Low Fermi energy and superconductivity in C_{60} compounds

E. Cappelluti[1], C. Grimaldi[2], L. Pietronero[1] and S. Strässler[2]

[1] Dipartimento di Fisica, Università "La Sapienza", P.le Aldo Moro 2, 00185 Roma, Italia

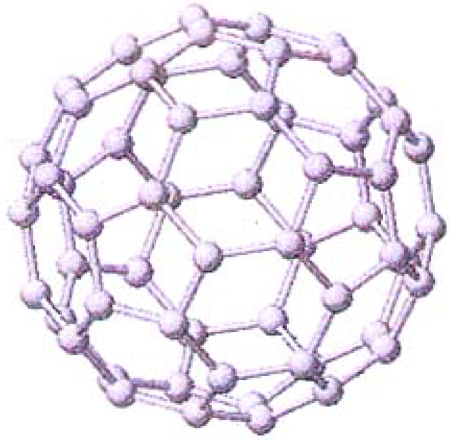
[2] École Polytechnique Fédérale, Départment de microtechnique IPM, CH-1015 Lausanne, Switzerland

Outline

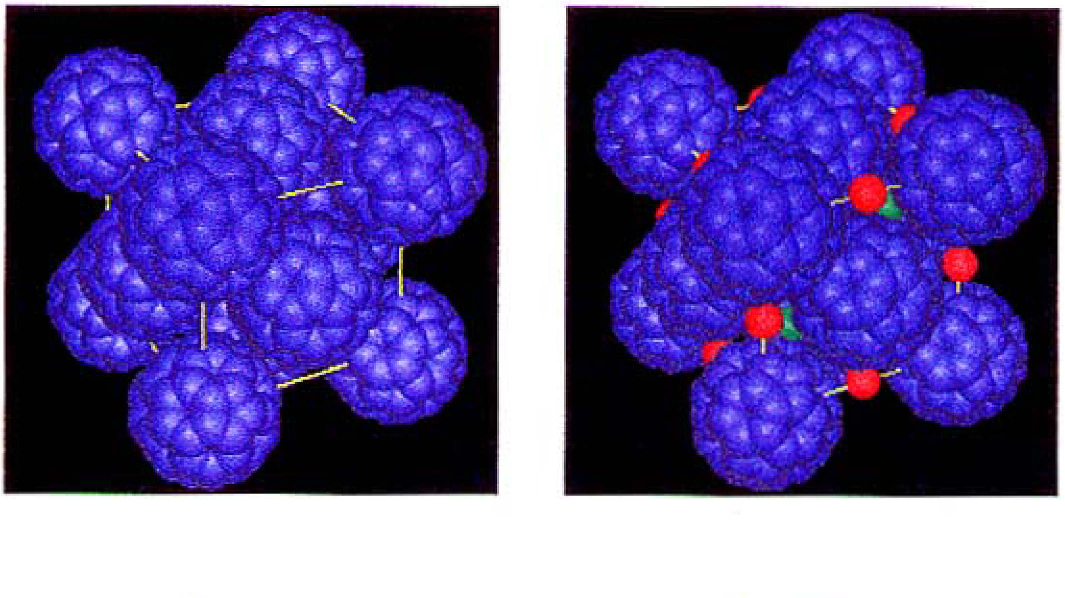
- High-T_c superconductivity in C_{60}
- Fullerenes: low carrier density; small Fermi energy
- Electron-phonon: breakdown of adiabatic hypothesis
- Nonadiabatic superconductivity
- Nonadiabatic Fermi liquid

