Development of density functional theory for superconductors: recent progress

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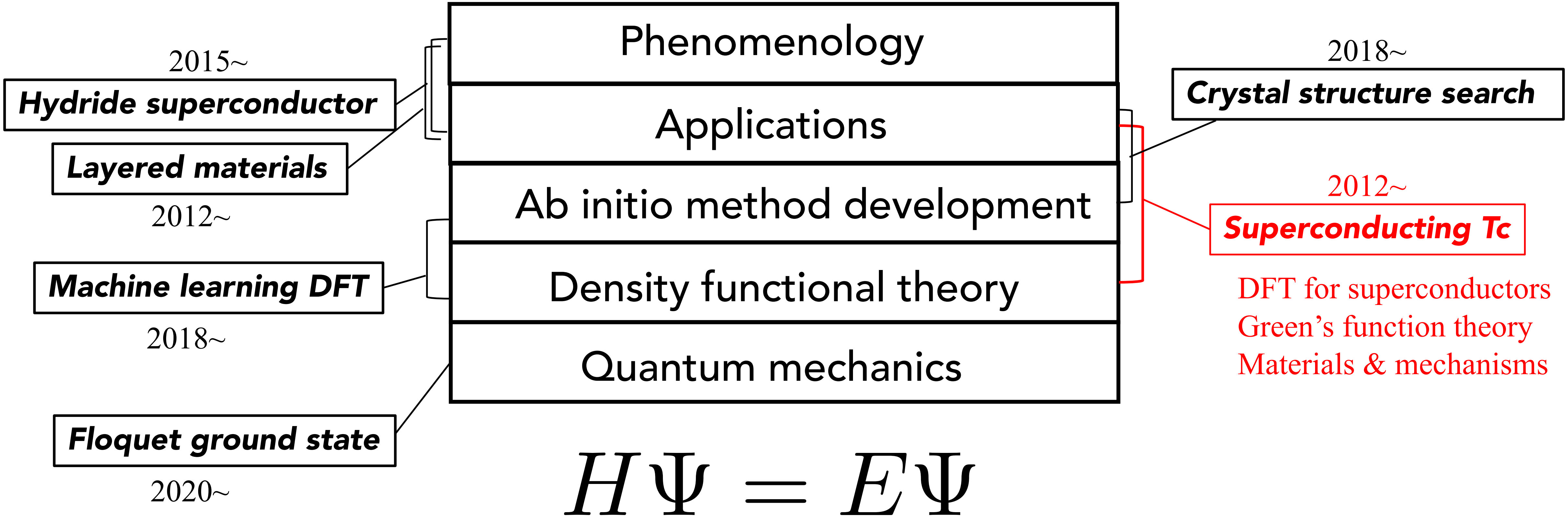
QST(量子科学技術研究開発機構)
(office in TITech, Ookayama)

RA and R. Arita, Phys. Rev. Lett. **111**, 057006 (2013);
K .Tsutsumi, Y. Hizume, M. Kawamura, RA, and S. Tsuneyuki, Phys. Rev. B **102**, 214515 (2020);
RA, Phys. Rev. B **105**, 104510 (2022).

Research scope

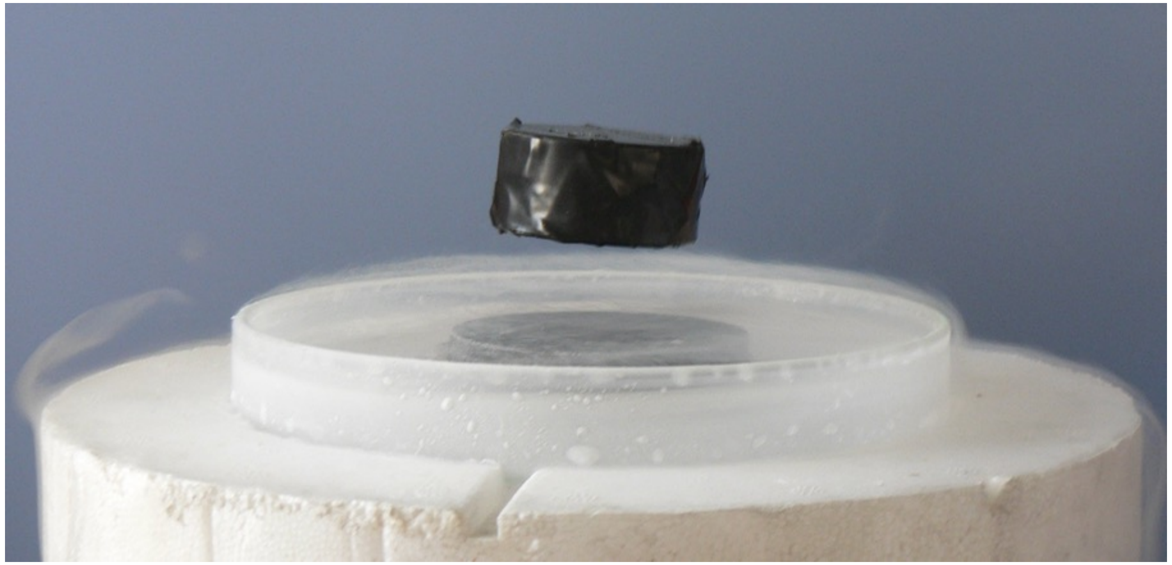
Mission: Making electronic materials accurately calculable



$$H\Psi = E\Psi$$

Superconductivity

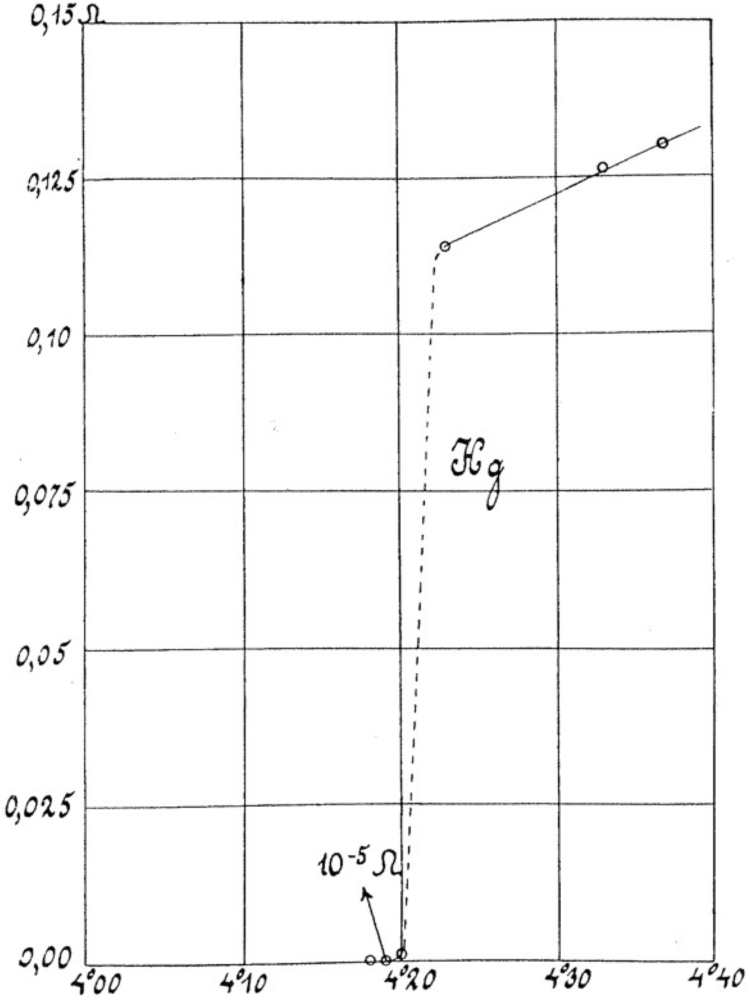
Perfect magnetic repulsion



From Wikipedia "superconductivity"

Zero resistivity

K. Onnes, Comm. Phys. Lab. Univ. Leiden; 124c (1911)



Superconductivity

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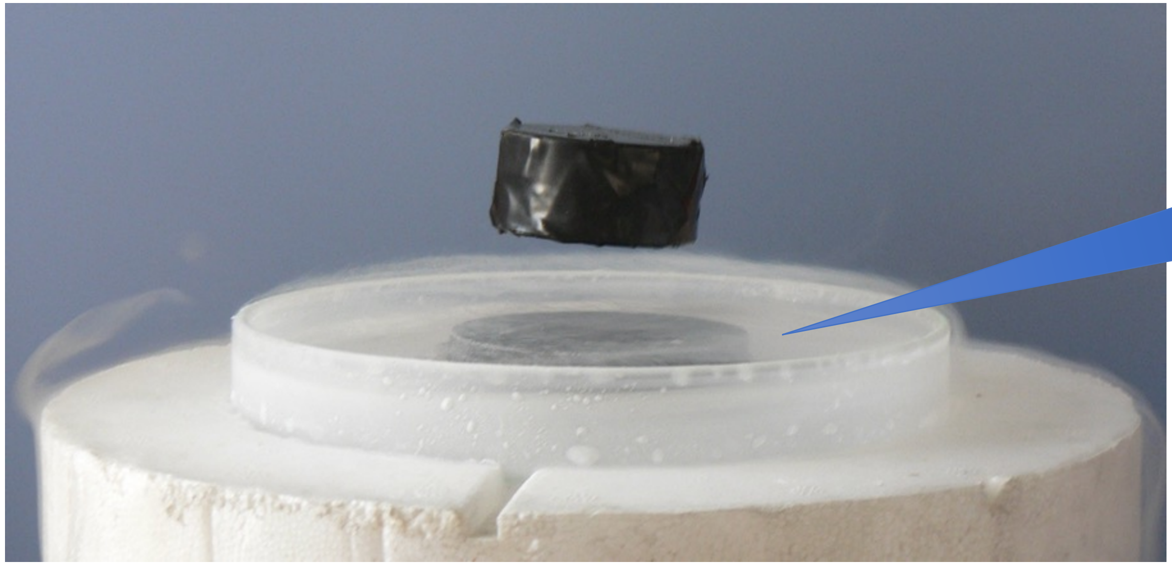
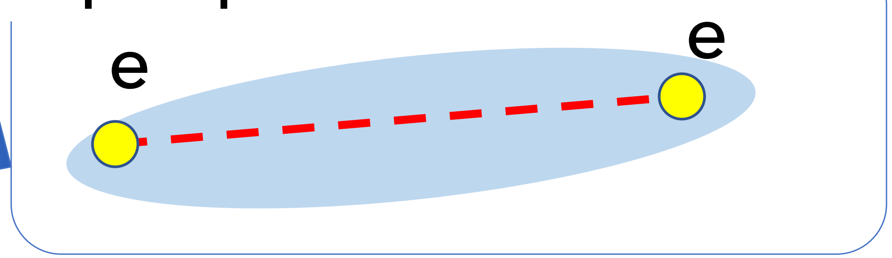
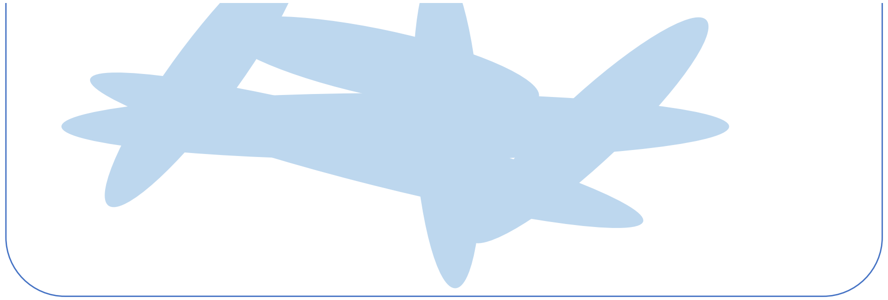


From Wikipedia “superconductivity”

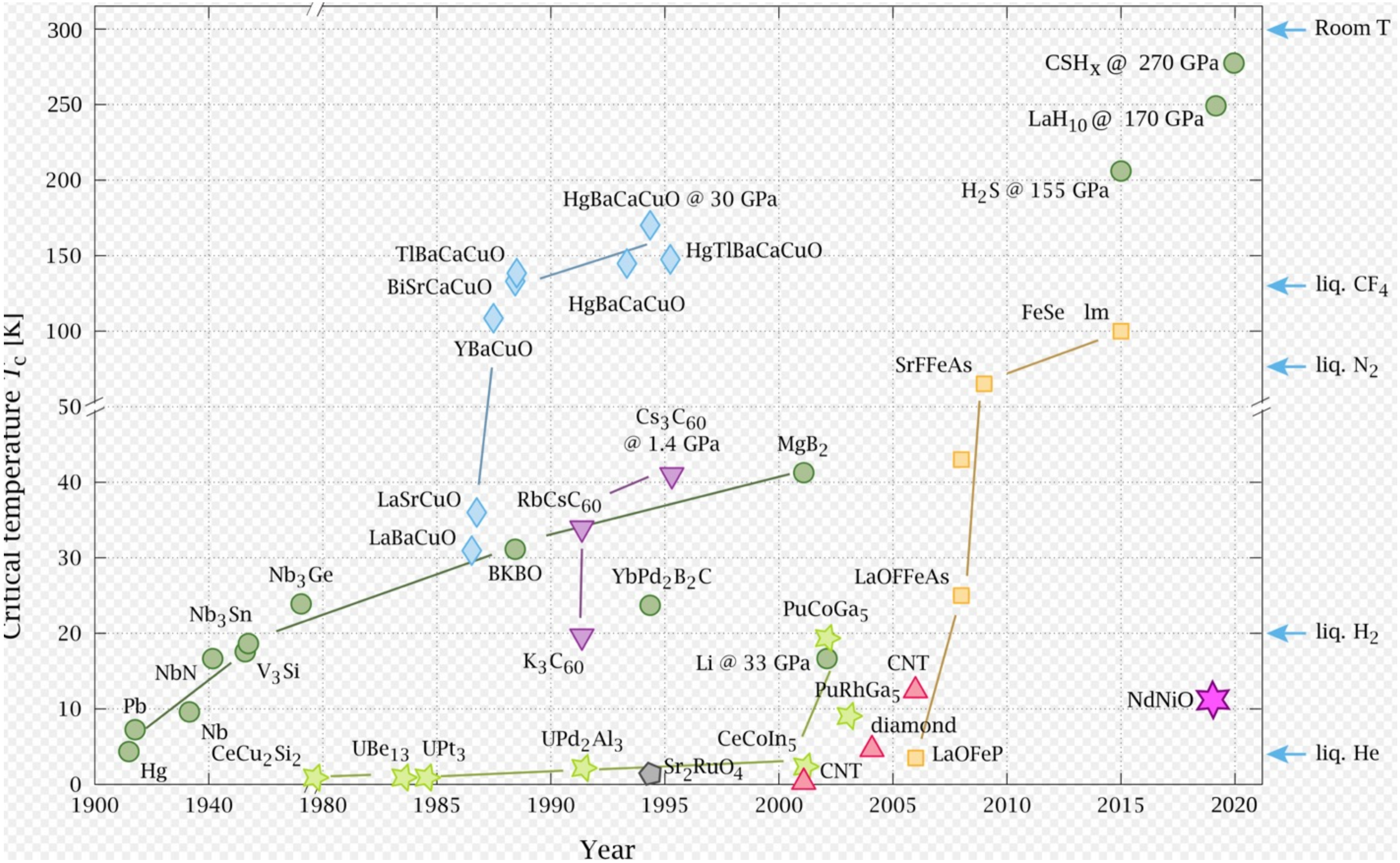
Cooper pair



Superposed wave functions



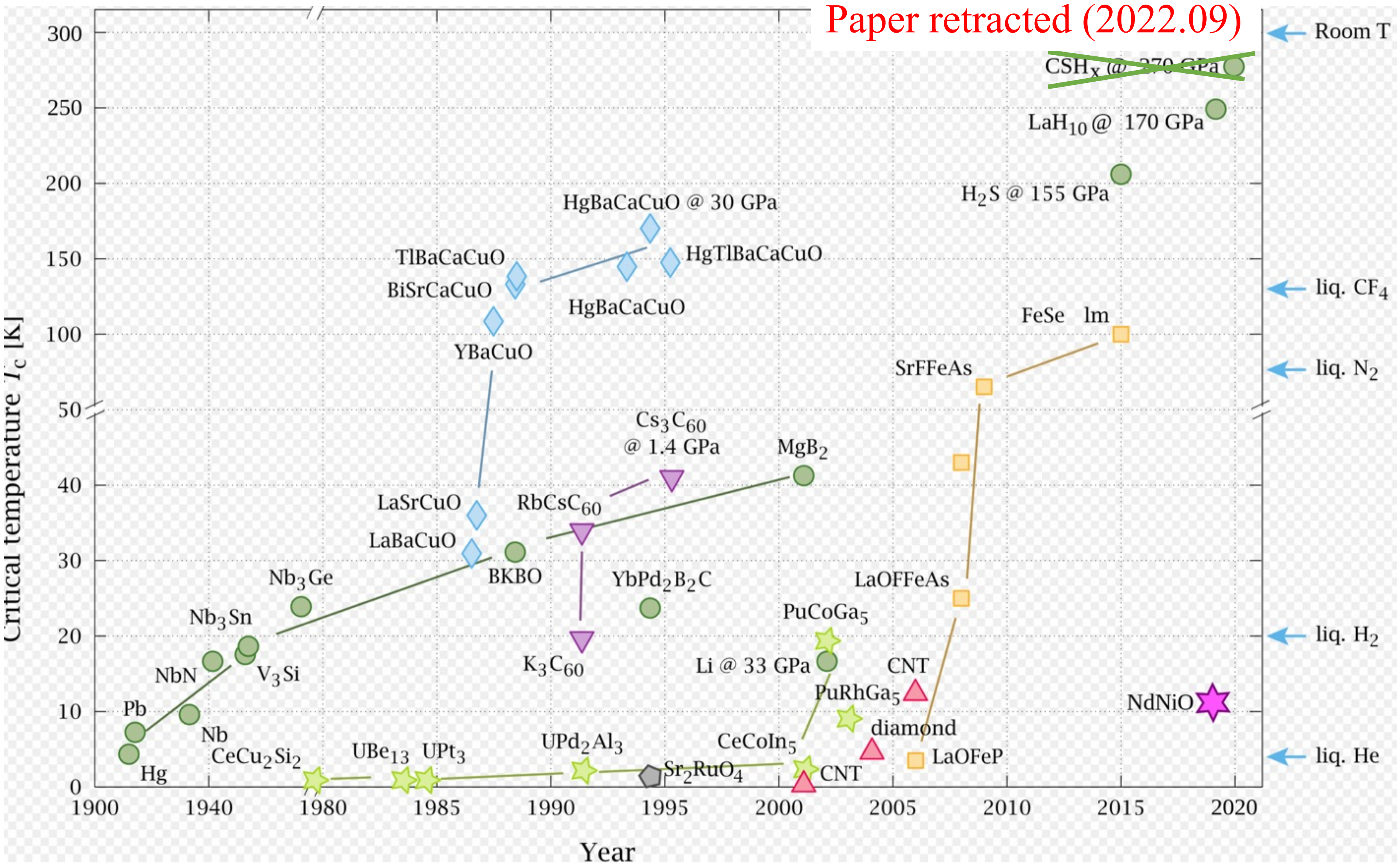
Superconductivity



- Pairing is caused by phonons

From Wikipedia “superconductivity”

