Beyond Quasiparticles: computing spectral functions

subtitle: ***Surprising errors of GW and Migdal;***
surprising success of "cumulants".

Philip B. Allen
Stony Brook University

GW =
Migdal =

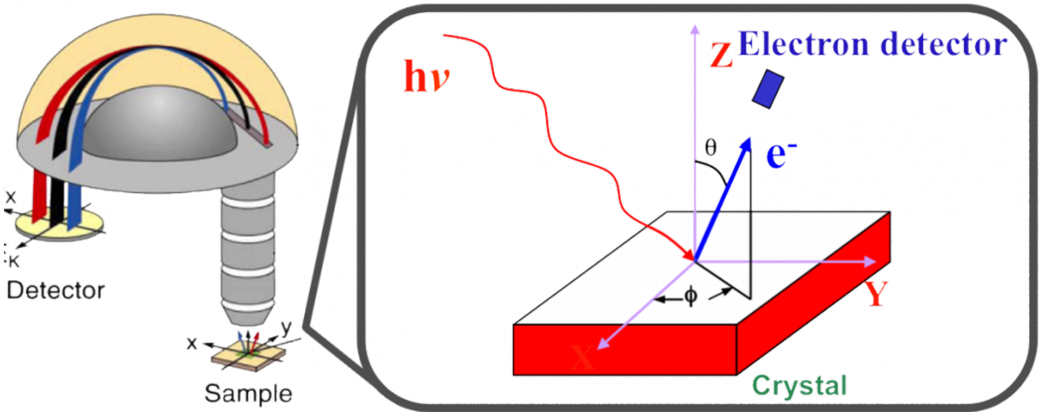
ES18 and PCTC18
Philadelphia, June 10-14, 2018

0. What's a quasiparticle (QP)?
1. Core levels with plasmon satellites
2. Valence levels with plasmon satellites
3. Valence levels with phonon satellites
   a. Franck-Condon
   b. Fröhlich polaron
   c. Low T metals

Q: What's a quasiparticle (QP)?
A: Who knows?

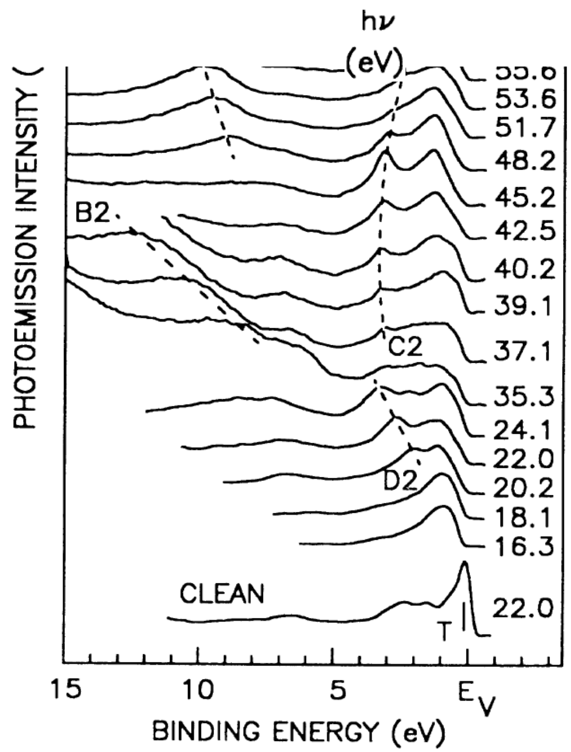
Quasiparticle Energies

Hole states in silicon measured by angle-resolved photoemission (**ARPES**)
Expt: D. H. Rich, T. Miller, G. E. Franklin, and T. -C. Chiang (PRB,1989)
Theory: J. R. Chelikowsky and M. L. Cohen, Phys. Rev. B 14, 556 (1976).

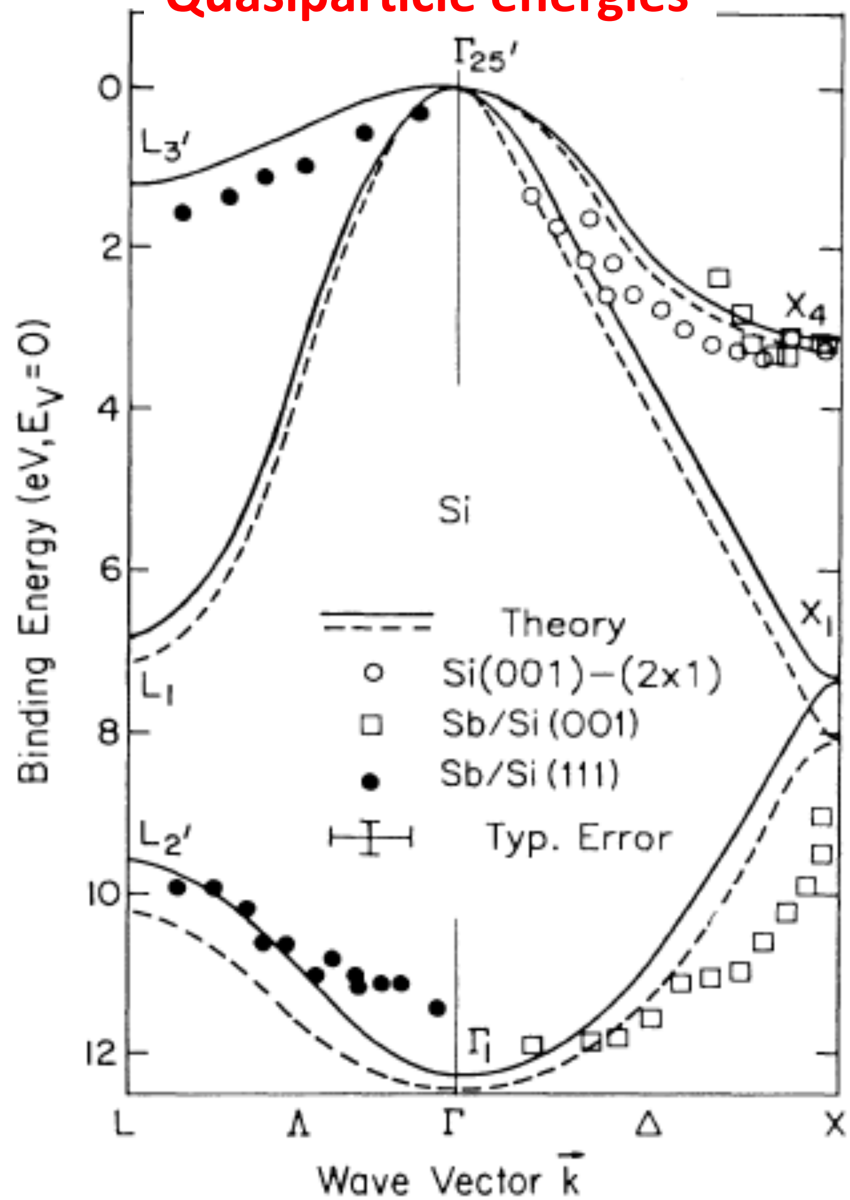


arpes.Stanford.edu

The question: Should we be analyzing the shape of the measured spectrum? Or just the peak positions (QP energies)?



Quasiparticle energies



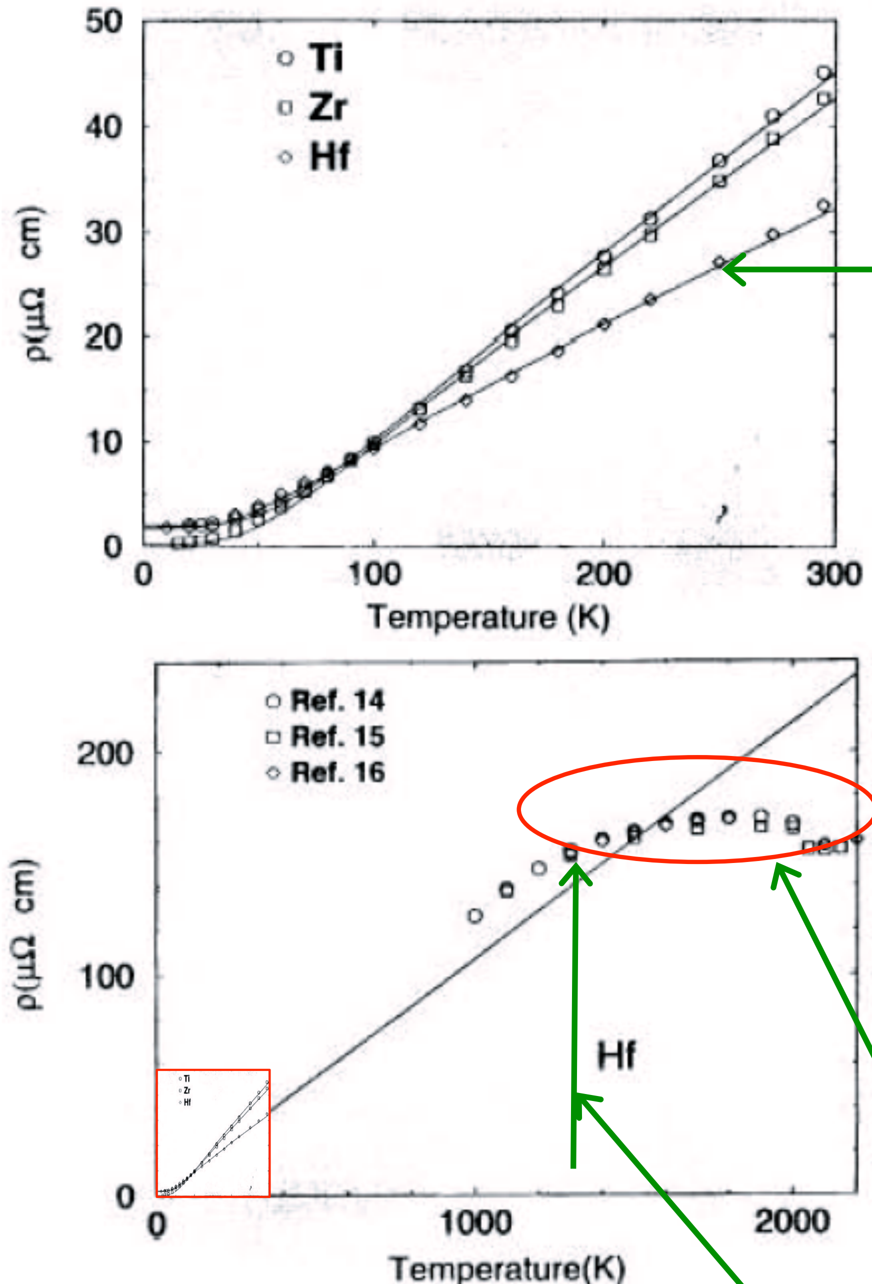
Q: Who knows what's a quasiparticle?
A: Landau knows.

a. Propagating particle/wave state (energy ε_k, equilibration rate $1/\tau_k$)

$$\varepsilon_k \gg 1/\tau_k$$

b. 1 to 1 correspondence with states of a non-interacting gas
c. Low energy properties of solids determined by distribution of QP'S.

"Resistivity Saturation"



Bloch-Grüneisen fits

1) Downward deviation from Bloch-Grüneisen seen when ℓ < 10Å

2) Quasiparticle picture works beautifully until it fails.