C_{60} Fullerene

Molecular structure

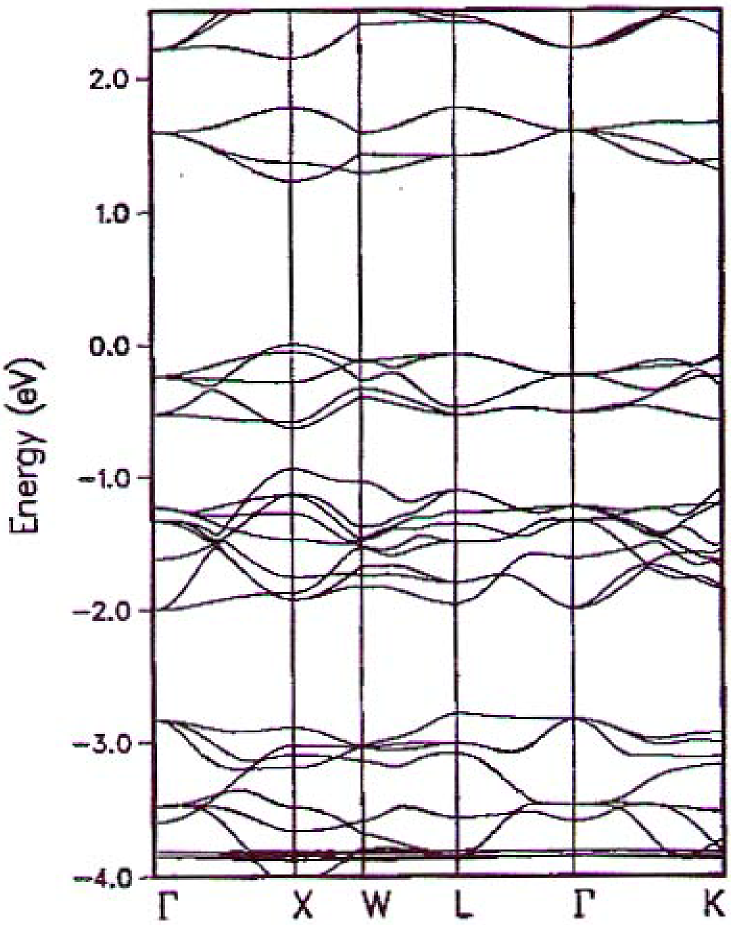


Crystal structure



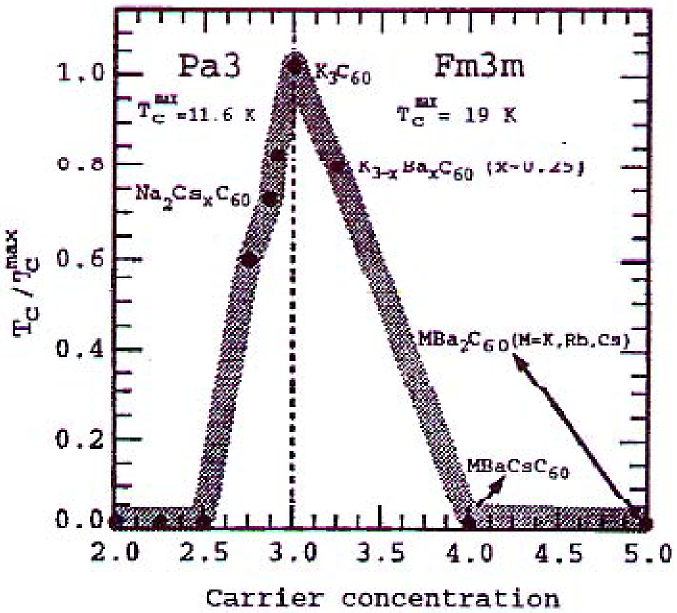
C_{60} A_3C_{60}

Electronic band structure

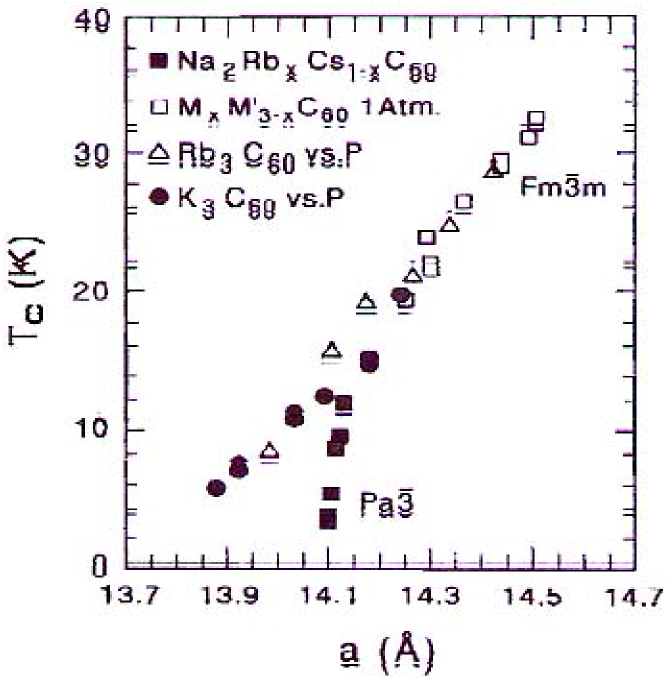


[S.C. Erwin, in Buckminsterfullerenes, eds. W.E. Billups and M.A. Ciufolini (1993)]

Phase diagram

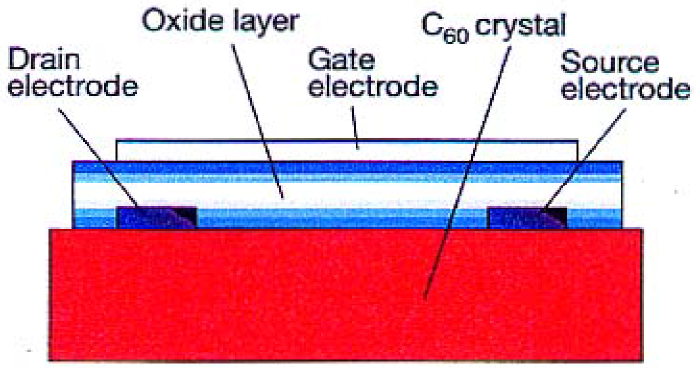


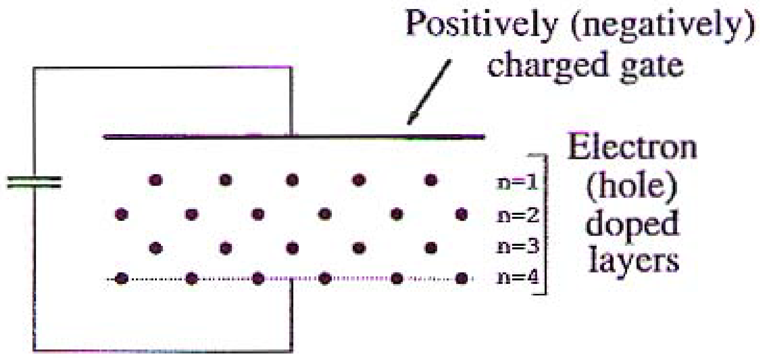
[Yildirim et al. Phys. Rev. Lett. 1996]



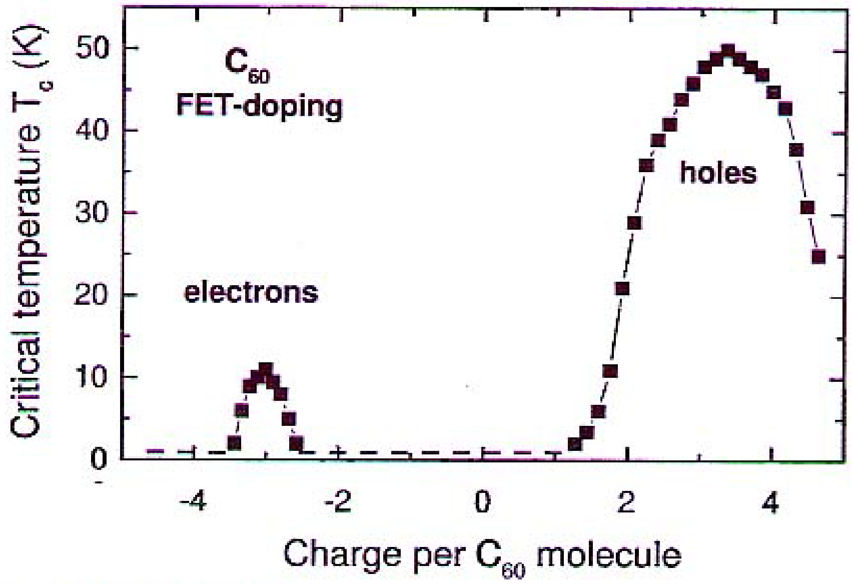
[Yildirim et al. Solid State Commun. 1995]

Field Effect Transistor doping

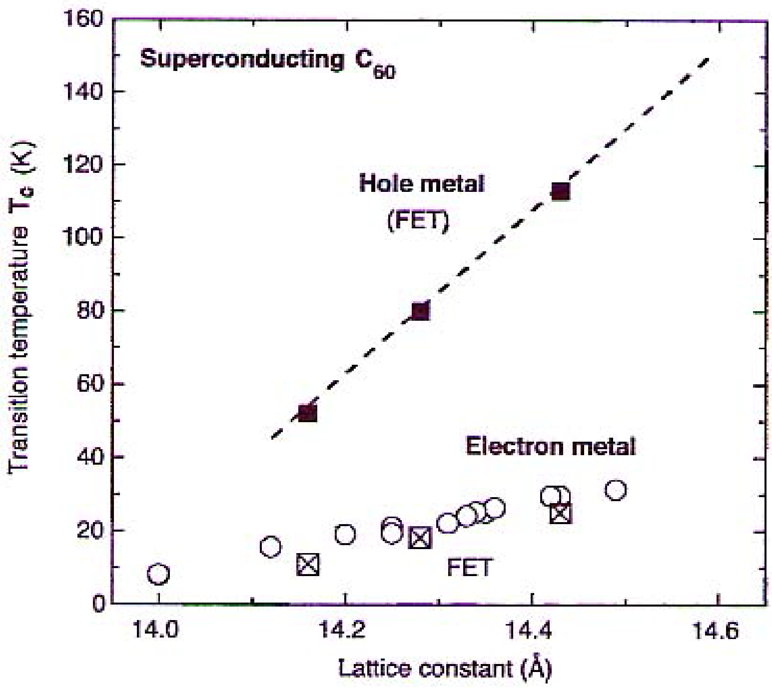




FET results



[Schön et al. Nature 2000]



[Schön et al. Science 2001]

Superconductivity in fullerenes

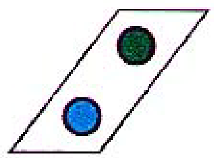
- $T_c^{max} = 117$ K! (so far ...)
  [BCS empirical limit $T_c \lesssim 20 - 25$ K]
- Strong electron-electron repulsion
  [Cooper pairing disfavoured]
- Small density of carriers
  [poor metal]

$\Rightarrow$ Superconductivity in fullerides
in spite of
these negative features ????