Superconductivity



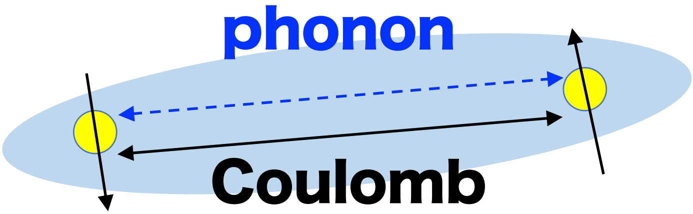
Pairing is caused by phonons

From Wikipedia “superconductivity”

Phonon mechanism for Cooper pairing

BCS theory (1957)

1, Cooper pairing

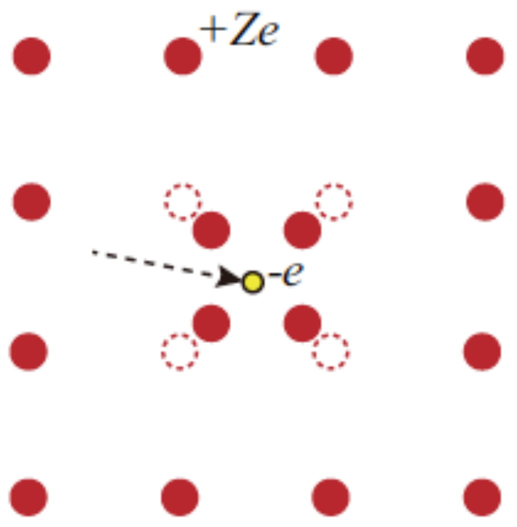


2, Overlapping pair wavefunctions

Phonon mechanism for Cooper pairing

Origin of the pairing interaction

An electron attract
ions (= exciting phonons)



Phonon mechanism for Cooper pairing

Origin of the pairing interaction

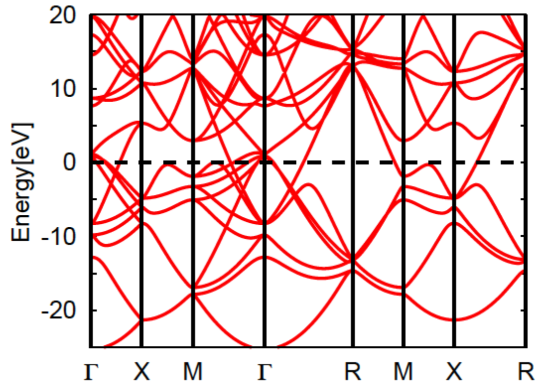
The electron flies away, but the ion distortion lasts, which in turn attracts another electron.
(=phonon absorption)

Essential ingredients of accurate SC theory

Electrons:

KS eigenstates are plausible single particle states.

Not exact



Essential ingredients of accurate SC theory

Electrons:

KS eigenstates are <u>plausible</u> single particle states.

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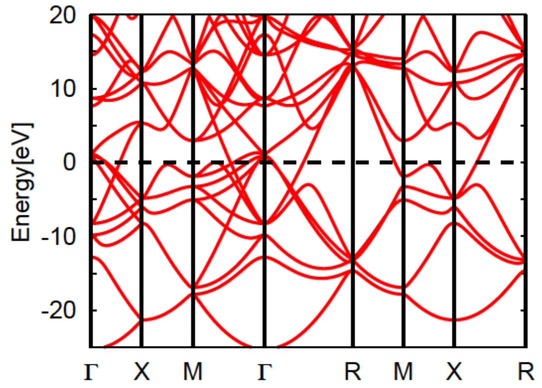

Phonons:

Exact harmonic eigenmodes are in principle available.

Essential ingredients of accurate SC theory

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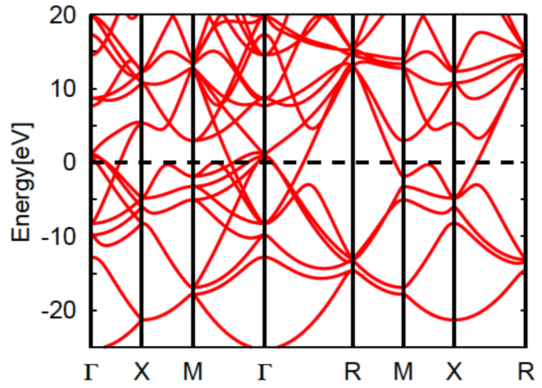

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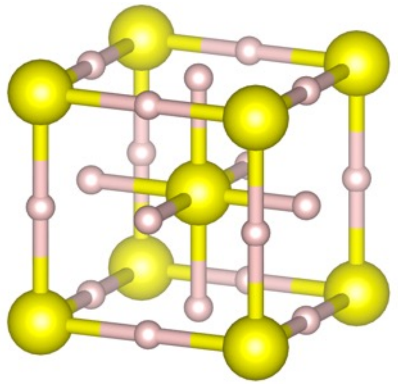

$$E = E(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3, \cdots) + [\delta\mathbf{R}_1, \delta\mathbf{R}_2, \cdots]\mathbf{D}\begin{bmatrix}\delta\mathbf{R}_1 \\ \delta\mathbf{R}_2 \\ \vdots\end{bmatrix} + O(\delta^3)$$

Migdal-Eliashberg theory for phonon SC

A. B. Migdal, Zh. Eksp. Theor. Fiz. 34, 1438 (1958); G. M. Eliashberg, *ibid*. **38**, 966 (1960);
J. R. Schrieffer, *Theory of superconductivity* (Benjamin, NY, 1964)

$$H = H_{\mathrm{el}} + H_{\mathrm{ph}} + H_{\mathrm{el-ph}} + H_{\mathrm{el-el}}$$

Green's function (amplitude of creation/annihilation process)

$$\mathbf{G}_{n\mathbf{k}}(t) = \begin{pmatrix} -\langle T c_{n\mathbf{k}\sigma}(t) c^{\dagger}_{n\mathbf{k}\sigma}(0)\rangle & -\langle T c_{n\mathbf{k}\sigma}(t) c_{n-\mathbf{k}-\sigma}(0)\rangle \\ -\langle T c^{\dagger}_{n-\mathbf{k}-\sigma}(t) c^{\dagger}_{n\mathbf{k}\sigma}(0)\rangle & -\langle T c^{\dagger}_{n-\mathbf{k}-\sigma}(t) c_{n-\mathbf{k}-\sigma}(0)\rangle \end{pmatrix}$$

$$\equiv \begin{pmatrix} G_{n\mathbf{k}}(t) & F_{n\mathbf{k}}(t) \\ F^{*}_{n\mathbf{k}}(t) & -G_{n\mathbf{k}-\sigma}(-t) \end{pmatrix}$$

When anomalous components (F) become nonzero,
system becomes superconducting.

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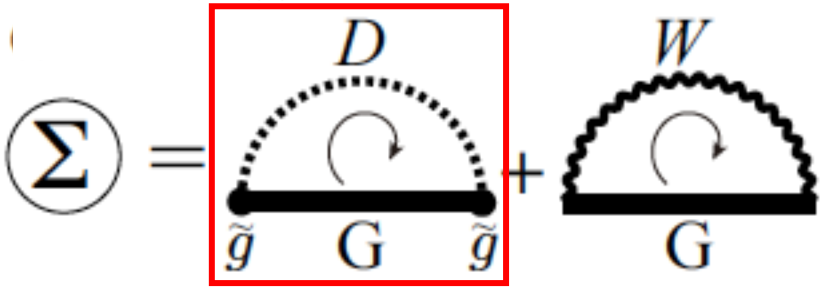
Dyson equation

$$\mathbf{G}^{-1} = \mathbf{G}_0^{-1} - \mathbf{\Sigma}$$

Approximate self energy



electron emit/absorb phonon

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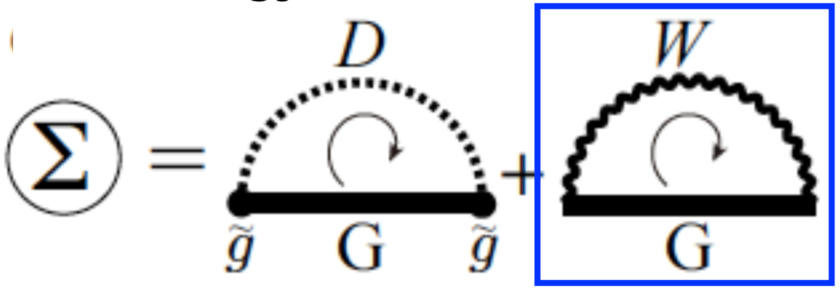


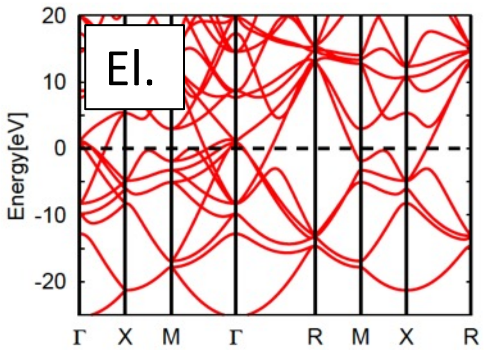
electrons interact via screened Coulomb

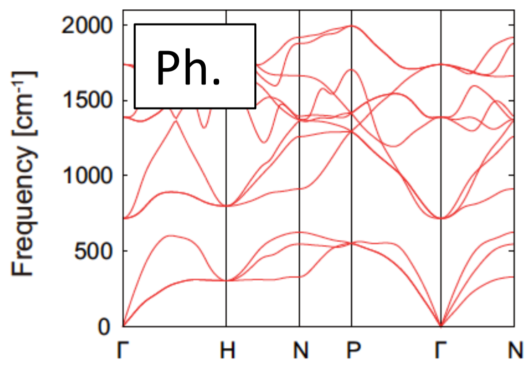
First-principles Migdal-Eliashberg theory

A. B. Migdal, Zh. Eksp. Theor. Fiz. 34, 1438 (1958); G. M. Eliashberg, *ibid*. **38**, 966 (1960);
J. R. Schrieffer, *Theory of superconductivity* (Benjamin, NY, 1964)

$$H = H_{\mathrm{el}} + H_{\mathrm{ph}} + H_{\mathrm{el-ph}} + H_{\mathrm{el-el}}$$

$$H_{\mathrm{el}} = H_{\mathrm{KS}}, \quad H_{\mathrm{ph}} = H_{\mathrm{KS}}^{\mathrm{phonon}}$$





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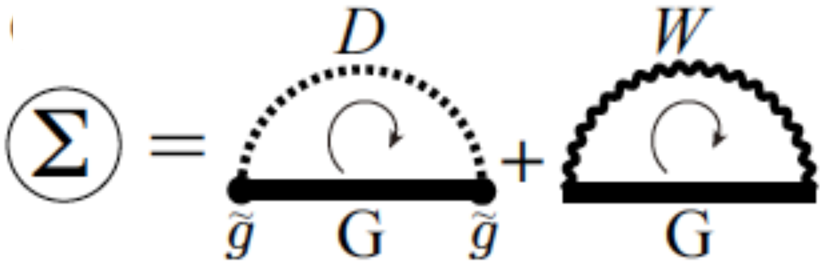

First-principles superconducting calc.

1, Calculate el and ph state by KS
2, Solve the Dyson eq.

First-principles Migdal-Eliashberg theory

A. B. Migdal, Zh. Eksp. Theor. Fiz. 34, 1438 (1958); G. M. Eliashberg, *ibid*. **38**, 966 (1960);
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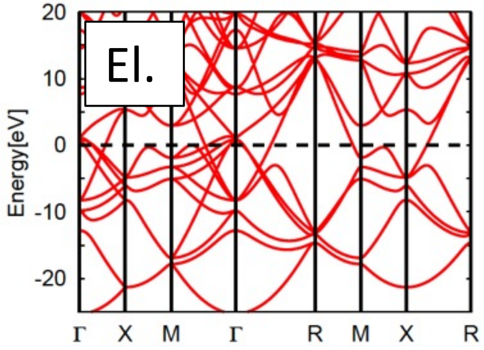
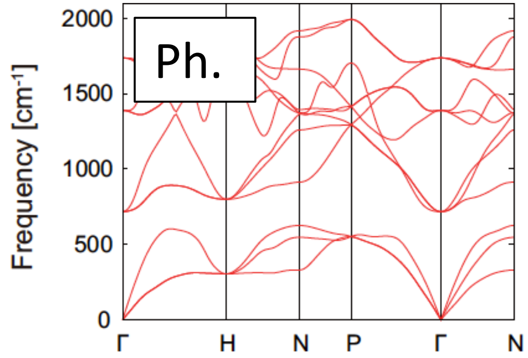
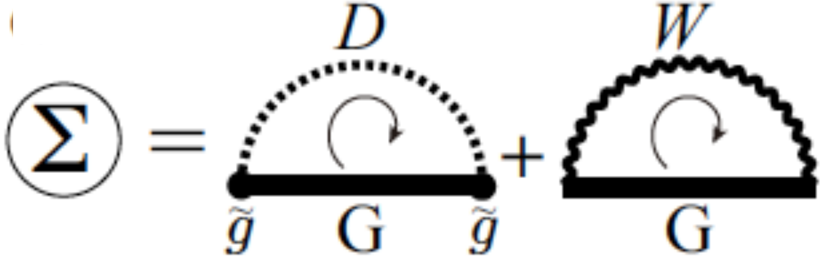




Dyson equation

$$\mathbf{G}^{-1} = \mathbf{G}_0^{-1} - \mathbf{\Sigma}$$

Approximate self energy



First-principles superconducting calc.

1. Calculate el and ph state by KS

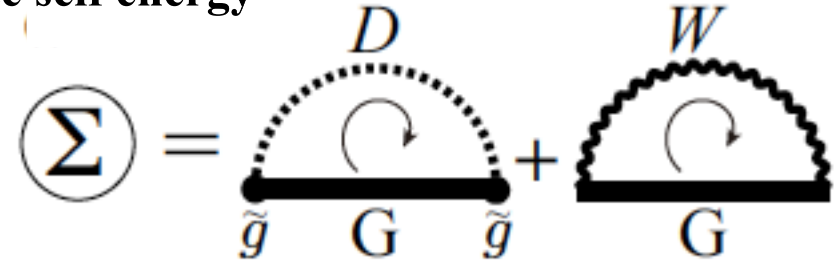
No one in this 60 years has faithfully solved ME equation with real KS basis!

What is difficult for faithful ME equations?

Dyson equation

$$\mathbf{G}^{-1} = \mathbf{G}_0^{-1} - \mathbf{\Sigma}$$

Approximate self energy



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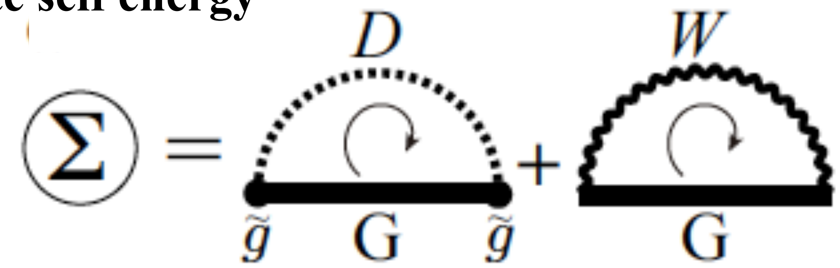
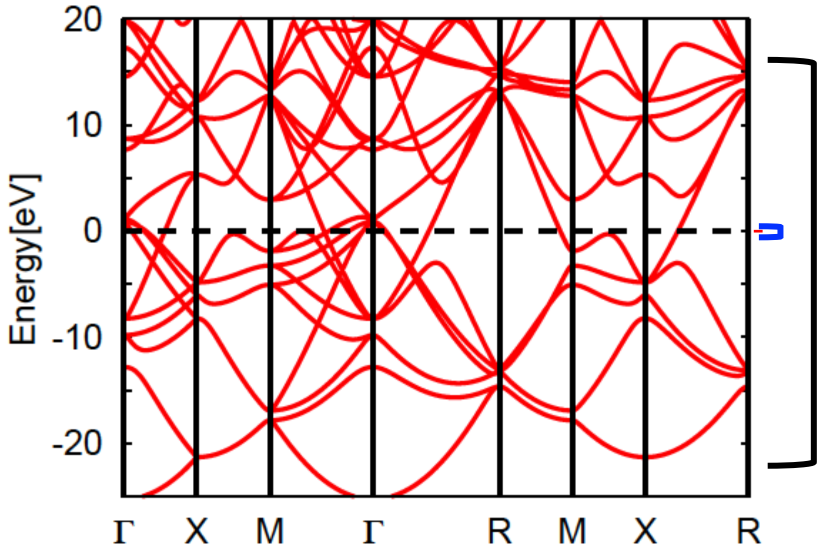
Approximate self energy



Phonon mediated pairing

El-El Coulomb repulsion



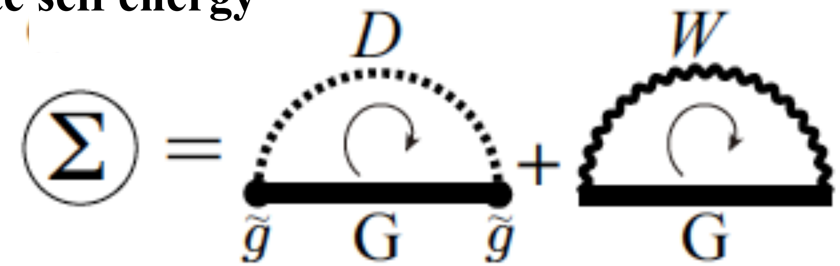
Alternative: DFT for superconductors

Luders, Marques, Gross et al., 2005.

