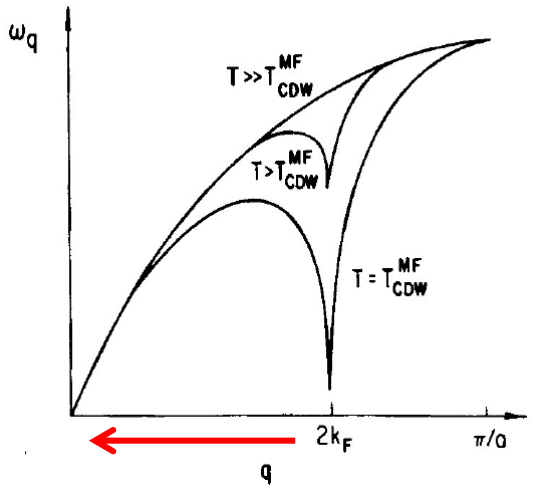


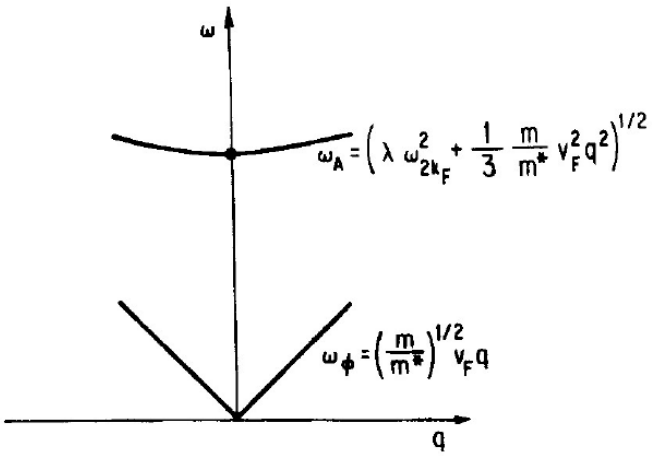


To our knowledge, such low energy scales of CDW amplitude modes (0.1 THz, 0.3 THz) were not seen/reported in any other CDW materials.

The low energy scale of amplitude mode (~4 cm-1) is hard to be detected by other techniques (e.g. Raman).

What is the reason behind?





George Gruner, Density waves in Solids, 1994

One possible scenario:

Because of the very small nesting wave vector $2k_F$, the acoustic phonon mode, which experiences a softening to zero frequency and triggers the CDW transition, also has very low energy scale.

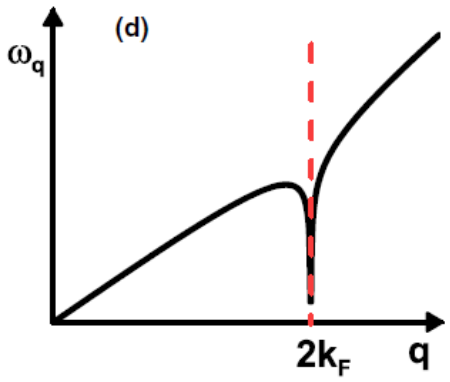
$\omega_{2KF} \approx v_{phonon} \cdot 2k_F$

At q=0, the amplitude mode energy

$\omega_A = \omega_{2KF} \cdot \lambda^{1/2}$.

very low energy scale of ω_A

It also explain why the lower CDW order has higher amplitude mode energy, because of bigger nesting wave vector.



Information about velocities of V_{F1} and V_{F2} two acoustic phonon branches which experience softening:

$$\frac{V_{F1}}{V_{F2}} = \frac{\omega_{2k_{F1}}/2k_{F1}}{\omega_{2k_{F2}}/2k_{F2}} \sim \frac{\nu_1/k_{F1}}{\nu_2/k_{F2}} \sim 2.2$$

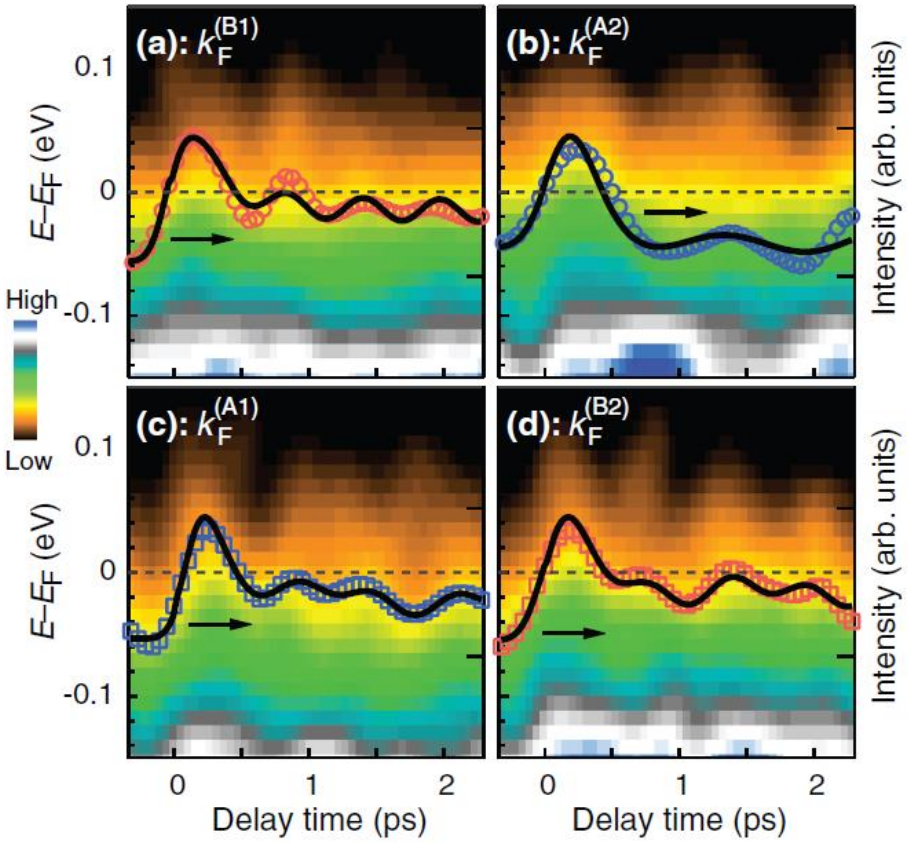
The in-plane one has higher velocity (about two times higher).

Conclusion

- **We clearly observe the energy gap formation below the CDW phase transition temperatures in optical conductivity.**
- **We observe two CDW amplitude modes with extremely low energy scales.**
- **The amplitude and relaxation time of the transient reflectivity remains unchanged across the two phase transitions**
- **Those unusual properties are closely linked to the extremely small nesting wave vectors of the two CDW orders.**
- **Possible effect of AM on low energy physical properties?**

Thank you !

K0.3MoO3


(a): $k_F^{(B1)}$
(b): $k_F^{(A2)}$
(c): $k_F^{(A1)}$
(d): $k_F^{(B2)}$
$E-E_F$ (eV)
0.1
0
-0.1
High
Low
Intensity (arb. units)
0
1
2
Delay time (ps)