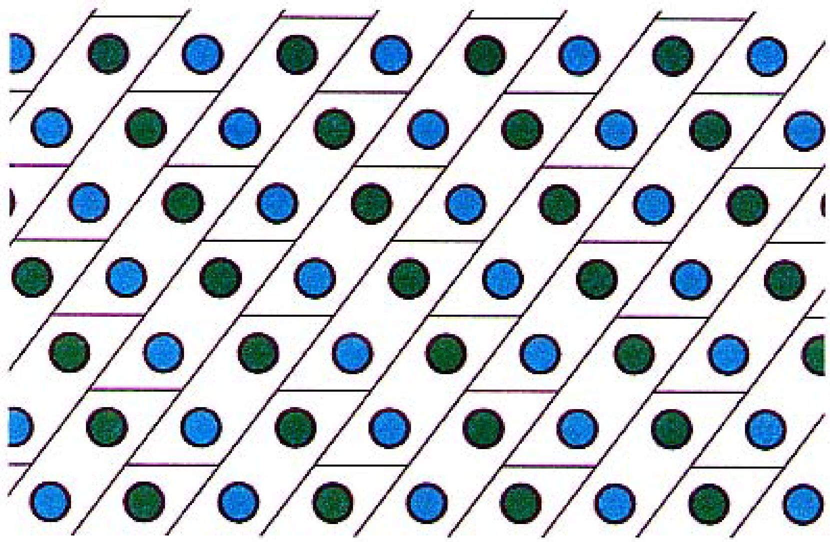
$\Rightarrow$ Unconventional superconductivity:
new context in which
these negative features favour pairing

Common metals

(a) Few atoms in unit cell



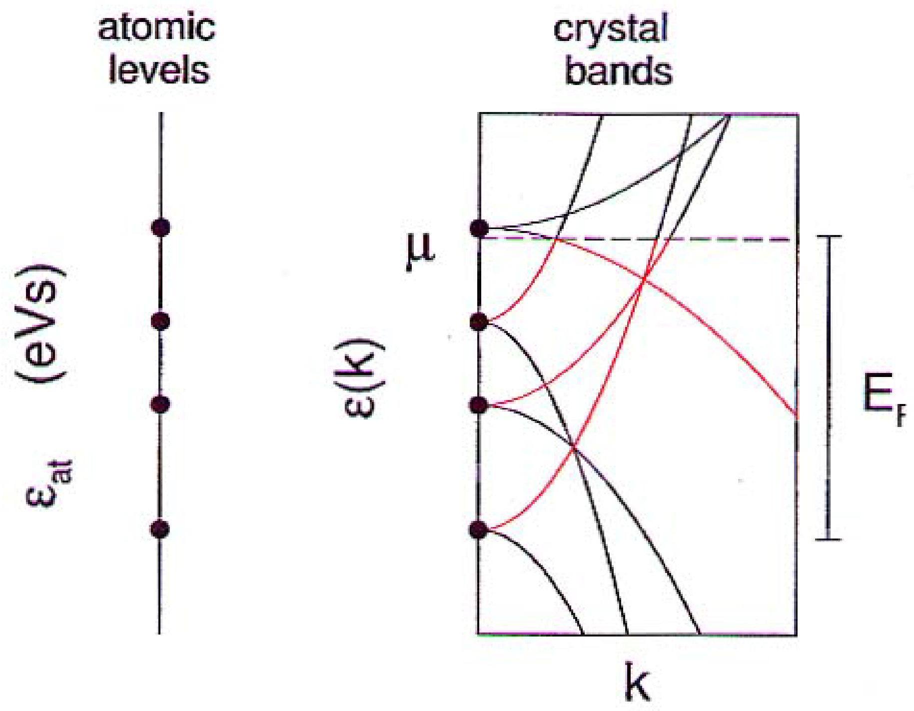
(b) Close packed unit cells



Electronic states

(a) High energy spaced atomic levels

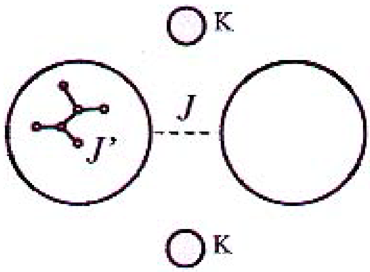
(b) Strong hybridization $\Rightarrow$ large bands