Dyson equation

$$\mathbf{G}^{-1} = \mathbf{G}_0^{-1} - \mathbf{\Sigma}$$

Approximate self energy



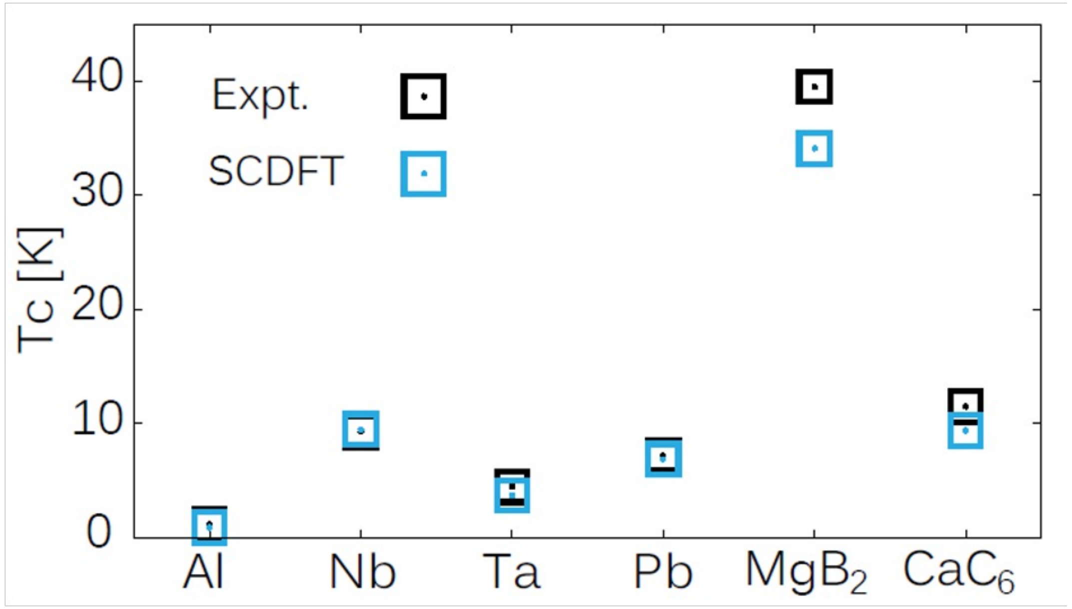
SCDFT gap equation (low cost)

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

Alternative: DFT for superconductors

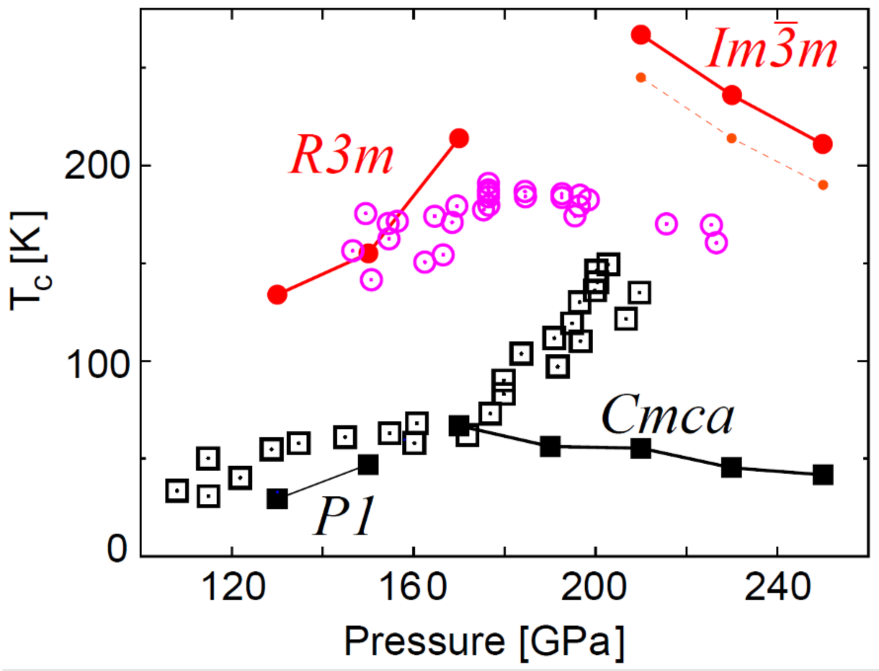
Typical phonon mediated superconductors

Luders, Marques, Gross et al., 2005; Floris et al., 2005; Sanna et al., 2007.



High-pressure sulfur hydride

RA. Kawamura, Tuneyuki, Nomura and Arita, Phys. Rev. B 91, 224513 (2015)



Extension to electronic fluctuations?

1, Extensions of DFT for superconductors

2, Faithful solution of the Migdal-Eliashberg eqs.

Extensions of DFT for superconductors

DFT for superconductors (SCDFT)

Oliveira, Gross, Kohn, Phys. Rev. Lett. 60, 2430 (1988);
Luders, Marques, Gross et al., Phys. Rev. B 72, 024545; 024546 (2005).

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

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

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)

DFT for superconductors (SCDFT)

Oliveira, Gross, Kohn, Phys. Rev. Lett. 60, 2430 (1988);
Luders, Marques, Gross et al., Phys. Rev. B 72, 024545; 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

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

DFT for superconductors (SCDFT)

Oliveira, Gross, Kohn, Phys. Rev. Lett. 60, 2430 (1988);
Luders, Marques, Gross et al., Phys. Rev. B 72, 024545; 024546 (2005).

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

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

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

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

$$\left[-\frac{\nabla_r^2}{2} + v_0^e(\boldsymbol{r}) - \mu\right] u_n(\boldsymbol{r}) - \int \Delta_0(\boldsymbol{r}, \boldsymbol{r}') v_n(\boldsymbol{r}') = E_n u_n(\boldsymbol{r})$$

$$-\left[-\frac{\nabla_r^2}{2} + v_0^e(\boldsymbol{r}) - \mu\right] v_n(\boldsymbol{r}) - \int \Delta_0^*(\boldsymbol{r}, \boldsymbol{r}') u_n(\boldsymbol{r}') = E_n v_n(\boldsymbol{r})$$

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

$v_0^e(\boldsymbol{r})$ $\Delta_0(\boldsymbol{r}, \boldsymbol{r}')$

$v_0^n(\underline{\boldsymbol{R}})$

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

The "gap" equation

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

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

Decoupling of dependencies

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

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

The "gap" equation

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

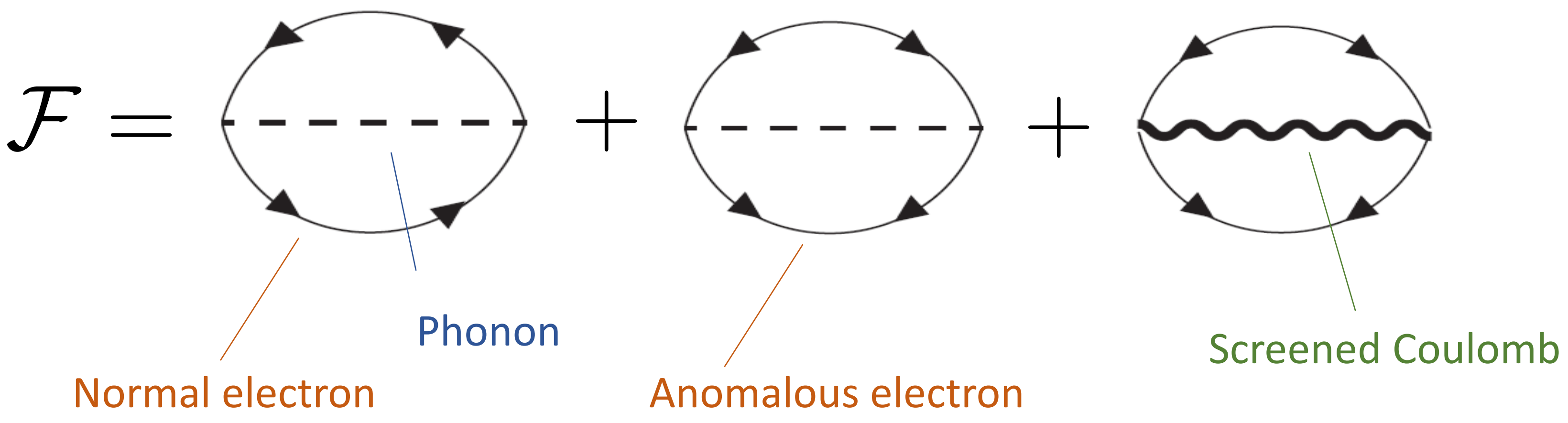
$$\Delta_{n\mathbf{k}} = -\mathcal{Z}_{n\mathbf{k}}\Delta_{n\mathbf{k}} - \frac{1}{2}\sum_{n'\mathbf{k}'}\mathcal{K}_{n\mathbf{k}n'\mathbf{k}'}\frac{\tanh[(\beta/2)E_{n'\mathbf{k}'}]}{E_{n'\mathbf{k}'}}\Delta_{n'\mathbf{k}'}$$

Exchange-correlation kernels

$$\mathcal{Z}, \mathcal{K} = \frac{\delta^2 \mathcal{F}}{\delta\chi\delta\chi}$$

Exchange-correlation free energy

Free energy for phonon-mediated ex-corr kernels



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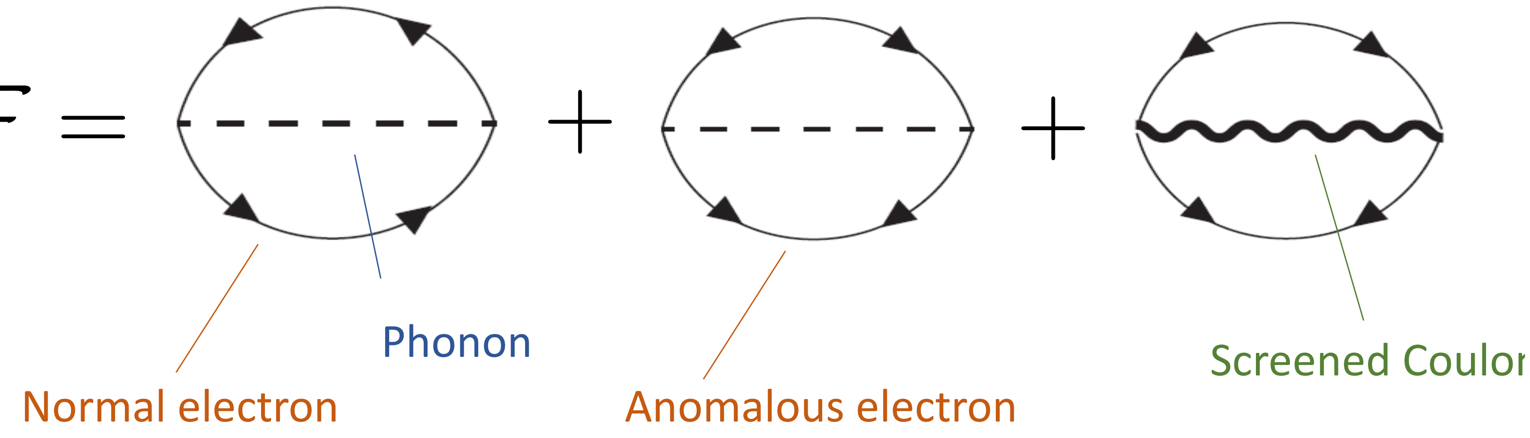
Exchange-correlation kernels

$$\mathcal{Z},\mathcal{K} = \frac{\delta^2\mathcal{F}}{\delta\chi\delta\chi}$$

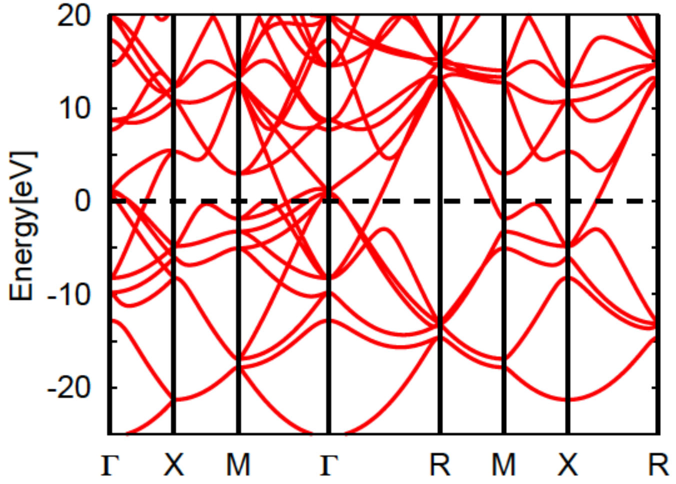
Exchange-correlation free energy

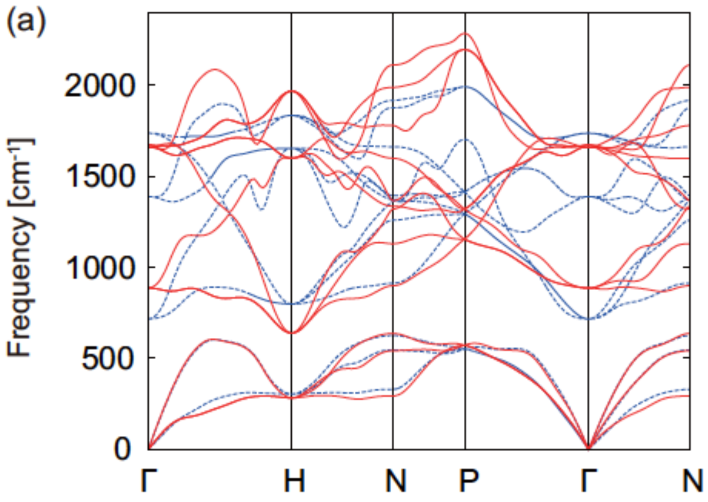
Free energy for phonon-mediated ex-corr kernels

$$\mathcal{F} =$$



Propagators by KS calc.





First-principles calculation method for superconducting T_c

Electron correlation

Conventional phonon SC

DFT for superconductors
Luders, Marques et al., 2005;
Sanna, Pellegrini, Gross 2020.

Migdal-Eliashberg theory
Sano, Koretsune, Tadano, RA, Arita, 2016;
Sanna *et al.*, 2018;
Wang *et al.*, 2020

Intermediate, ph+el. fluctuation

Plasma oscillation
RA and Arita, 2013;
Davydov et al., 2020.

Spin fluctuation
Essenberger et al., 2014.

Unconventional SC

Unprecedented
(cf. Downfolding)

First-principles calculation method for superconducting T_c

Conventional phonon SC

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Luders, Marques et al., 2005;
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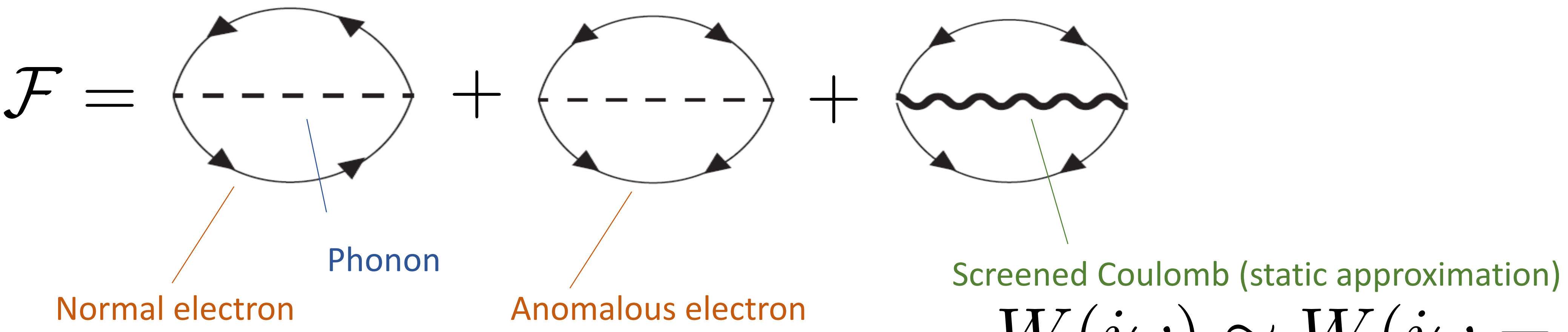
Extension to plasmon pairing

