Quantifying the impact of plasmon and paramagnon effects in "conventional" superconductors from first principles

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Thanks

In collaboration with

Univ. of Tokyo

Shinji Tsuneyuki Kentaro Tsutsumi Mitsuaki Kawamura

RIKEN

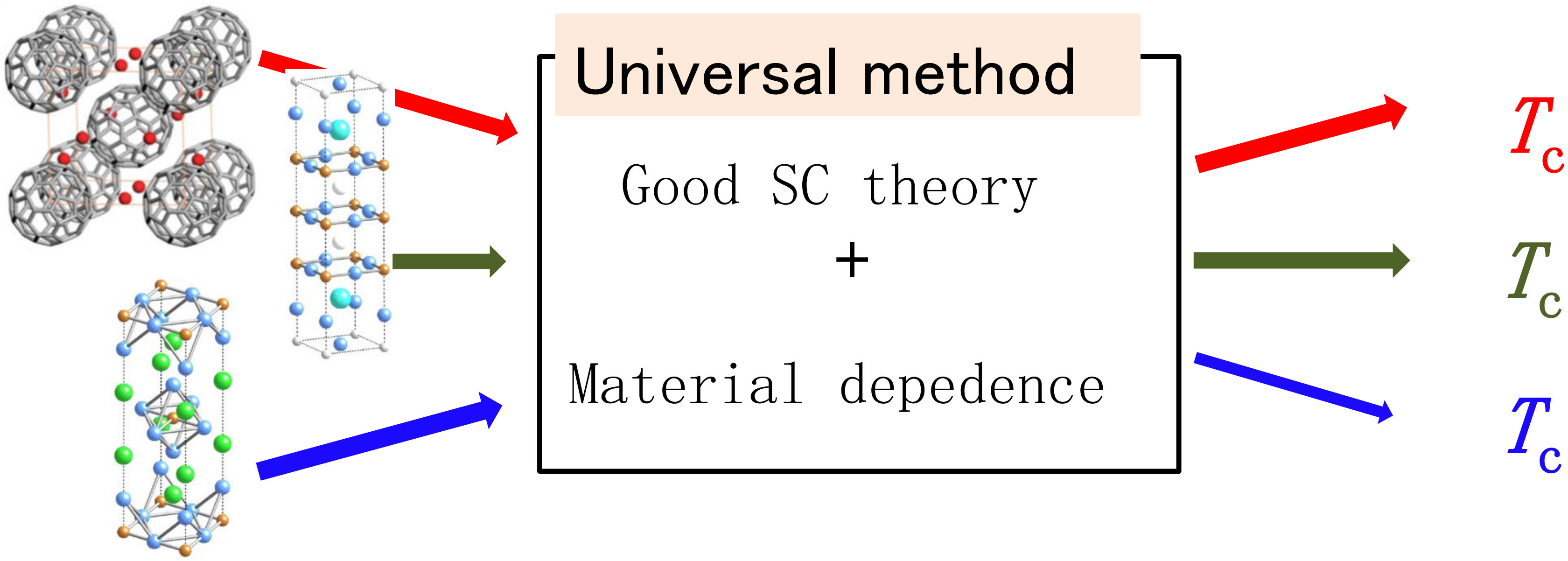
Ryotaro Arita (My PhD supervisor)

Thanks

On the shoulder of:

Hardy Gross

Superconducting Tc from first principles



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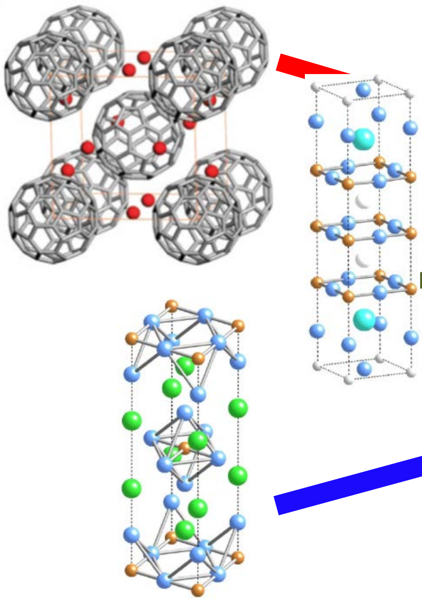


Universal method

Good SC theory

+

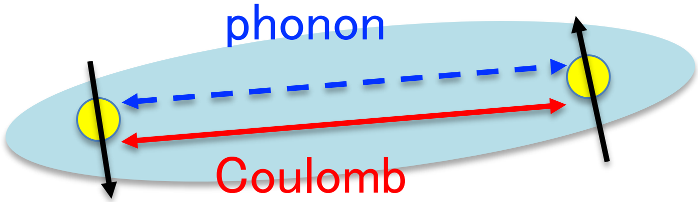
Material depedence

T_c

T_c

T_c

Cooper pair

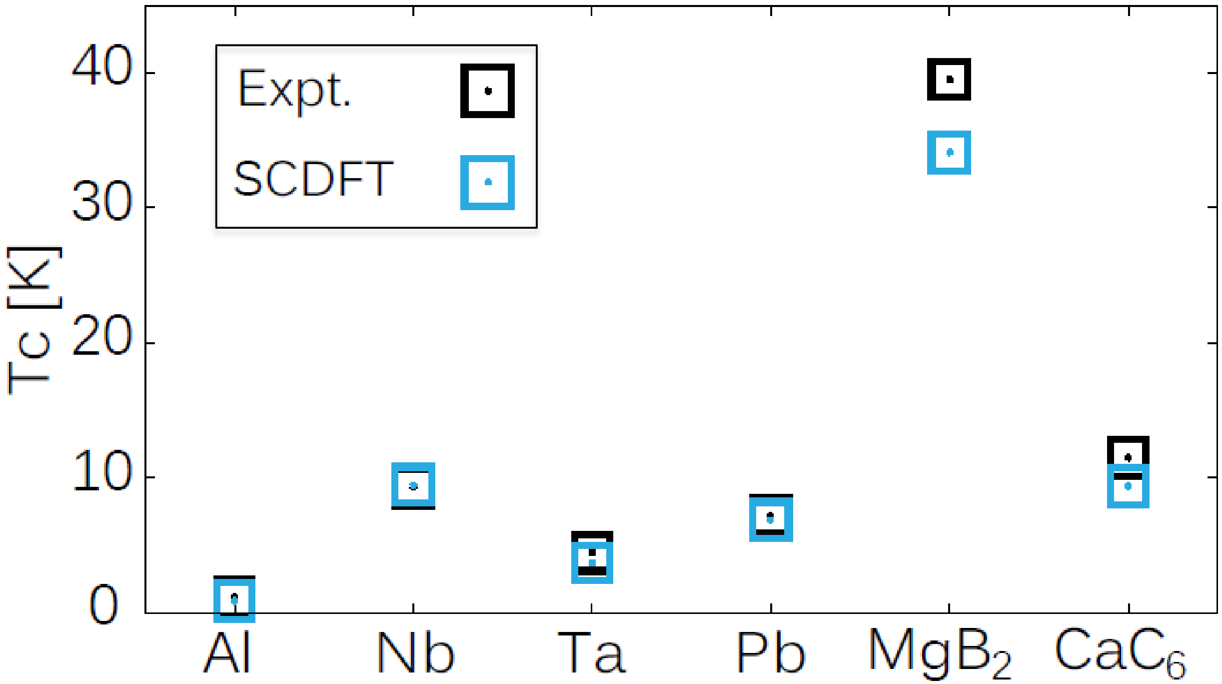


How can we describe the competition accurately?

Superconducting Tc from first principles

<u>It's possible.</u>

Marques *et al*., PRB(2005); Floris *et al*., PRL (2005); Sanna *et al*., PRB (2007).



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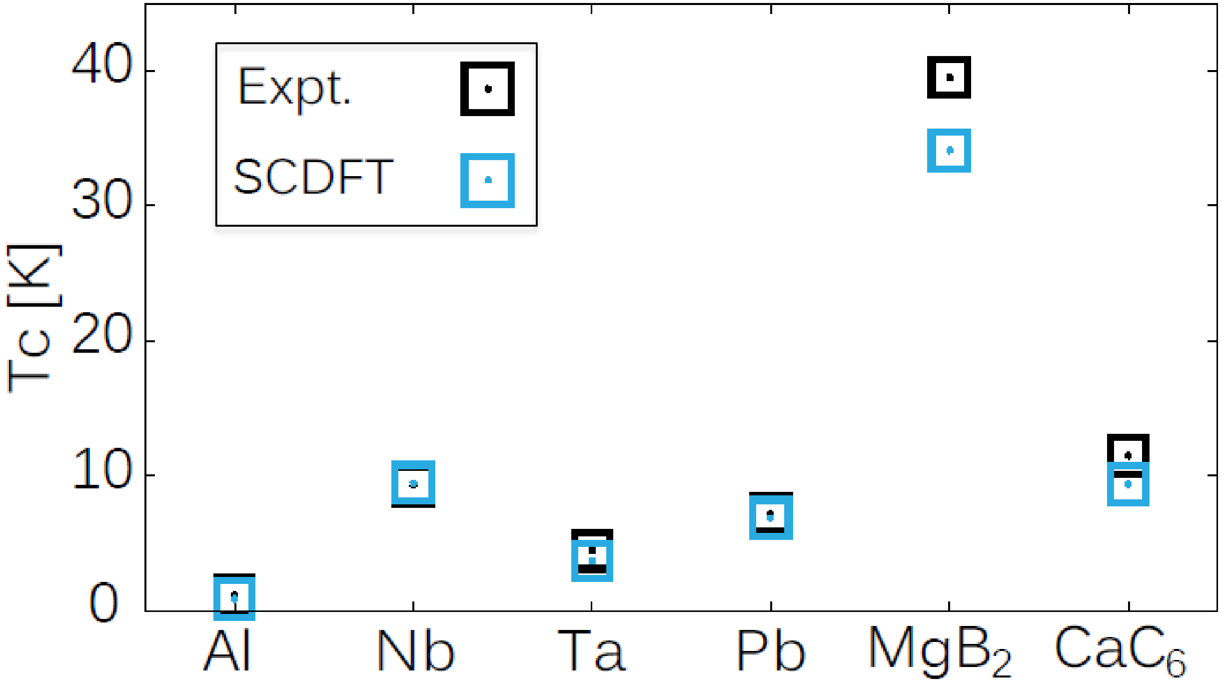
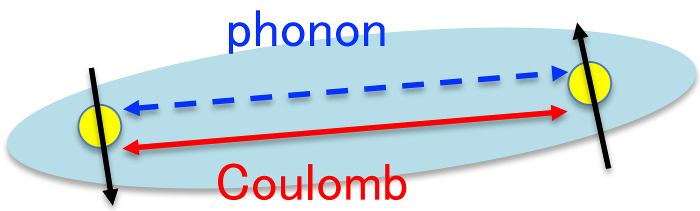