3) High T failure is common in ordinary metals, BCS superconductors

"Saturation"

Temperature where mfp is (predicted to be) 10Å

Successful quasiparticle theories

a. Equilibrium

* $$C_{\rm el}(T) = \frac{d}{dT}\sum_k E_k f(E_k) \rightarrow \gamma T$$

$$C_{\rm vib}(T) = \frac{d}{dT}\sum_k \omega_Q n(\omega_Q) \rightarrow AT^3$$

f (and n) are equilibrium Fermi-Dirac (and Bose-Einstein) distributions

b. Nonequilibrium

** $$\sigma(T) = \frac{1}{\rho(T)} = \frac{d}{d\vec{E}}(-e)\sum_k \vec{v}_k F_k \rightarrow \frac{1}{\rho_1 T} \quad \text{(high T)}$$

$$\kappa(T) = \frac{d}{d\vec{\nabla}T}\sum_Q \hbar\omega_Q \vec{v}_Q N_Q \rightarrow \frac{\kappa_1}{T} \quad \text{(high T)}$$

F (and N) are non-equilibrium Fermi-Dirac (and Bose-Einstein) distributions

Glitch (rarely mentioned): The QP's of * differ from those of **.

** Resistivity uses band energies ε_k

* Specific heat uses renormalized band energies $E_k=\varepsilon_k/(1+\lambda_k)$

Spectral Function
(retarded Green's function)

using Dyson equation:

$$G(k,\omega) = \frac{1}{\omega - \epsilon_k - \Sigma(k,\omega)}$$

$$A(k,\omega) \equiv -\frac{1}{\pi}\mathrm{Im}G_R(k,\omega)$$

The simplest version of a quasiparticle:

$$\mathcal{R}e\Sigma(k,\omega=\epsilon_k) = \Delta\epsilon_k$$

Σ = **"self-energy"**

$$\mathcal{I}m\Sigma(k,\omega=\epsilon_k) = -1/2\tau_k$$

$$A(k,\omega) \approx \frac{1/2\pi\tau_k}{(\omega - \epsilon_k - \Delta\epsilon_k)^2 + (1/2\tau_k)^2}$$

Lorentzian line-shape;
~ **"quasiparticle" peak**

$$1 = \int_{-\infty}^{\infty} d\omega A(k,\omega)$$

Total "spectral weight" = 1. Always true, **but** the QP peak **never** has all the spectral weight.

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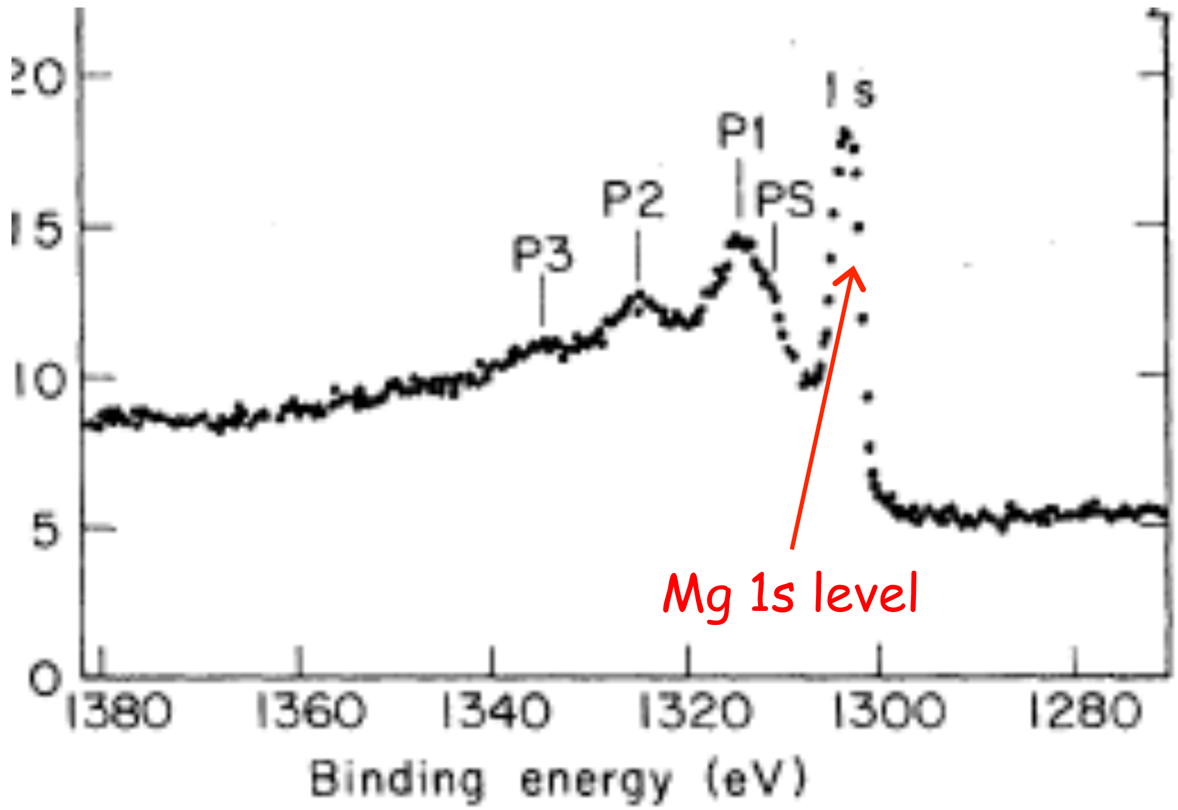
Beyond quasiparticles: 1. core level plasmon satellites

PHYSICAL REVIEW B VOLUME 11, NUMBER 2 15 JANUARY 1975

Many-body effects in x-ray photoemission from magnesium*

L. Ley,† F. R. McFeely, S. P. Kowalczyk, J. G. Jenkin,‡ and D. A. Shirley



Plasmon satellites, $E = E_{1s} - n\omega_p$ (plasmon emission)
Binding energies are given relative to E_F.

Langreth's theory

Singularities in the X-Ray Spectra of Metals

Phys. Rev. B1, 471 (1970)

DAVID C. LANGRETH

Model Hamiltonian
dispersionless core hole
plasmons modeled as pure Bosons

$$\mathcal{H} = \epsilon c^\dagger c + \sum_q c c^\dagger g_q (a_q + a_q^\dagger) + \sum_q \omega_q a_q^\dagger a_q$$

$$f_q = g_q / \omega_q$$

Exact solution in time domain:

$$\langle 0 | \exp(i\tilde{H}t) | 0 \rangle = e^{-i\Delta\epsilon t} e^{-\Sigma_q f_q^2} \exp\left[\sum_q f_q^2 e^{i\omega_q t}\right]$$

Cumulant

Transform to frequency domain:

$$\sum_{n=0}^{\infty} (e^{-a} a^n / n!) \delta(\omega - \epsilon - a\omega_p + n\omega_p)$$