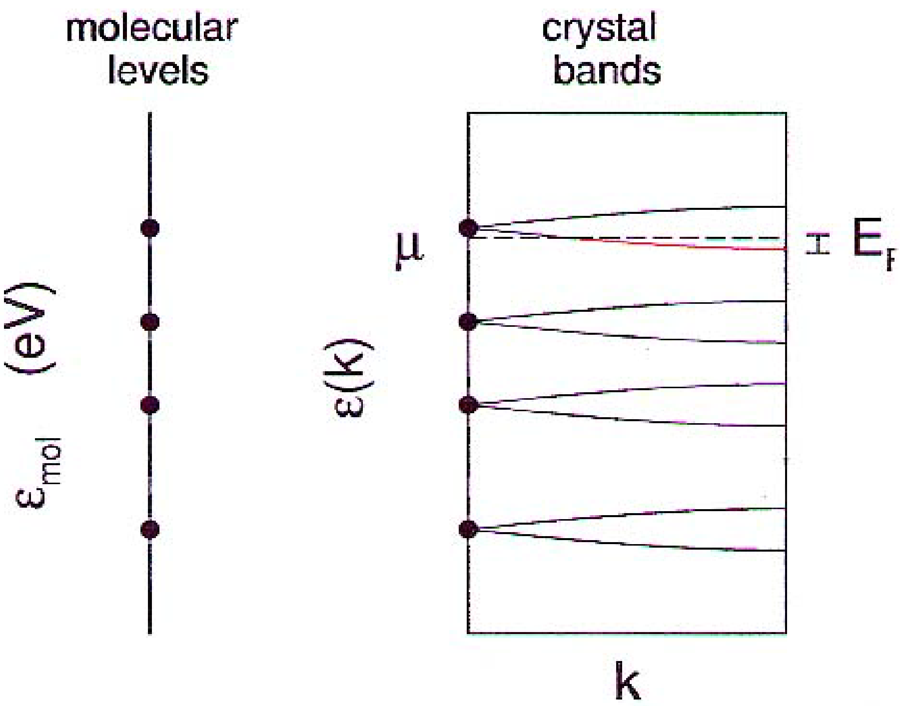
$\Rightarrow$ Large carrier density

$\Rightarrow$ Large Fermi energy E_F

Fullerenes



- Large cluster unit block
- Weak hybridization

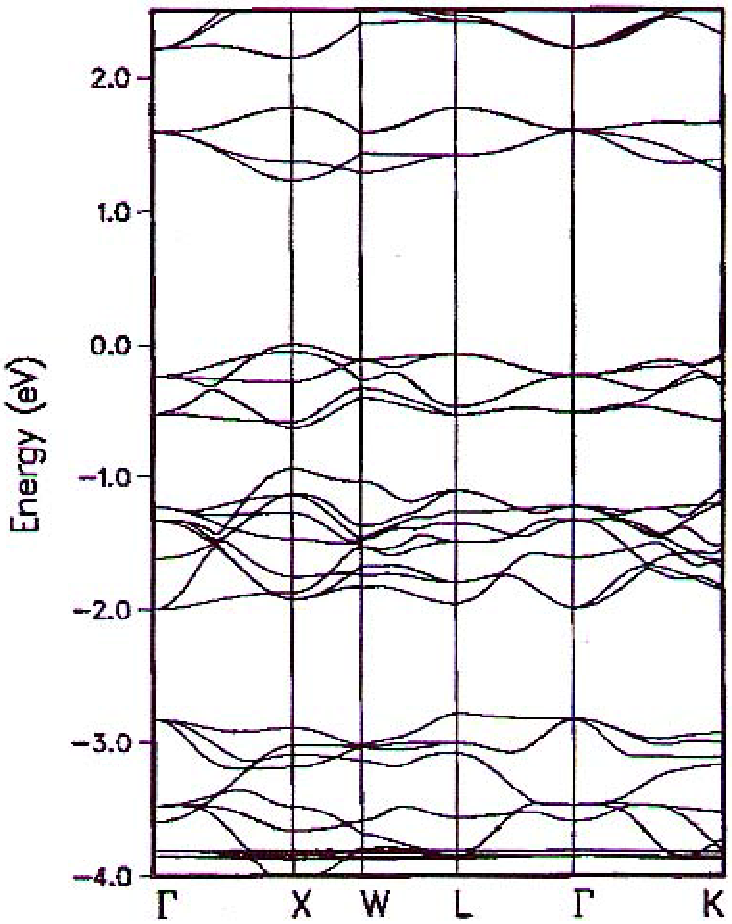


⇒ Small carrier density

⇒ Narrow bands

⇒ Small Fermi energy E_F

Band structure C_{60}



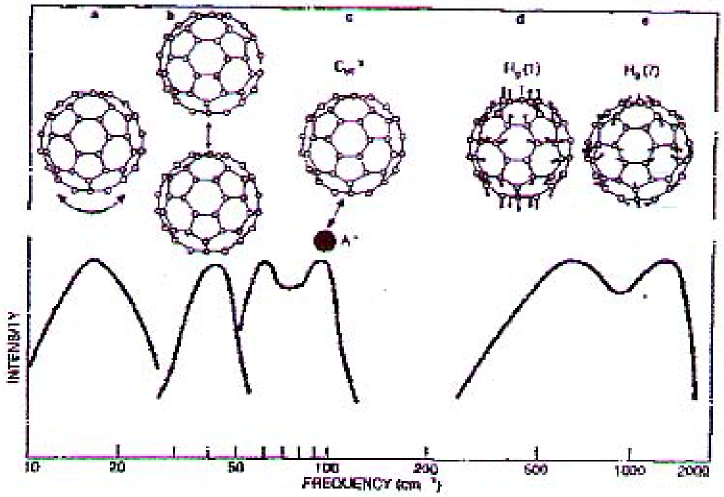
[S.C. Erwin, in Buckminsterfullerenes, eds. W.E. Billups and M.A. Ciufolini (1993)]

Narrow bandwidths $W \sim 0.5 - 0.6$ eV

Small Fermi energies $E_F \sim 0.2 - 0.3$ eV

Nonadiabaticity

- Phonon frequencies $\omega_{\text{ph}} \sim 80 - 200$ meV



A.F. Hebard et al., Phys. Today (1992)

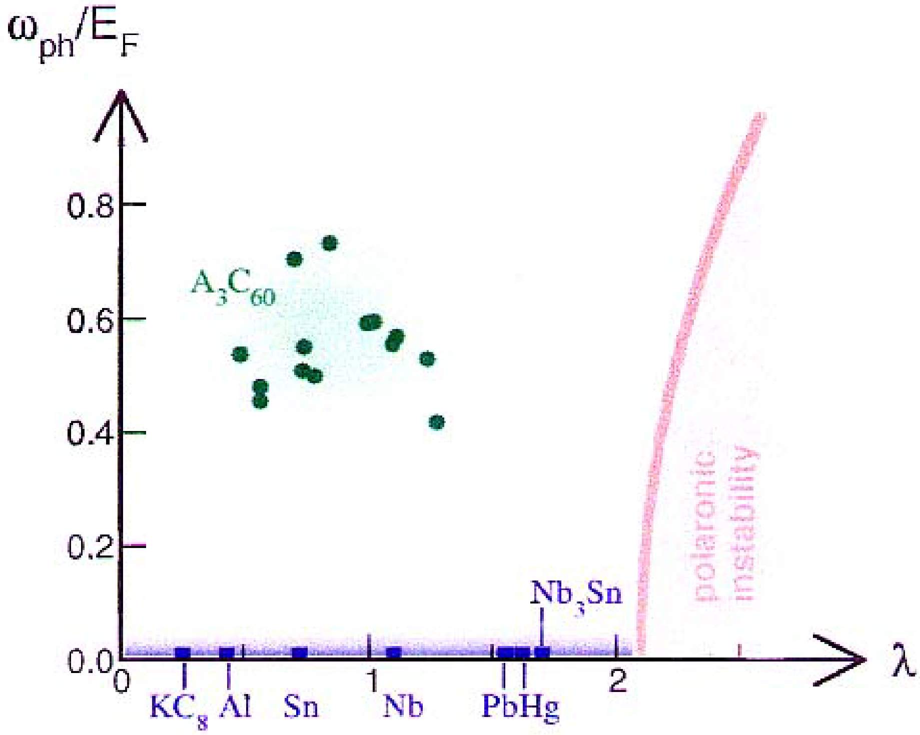
- Fermi energies $E_{\text{F}} \sim 0.25 - 0.35$ eV

⇨ $$\omega_{\text{ph}}/E_{\text{F}} \simeq 0.4 - 0.8$$

The adiabatic principle (Born-Oppenheimer) breaks down

Nonadiabatic metal

Adiabatic phase diagram



$N(E_F) \simeq 8$ states/eV C_{60} $\qquad E_F \simeq 0.25$ eV

Data collected by LDA calculations:

Schluter et al., Journ. Phys. Chem. Sol., 1992

Schluter et al., Phys. Rev. Lett., 1992

Varma et al., Science, 1991

Faulhaber et al., Phys. Rev. B, 1993

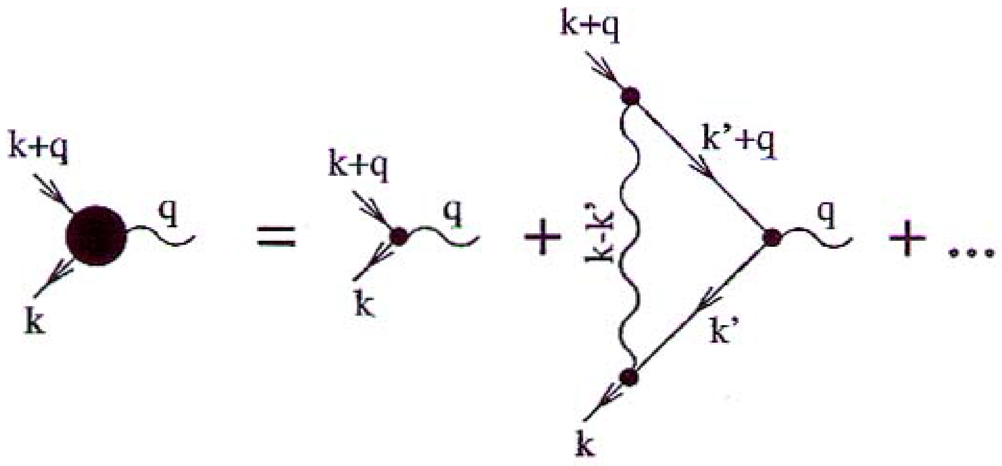
Antropov et al., Phys. Rev. B, 1993

Breda et al., Chem. Phys. Lett., 1998

Onida et al., Europhys. Lett., 1992

Adiabatic hypothesis

- Vertex function



$$\Lambda = 1 + P + \ldots$$

Migdal's theorem

$$P \sim \lambda \frac{\omega_{\text{ph}}}{E_{\text{F}}}$$

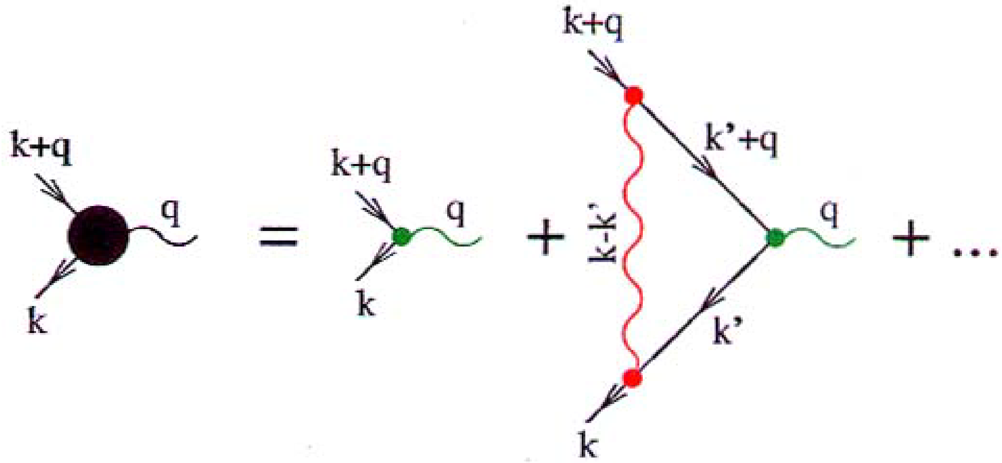
- Conventional ME superconductors ($\omega_{\text{ph}} \ll E_{\text{F}}$):

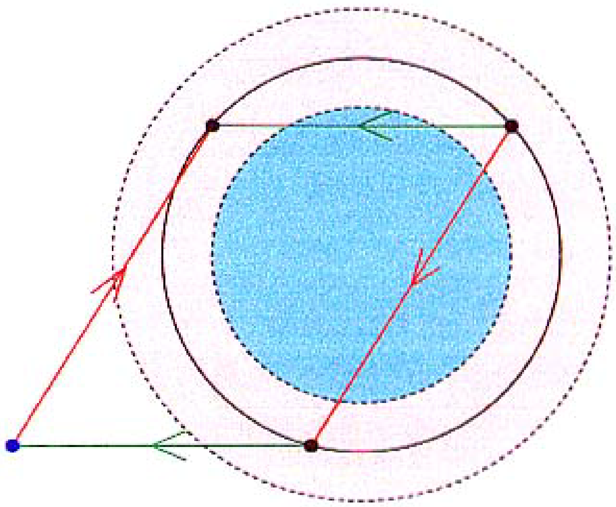
  $P \ll 1 \qquad \Rightarrow \qquad \Lambda = 1$

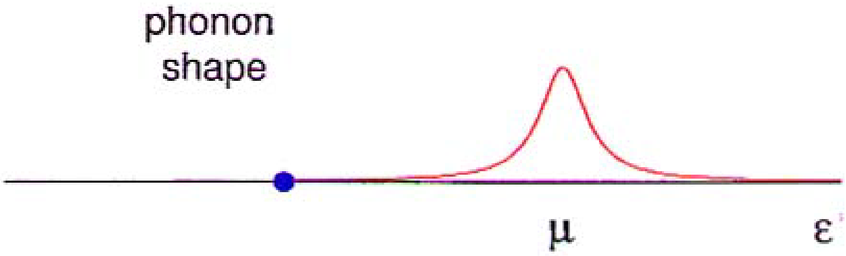
- Fullerenes ($\omega_{\text{ph}} \sim E_{\text{F}}$)

  $P \sim 1 \qquad \Rightarrow \qquad \Lambda = 1 + P$

Vertex interferences



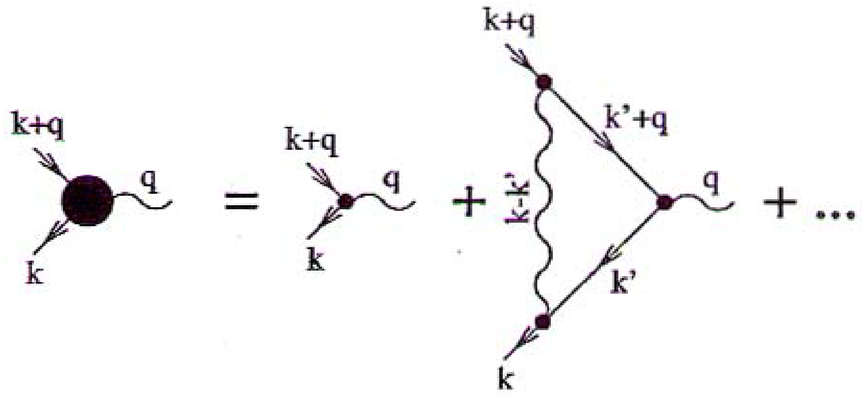




$$D(\omega) = \frac{\omega_{\mathsf{ph}}}{\omega_{\mathsf{ph}}^2 + \omega^2} \quad \Rightarrow \quad D(E_{\mathsf{F}})E_{\mathsf{F}} \sim \frac{\omega_{\mathsf{ph}}}{E_{\mathsf{F}}^2}E_{\mathsf{F}} \sim \frac{\omega_{\mathsf{ph}}}{E_{\mathsf{F}}}$$