…by DFT for superconductors.

Superconducting Tc from first principles

Cooper pair



Role of e-e Coulomb interaction:

-Roughly, static repulsion

-More accurately, as a medium of ***fluctuations***

-dynamical charge fluctuation

-spin fluctuation

SCDFT formulation

Ab initio Hamiltonian for normal state electrons

$$H = T_e + U_{ee} + V_e$$

T_e :Electrons, kinetic term V_e : one-body potential term

U_{ee} :e-e, interaction term

$$n(\boldsymbol{r}) = \sum_{\sigma} \langle \hat{\Psi}_{\sigma}^{\dagger}(\boldsymbol{r}) \hat{\Psi}_{\sigma}(\boldsymbol{r}) \rangle$$

Normal-state Kohn-Sham Eq.

$$\left[-\frac{\nabla^2}{2} + v_0^e(\boldsymbol{r}) - \mu \right] \varphi(\boldsymbol{r}) = \epsilon_i \varphi(\boldsymbol{r})$$

Kohn-Sham potential (functional of electron density)

SCDFT formulation

L. N. Oliveira, E. K. U. Gross, and W. Kohn, PRL **60**, 2430 (1988);
T. Kreibich and E. K. U. Gross, PRL **86**, 2984 (2001);
M. Lueders, *et al.*, PRB **72,** 024545 (2005); M. A. L. Marques *et al.*, PRB **72**, 024546 (2005)

Ab initio Hamiltonian for superconductivity

$$H = T_e + U_{ee} + T_n + U_{nn} + U_{en} (+\Delta)$$

T_e	:Electrons, kinetic term	T_n	:nuclei, kinetic term
U_{ee}	:e-e, interaction term	U_{nn}	:n-n, interaction term
U_{en}	:e-n, interaction term	Δ	:gauge-symmetry breaking term

$$n(\boldsymbol{r}) = \sum_{\sigma} \langle \hat{\Psi}^{\dagger}_{\sigma}(\boldsymbol{r}) \hat{\Psi}_{\sigma}(\boldsymbol{r}) \rangle$$:electron normal density

$$\Gamma(\underline{\boldsymbol{R}}) = \langle \hat{\Phi}^{\dagger}(\underline{\boldsymbol{R}}) \hat{\Phi}(\underline{\boldsymbol{R}}) \rangle$$:nuclei density

$$\chi(\boldsymbol{r}, \boldsymbol{r}') = \langle \hat{\Psi}_{\uparrow}(\boldsymbol{r}) \hat{\Psi}_{\downarrow}(\boldsymbol{r}') \rangle$$:electron anomalous density

SCDFT formulation

$$H = T_e + U_{ee} + T_n + U_{nn} + U_{en} (+\Delta)$$

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Kohn-Sham Bogoliubov-deGennes Eq. + Born-Oppenheimer Eq.

$$\left[-\frac{\nabla^2_{\boldsymbol{r}}}{2} + v^e_0(\boldsymbol{r}) - \mu \right] u_n(\boldsymbol{r}) - \int \Delta_0(\boldsymbol{r}, \boldsymbol{r}') v_n(\boldsymbol{r}') = E_n u_n(\boldsymbol{r})$$

$$-\left[-\frac{\nabla^2_{\boldsymbol{r}}}{2} + v^e_0(\boldsymbol{r}) - \mu \right] v_n(\boldsymbol{r}) - \int \Delta^*_0(\boldsymbol{r}, \boldsymbol{r}') u_n(\boldsymbol{r}') = E_n v_n(\boldsymbol{r})$$

$$\left[\sum_\alpha -\frac{\nabla^2_{R_\alpha}}{2} + v^n_0(\underline{\boldsymbol{R}}) \right] \Phi(\underline{\boldsymbol{R}}) = \mathcal{E}_n \Phi(\underline{\boldsymbol{R}})$$

$v^e_0(\boldsymbol{r})$ $\Delta_0(\boldsymbol{r}, \boldsymbol{r}')$

$v^n_0(\underline{\boldsymbol{R}})$

{n, χ, Γ} dependent Kohn-Sham potentials

SCDFT formulation

Self-consistent KS-BdG Eq. + BO Eq.

M. Lueders, *et al.*, PRB ***72,*** 024545 (2005)

Decoupling of dependencies

$$v_0^e([n,\chi,\Gamma];\mathbf{r}) \approx v_0^e([n^{\mathrm{GS}},\Gamma_{\underline{\mathbf{R}}_0}];\mathbf{r})$$
$$v_0^n([n,\chi,\Gamma];\underline{\mathbf{R}}) \approx v_0^n([n^{\mathrm{GS}},\Gamma];\underline{\mathbf{R}}).$$

SCDFT formulation

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Successive calculations

1, Normal-state Kohn-Sham Eq.

$$\left[-\frac{\nabla^2}{2} + v_0^e(\boldsymbol{r}) - \mu\right]\varphi_i(\boldsymbol{r}) = \epsilon_i\varphi_i(\boldsymbol{r})$$

2, Normal-state BO Eq.
(Harmonic level in practice)

$$\left[\sum_\alpha -\frac{\nabla^2_{R_\alpha}}{2} + v_0^n(\underline{\boldsymbol{R}})\right]\Phi(\underline{\boldsymbol{R}}) = \mathcal{E}_n\Phi(\underline{\boldsymbol{R}})$$

3, Equation for anomalous density

Determination of anomalous density

Sham-Schlueter connection PRL **51**, 1888 (1983)

The interacting-system densities must be identical to those of KS-BdG equations

$$H = H^{\mathrm{KS-BdG}} + H^{\mathrm{ph}} + H^{\mathrm{e-ph}} + H^{\mathrm{e-e}} - H_{\mathrm{Hxc}}$$

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$$H = H^{\text{KS-BdG}} + H^{\text{ph}} + H^{\text{e-ph}} + H^{\text{e-e}} - H_{\text{Hxc}}$$

Dyson equation (Nambu 2x2 notation)

$$\mathbf{G}^{-1} = \mathbf{G}^{-1}_{\text{KS-BdG}} - \boldsymbol{\Sigma}$$

$$\chi(\mathbf{r},\mathbf{r}') = -\frac{1}{\beta}\sum_n e^{i\omega_n 0^+} F(\mathbf{r},\mathbf{r}',\omega_n) = -\frac{1}{\beta}\sum_n e^{i\omega_n 0^+} F_{\text{KS-BdG}}(\mathbf{r},\mathbf{r}',\omega_n)$$

$$n(\mathbf{r}) = \frac{1}{\beta}\sum_n e^{i\omega_n 0^+} G(\mathbf{r},\mathbf{r},\omega_n) = \frac{1}{\beta}\sum_n e^{i\omega_n 0^+} G_{\text{KS-BdG}}(\mathbf{r},\mathbf{r},\omega_n)$$

Taylor expansion gives us linear equation for Δ_{xc}.