$$a = \sum_q f_q^2 = \omega_p^{-2} \sum_q g_q^2$$

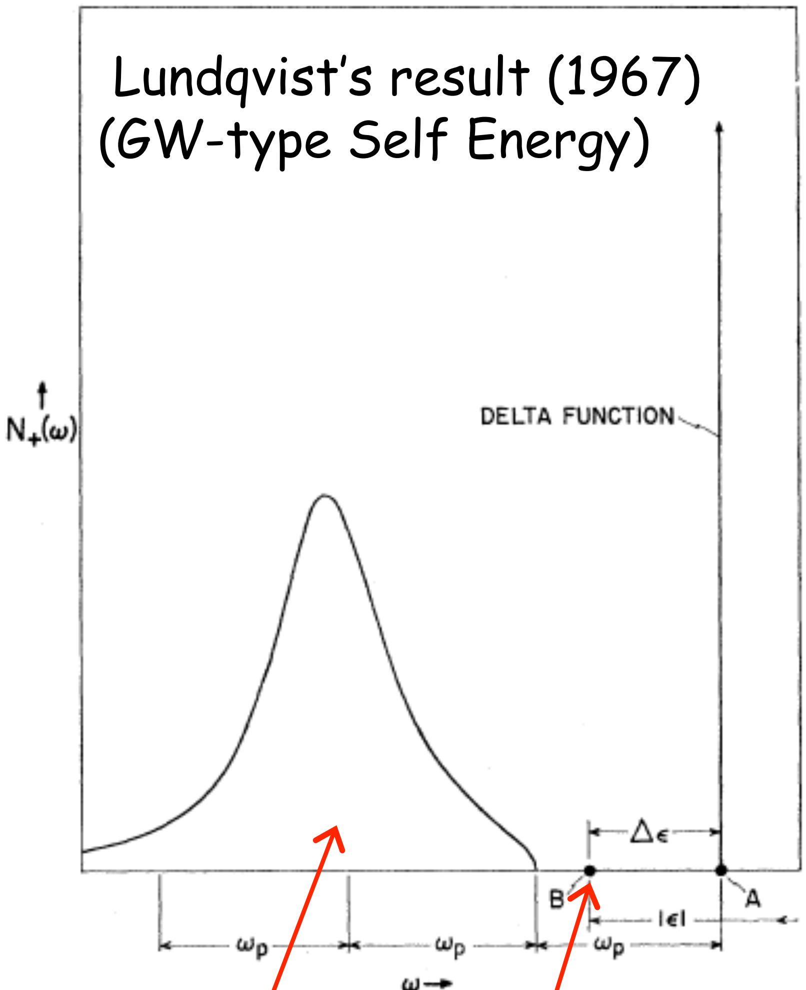
Plasmon satellites

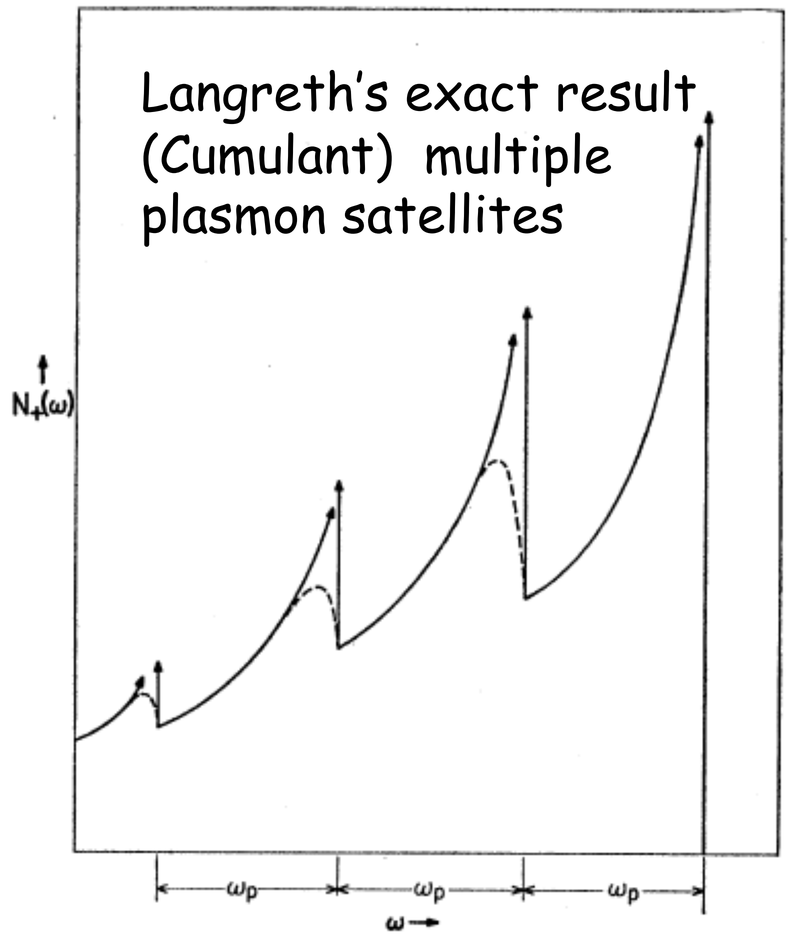
Renormalized core hole energy

GW versus cumulant

Singularities in the X-Ray Spectra of Metals

DAVID C. LANGRETH





Renormalized core energy E_c

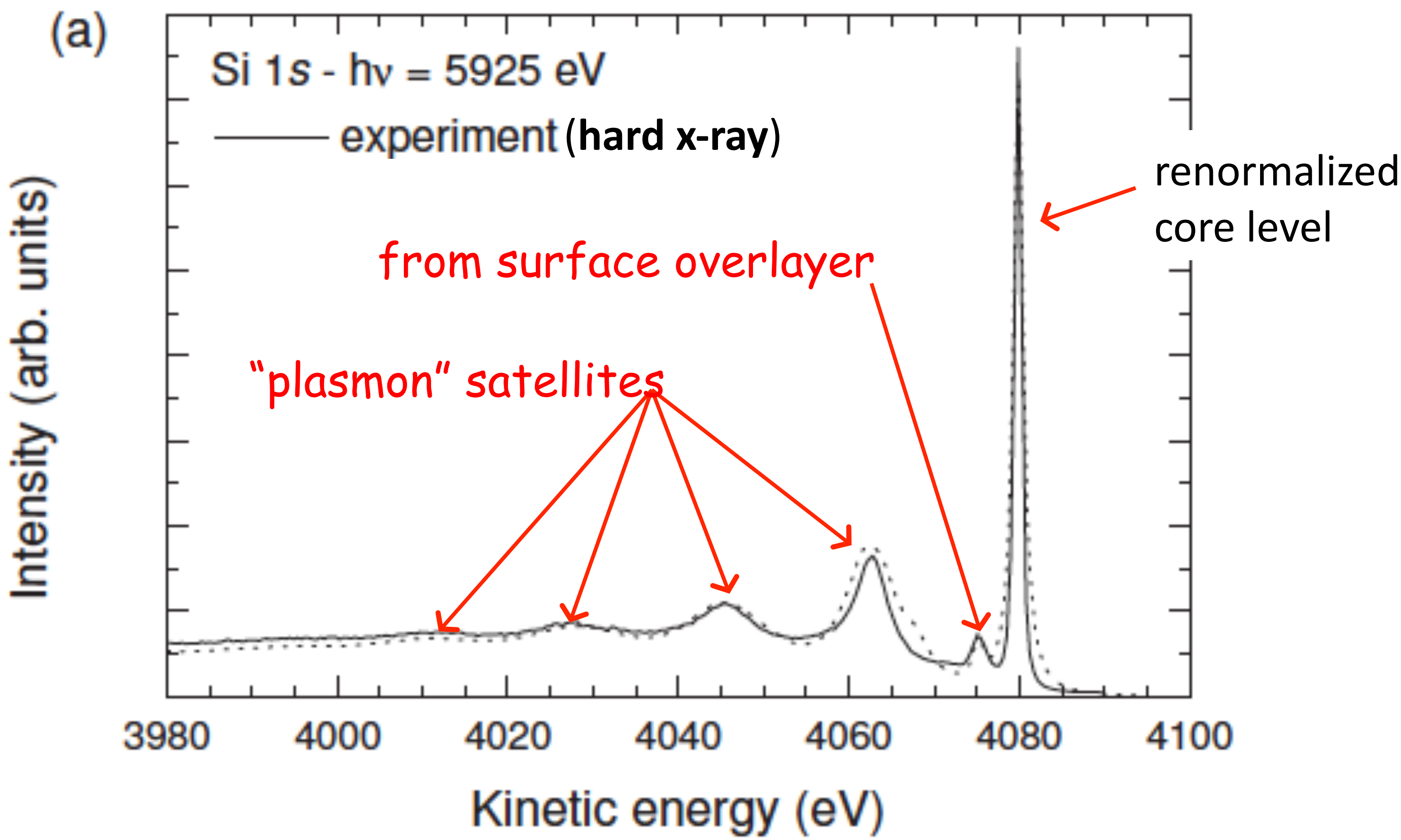
Only one "plasmon" (e-h pairs) satellite, plasmon peak at $E_c - f\omega_p$ with $f > 1$

Not just metals.

PHYSICAL REVIEW B **76**, 085422 (2007)

Comparison of hard and soft x-ray photoelectron spectra of silicon

F. Offi,[1] W. S. M. Werner,[2] M. Sacchi,[3] P. Torelli,[4] M. Cautero,[5] G. Cautero,[5] A. Fondacaro,[1,*] S. Huotari,[6] G. Monaco,[6] G. Paolicelli,[4] W. Smekal,[2] G. Stefani,[1] and G. Panaccione[7]



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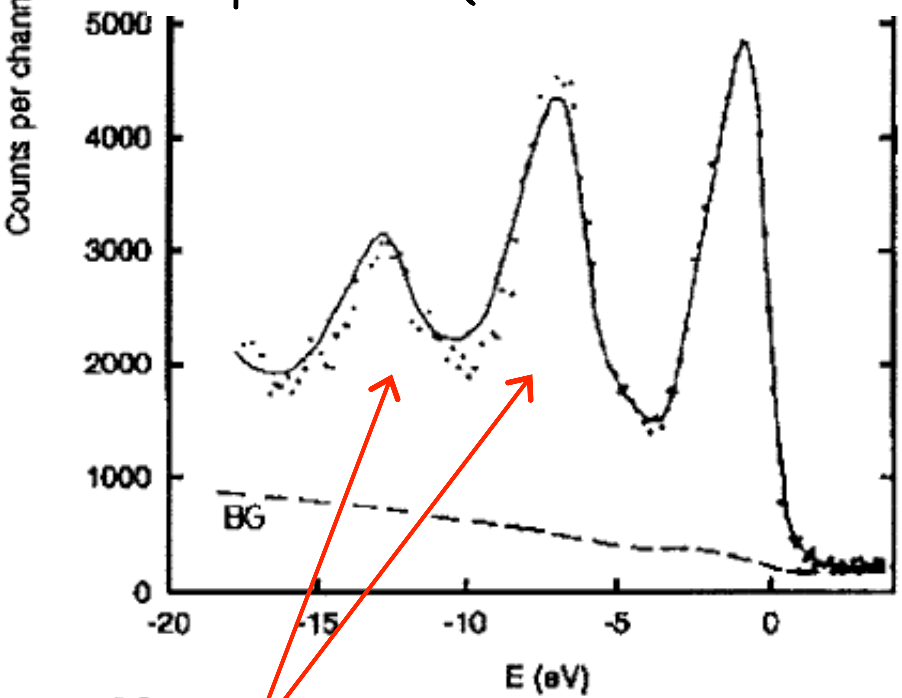
Beyond quasiparticles: 2. valence electron plasmon satellites

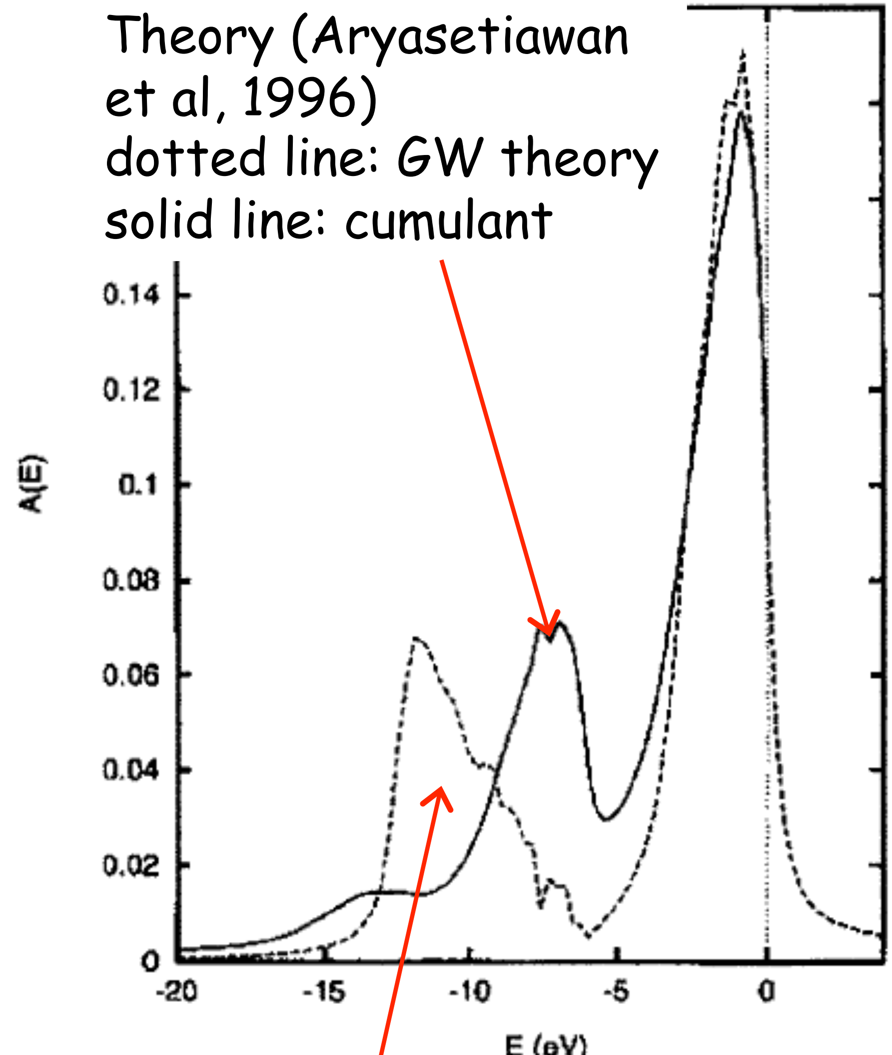
Sodium metal
photoemission from valence states

Experiment (Steiner et al. 1979)



"plasmon" satellites

Theory (Aryasetiawan et al, 1996)
dotted line: GW theory
solid line: cumulant



GW gives a single "e-h pair" satellite separated too far from the QP part

"Ad hoc" cumulant formula, derived

$$G(k,\omega) = \frac{1}{G_0^{-1} - \Sigma} = G_0 + G_0 \Sigma G_0 + \ldots$$ Dyson expansion

$$G(k,t) = G_0 e^{C(k,t)} = G_0 + G_0 C + \ldots$$ Cumulant expansion

First order expansions.
Aryasetiawan-Hedin-Karlsson (1996) methodology:

1. Use a theory that gives a **good QP energy**.
2. Use the **corresponding** Σ to construct **C(t)** and **G(t)**.
3. Use the resulting **G(k,ω)** to get **A(k,ω)**.
4. **Use a retarded Green's function & Σ.**

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Beyond quasiparticles: 3. Phonon satellites