Beyond Migdal's theorem

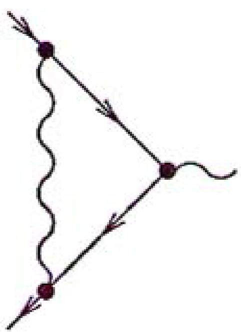
- Vertex function



Expansion parameter: $P = \lambda\omega_{\text{ph}}/E_F$

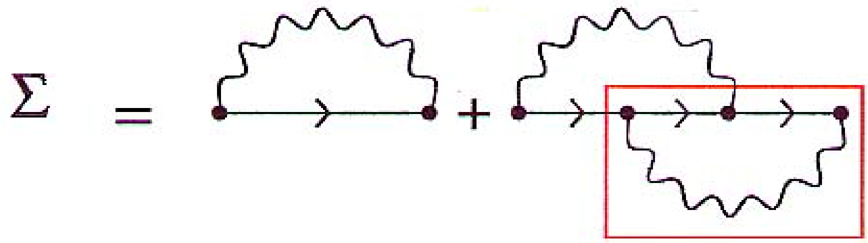
- Migdal-Eliashberg: nonperturbative in λ
- Nonadiabatic theory: nonperturbative in λ, perturbative in $\lambda\omega_{\text{ph}}/E_F$
  $\Rightarrow$ counting of vertex diagrams

First order in vertex

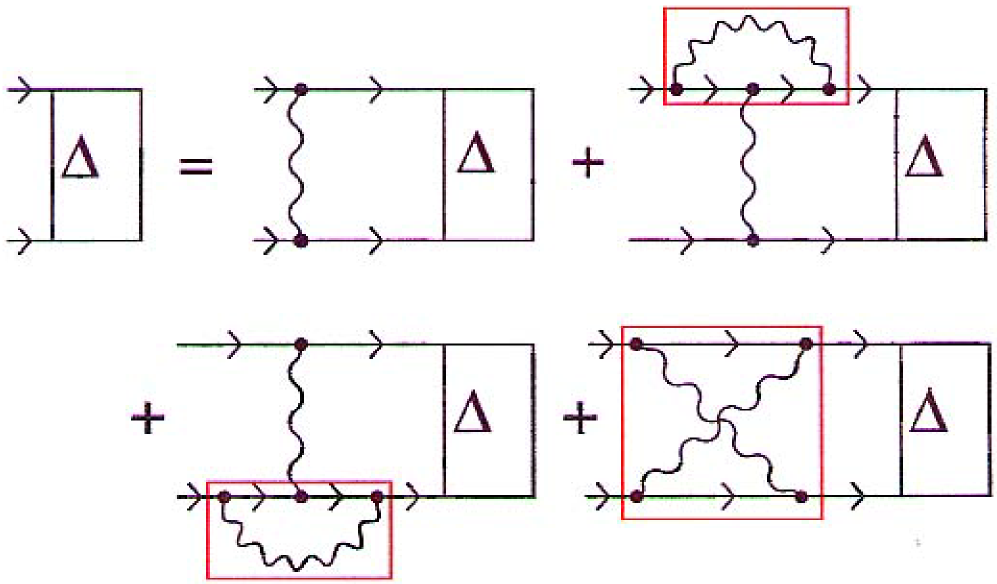


Nonadiabatic channels

- Self-energy



- Superconducting Cooper pairing



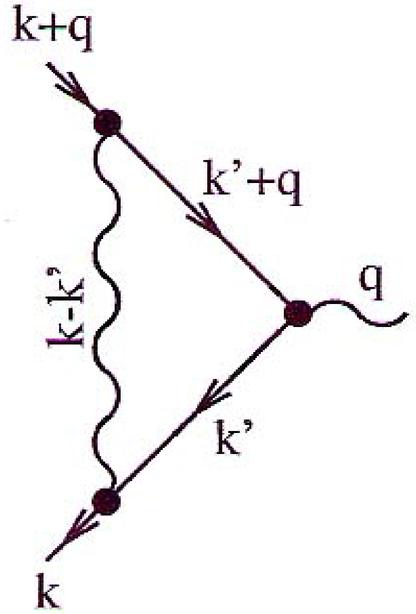
Nonadiabatic metal parameters

- Not a parameter renormalization
  Es.: Electronic mass $m^*(\lambda) \not\longrightarrow m^*(\lambda')$

$$
\begin{aligned}
m^*(\lambda) &\longrightarrow m^*(\lambda, \omega_{\mathrm{ph}}/E_{\mathrm{F}}) \\
t_c(\lambda) &\longrightarrow t_c(\lambda, \omega_{\mathrm{ph}}/E_{\mathrm{F}}) \\
\chi_{\mathrm{P}} &\longrightarrow \chi_{\mathrm{P}}(\lambda, \omega_{\mathrm{ph}}/E_{\mathrm{F}})
\end{aligned}
\qquad \left[t_c = \frac{T_c}{\omega_{\mathrm{ph}}} \right]
$$

- Different nonadiabatic effects
  on different physical quantities

Key element: vertex function

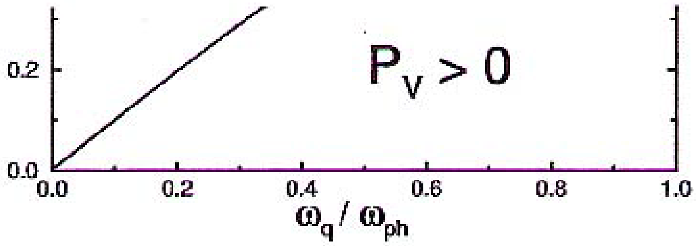


$$P(\mathbf{k}, \mathbf{q}; m, n) = -T \sum_{l} \sum_{\mathbf{q}} |g(\mathbf{k} - \mathbf{k}')|^2 D(m - l)$$
$$\times G(\mathbf{k}' + \mathbf{q}, l + n) G(\mathbf{k}', l).$$

Smooth dependence on the external electronic momenta-frequencies $\mathbf{k}$-ω_m

Complex structure in the exchanged phonon momenta-frequencies $\mathbf{q}$-ω_n

Sign of vertex corrections



- Static limit $\mathbf{q} \to 0,\ \omega = 0$

$$\lim_{\mathbf{q}\to 0, \omega=0} P < 0$$

- Dynamic limit $\omega \to 0,\ \mathbf{q} = 0$

$$\lim_{\omega\to 0, \mathbf{q}=0} P > 0$$

Grabowski and Sham 1984: $\lim_{\omega=0} P < 0$

Electronic correlation

- Metals: long range el-el interaction screened; short-range (on-site)?