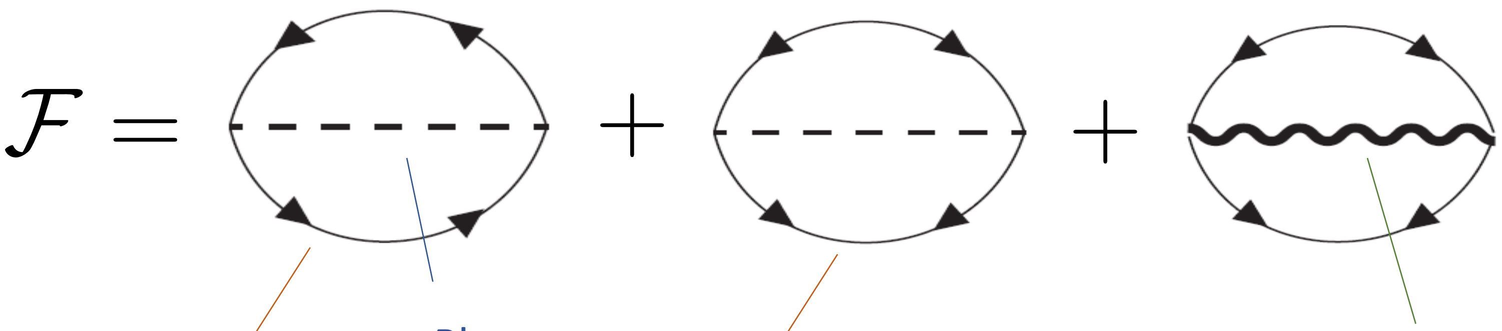
Free energy



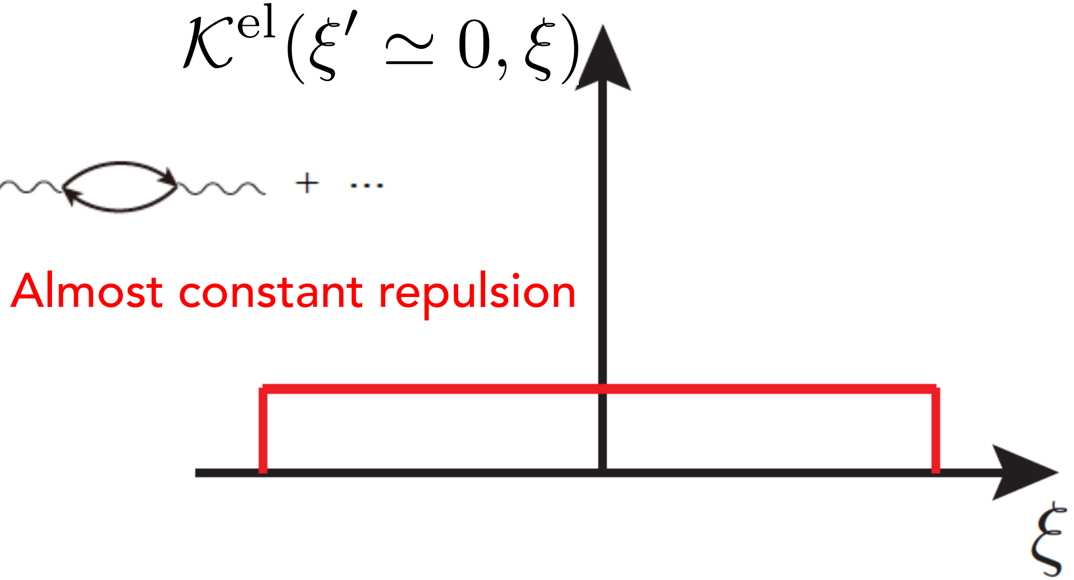
$$W(i\omega) \simeq W(i\omega = 0)$$

Extension to plasmon pairing

Free energy



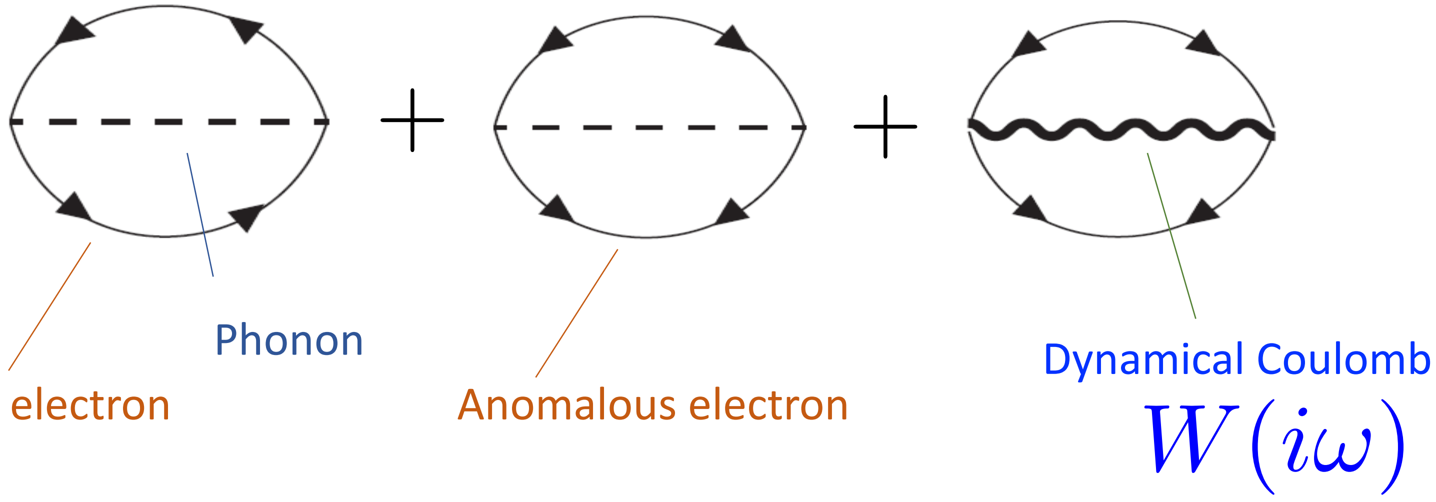
$$W(i\omega) \simeq W(i\omega = 0)$$

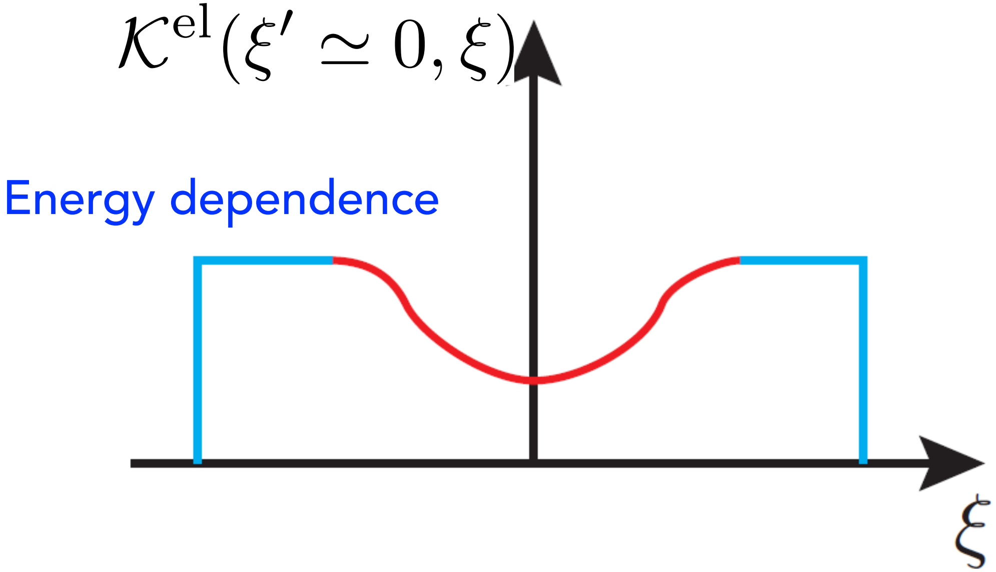


Extension to plasmon pairing

Free energy

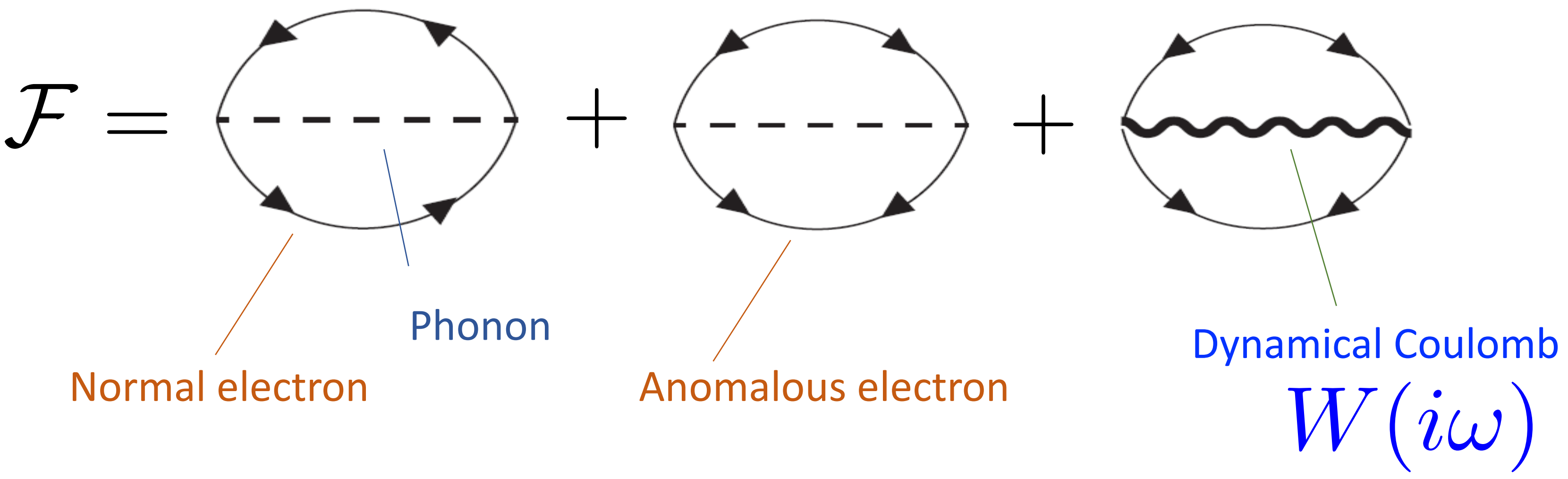
$\mathcal{F} =$

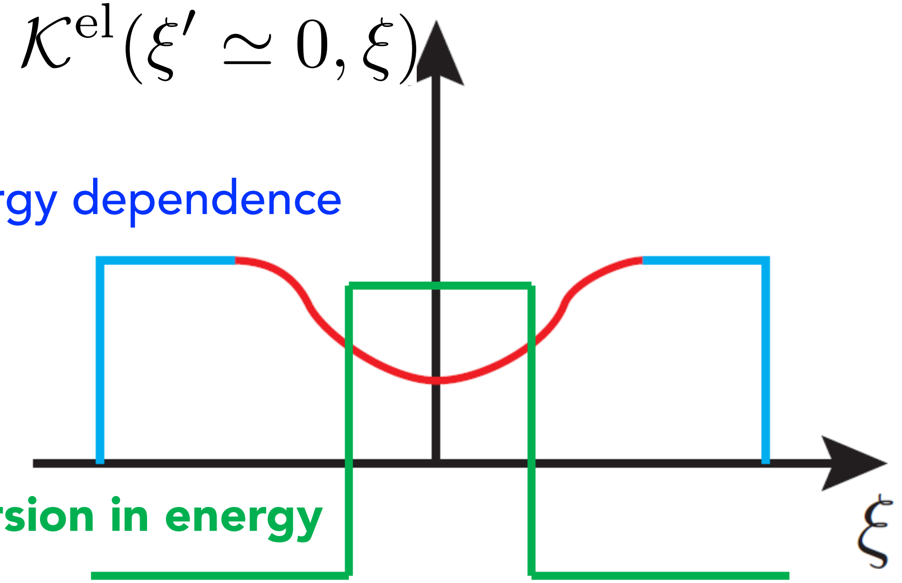




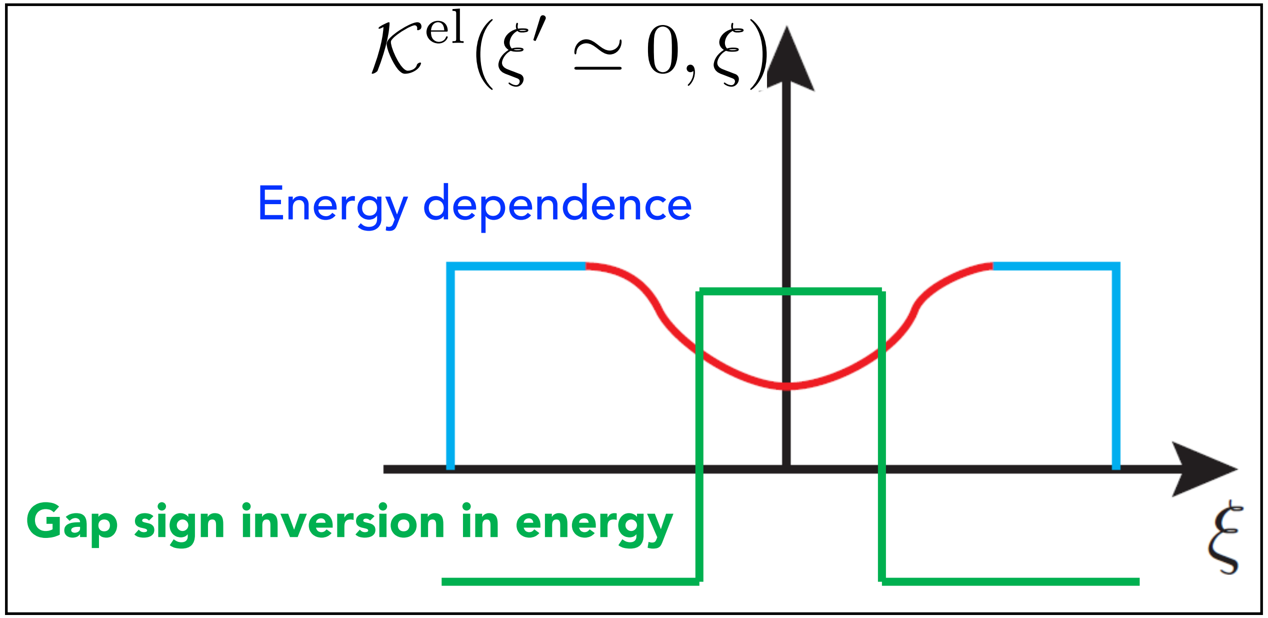
Extension to plasmon pairing

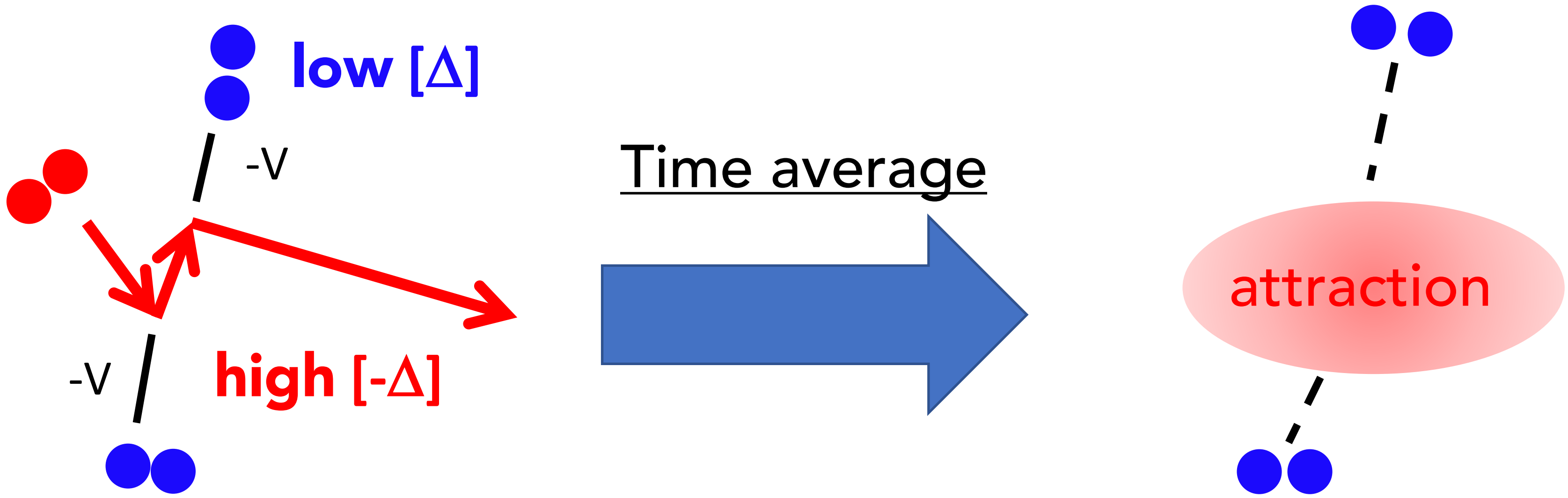
Free energy





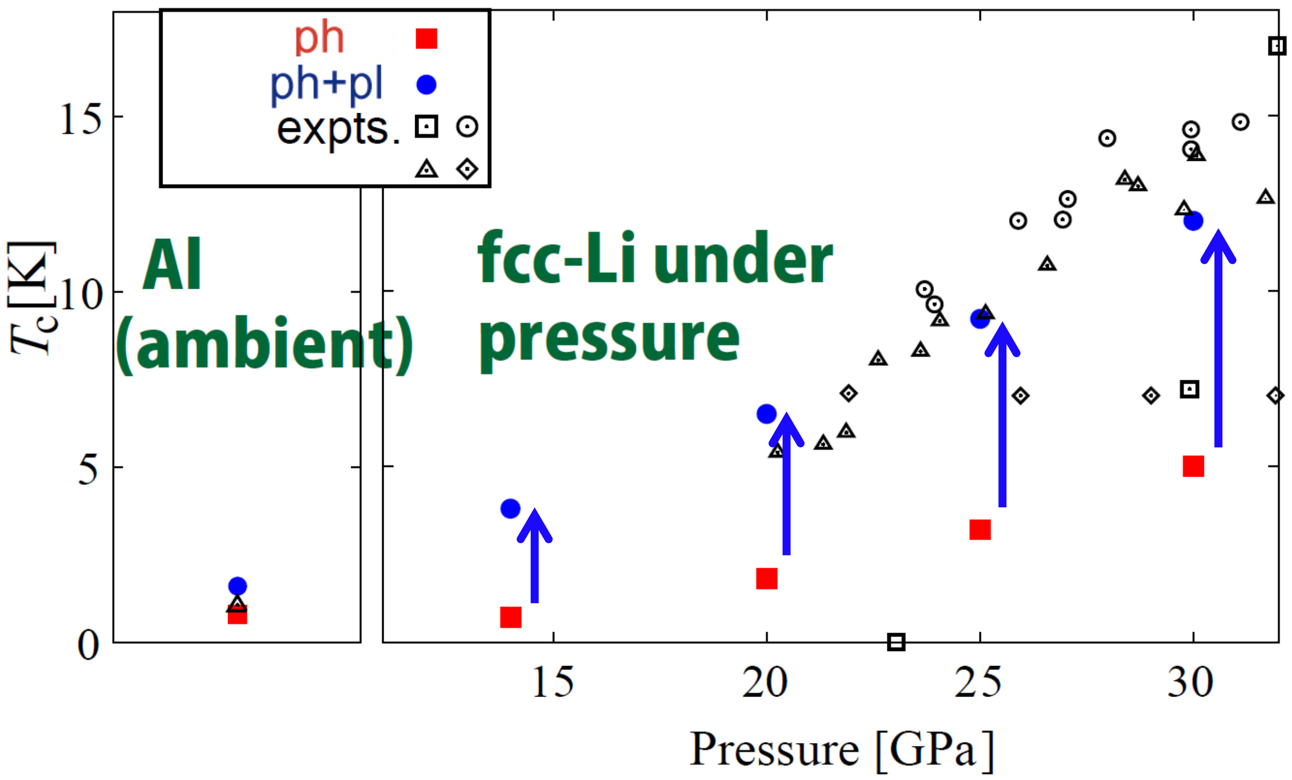
Y. Takada, J. Phys. Soc. Jpn. **45**, 786 (1978)


$\mathcal{K}^{\rm el}(\xi' \simeq 0, \xi)$
Energy dependence
Gap sign inversion in energy
ξ


low [Δ]
-V
-V
high [-Δ]
Time average
attraction

Extension to plasmon pairing

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



Plasmon pairing cooperates with phonons, enhancing Tc.