(only seen, so far, for doped electrons or holes)

a) Franck-Condon satellites (self-trapped; no quasiparticle)
b) Fröhlich polaron satellites
c) Metals: Low T mass renormalization (no satellites yet)

PRL 106, 137602 (2011) PHYSICAL REVIEW LETTERS

a) Franck-Condon

Properties of Hydrogen Terminated Diamond as a Photocathode

J. D. Rameau,[1] J. Smedley,[1] E. M. Muller,[2] T. E. Kidd,[1,3] and P. D. Johnson[1]

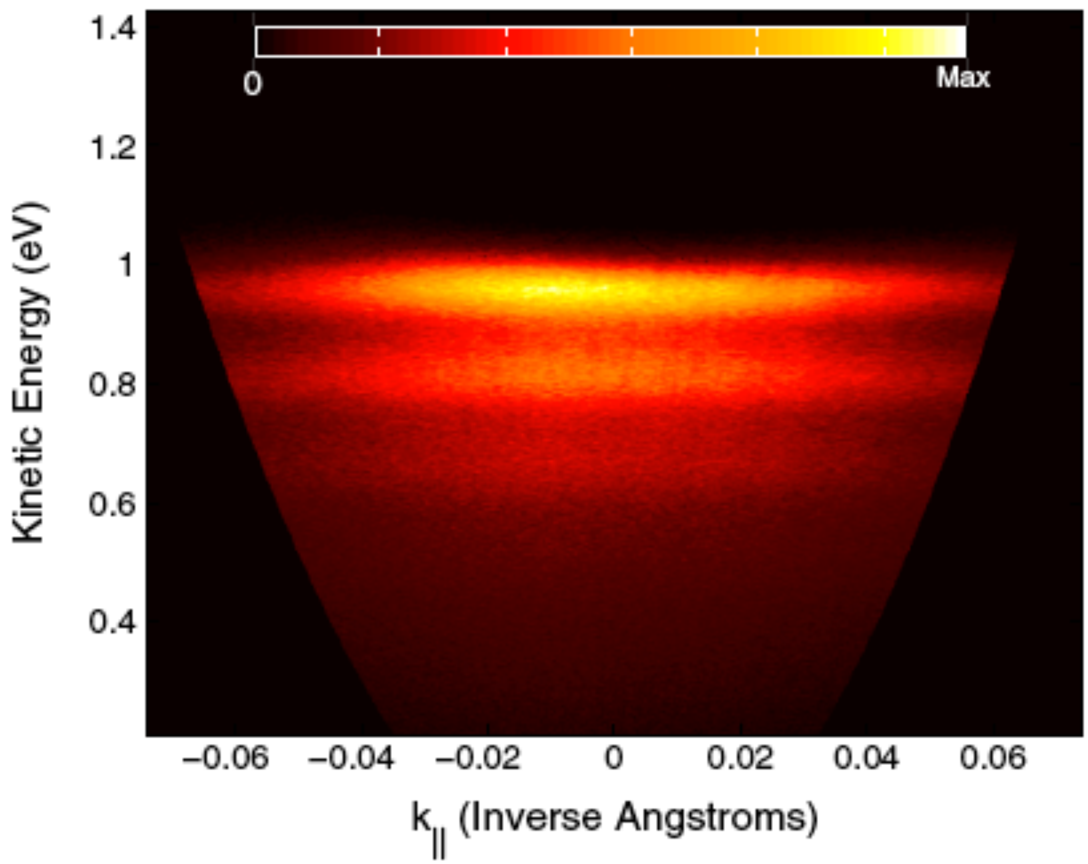
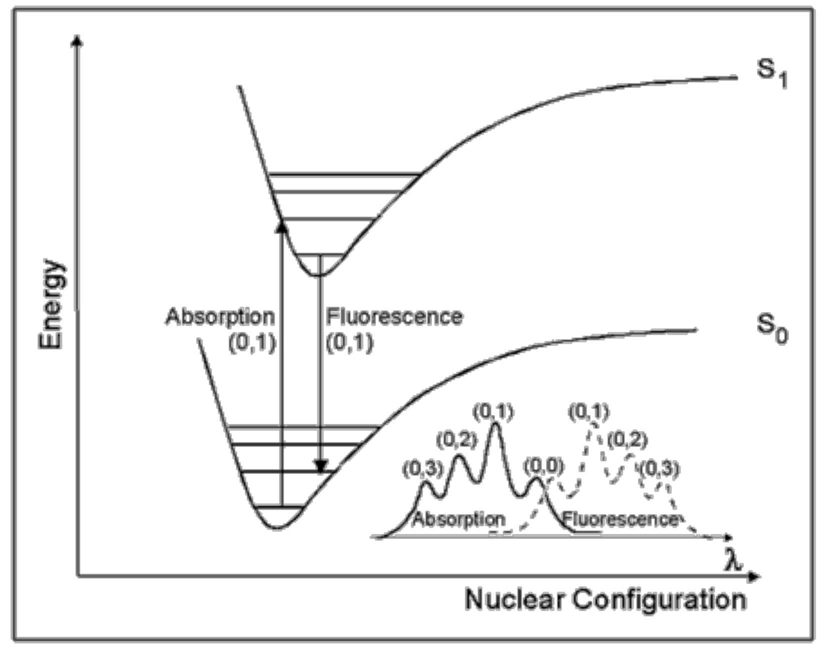


FIG. 2 (color online). ARPES spectrum acquired using the 6.01 eV laser. The color bar shows relative intensity. The overall "bowl" shape of the spectrum results from scaling between emission angle and $k_{||}$.

Franck-Condon Energy Level Diagram



http://photobiology.info/
Photochem.html

Properties of Hydrogen Terminated Diamond as a Photocathode
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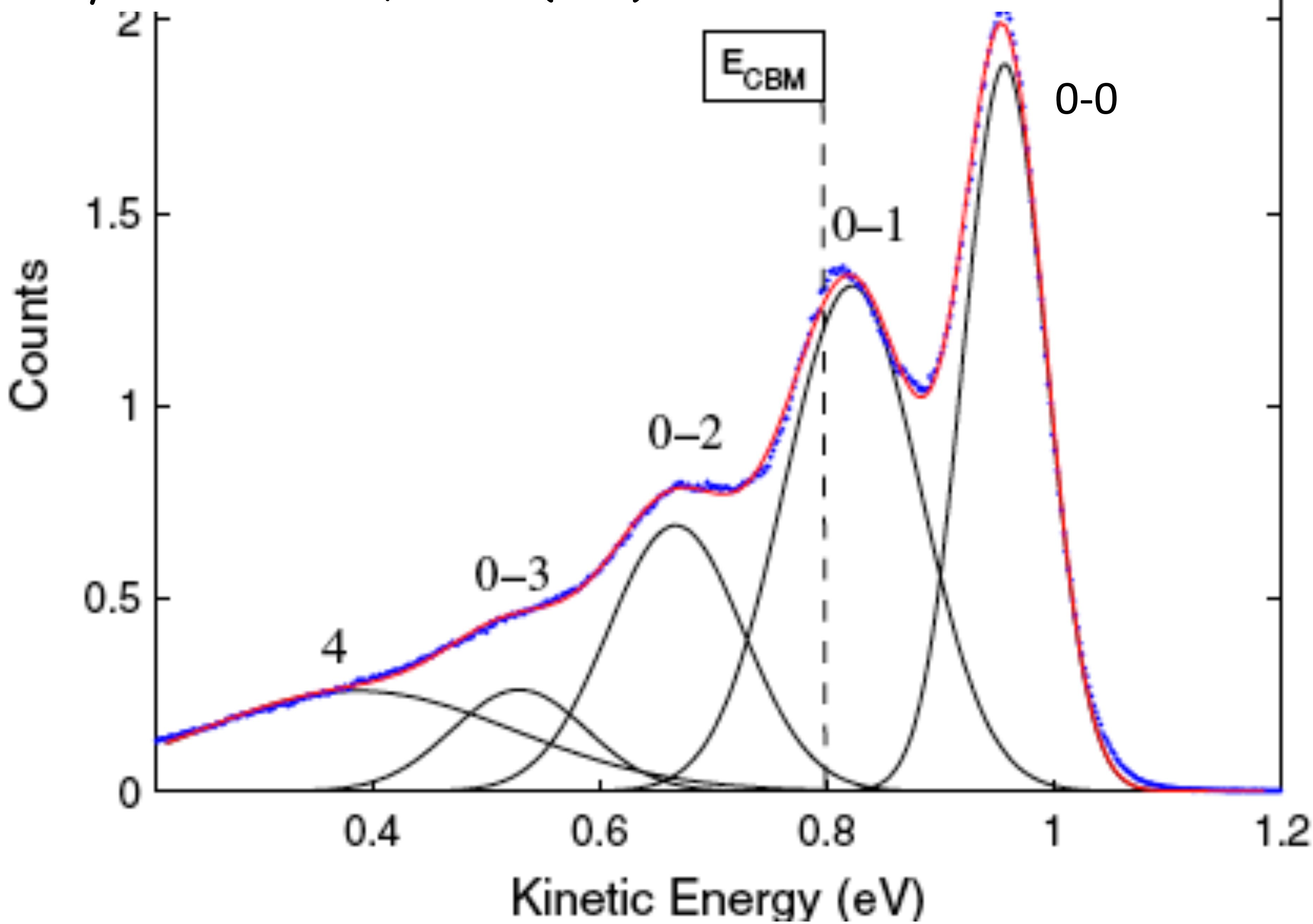

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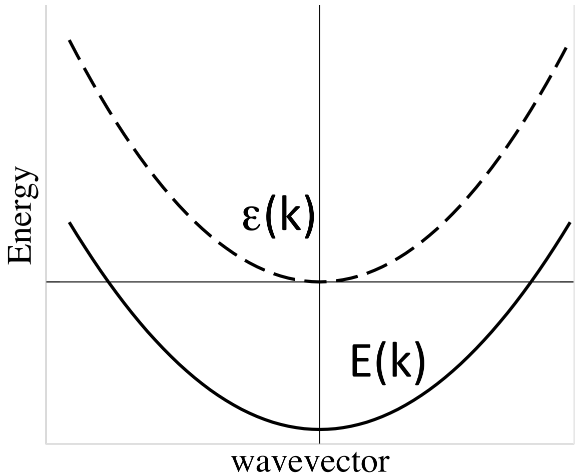
2. Fröhlich polaron (A simplified model, often applied only to **single electrons** at the bottom of the conduction band.)