Simplest (Hubbard) model

$$H = \sum_{\mathbf{k},\sigma} \epsilon(\mathbf{k}) c^{\dagger}_{\mathbf{k},\sigma} c_{\mathbf{k},\sigma} + U \sum_{i} n_{i,\uparrow} n_{i,\downarrow}$$

- Competition between propagating $[\epsilon(\mathbf{k})]$ and local $[U]$ terms
- Half-filling (one electron per site) for $U \gg E_{\mathsf{F}}$: no propagation (strong correlation)
- At low density $n_i \ll 1$: weak correlation no matter U/E_{F}

Strongly correlated systems

- Textbook example

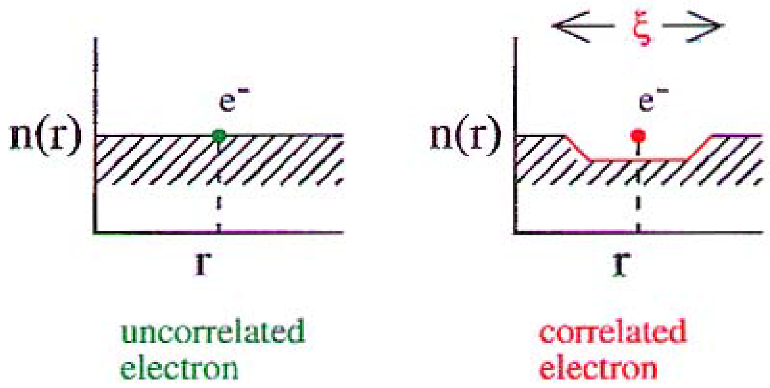
	low density	high density
$U \ll E_F$		
$U \gg E_F$		$\surd$

- More complex structures

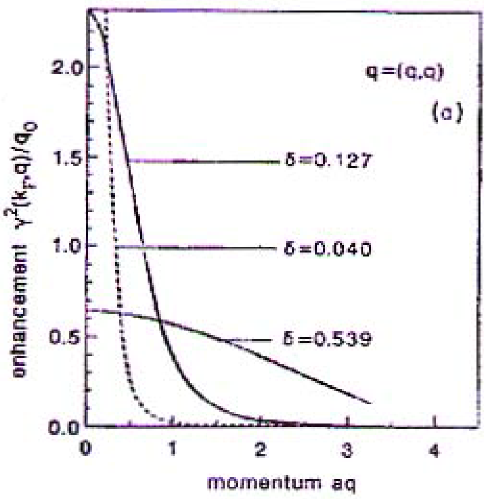
	$k_F \ll \frac{\pi}{2a}$	$k_F \simeq \frac{\pi}{2a}$
large E_F		
small E_F		$\surd$

Electron-phonon interaction in strongly correlated metals

- Narrow bands $\Rightarrow E_F \sim U$
  $\Rightarrow$ strong correlation



Large correlation hole induces small q-momenta selection: $q_c \propto 1/\xi$



[M. Kulić and R. Zeyher, PRB1994]

High T_c in nonadiabatic metals

Low charge carrier density

⇓

small E_F

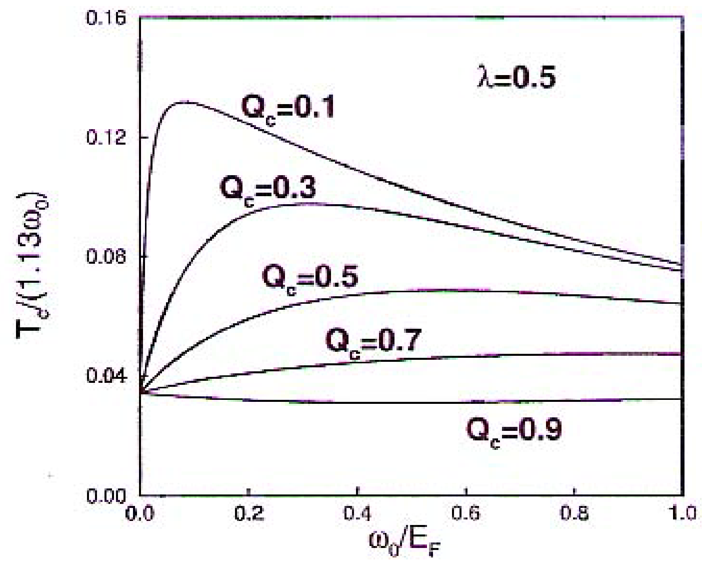
⇓

nonadiabatic regime

(+ strong correlation)

⇓

enhancement of superconducting pairing

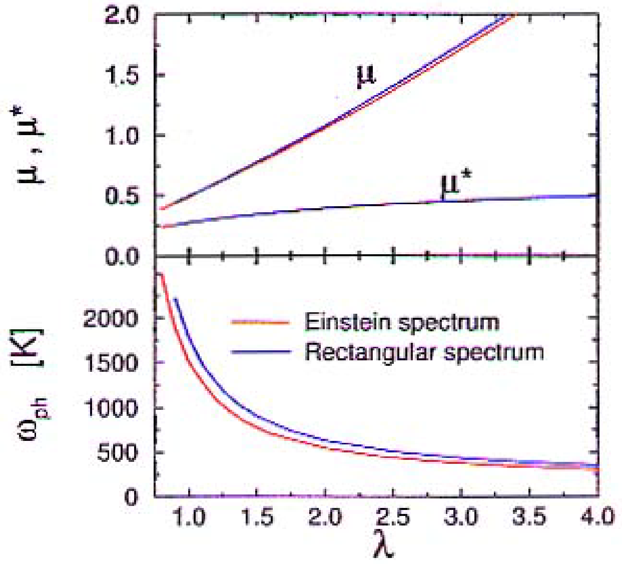


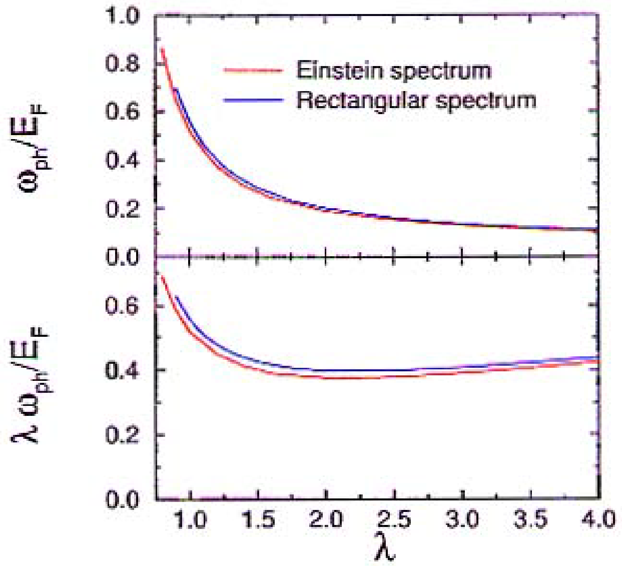
High-T_c: conventional parameters in a new theory