The "gap" equation

Lüders, Marques, Gross et al., PRB **72**, 024545; 024546 (2005).

$$\Delta_{n\mathbf{k}} = -\mathcal{Z}_{n\mathbf{k}}\Delta_{n\mathbf{k}} - \frac{1}{2}\sum_{n'\mathbf{k}'}\mathcal{K}_{n\mathbf{k}n'\mathbf{k}'}\frac{\tanh[(\beta/2)E_{n'\mathbf{k}'}]}{E_{n'\mathbf{k}'}}\Delta_{n'\mathbf{k}'}$$

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$$\mathbf{\Sigma} \simeq \mathbf{\Sigma}^{\mathrm{ph}} + \mathbf{\Sigma}^{\mathrm{el}} - \mathbf{\Sigma}^{\mathrm{Hxc}}(v_{\mathrm{Hxc}}, \Delta_{\mathrm{xc}})$$

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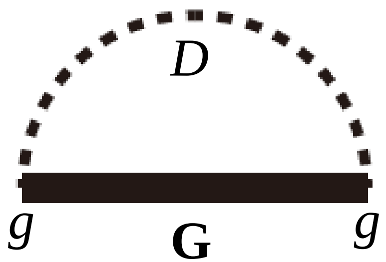
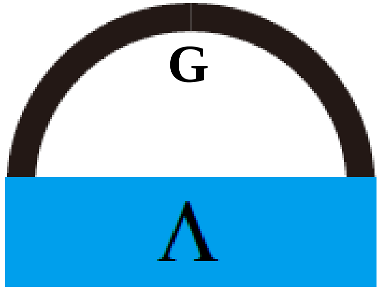


Phonon propagator



el-el interaction

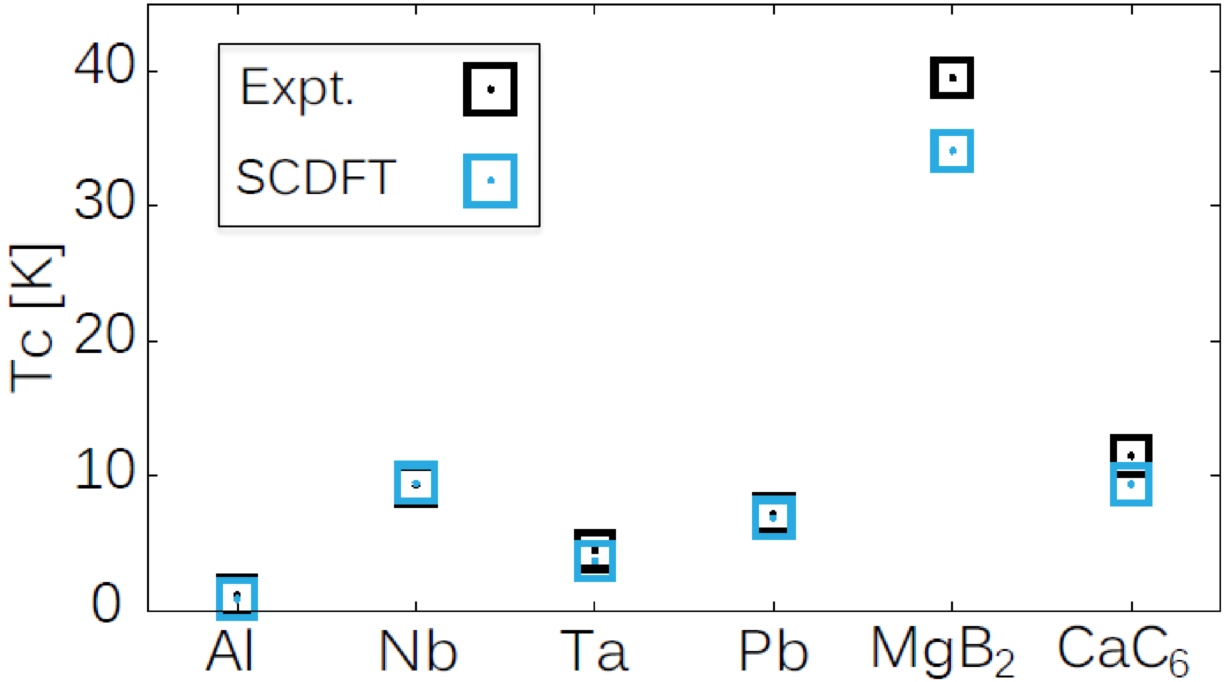
$$Z = Z^{\mathrm{ph}}(\mathbf{G}, D, g) + Z^{\mathrm{el}}(\mathbf{G}, \Lambda) - Z^{\mathrm{Hxc}}$$

$$K = K^{\mathrm{ph}}(\mathbf{G}, D, g) + K^{\mathrm{el}}(\mathbf{G}, \Lambda)$$

Minimum: static metallic screening

$$\Lambda(\mathbf{q},\omega) = W(\mathbf{q},\omega) \simeq W(\mathbf{q},\omega=0) \simeq \begin{cases} W^{\mathrm{T-F}}(\mathbf{q}) \\ W^{\mathrm{RPA}}(\mathbf{q}) \end{cases}$$

Marques *et al*., PRB(2005); Floris *et al*., PRL (2005); Sanna *et al*., PRB (2007).



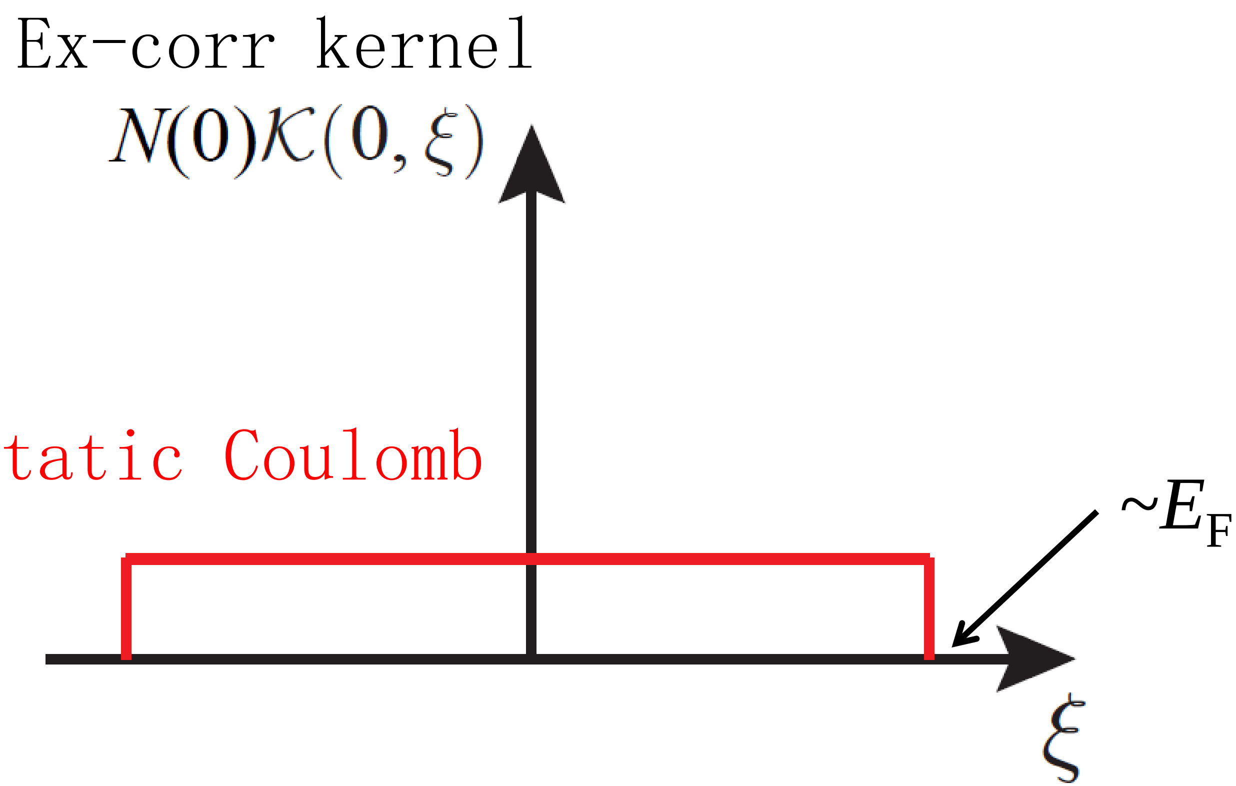
Accurate for typical phonon-mediated SCs.

What is missing in static *W*

RPA-screened interaction

$W(\mathbf{q}, w)=$ 

Static case



What is missing in static *W*

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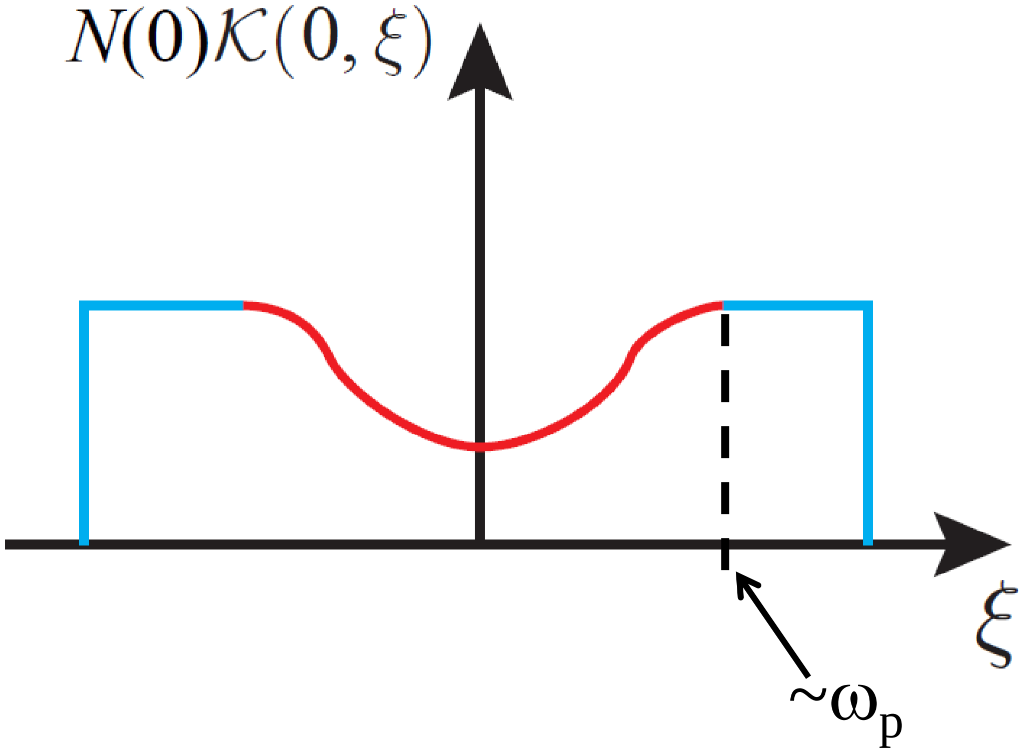
Dynamical case