To **lowest order** in the Fröhlich coupling constant α (Rayleigh-Schrödinger, "RS") :

$$\varepsilon(k=0) = 0$$

$$E(k=0) = -\alpha\hbar\omega_{LO}$$

$$E(k) = -\alpha\hbar\omega_{LO} + \varepsilon(k)/(1+\alpha/6)$$

$\Sigma(k,\omega)$ =  "Migdal"

$$\Sigma(k,\varepsilon_{k=0}) = -\alpha\hbar\omega_{LO} \text{ (R-S)}$$

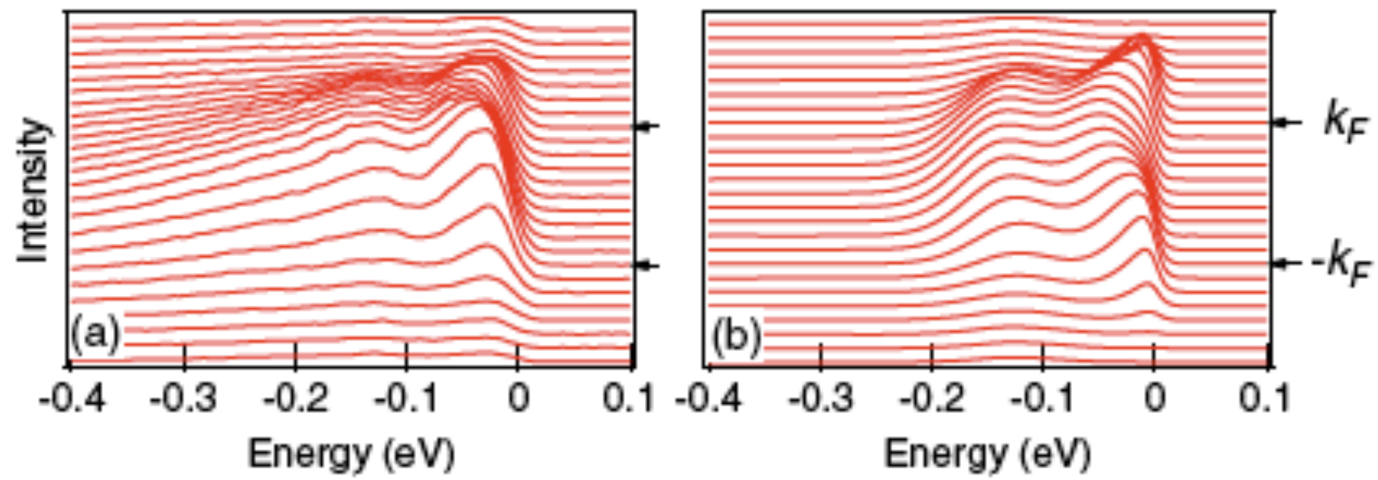
Material	α	Material	α
InSb	0.023	KI	2.5
InAs	0.052	TlBr	2.55
GaAs	0.068	KBr	3.05
GaP	0.20	RbI	3.16
CdTe	0.29	$Bi_{12}SiO_{20}$	3.18
ZnSe	0.43	CdF_2	3.2
CdS	0.53	KCl	3.44
AgBr	1.53	CsI	3.67
AgCl	1.84	$SrTiO_3$	3.77
α-Al_2O_3	2.40	RbCl	3.81

Devreese, in Encyclopedia of Physics (2005)

PRL 110, 196403 (2013) PHYSICAL REVIEW LETTERS week ending 10 MAY 2013

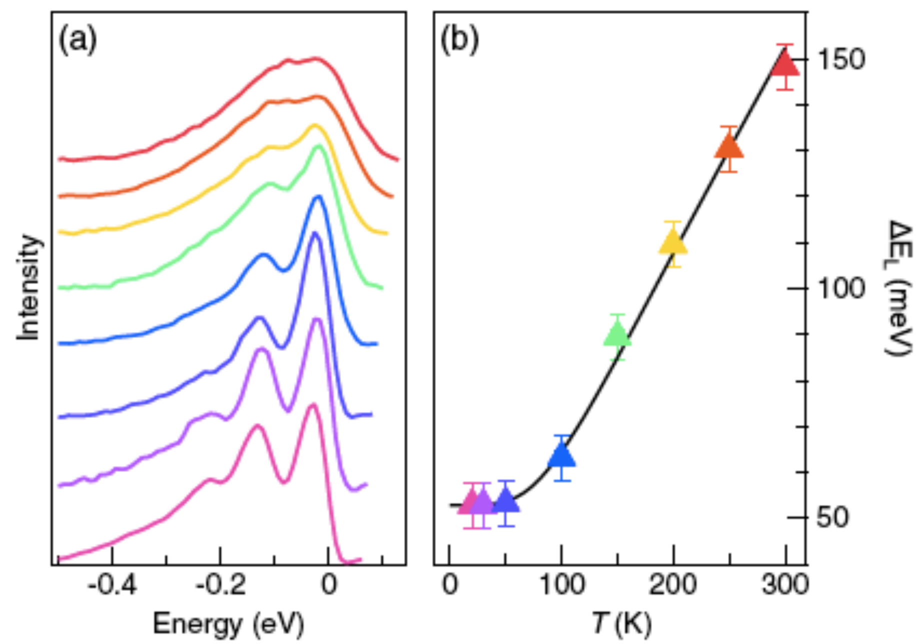
Tunable Polaronic Conduction in Anatase TiO_2

S. Moser,[1,2] L. Moreschini,[1] J. Jaćimović,[2] O. S. Barišić,[3] H. Berger,[2] A. Magrez,[2] Y. J. Chang,[1,4] K. S. Kim,[1]
A. Bostwick,[1] E. Rotenberg,[1] L. Forró,[2] and M. Grioni[2]



ARPES, Lightly electron-doped TiO_2
(anatase form)

Phonon satellites are T-dependent,
as is the band energy.

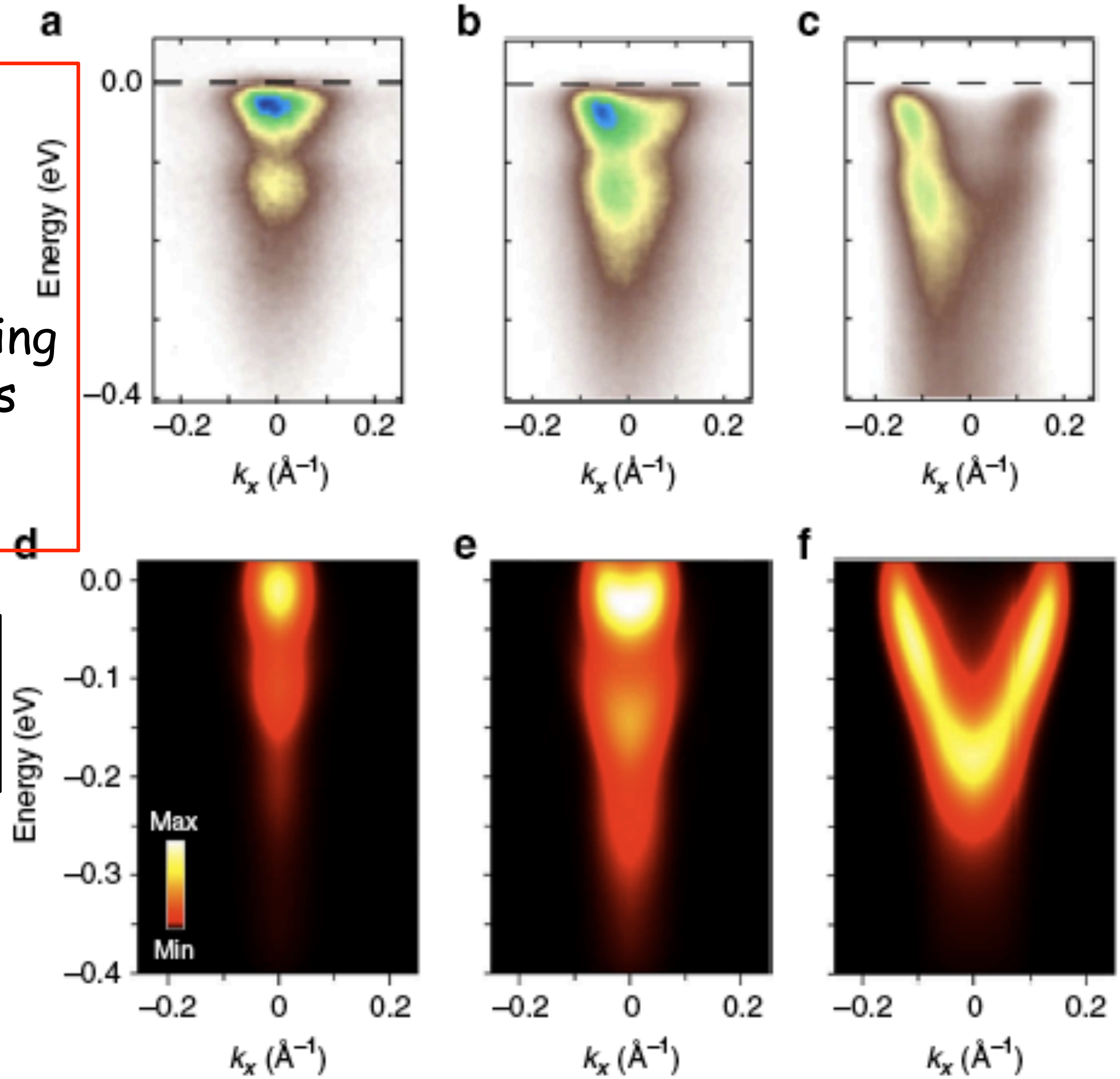


Origin of the crossover from polarons to Fermi liquids in transition metal oxides
C Verdi, F Caruso, F Giustino - Nature communications, 2017

Measured ARPES
in anatase TiO_2
(Moser *et al.*, 2013)

With increasing n-doping
the spectrum evolves
from "polaron" gas
to "metal".

"*ab initio*" computations
using cumulants
(Verdi *et al.*, 2017)