This enhancement is more or less ubiquitous.

Cf. hydrides: RA et al., Phys. Rev. B 91, 224513 (2015).

Spin fluctuation

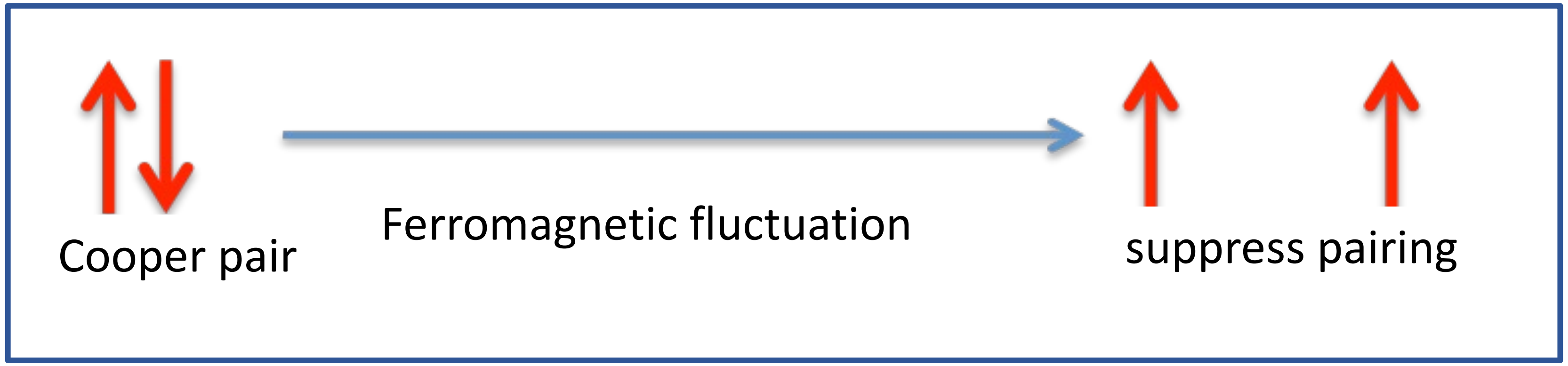
Unconventional：Possible origin of pairing

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

N. F. Berk and J. R. Schrieffer, Phys. Rev. Lett. 17, 433 (1966);
H. Rietschel and H. Winter, Phys. Rev. Lett. **43**, 1256 (1979).



Ex. effect is significant in transition metals having *d*-electrons

Interaction via spin and charge fluctuations

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

RPA (no spin dep.)

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

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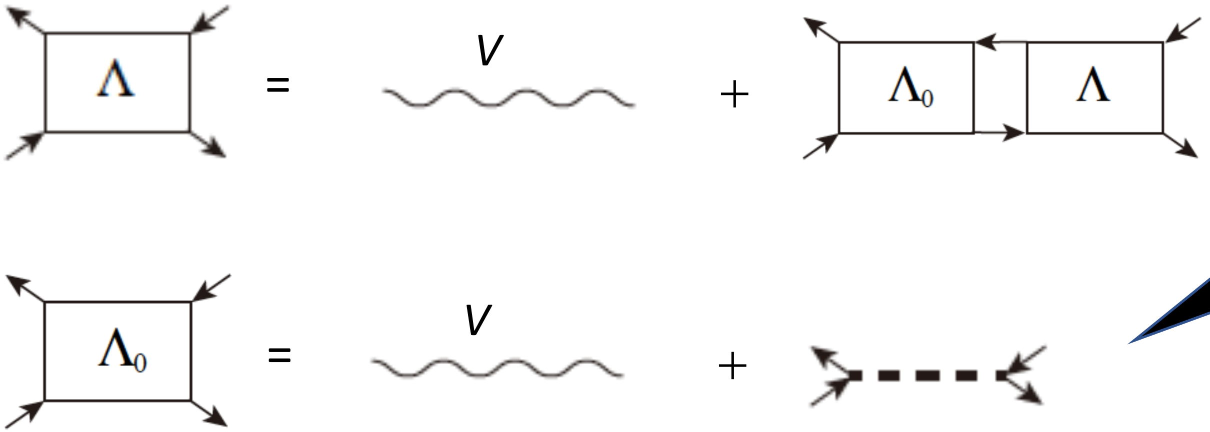
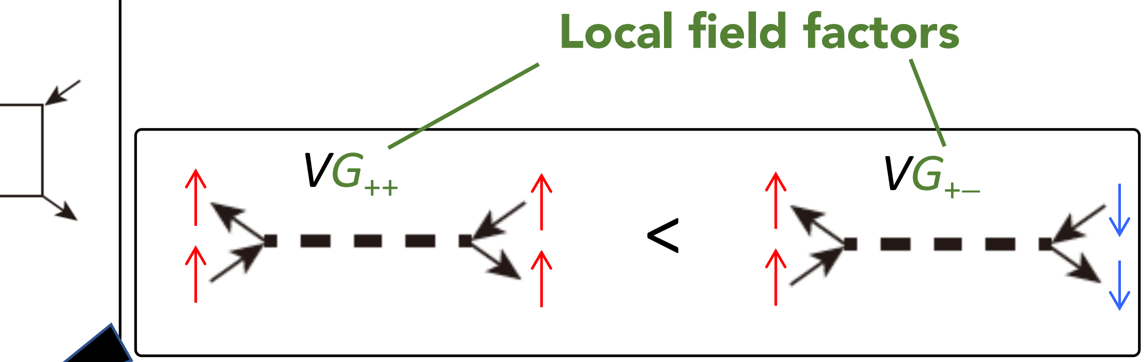
G. Vignale and K. S. Singwi, PRB **32**, 2156 (1985); F. Essenberger et al., PRB **90**, 214504 (2014)

Spin dependent interaction





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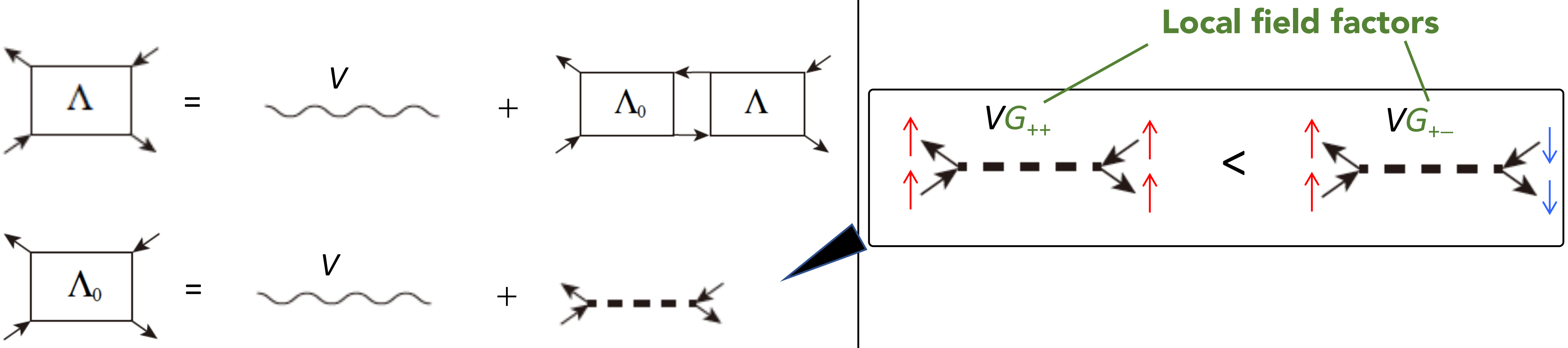
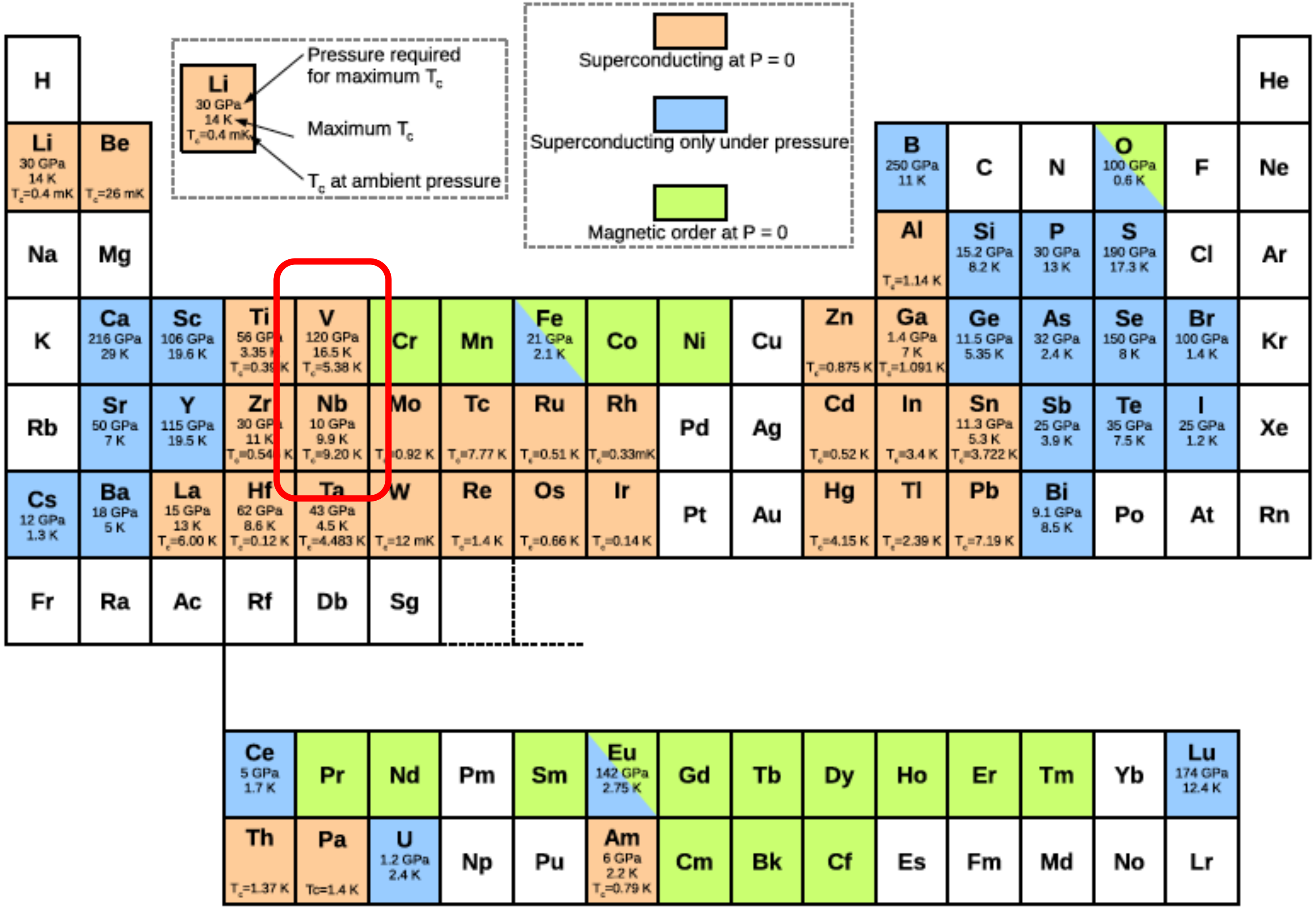


$$V(G_{++}+G_{+-}) \simeq -\frac{\delta^2 E_{\rm xc}^{\rm LSDA}}{\delta n \delta n} \qquad V(G_{++}-G_{+-}) \simeq \frac{\delta^2 E_{\rm xc}^{\rm LSDA}}{\delta m \delta m}$$

Application to elemental superconductors

J. J. Hamlin, Physica C 514, 59 (2015).



Calculated data from
S. Y. Savrasov and D. Y. Savrasov,
Phys. Rev. B 54, 16487 (1996)

	V	Nb
λ	1.19	1.26
$\omega_{\ln}(\mathrm{K})$	245	185
$T_c^{\exp}(\mathrm{K})$	5.40	9.25

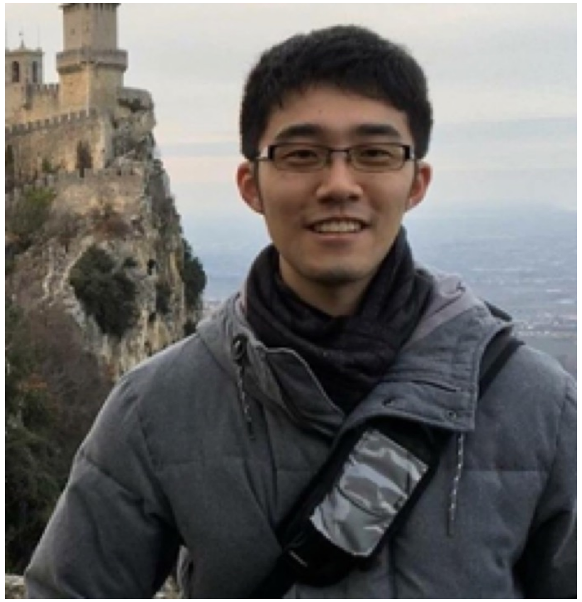
Tc (Nb)>Tc (V) by spin fluctuations?

H. Rietschel and H. Winter,
Phys. Rev. Lett. **43**, 1256 (1979).

Application to elemental superconductors

K .Tsutsumi, Y. Hizume, M. Kawamura, RA, and S. Tsuneyuki, Phys. Rev. B **102**, 214515 (2020)

Kentaro Tsutsumi



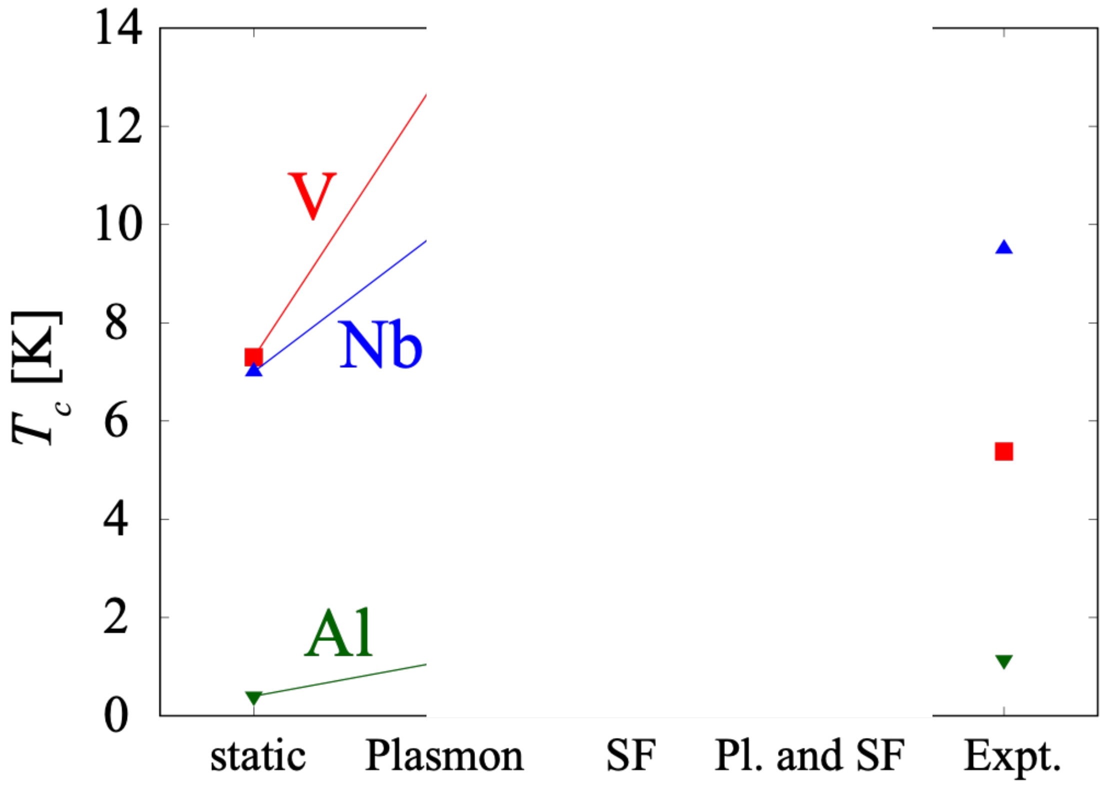
Yuma Hizume

Mitsuaki Kawamura (ISSP)

c.f.: M. Kawamura, Y. Hizume and T. Ozaki, Phys. Rev. B **101**, 134511 (2020)

Application to elemental superconductors

K .Tsutsumi, Y. Hizume, M. Kawamura, R. Akashi, and S. Tsuneyuki, Phys. Rev. B **102**, 214515 (2020)