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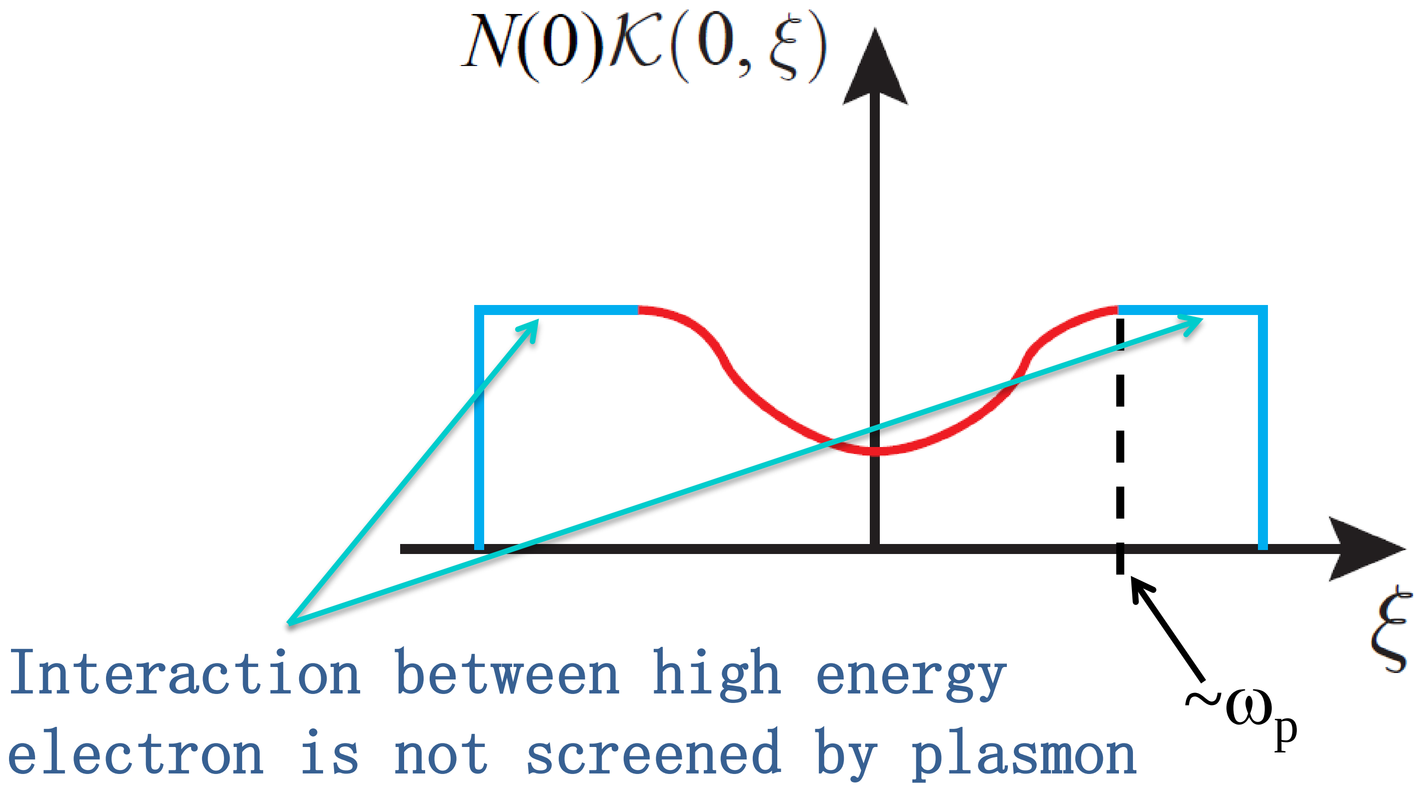
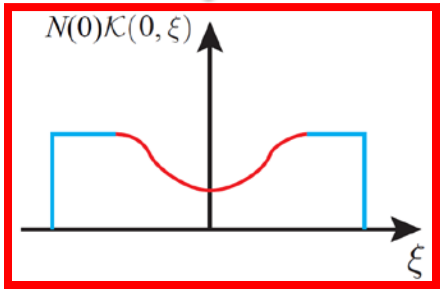


Effect of high-energy repulsion on T_c

$$\Delta(\xi) = -\frac{1}{2}N(0)\int d\xi' \underline{\mathcal{K}(\xi,\xi')}\frac{\tanh[(\beta/2)\xi']}{\xi'}\Delta(\xi')$$



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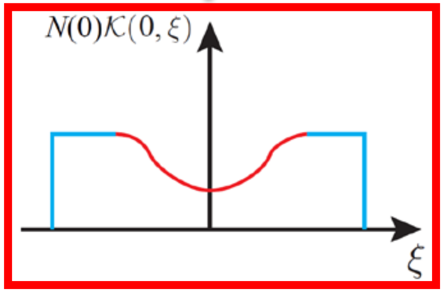

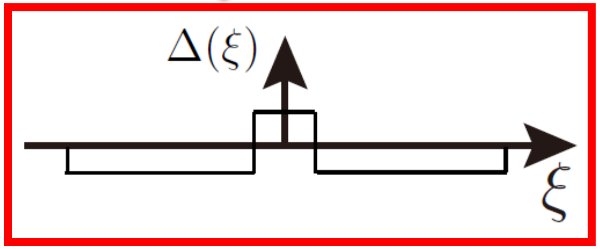

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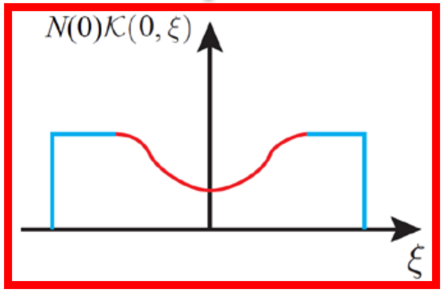

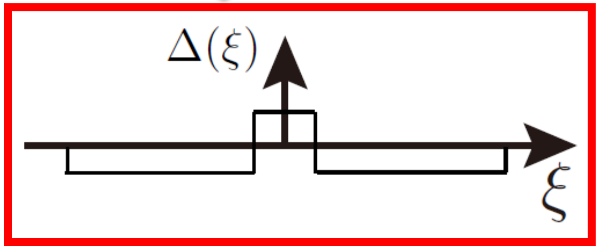

Large & Negative

$K(0,\xi)\Delta(\xi)$

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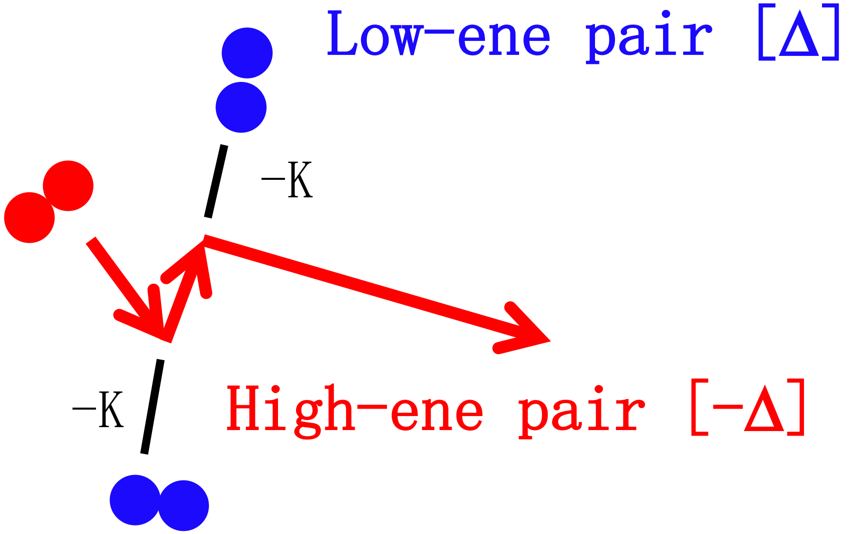