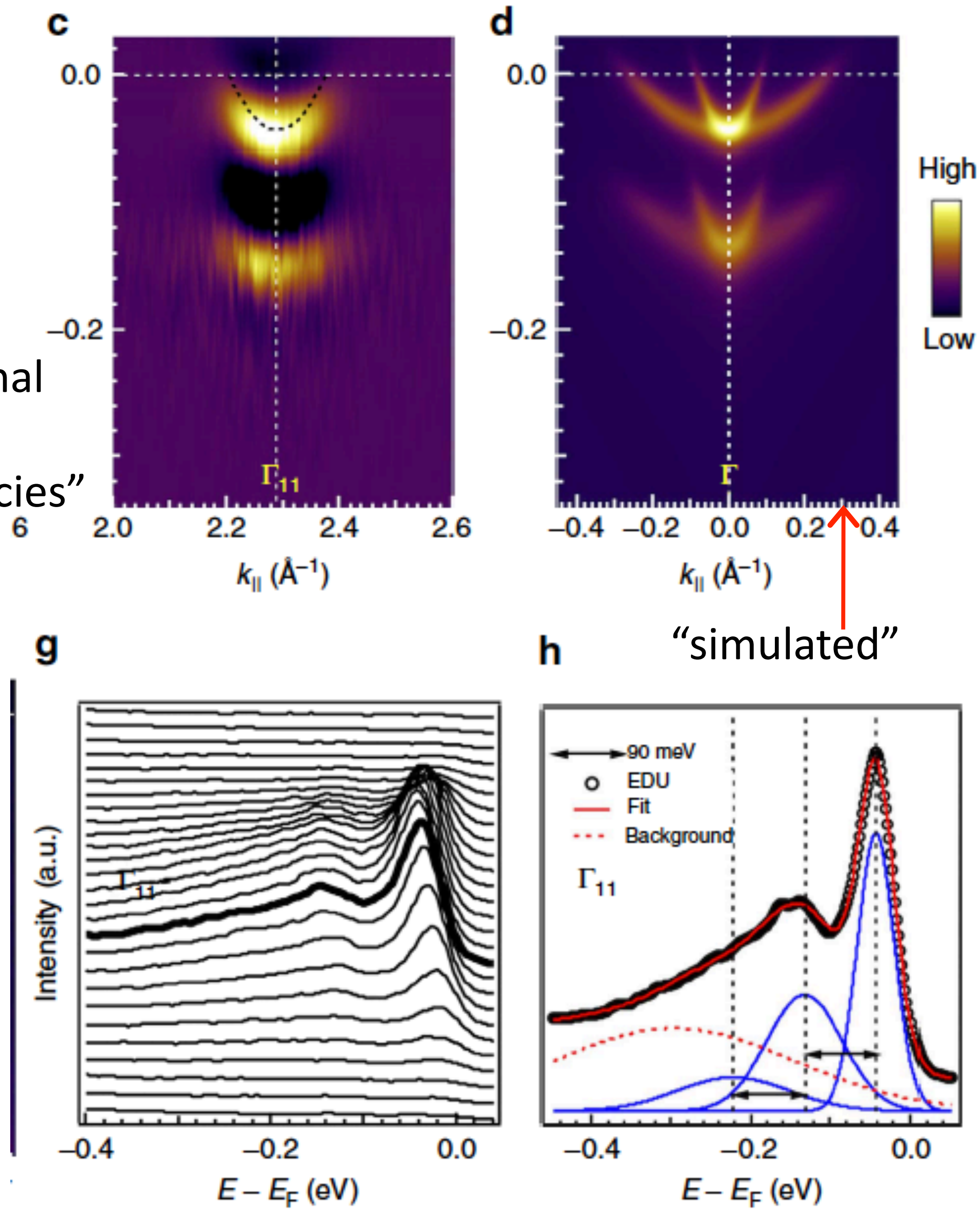


Observation of a two-dimensional liquid of Fröhlich polarons at the bare $SrTiO_3$ surface

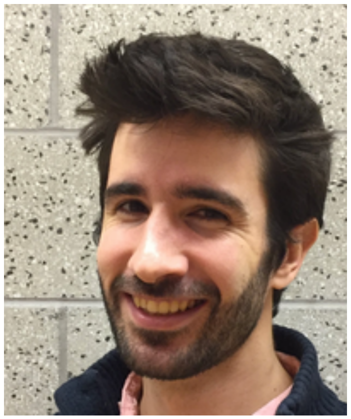
Chaoyu Chen, José Avila […] Maria C. Asensio

Nature Communications **6,** 8585 2015

"these Fröhlich polarons are two-dimensional and only exist with inversion symmetry breaking by two-dimensional oxygen vacancies"



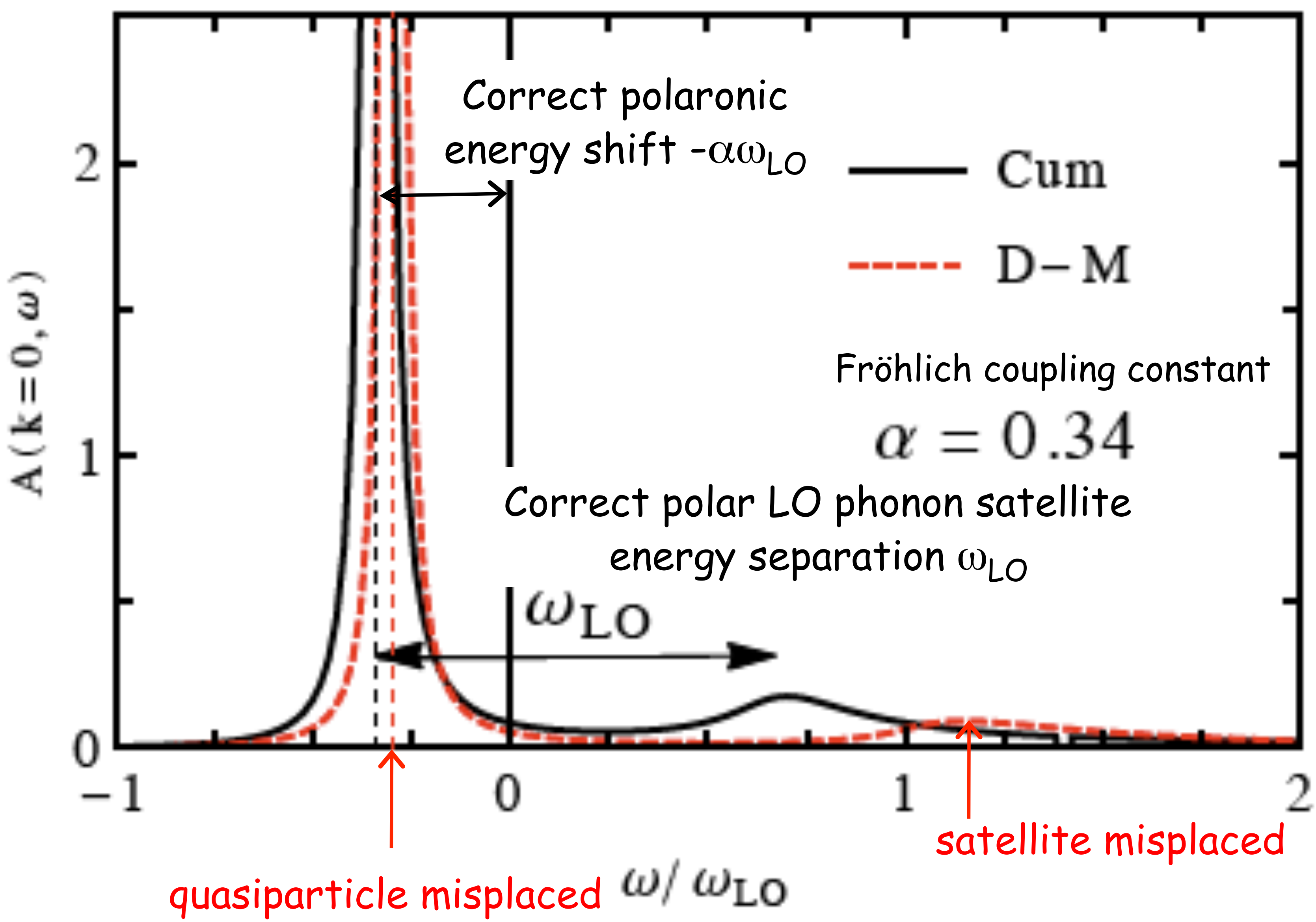
J. P. Nery, P. B. Allen, G. Antonius, L. Reining, A. Miglio, and X. Gonze
Quasiparticles and phonon satellites in spectral functions of semiconductors and insulators: Cumulants applied to the full first-principles theory and the Fröhlich polaron
Phys. Rev. B 97, 115145 (2018)