○	○	○	○	**Phonon**
○	○	○	○	**stat. Co**
-	○	-	○	**Plasmon**
-	-	○	○	**SF**

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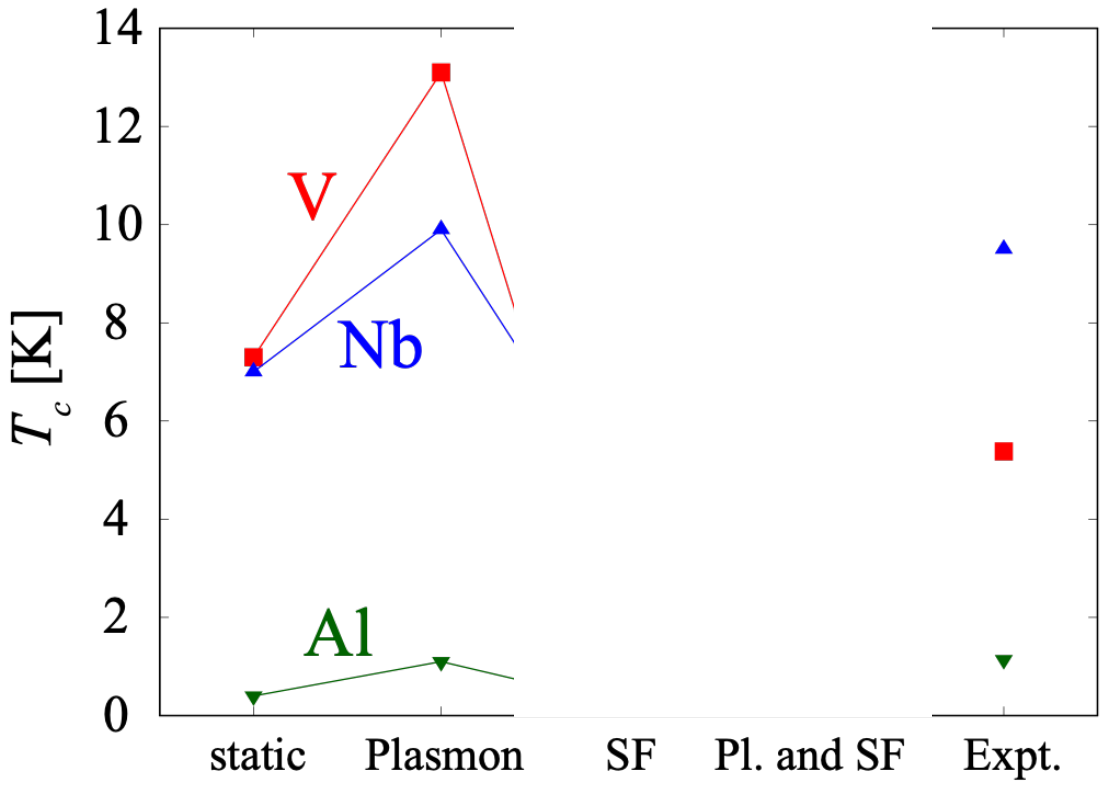

○	○	○	○	**Phonon**
○	○	○	○	**stat. Co**
-	○	-	○	**Plasmon**
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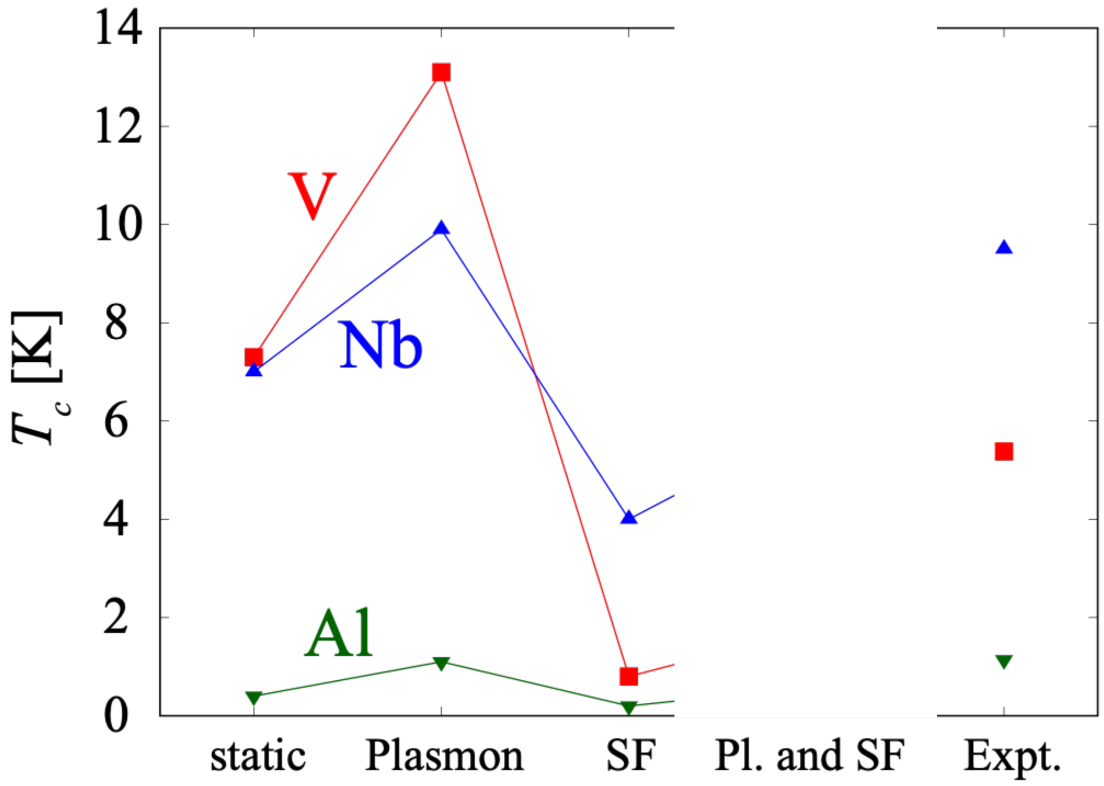

○	○	○	○	**Phonon**
○	○	○	○	**stat. Co**
-	○	-	○	**Plasmon**
-	-	○	○	**SF**

Application to elemental superconductors

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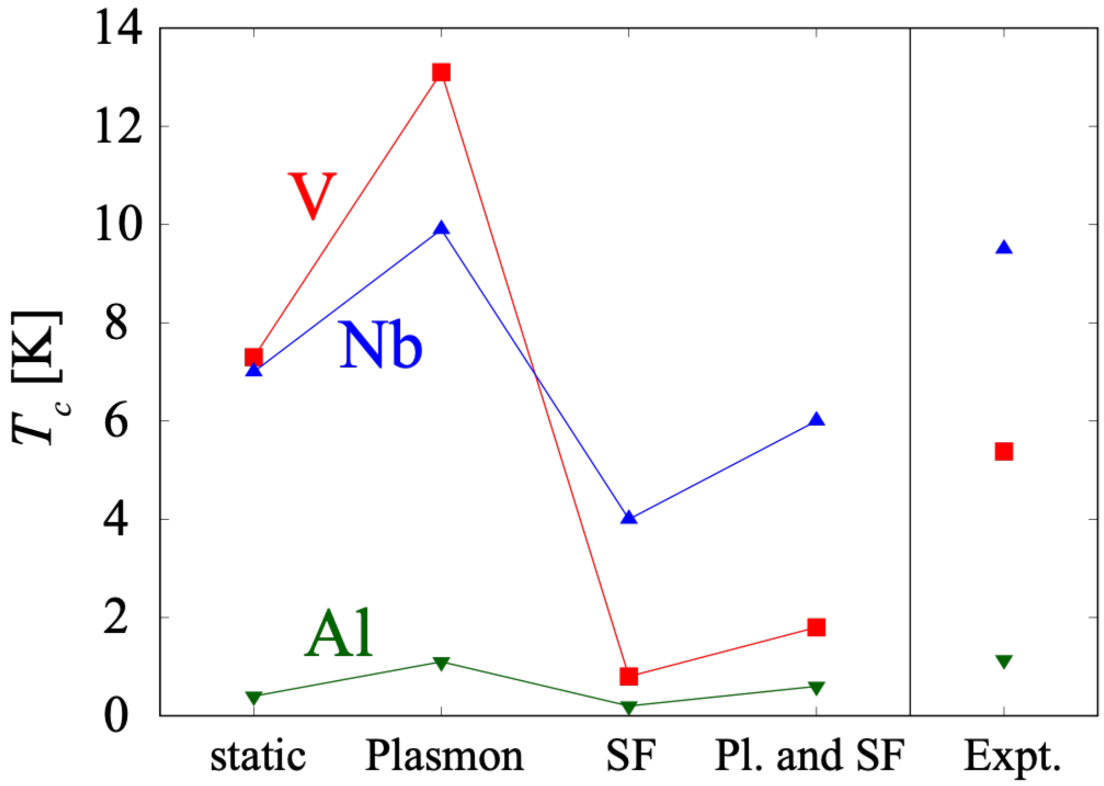


○	○	○	○	**Phonon**
○	○	○	○	**stat. Co**
-	○	-	○	**Plasmon**
-	-	○	○	**SF**

Spin fluctuation effect is indeed responsible for Tc (Nb)> Tc (V).

Faithful solution of the Migdal-Eliashberg eqs.

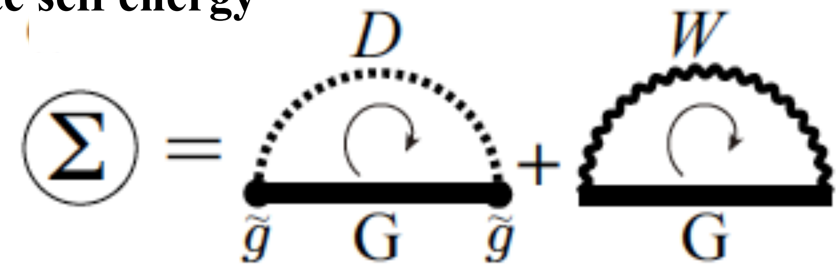
Alternative: DFT for superconductors

Luders, Marques, Gross et al., 2005;

Dyson equation

$$\mathbf{G}^{-1} = \mathbf{G}_0^{-1} - \mathbf{\Sigma}$$

Approximate self energy



SCDFT gap equation (low cost)

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

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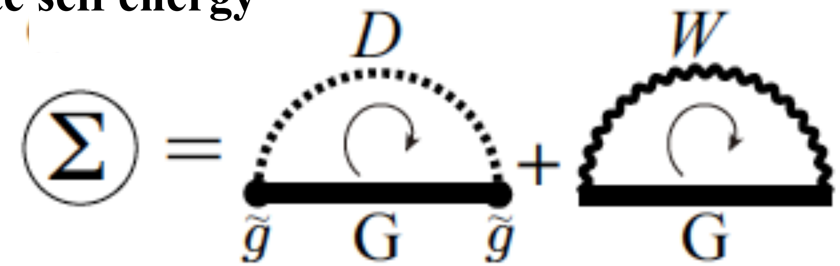

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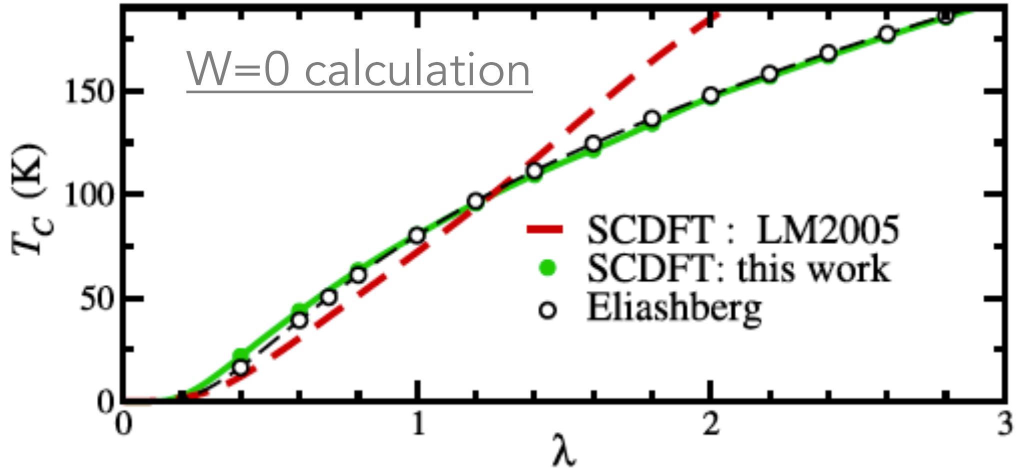
Sanna, Pellegrini, Gross, Phys. Rev. Lett. **125**, 057001 (2020)



Agreement is not perfect
(and yet under investigation)

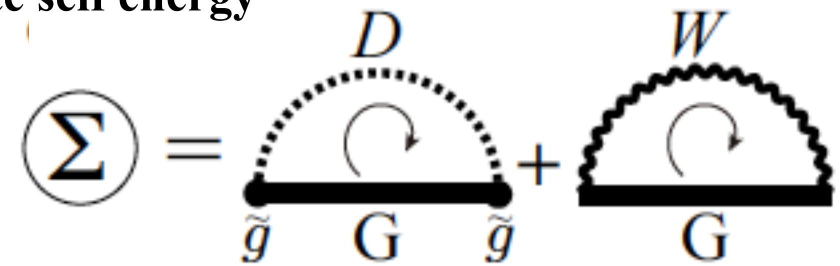
Alternative: DFT for superconductors

Luders, Marques, Gross et al., 2005;

Dyson equation

$$\mathbf{G}^{-1} = \mathbf{G}_0^{-1} - \mathbf{\Sigma}$$

Approximate self energy



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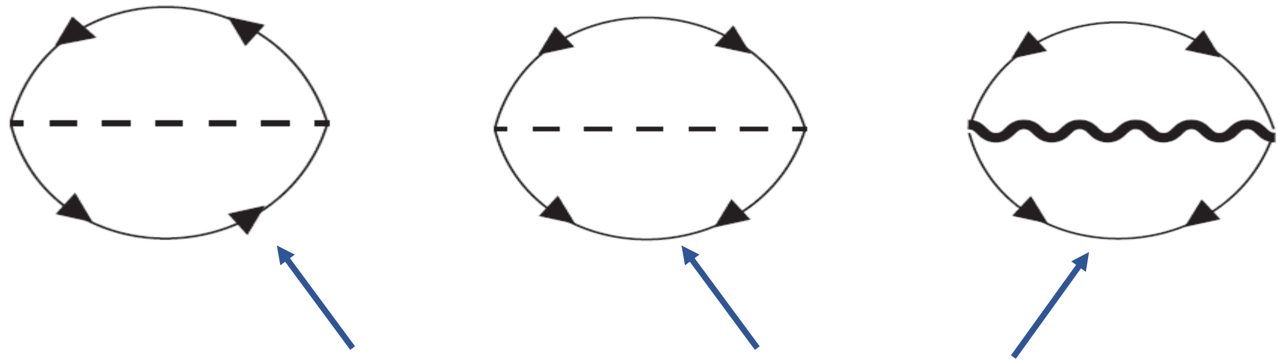
$$\Delta_{n\mathbf{k}} = -\mathcal{Z}_{n\mathbf{k}}\Delta_{n\mathbf{k}} - \frac{1}{2}\sum_{n'\mathbf{k}'} \mathcal{K}_{n\mathbf{k}n'\mathbf{k}'} \frac{\tanh[(\beta/2)E_{n'\mathbf{k}'}]}{E_{n'\mathbf{k}'}}\Delta_{n'\mathbf{k}'}.$$

<u>SCDFT ex-corr free energy</u>

$$\mathcal{F} =$$



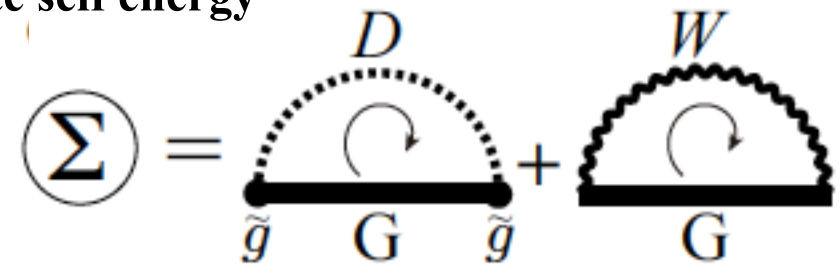
Unrenormalized KS propagators!

What is difficult for faithful ME equations?