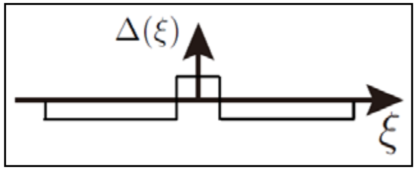

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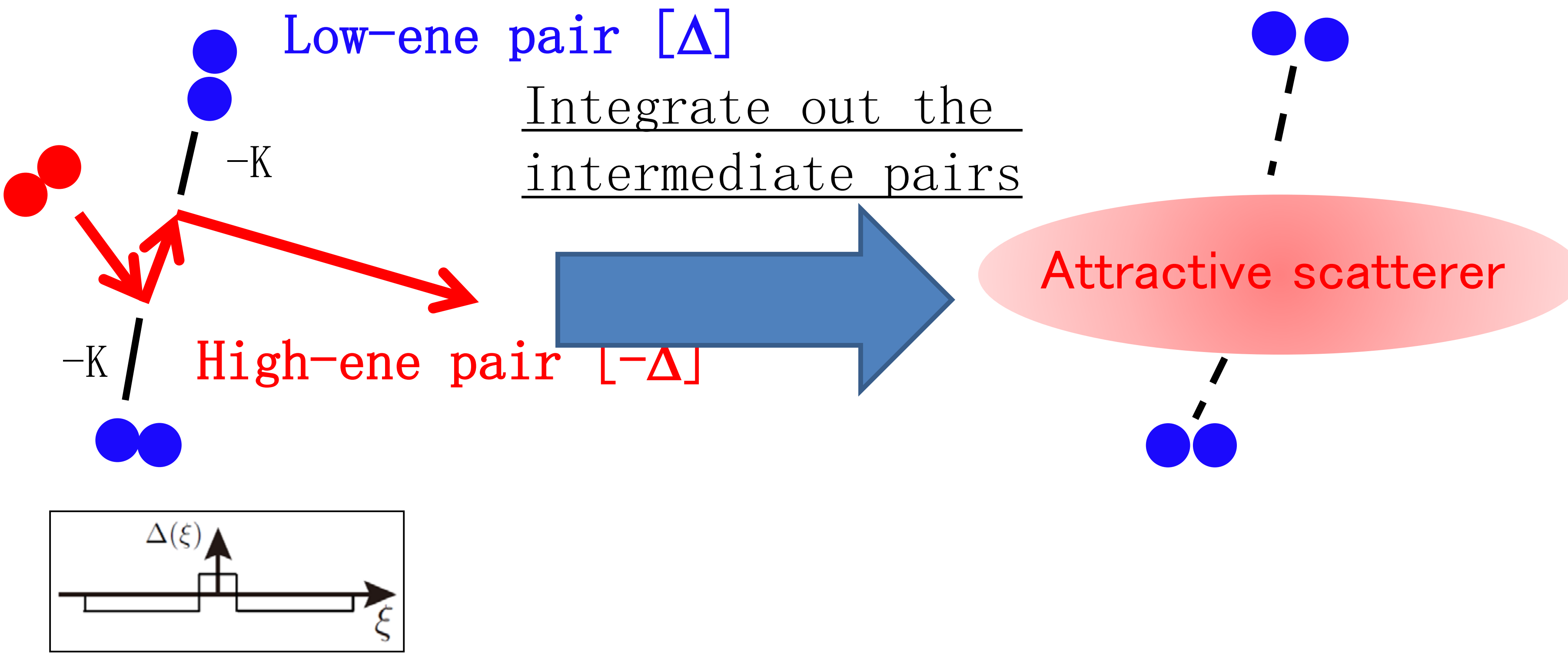
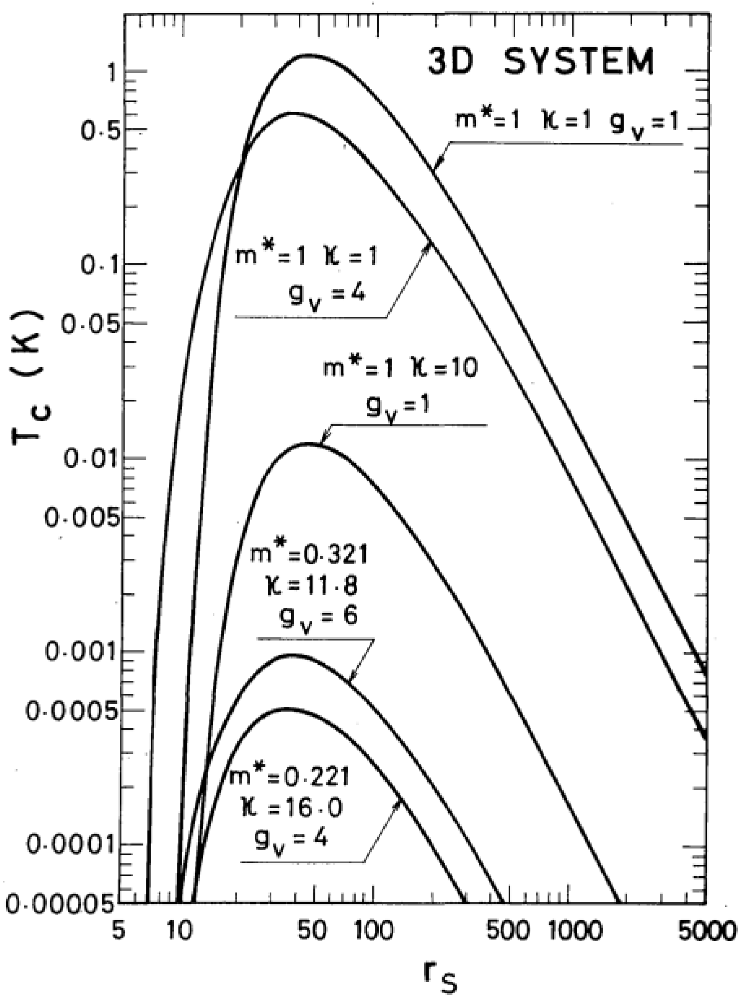


Plasmon superconducting mechanism

Takada, 1978; Rietschel and Sham, 1983

Superconductivity in dilute electron gas
Y. Takada, JPSJ **45**, 786 (1978).



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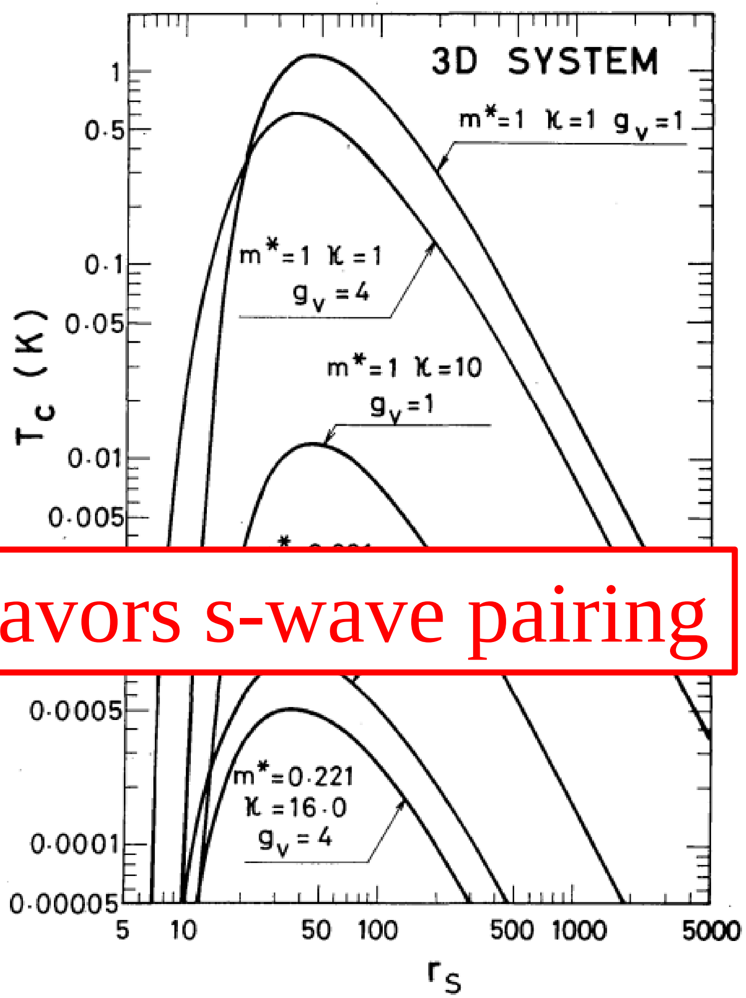