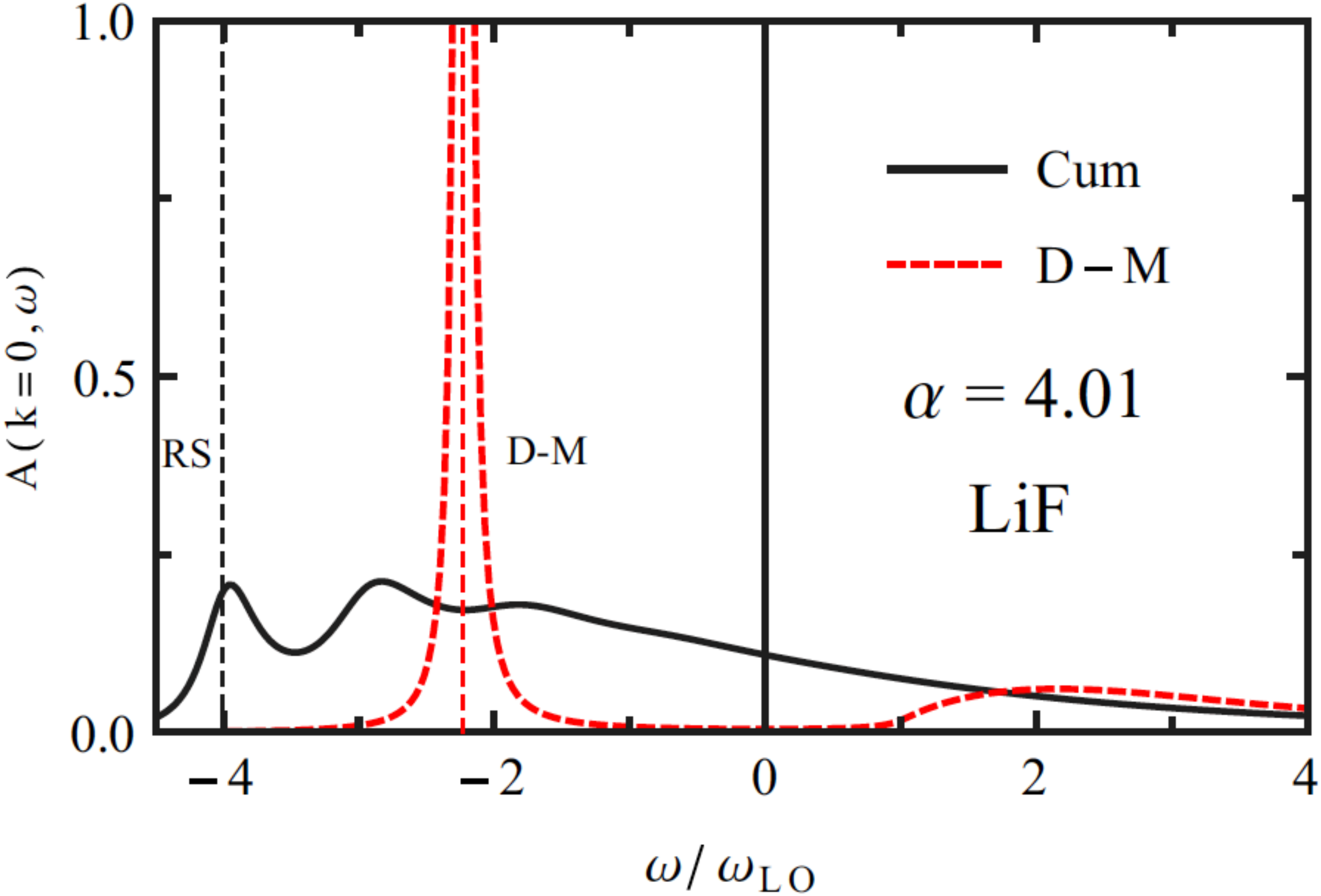


Jean Paul Nery
PhD May 18

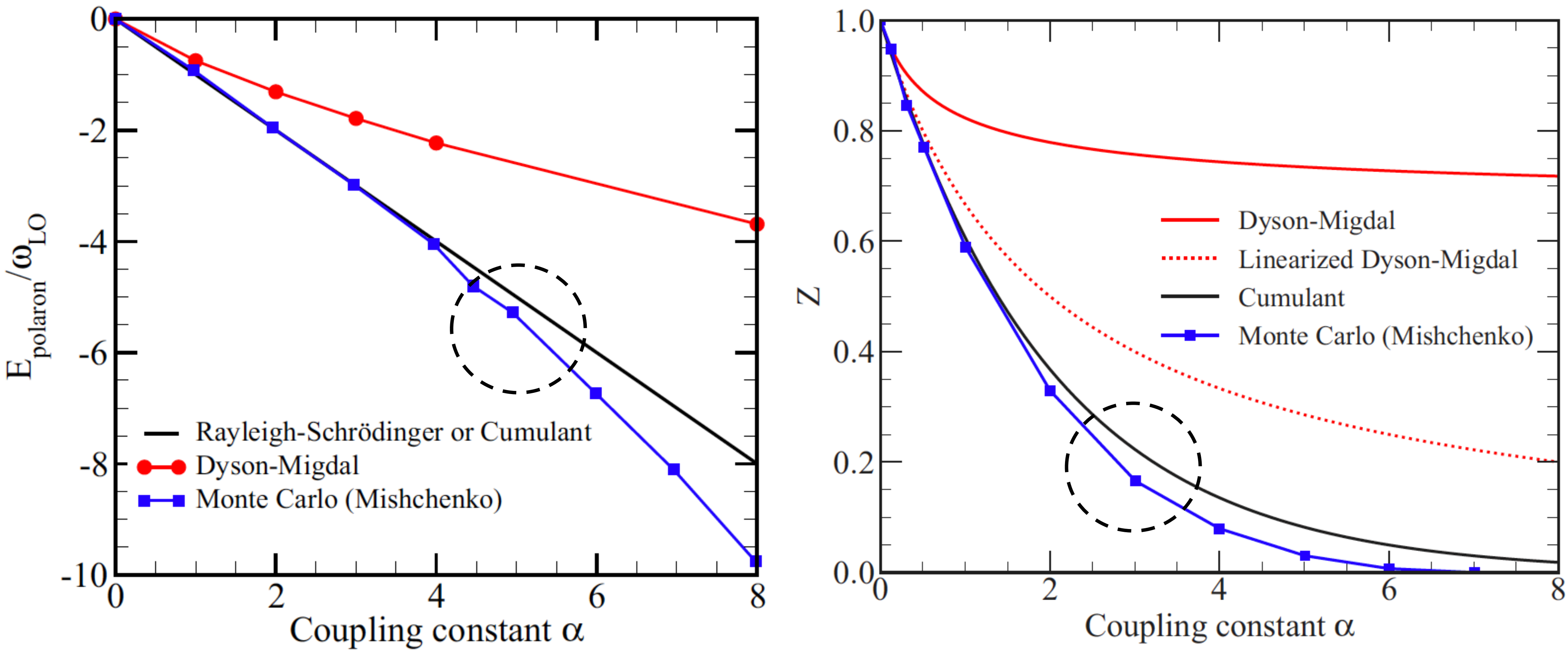
Lowest order $\Sigma(k,\omega)$ ("Migdal" including Fröhlich and "DW") used to construct lowest order C (Cumulant)

Analog of GW self-energy is "Migdal" self-energy.
the usual $A=-(1/\pi)$ Im G and $G^{-1}=\omega-\varepsilon_k-\Sigma$ screws up!





1.0
0.5
0.0
A (k = 0 , ω)
RS
D-M
Cum
D – M
α = 4.01
LiF
– 4
– 2
0
2
4
ω / ω LO



Computational Empiricism, not theory.
Dyson-Migdal is way off!
Cumulant is remarkably good.

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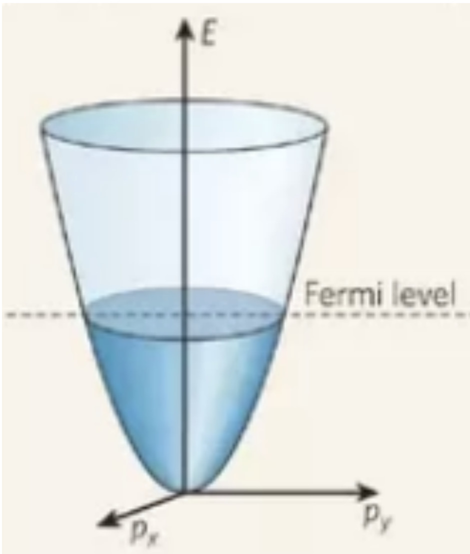
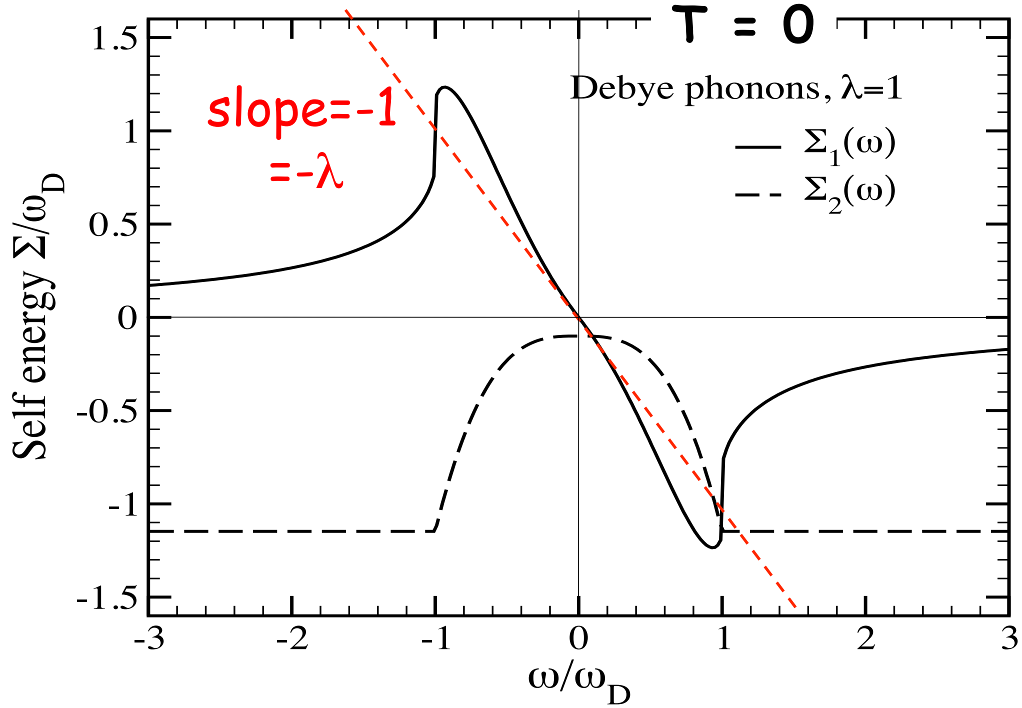
3. Metals: Migdal self energy (the "real" Migdal)

$$A(k,\omega) = -\frac{1}{\pi}\mathcal{I}mG(k,\omega)$$ where $$G(k,\omega) = \frac{1}{\omega - \epsilon_k - \Sigma(k,\omega)}$$

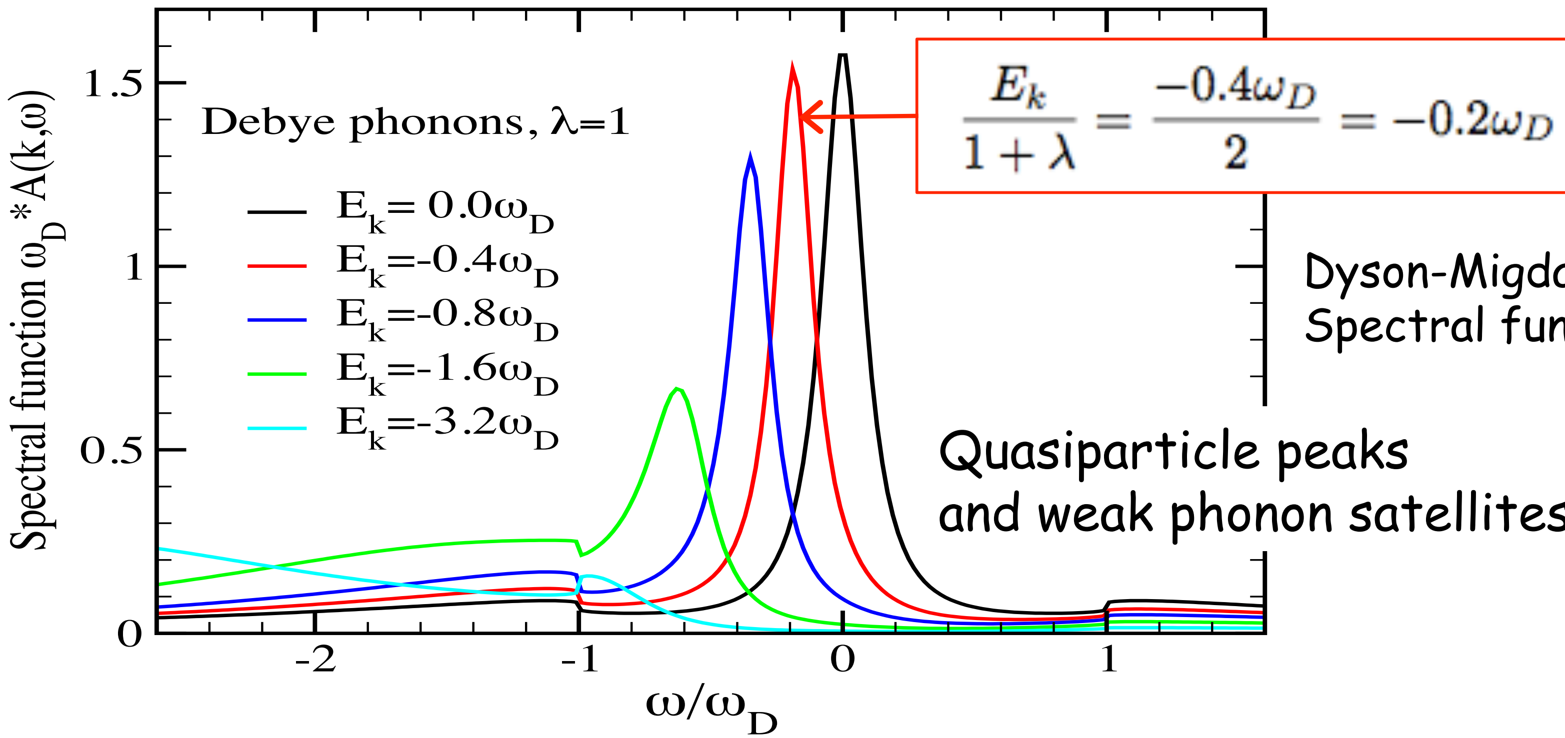
$\Sigma(k,\omega)$ =  $= \sum_Q |M_{k,k-Q}|^2 \left\{ \frac{1 - f_{k-Q} + n_Q}{\omega + i\eta - \epsilon_{k-Q} + \omega_Q} + \frac{f_{k-Q} + n_Q}{\omega + i\eta - \epsilon_{k-Q} + \omega_Q} \right\}$

Important at low T (esp. for superconductivity)
ω_Q cannot be dropped.
The process is DYNAMICAL.