Dyson equation

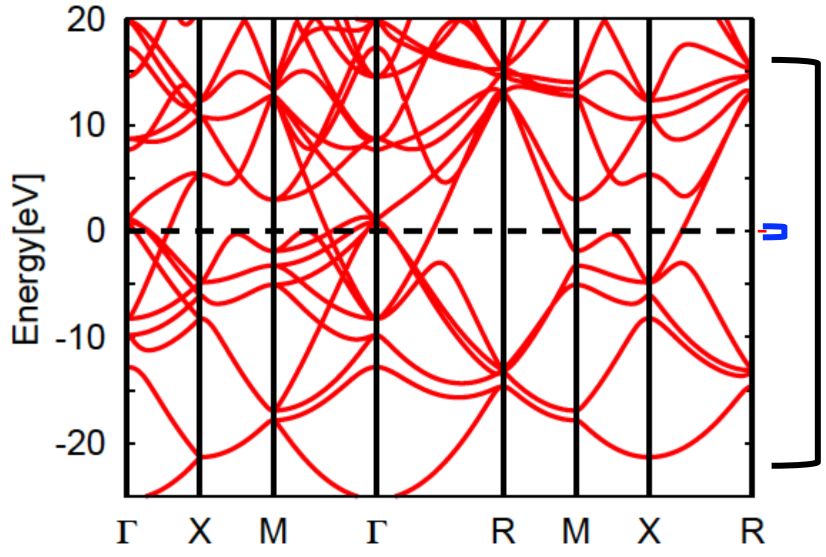
$$\mathbf{G}^{-1} = \mathbf{G}_0^{-1} - \mathbf{\Sigma}$$

Approximate self energy



Phonon mediated pairing

El-El Coulomb repulsion



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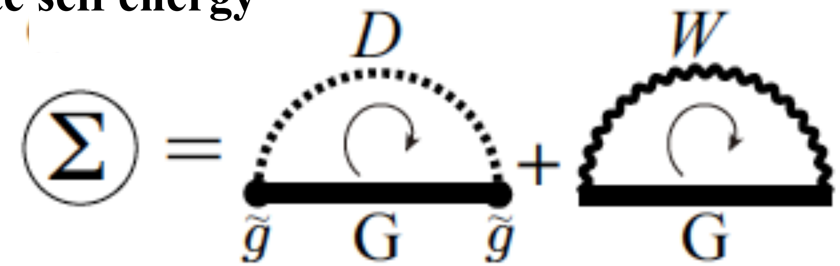
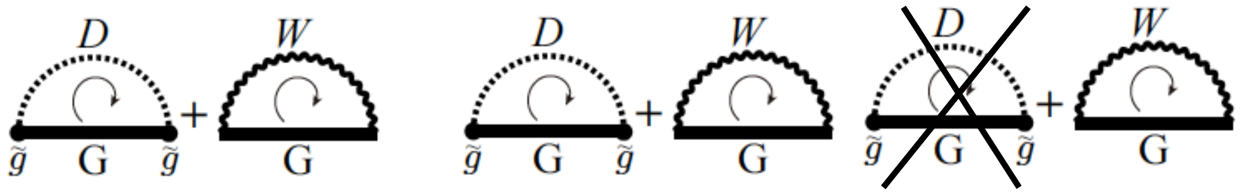
Approximate self energy



Decomposition to Pauli matrices

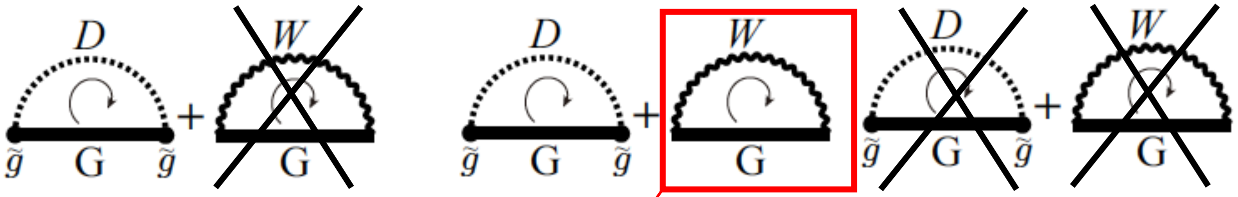
$$\mathbf{\Sigma}_{n\mathbf{k}}(i\omega_j) \equiv i\omega_j(1 - Z_{n\mathbf{k}}(i\omega_j))\sigma_0 + \phi_{n\mathbf{k}}(i\omega_j)\sigma_1 + \chi_{n\mathbf{k}}(i\omega_j)\sigma_3$$



Physically small (Schrieffer, 1964)

1, Static (instantaneous) screening

$$\Sigma_{n\mathbf{k}}(i\omega_j) \equiv \underline{i\omega_j(1 - Z_{n\mathbf{k}}(i\omega_j))\sigma_0} + \underline{\phi_{n\mathbf{k}}(i\omega_j)\sigma_1} + \underline{\chi_{n\mathbf{k}}(i\omega_j)\sigma_3}$$

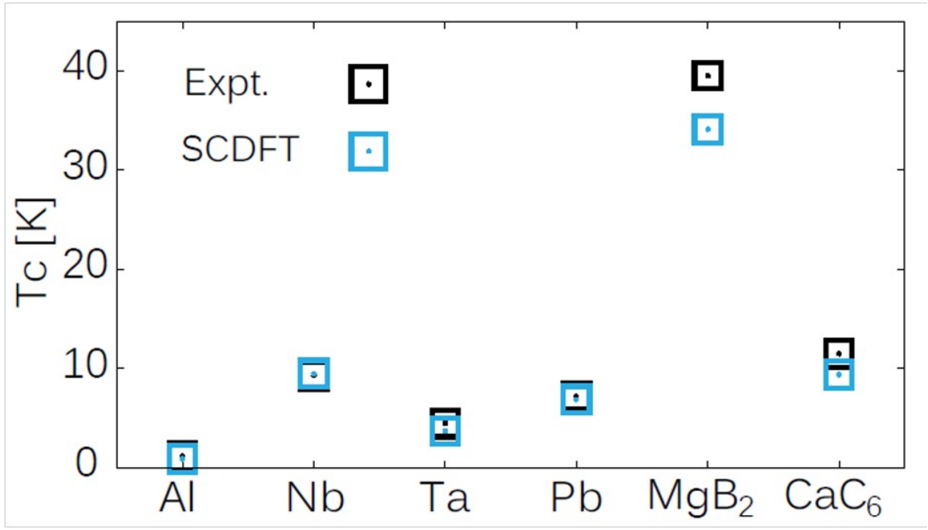


$$W(\omega) \simeq W(\omega = 0)$$

Corresponding static SCDFT

Luders, Marques, Gross et al., 2005

Sanna Pellegrini, Gross, 2020



Direct solution

Sano, Koretsune, Tadano, RA, Arita, 2016

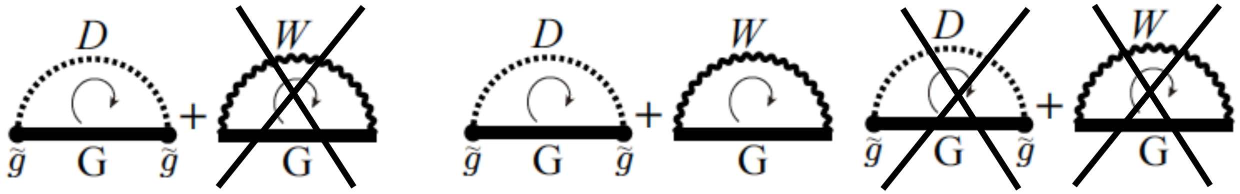
Sanna, Gross et al., 2018

Wang, Arita et al., 2020

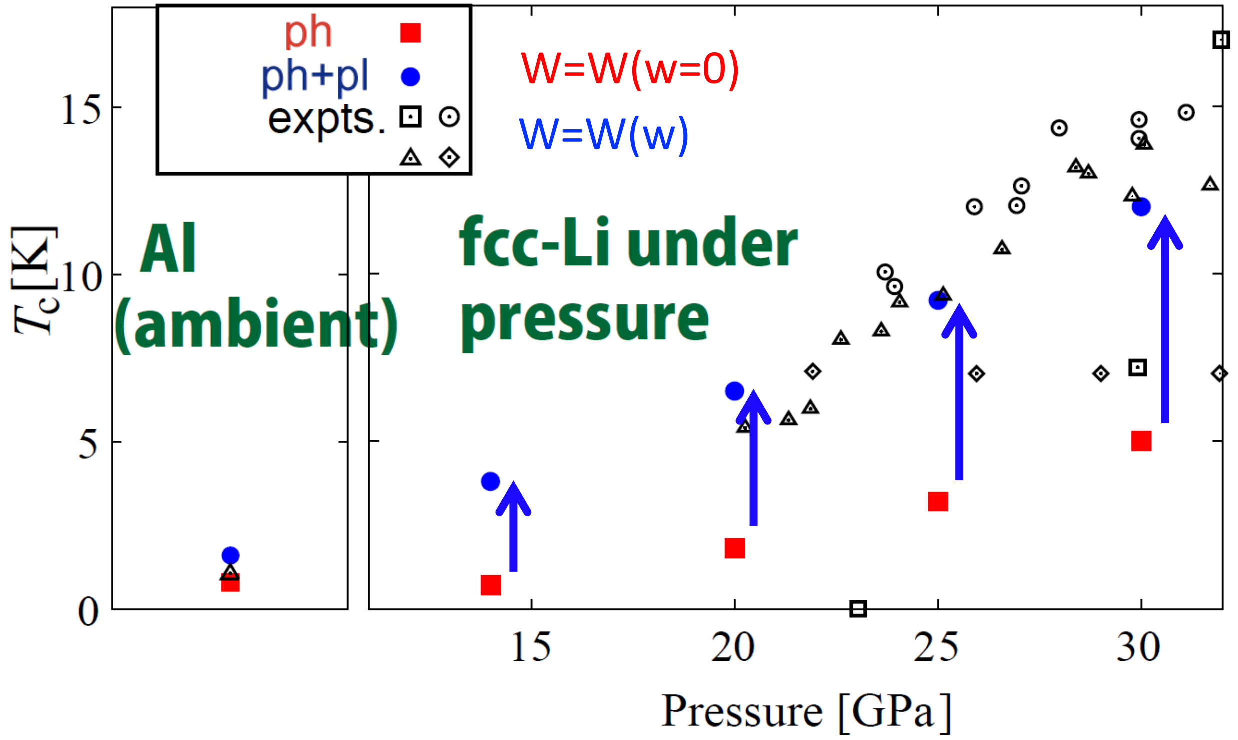
2, +Plasmon assisted superconductivity

RA and R. Arita, Phys. Rev. Lett. 111, 057006 (2013); J. Phys. Soc. Jpn. 83, 061016 (2014); Y. Takada, J. Phys. Soc. Jpn. **45**, 786 (1978)

$$\Sigma_{n\mathbf{k}}(i\omega_j) \equiv \underline{i\omega_j(1 - Z_{n\mathbf{k}}(i\omega_j))\sigma_0} + \underline{\phi_{n\mathbf{k}}(i\omega_j)}\sigma_1 + \underline{\chi_{n\mathbf{k}}(i\omega_j)}\sigma_3$$



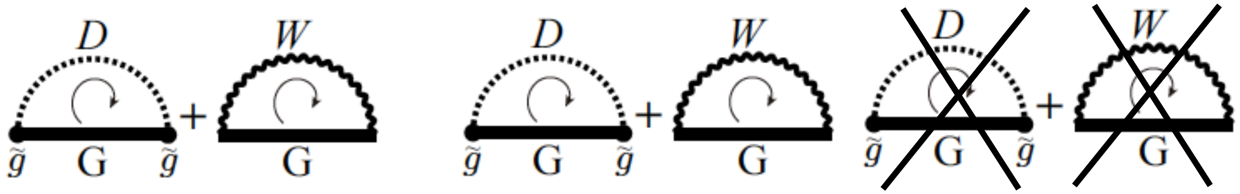
Superconducting DFT result



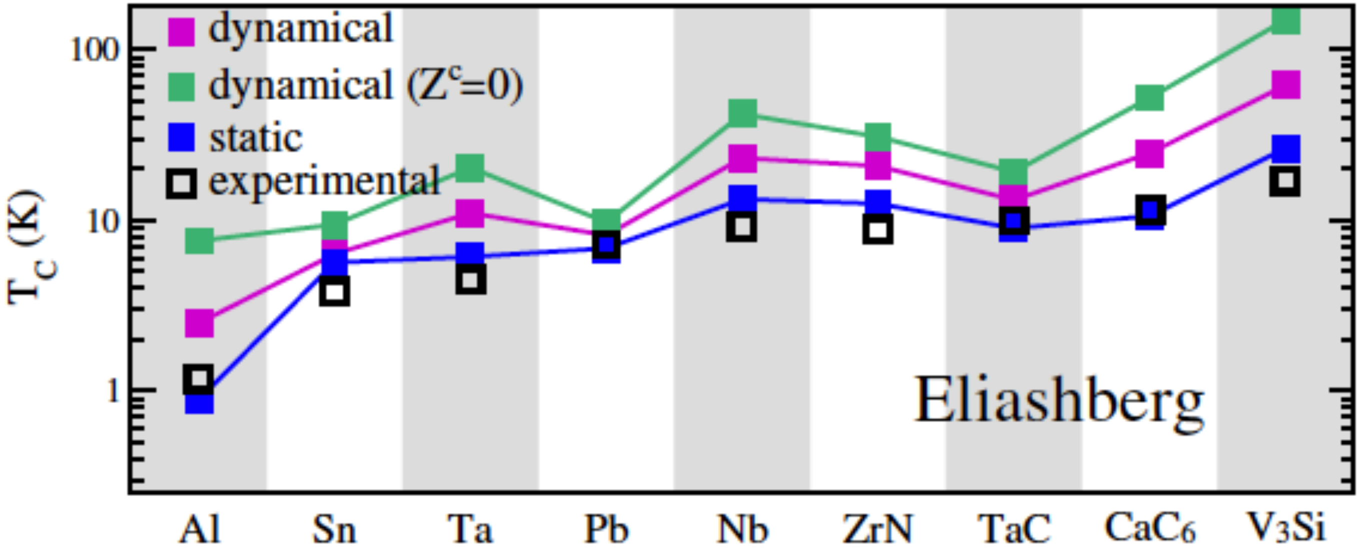
3, +Plasmonic self energy (Z term)

Davydov, Sanna, Gross et al., 2020

$$\boldsymbol{\Sigma}_{n\mathbf{k}}(i\omega_j) \equiv \underline{i\omega_j(1 - Z_{n\mathbf{k}}(i\omega_j))}\sigma_0 + \underline{\phi_{n\mathbf{k}}(i\omega_j)}\sigma_1 + \underline{\chi_{n\mathbf{k}}(i\omega_j)}\sigma_3$$



A. Davydov *et al.*, Phys. Rev. B **102**, 214508 (2020)

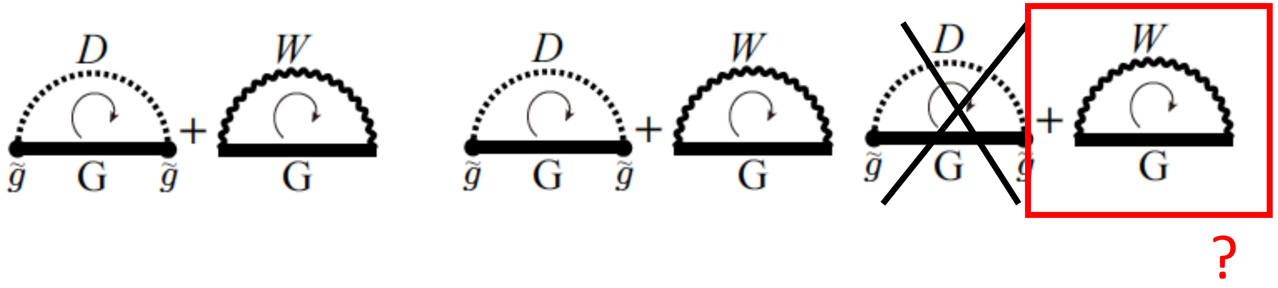


The Tc enhancement is cancelled partially.

3, +Plasmonic self energy (Z term)

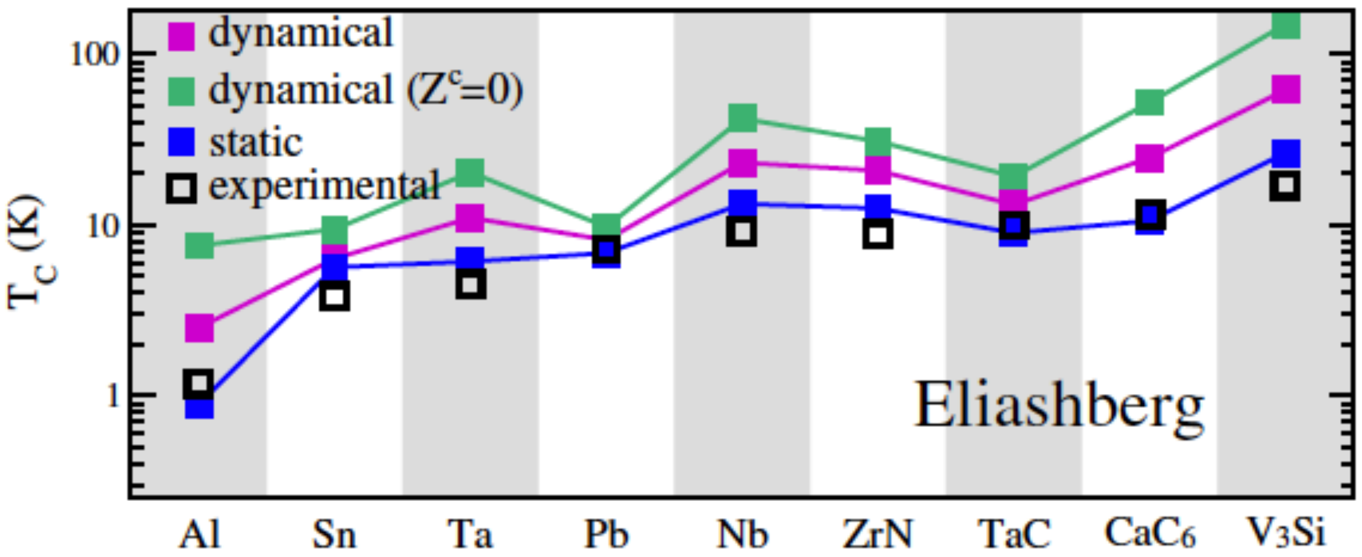
Davydov, Sanna, Gross et al., 2020

$$\Sigma_{n\mathbf{k}}(i\omega_j) \equiv i\omega_j(1 - Z_{n\mathbf{k}}(i\omega_j))\sigma_0 + \phi_{n\mathbf{k}}(i\omega_j)\sigma_1 + \chi_{n\mathbf{k}}(i\omega_j)\sigma_3$$



Numerically demanding

A. Davydov *et al*., Phys. Rev. B **102**, 214508 (2020)



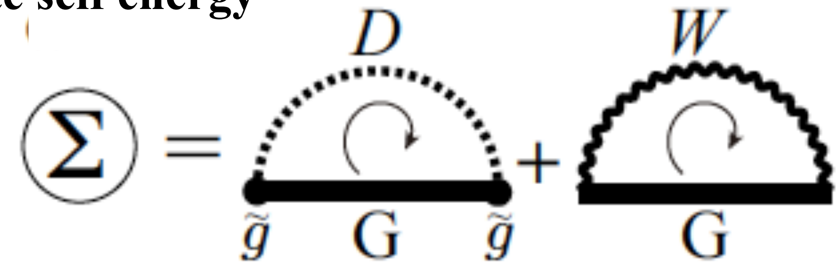
Issue summary

ME theory

Dyson equation

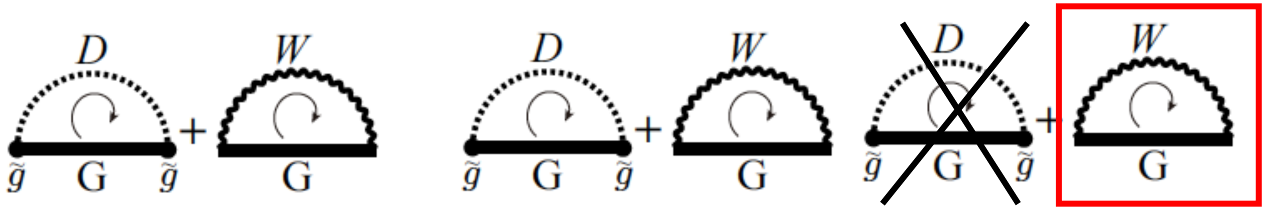
$$\mathbf{G}^{-1} = \mathbf{G}_0^{-1} - \mathbf{\Sigma}$$

Approximate self energy



No one solved this equation with first-principles Kohn-Sham states.

$$\Sigma_{n\mathbf{k}}(i\omega_j) \equiv \underline{i\omega_j(1 - Z_{n\mathbf{k}}(i\omega_j))}\sigma_0 + \underline{\phi_{n\mathbf{k}}(i\omega_j)}\sigma_1 + \underline{\chi_{n\mathbf{k}}(i\omega_j)}\sigma_3$$



The chi-W term is extremely demanding.