Favors s-wave pairing

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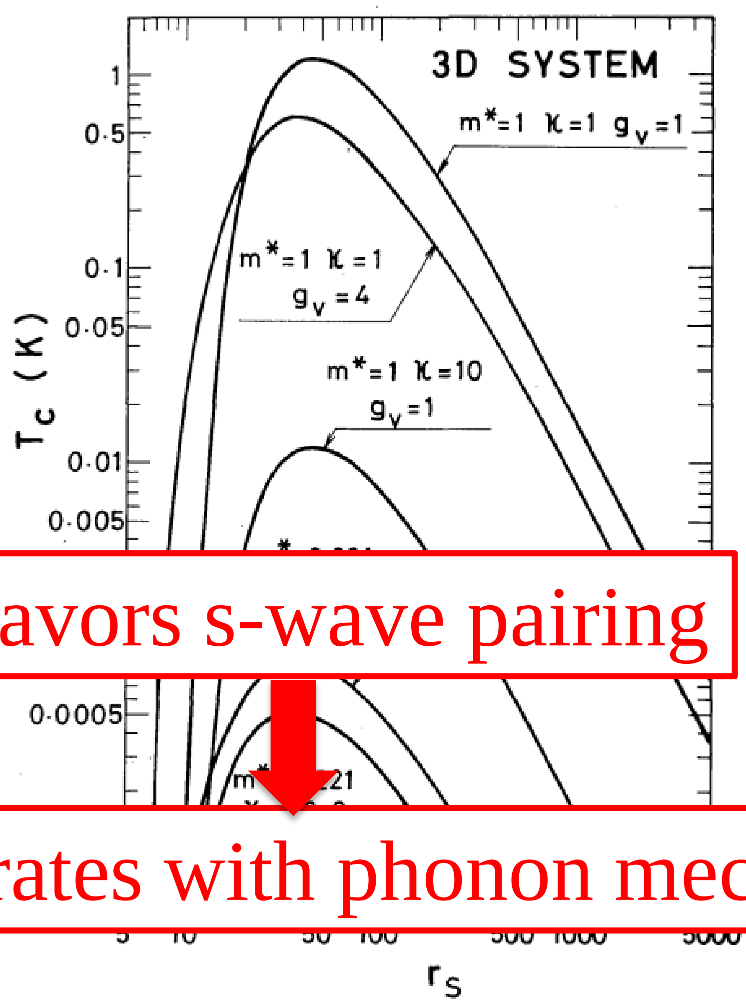
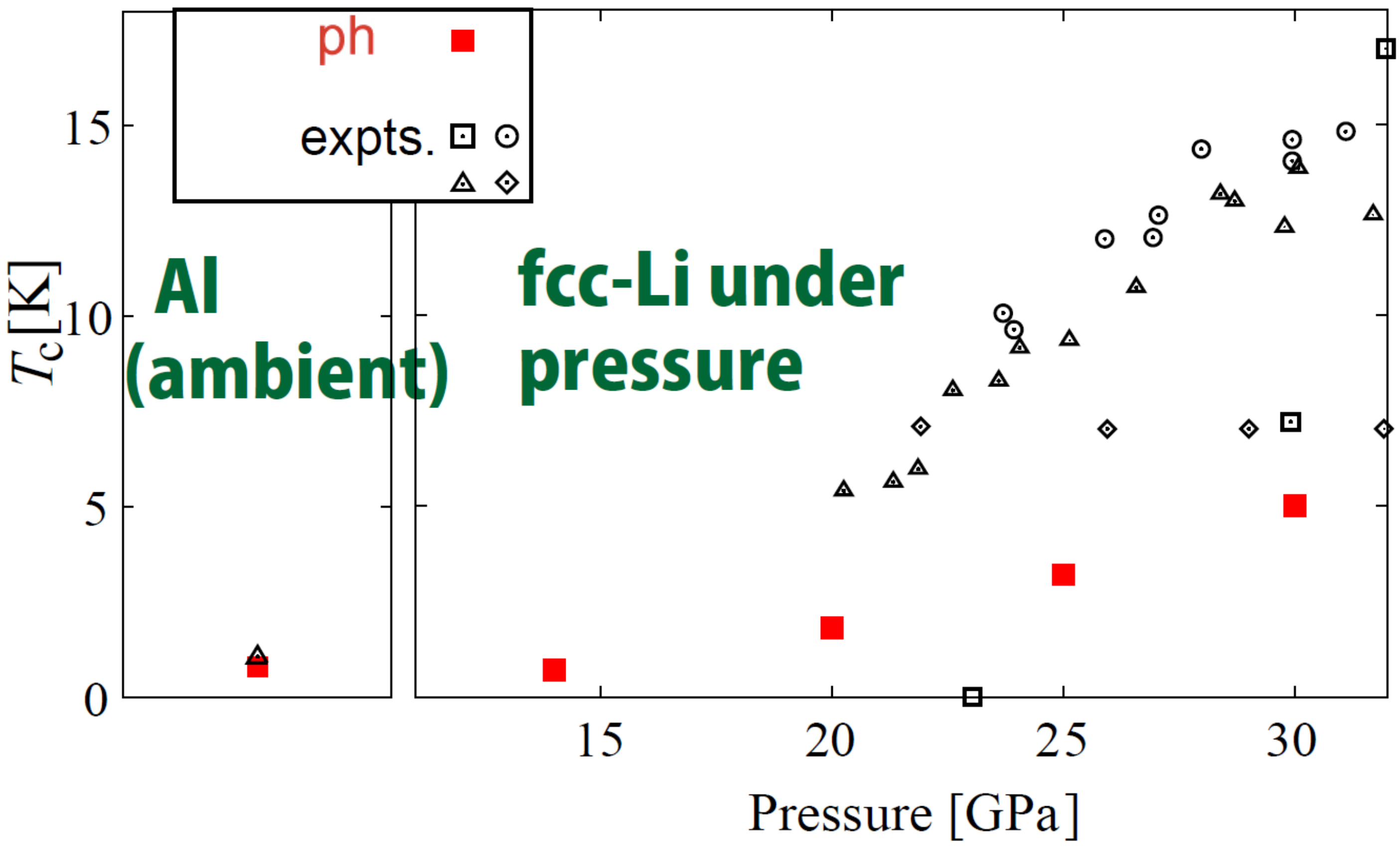


Favors s-wave pairing

Co-operates with phonon mechanism

Plasmon-phonon cooperation

$$\Delta_{n\mathbf{k}} = -\mathcal{Z}_{n\mathbf{k}}\Delta_{n\mathbf{k}} - \frac{1}{2}\sum_{n'\mathbf{k}'}\mathcal{K}_{n\mathbf{k}n'\mathbf{k}'}\frac{\tanh[(\beta/2)E_{n'\mathbf{k}'}]}{E_{n'\mathbf{k}'}}\Delta_{n'\mathbf{k}'}.$$



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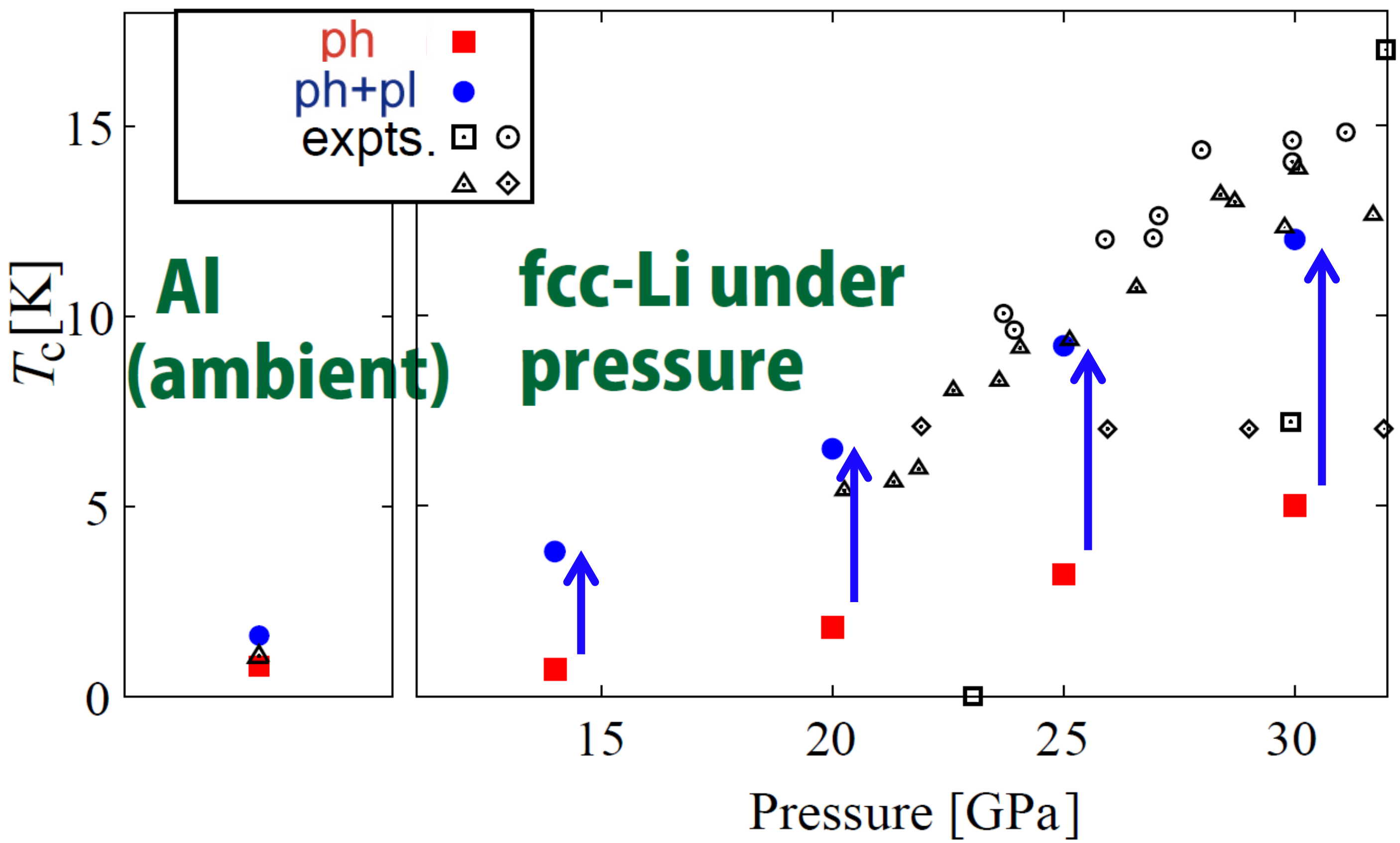


RA and R. Arita, PRL **111**, 057006 (2013); J. Phys. Soc. Jpn. **83**, 061016 (2014)