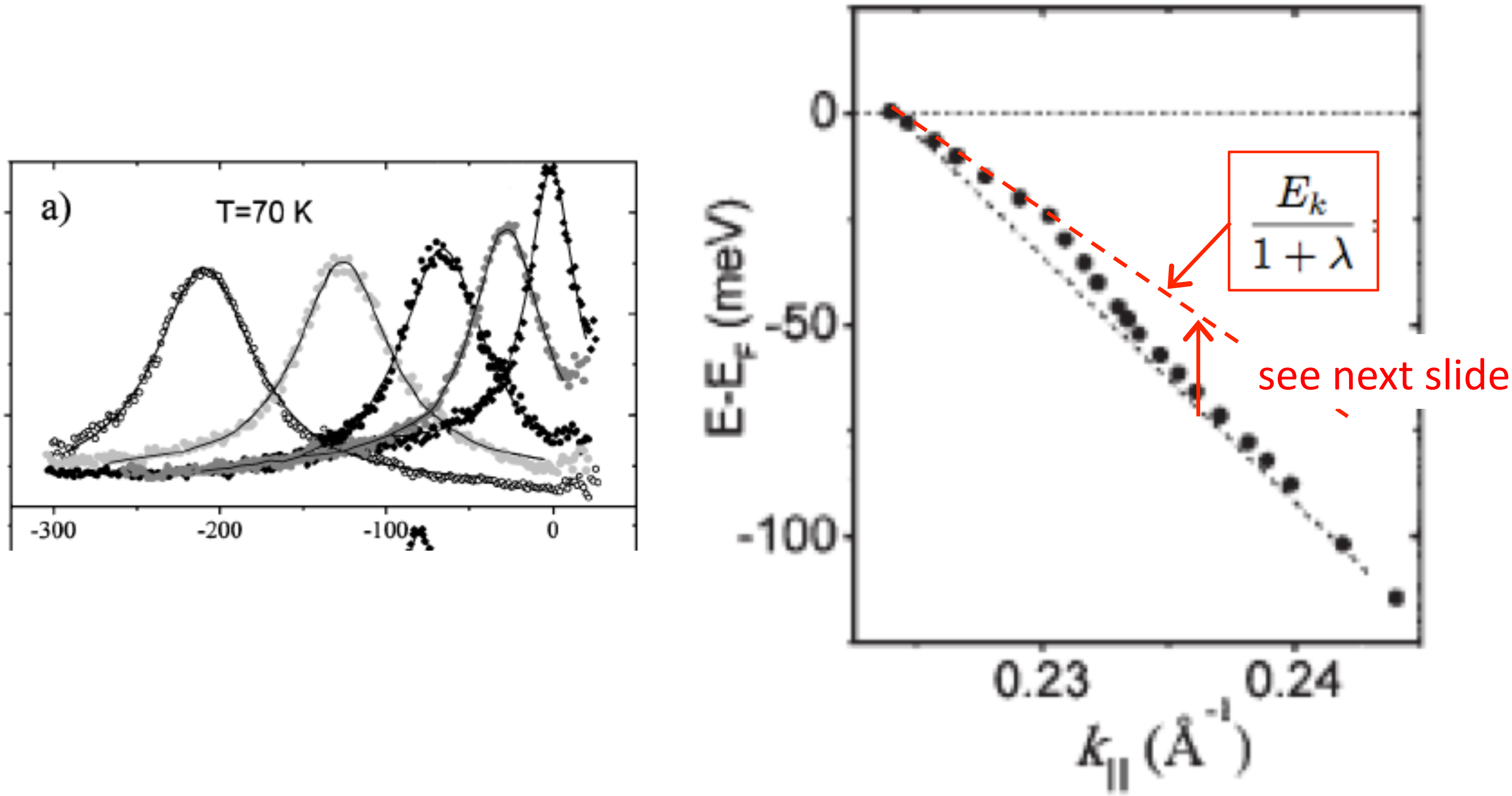


Debye-Migdal
self energy
λ = 1



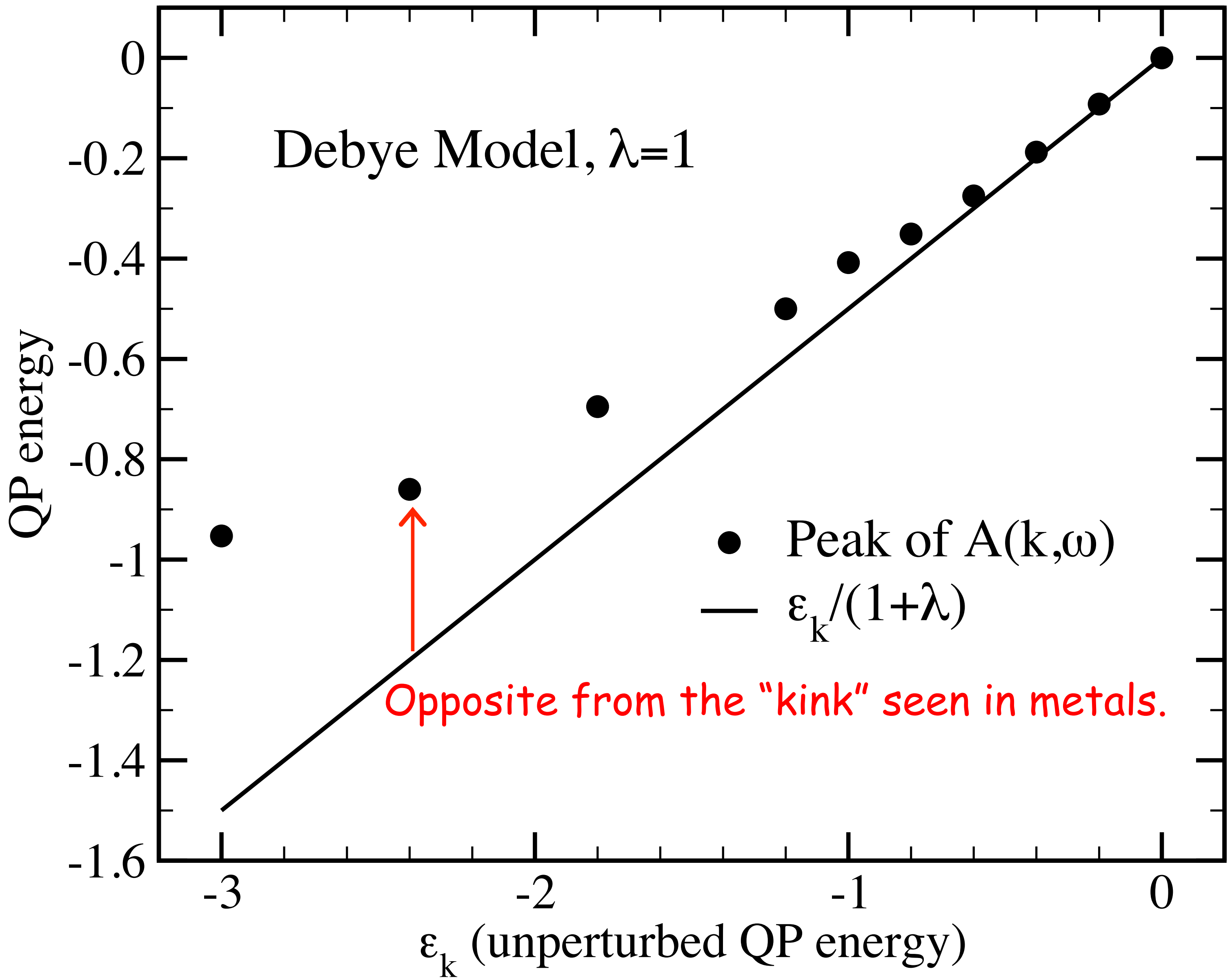
Dyson-Migdal
Spectral function

Valla, Fedorov, Johnson, and Hulbert, 1999
Many-body effects in angle-resolved photoemission (ARPES): quasiparticle energy and lifetime of a **Mo(110) surface state** *Phys. Rev. Lett.* ***83*** 2085.



Migdal self-energy and Dyson Eqn. work for $\varepsilon_k < \omega_D$ **QP**'s in metals.

Phonon satellite structure not resolved.


Debye Model, λ=1
Peak of A(k,ω)
ε_k/(1+λ)
Opposite from the "kink" seen in metals.
QP energy
0
-0.2
-0.4
-0.6
-0.8
-1
-1.2
-1.4
-1.6
-3
-2
-1
0
ε_k (unperturbed QP energy)

PHYSICAL REVIEW B **90**, 195135 (2014)

Cumulant expansion for phonon contributions to the electron spectral function

S. M. Story,[1] J. J. Kas,[1] F. D. Vila,[1] M. J. Verstraete,[2] and J. J. Rehr[1]

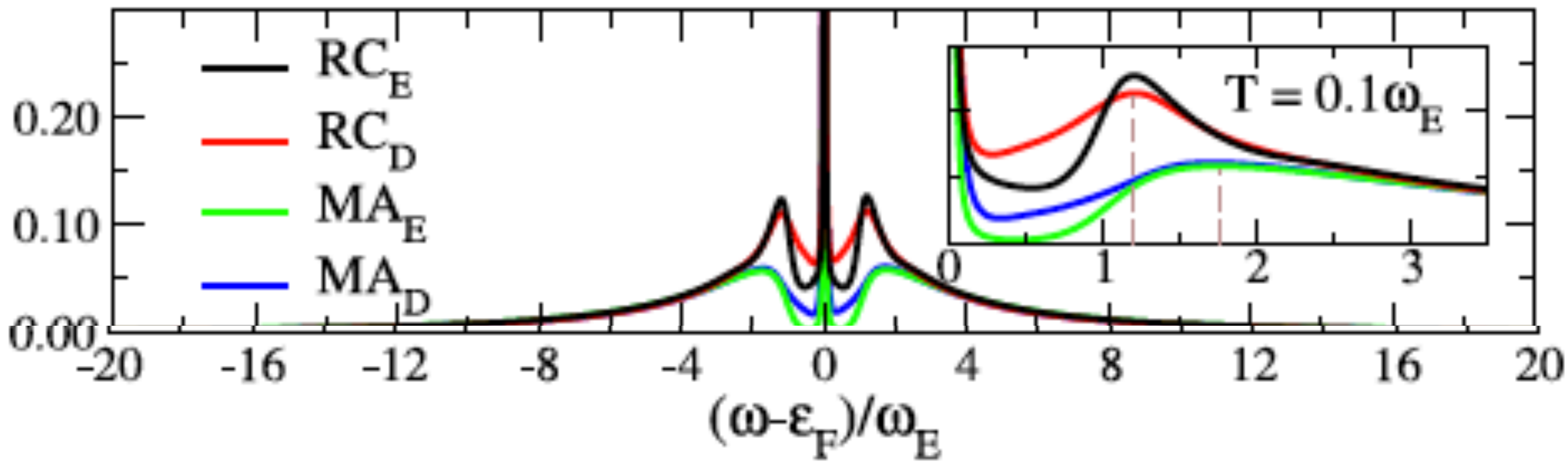


FIG. 5. (Color online) Comparison of spectral function from the RC and MA methods using the Einstein and Debye models at strong coupling (i.e., $\lambda = 1.6$) near the Fermi level ($\varepsilon_k = \varepsilon_F + 0.01\omega_E$) for

RC = retarded cumulant
MA = Migdal approximation

SUMMARY

1. Quasiparticles – We know them when we see them.
2. Computation by DFT is an essential part of QP theory.
3. Features beyond QP ("satellites") are being resolved.
4. $\Sigma(k,\omega)$ & $A(k,\omega)$ need improved theory; e.g. Cumulants.
5. May have benefits beyond spectral functions; *e.g.* resistivity saturation?

Personal opinion:
We are doing "computational experiments".
We need more experimenters at this meeting.

Thank You!

Thank you
Andrew & Joe!