So is chi-W relevant?

Let's see a very simple system

RA, Phys. Rev. B **105**, 104510 (2022)

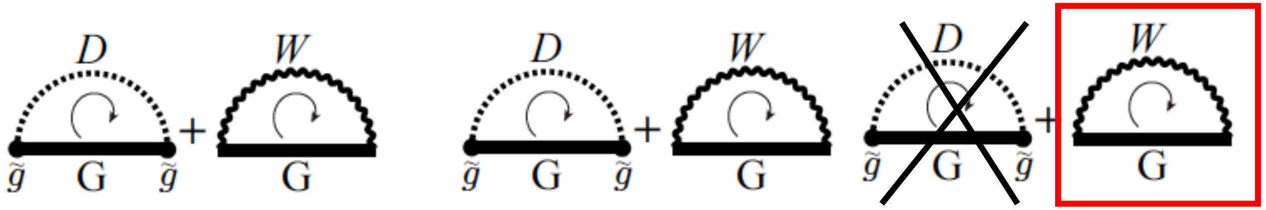
Uniform electron gas (UEG)

-Ions distribute as a constant medium
-Electron wavefunctions are all plane waves
-Good approximation for nearly uniform metals

What I have done:

Solved the Eliashberg equations for UEG with much reduced computational cost, to see the limiting behavior of the chi-term.

$$\mathbf{\Sigma}_{n\mathbf{k}}(i\omega_j) \equiv \underline{i\omega_j(1 - Z_{n\mathbf{k}}(i\omega_j))}\sigma_0 + \underline{\phi_{n\mathbf{k}}(i\omega_j)}\sigma_1 + \underline{\chi_{n\mathbf{k}}(i\omega_j)}\sigma_3$$



Model system

RA, Phys. Rev. B **105**, 104510 (2022)

Electron: Uniform gas

$$H_{\mathrm{el}} = \sum_{\mathbf{k}\sigma} \left(\frac{k^2}{2m} - \mu \right) c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} + \sum_{\mathbf{k}_1 \mathbf{k}_2 \sigma_1 \sigma_2} V(\mathbf{q}) c^{\dagger}_{\mathbf{k}_1+\mathbf{q}\sigma_1} c^{\dagger}_{\mathbf{k}_2-\mathbf{q}\sigma_2} c_{\mathbf{k}_2\sigma_2} c_{\mathbf{k}_1\sigma_1}$$

Phonon: Einstein model

elph coupling

$$|g^{\nu}_{k,k'}|^2 = \frac{\lambda \omega_{\mathrm{E}}}{2N_{\mathrm{F}}}$$

ph propagator

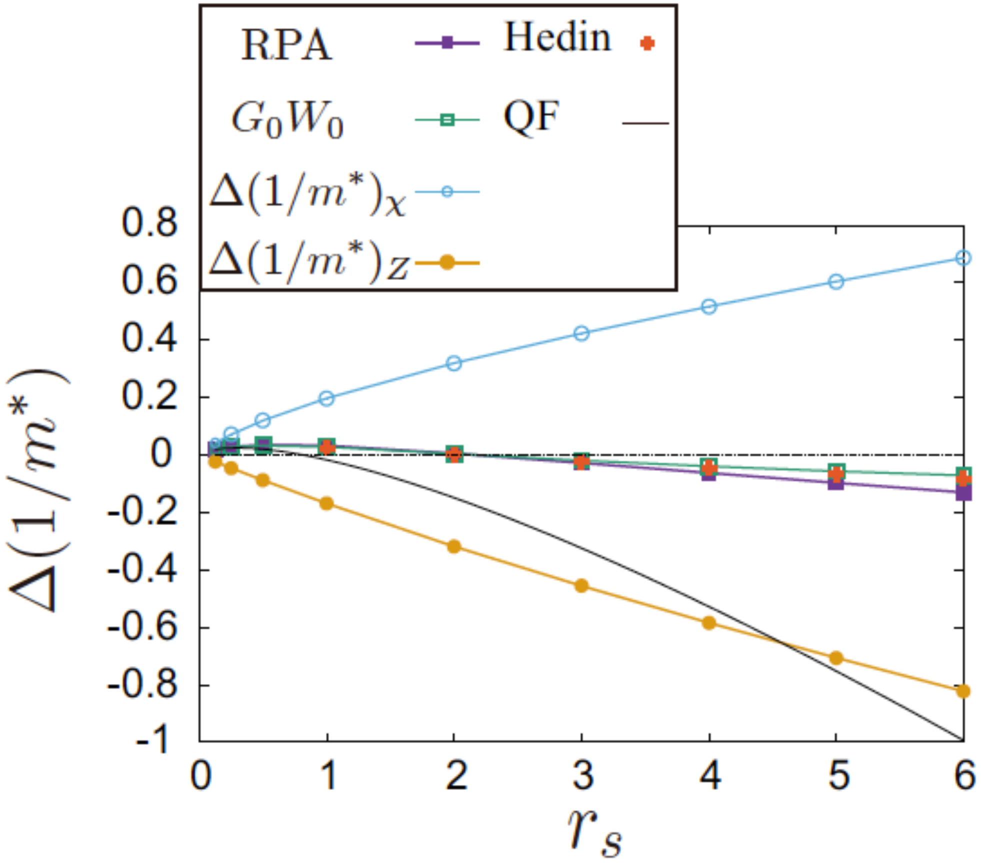
$$D_{\nu}(k-k', i(\omega_j - \omega_{j'})) = -\frac{2\omega_{\mathrm{E}}}{(\omega_j - \omega_{j'})^2 + \omega_{\mathrm{E}}^2}$$

W^{el}: Random phase approximation

$$W^{\mathrm{el}} = \frac{V(k-k')}{\varepsilon^{\mathrm{RPA}}(k-k', i(\omega_j - \omega_{j'}))}$$

Result: Inverse effective mass

RA, Phys. Rev. B **105**, 104510 (2022)



Z and χ corrections cancel with each other.

Well known in GW community

L. Hedin, Phys. Rev. **139**, A796 (1965); . . .

Result: Pairing strength (Eliashberg's largest eigenvalue)

RA, Phys. Rev. B **105**, 104510 (2022)

$$\phi = \phi^{\mathrm{ph}}[Z, \chi, \phi] + \phi^{\mathrm{el}}[Z, \chi, \phi]$$
$$\simeq \mathbf{\Lambda}\phi$$

Parameter setting

$$\lambda = 0.5$$
$$\omega_{\mathrm{E}} = 0.01\omega_{\mathrm{plasmon}}(r_s)$$
$$T = 0.05\omega_{\mathrm{E}}(r_s)$$

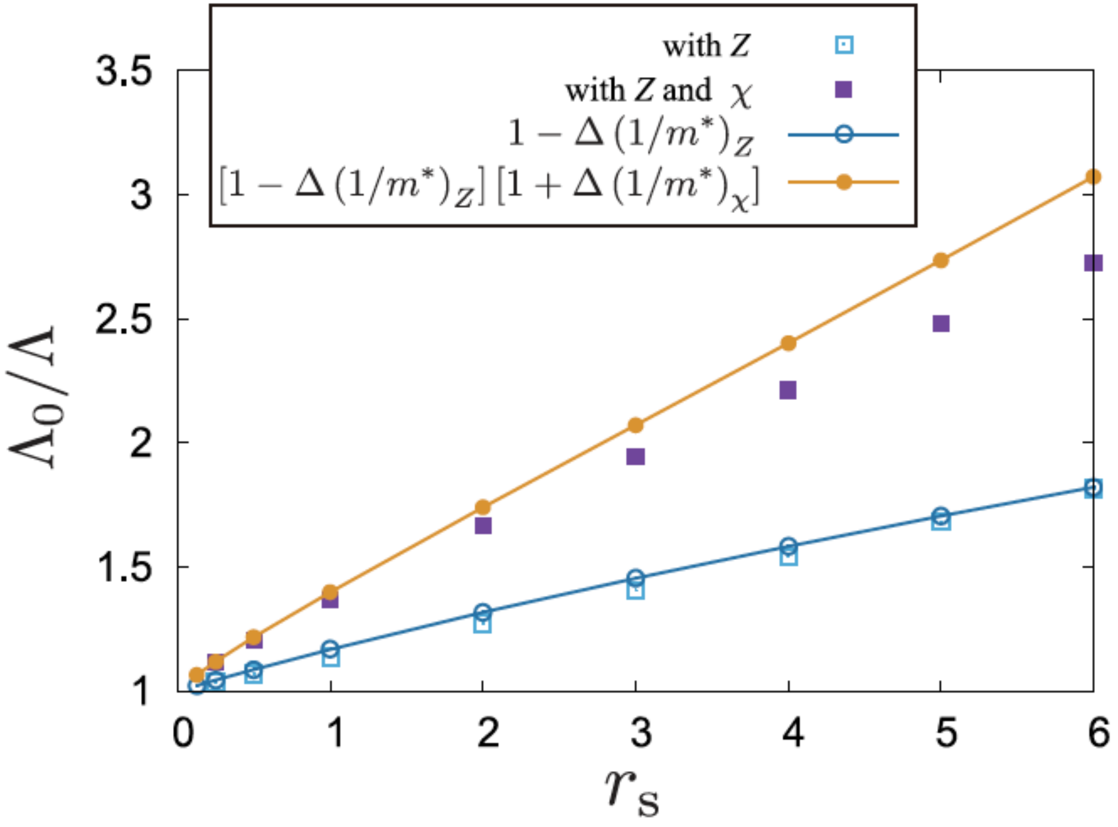
Result: Pairing strength (Eliashberg's largest eigenvalue)

RA, Phys. Rev. B **105**, 104510 (2022)

$$\phi = \phi^{\mathrm{ph}}[Z,\chi,\phi] + \phi^{\mathrm{el}}[Z,\chi,\phi]$$
$$\simeq \mathbf{\Lambda}\phi$$

Parameter setting

$$\lambda = 0.5$$
$$\omega_{\mathrm{E}} = 0.01\omega_{\mathrm{plasmon}}(r_s)$$
$$T = 0.05\omega_{\mathrm{E}}(r_s)$$

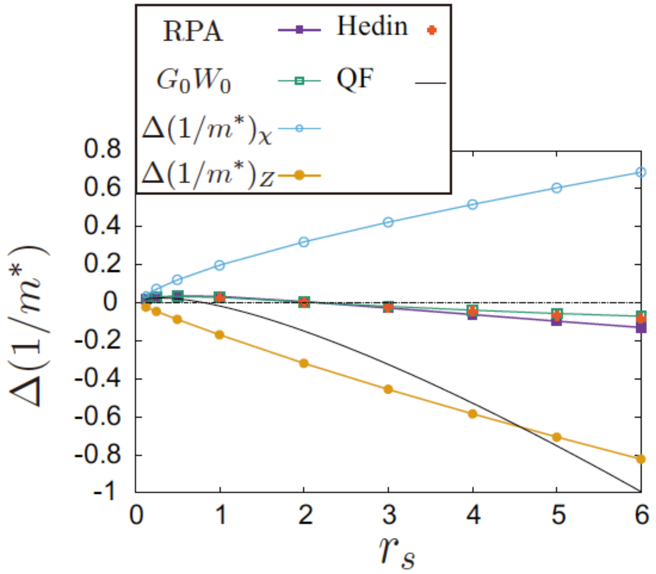


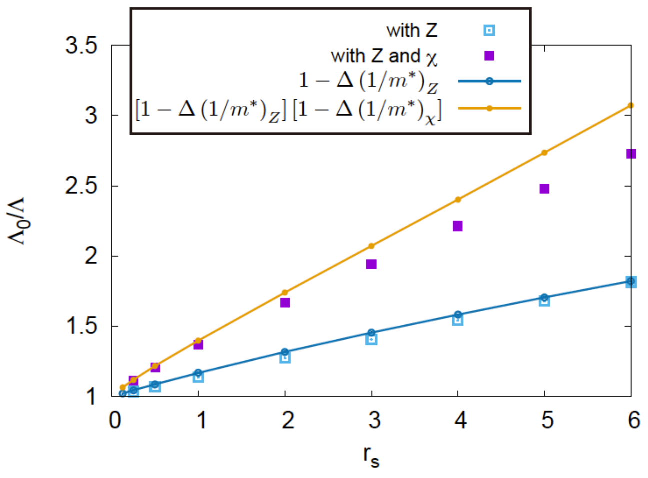
Z and χ corrections both suppress the pairing.

$Tc \sim \exp(-\Lambda)$

Results summary

RA, Phys. Rev. B **105**, 104510 (2022)





Mass

$$m^* = \frac{1-\Delta_Z}{1+\Delta_\chi}$$

Pairing

$$\frac{\Lambda_0}{\Lambda} = (1-\Delta_Z)\,(1+\Delta_\chi)$$

$$\Delta_Z = 1 - Z(k{=}k_\mathrm{F}, i\omega{=}0)$$

$$\Delta_\chi = \frac{1}{k_\mathrm{F}} \left.\frac{\partial \chi(k, i\omega=0)}{\partial k}\right|_{k=k_\mathrm{F}}$$

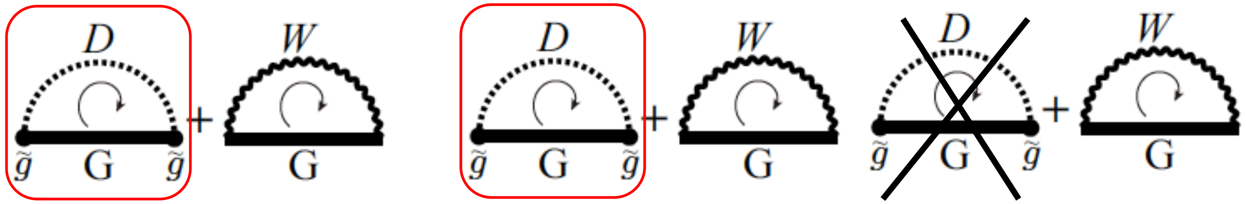
Both are ~O(0.1)

Departure of mass and pairing

RA, Phys. Rev. B **105**, 104510 (2022)

In the phononic case, they depend on the same factor

$$\boldsymbol{\Sigma}_{n\mathbf{k}}(i\omega_j) \equiv i\omega_j(1 - Z_{n\mathbf{k}}(i\omega_j))\sigma_0 + \phi_{n\mathbf{k}}(i\omega_j)\sigma_1 + \chi_{n\mathbf{k}}(i\omega_j)\sigma_3$$



Mass

$$m^* \simeq 1 + \lambda$$

termed "mass enhancement"

Pairing

$$\frac{\Lambda_0}{\Lambda} = 1 + \lambda$$

Cf. McMillan Tc formula

McMillan, Phys. Rev. 167, 331 (1968);
Allen and Dynes, Phys. Rev. B **12**, 905 (1975)

$$T_c \sim \exp\left[-\frac{1+\lambda}{\lambda - \mu^*}\right]$$

Departure of mass and pairing

RA, Phys. Rev. B **105**, 104510 (2022)

Phononic part (chi=0)

Mass

$$m^* \simeq 1 + \lambda$$

Pairing

$$\frac{\Lambda_0}{\Lambda} = 1 + \lambda$$

Coulomb part

Mass

$$m^* = \frac{1 - \Delta_Z}{1 + \Delta_\chi}$$

Pairing

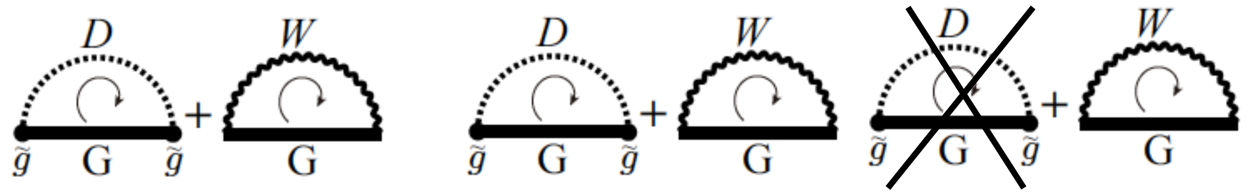
$$\frac{\Lambda_0}{\Lambda} = (1 - \Delta_Z)\,(1 + \Delta_\chi)$$

Pairing renormalization is actually different from mass renorm.
Coulomb pairing suppression is not detected as mass renormalization.

Results summary

RA, Phys. Rev. B **105**, 104510 (2022)

$$\Sigma_{n\mathbf{k}}(i\omega_j) \equiv \underline{i\omega_j(1 - Z_{n\mathbf{k}}(i\omega_j))}\sigma_0 + \underline{\phi_{n\mathbf{k}}(i\omega_j)}\sigma_1 + \underline{\chi_{n\mathbf{k}}(i\omega_j)}\sigma_3$$



	Static scr.	+phi_W ("plasmon")	+Z_W ("pl. self energy")	+chi_W ("pl. self energy")
Effective mass	…	…	+	-
Pairing strength	…	+	-	-

-For accurate Tc both Z_W and chi_W are necessary.
-Pairing weakening by Z_W and chi_W are invisible from mass.
-Static screening approximation is accidentally accurate?

Conclusions

RA and R. Arita, Phys. Rev. Lett. **111**, 057006 (2013);
K .Tsutsumi, Y. Hizume, M. Kawamura, RA, and S. Tsuneyuki, Phys. Rev. B **102**, 214515 (2020);
RA, Phys. Rev. B **105**, 104510 (2022).

Plasmon and spin fluctuation effects are incorporated in SCDFT.
-they ubiquitously enhance/suppress Tc of phonon superconductors

Eliashberg analysis shows that the Coulomb self energy effects cannot be ignored.
-it hardly affects effective mass, but significantly reduces Tc
-plasmonic pairing may be largely canceled?

Reviews on other topics

DFT fundamentals

Y. Nomura and RA, arXiv:2210.07647

Machine-learning DFT

R. Nagai and RA, arXiv:2206.15370