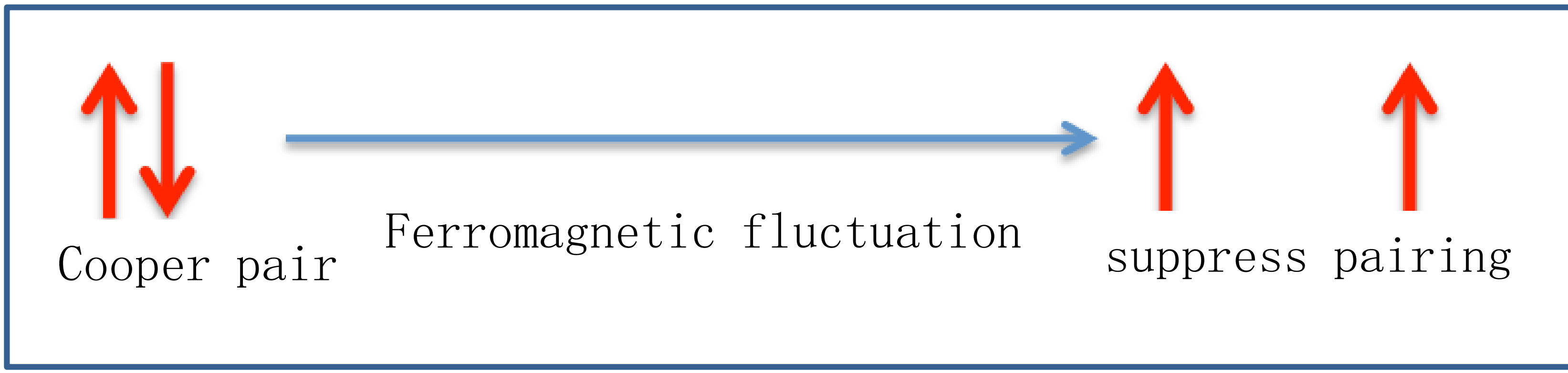
Spin fluctuation effect

Unconventional: Possible origin of pairing

D. J. Scalapino, Rev. Mod. Phys. 84, 1383 (2012);
F. Essenberger, et al., PRB **94**, 014503 (2016)

Conventional:

- Suppression of pairing due to exchange effect
- Ex. effect is significant in transition metals having *d*-electr



The effect of SF in V, Nb[1][2] and some other materials[1] has been explored using empirical parameters

[1] M. Wierzbowska, The Eur. Phys. J. B **48**, 207 (2005).
[2] H. Rietschel and H. Winter, Phys. Rev. Lett. **43**, 1256 (1979).

Incorporating spin fluctuation

F. Essenberger, et al. PRB **90**, 214504 (2014); **94**, 014503 (2016)

$W(\mathbf{q}, w)=$ 

The RPA-screened interaction is spin-independent.

Coulomb

V

Incorporating spin fluctuation

F. Essenberger, et al. PRB **90**, 214504 (2014); **94**, 014503 (2016)

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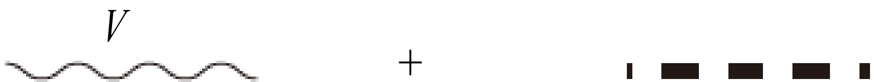
The RPA-screened interaction is spin-independent.

Coulomb | Spin-dependent interaction



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$W(\mathbf{q}, \mathrm{w})=$ 

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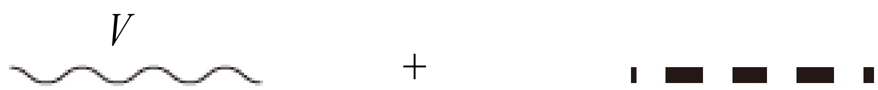
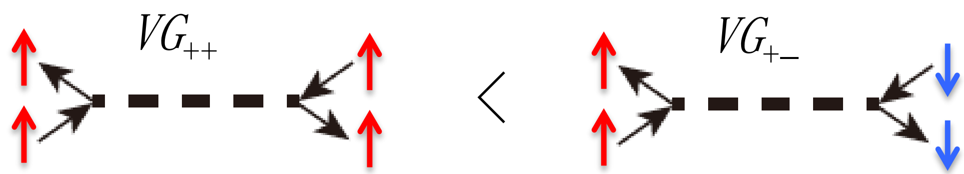
Coulomb | Spin-dependent interaction





$$VG_s = VG_{++} + VG_{+-}$$

$$VG_a = VG_{++} - VG_{+-}$$

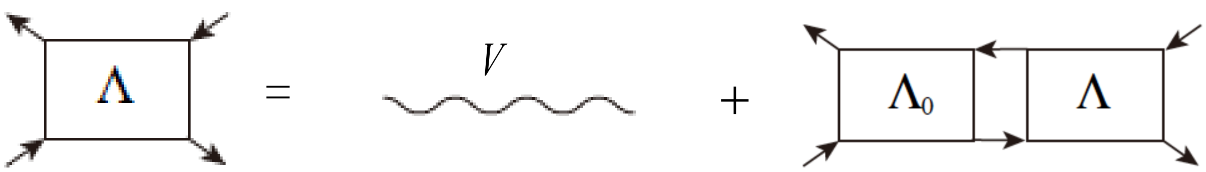
El-el interaction: the "dangerous" term

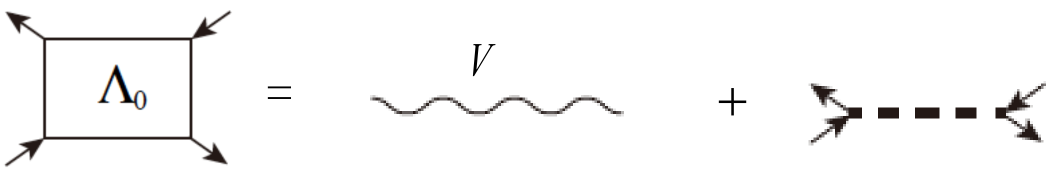
G. Vignale and K. S. Singwi, PRB **32**, 2156 (1985)

Sum up the "dangerous" terms in small (**q**, w) limit

1) long-range character of bare Coulomb interaction
2) gapless particle-hole excitation

"Dangerous" interaction terms (including RPA)





El-el interaction: the “dangerous” term

C. A. Kukkonen and A. W. Overhauser, PRB 20, 550 (1979).
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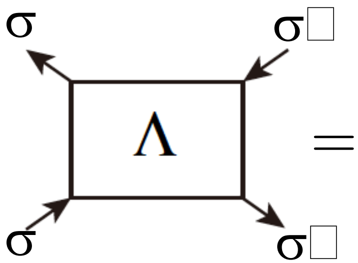

$$= V + [V(1 - G_s)]\chi_{\mathrm{C}}[V(1 - G_s)] + \sigma\sigma'[VG_a]\chi_{\mathrm{SL}}[VG_a]$$

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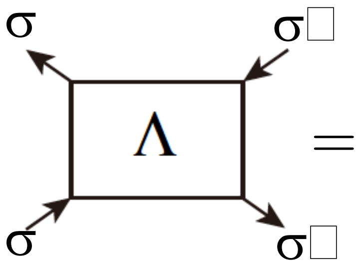


$$= V + [V(1-G_s)]\chi_{\mathrm{C}}[V(1-G_s)] + \sigma\sigma'[VG_a]\chi_{\mathrm{SL}}[VG_a]$$

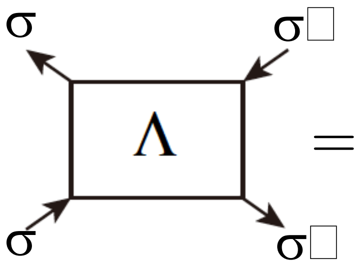
In ALDA,

$$(V + K_{\mathrm{xc}})\chi_{\mathrm{C}}(V + K_{\mathrm{xc}}) \qquad I_{\mathrm{xc}}\chi_{\mathrm{SL}}I_{\mathrm{xc}}$$

$$[K_{\mathrm{xc}}]_{\mathbf{r},\mathbf{r}'} = \frac{\delta^2 E_{\mathrm{xc}}}{\delta n(\mathbf{r})\delta n(\mathbf{r}')} \qquad [I_{\mathrm{xc}}]_{\mathbf{r},\mathbf{r}'} = \frac{\delta^2 E_{\mathrm{xc}}}{\delta m(\mathbf{r})\delta m(\mathbf{r}')}$$

El-el interaction: the “dangerous” term

C. A. Kukkonen and A. W. Overhauser, PRB 20, 550 (1979).
G. Vignale and K. S. Singwi, PRB **32**, 2156 (1985)



$$= V + [V(1 - G_s)]\chi_{\mathrm{C}}[V(1 - G_s)] + \sigma\sigma'[VG_a]\chi_{\mathrm{SL}}[VG_a]$$

In ALDA,

$$(V + K_{\mathrm{xc}})\chi_{\mathrm{C}}(V + K_{\mathrm{xc}}) \qquad I_{\mathrm{xc}}\chi_{\mathrm{SL}}I_{\mathrm{xc}}$$

=Ch. fluctuation mediated interaction

=***longitudinal*** Sp. Fluctuation mediated interaction

El-el interaction: the "dangerous" term

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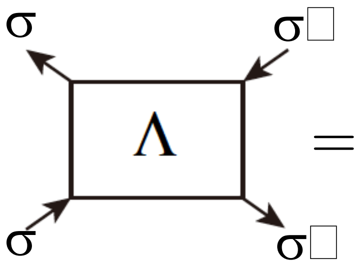


$$= V + [V(1-G_s)]\chi_C[V(1-G_s)] + \sigma\sigma'[VG_a]\chi_{SL}[VG_a]$$

In ALDA,

$$(V+K_{xc})\chi_C(V+K_{xc})$$

=Ch. fluctuation mediated interaction

$$I_{xc}\chi_{SL}I_{xc}$$

=***longitudinal*** Sp. Fluctuation mediated interaction

w. transverse fluctuation

$$\Lambda^{SF}=3I_{xc}\chi_{SL}I_{xc}$$

Kernels originating from SF

SCDFT gap equation with the effect of SF

$$\Delta_k^{\mathrm{xc}} = -Z_k \Delta_k^{\mathrm{xc}} - \sum_{k'} K_{kk'} \frac{\tanh\left[(\beta/2)E_{k'}\right]}{2E_{k'}} \Delta_{k'}^{\mathrm{xc}}$$

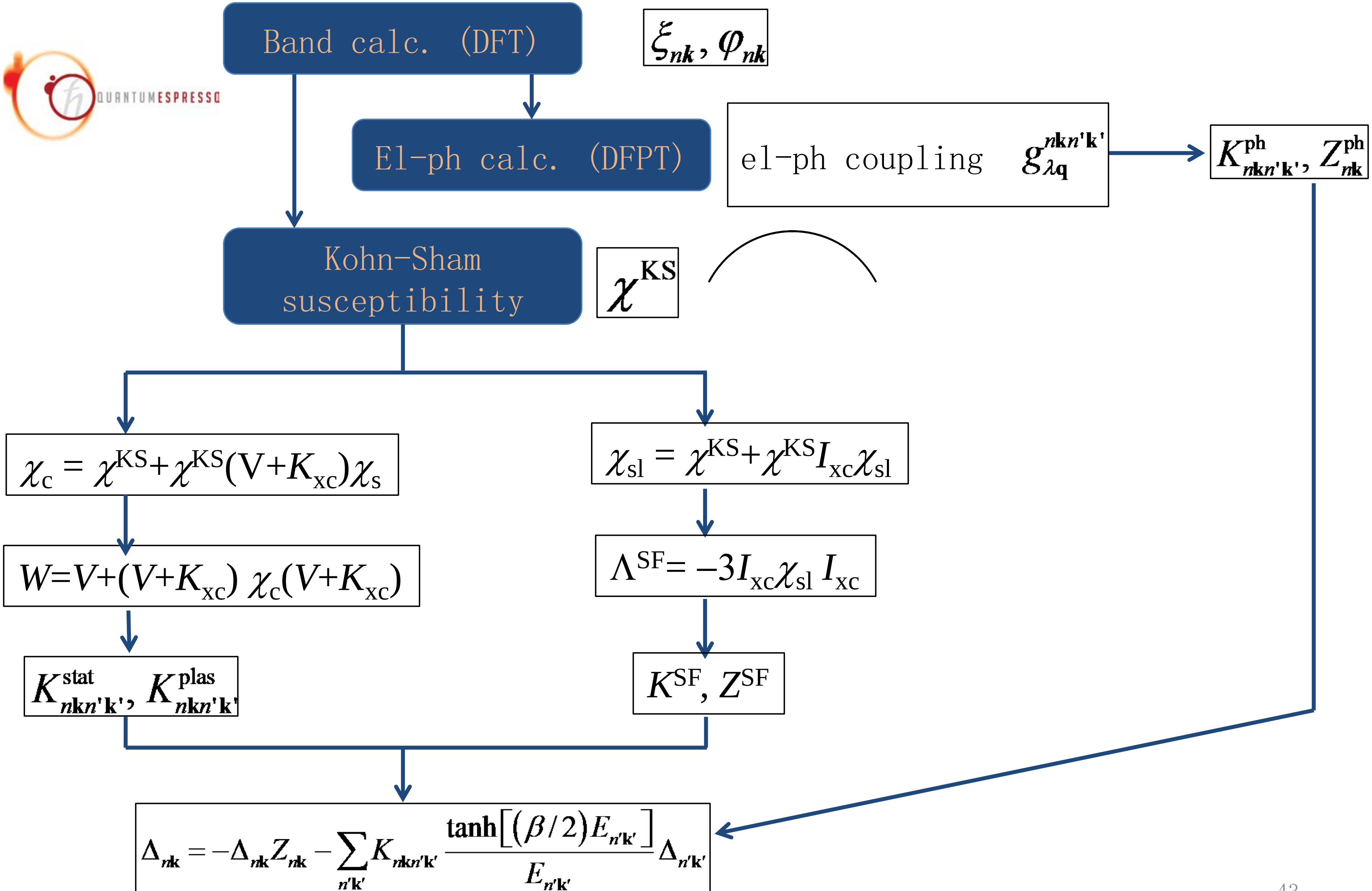
$$Z_k = Z_k^{\mathrm{ph}} + Z_k^{\mathrm{SF}}$$

$$K_{kk'} = K_{kk'}^{\mathrm{ph}} + K_{kk'}^{\mathrm{el}} + K_{kk'}^{\mathrm{SF}}$$

$$K_{kk'}^{\mathrm{SF}} = K_{kk'}^{\mathrm{SF}}(\mathbf{G}, \Lambda^{\mathrm{SF}}) \qquad Z_k^{\mathrm{SF}} = Z_k^{\mathrm{SF}}(\mathbf{G}, \Lambda^{\mathrm{SF}})$$

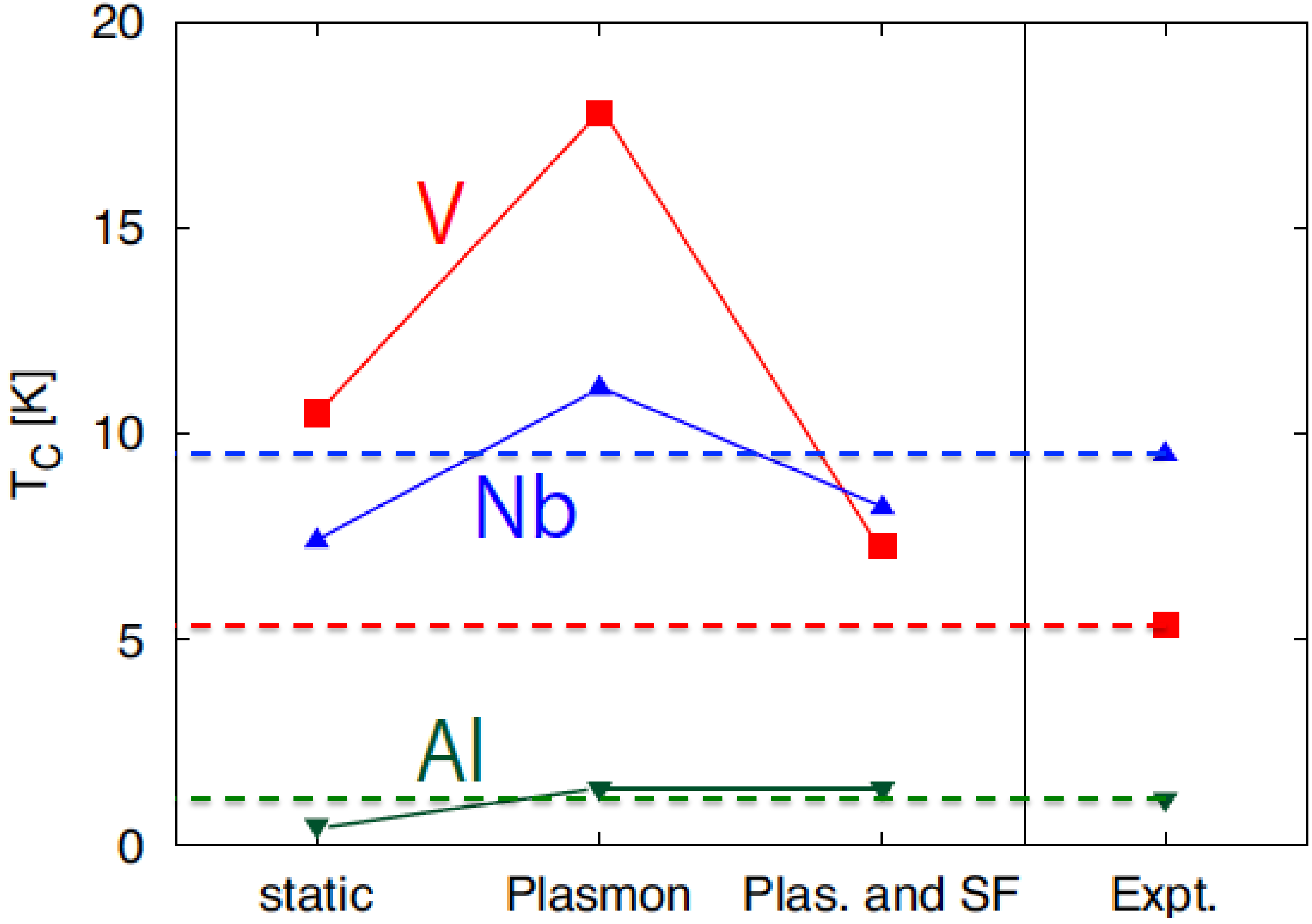
[1] F. Essenberger *et al.*, PRB **90**, 214504 (2014).
[2] M. Marques, Ph.D. thesis, 1998.

Procedure of calculation



Comparison of SCDFT & expt. T_C

K. Tsutsumi, M. Kawamura, RA, and S. Tsuneyuki in preparation



Summary

R. AKASHI and R. Arita, PRL **111**, 057003 (2013);
K. Tsutsumi, M.Kawamura, R. AKASHI, and S.Tsuneyuki in preparation

- First-principles calculation of T_C including the effects of ***dynamical charge*** and ***spin*** fluctuations

- Plasmon effect generally co-operates with phonon, whereas spin fluctuation competes.

- Strong electronic localization leads to significant T_C reduction due to SF

- Agreement with expt. T_C becomes better in wider range of materials

- Application to other materials is desired.