Superconductivity Rb_3C_{60}

Inconsistency of ME theory

- $T_c = 30$ K;
- $\alpha_{T_c} = 0.21$

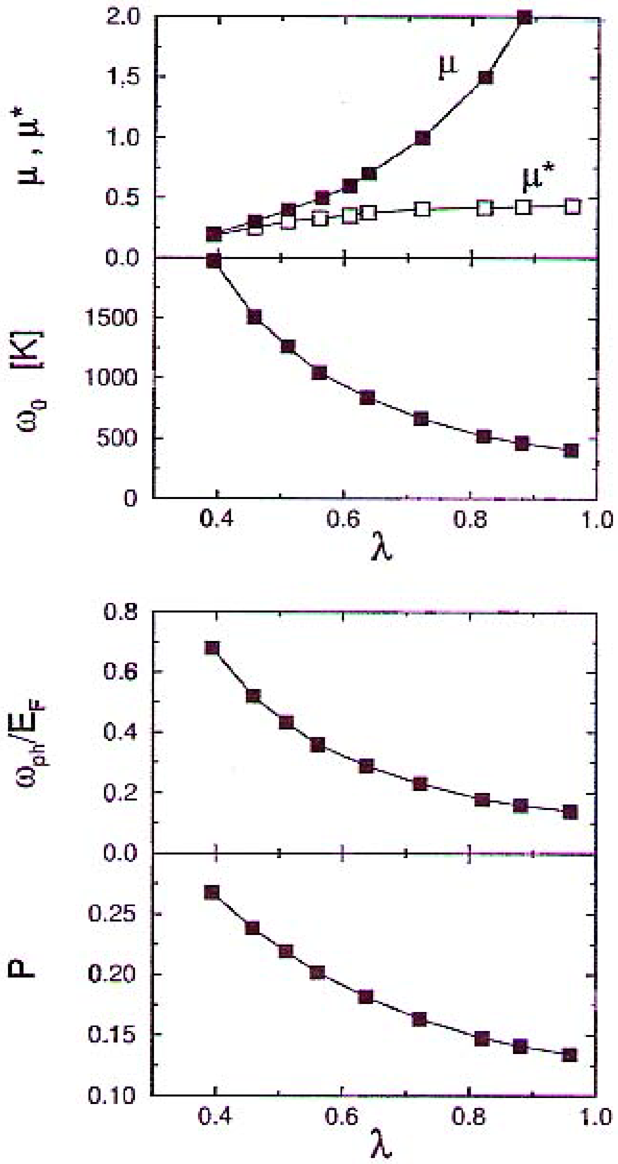




Nonadiabatic analysis

Rb_3C_{60}

- $T_c = 30$ K;
- $\alpha_{T_c} = 0.21$



Nonadiabatic characteristic features

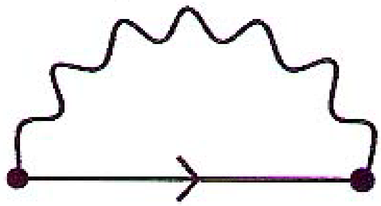
Experimental tests of nonadiabatic effects:

anomalous features not expected in conventional theory

In general, isotope effects provide important informations about nonadiabatic interaction

- ⇨ Isotope effect on quantities expected not to have it

Effective mass m^*

Σ = 

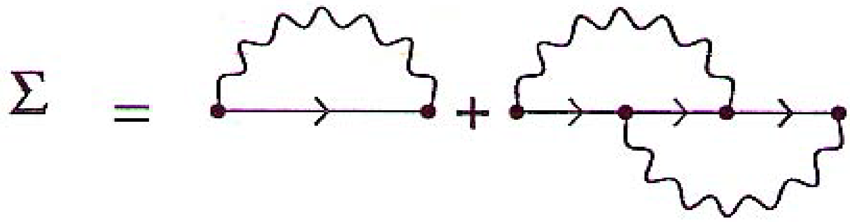
$$\frac{m^*}{m} = 1 - \left.\frac{\Sigma(i\omega_n)}{i\omega_n}\right|_{n=0}$$

In Migdal-Eliashberg: $m^*/m = 1 + \lambda$

$\Rightarrow$ no isotope effect

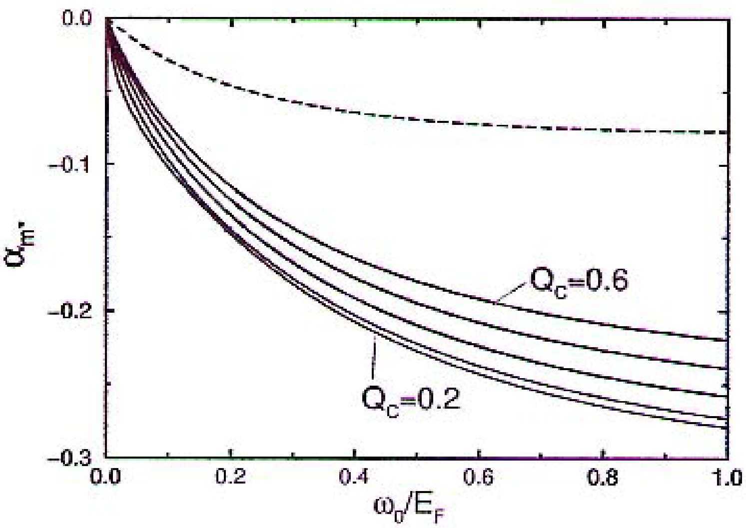
No more true in the nonadiabatic theory

Nonadiabatic effective mass m^*



- $m^*(\lambda) \longrightarrow m^*(\lambda, \omega_{\mathrm{ph}}/E_F)$

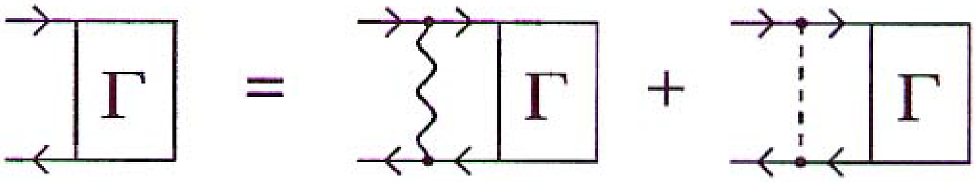
Finite isotope effect



Pauli spin susceptibility χ

$$\chi = -\lim_{q\to 0} \mu_B^2 T \sum_{\mathbf{k},n} G(\mathbf{k},\omega_n) G(\mathbf{k}+\mathbf{q},\omega_n) \times \Gamma_s(\mathbf{k}+\mathbf{q},\mathbf{k};\omega_n)$$

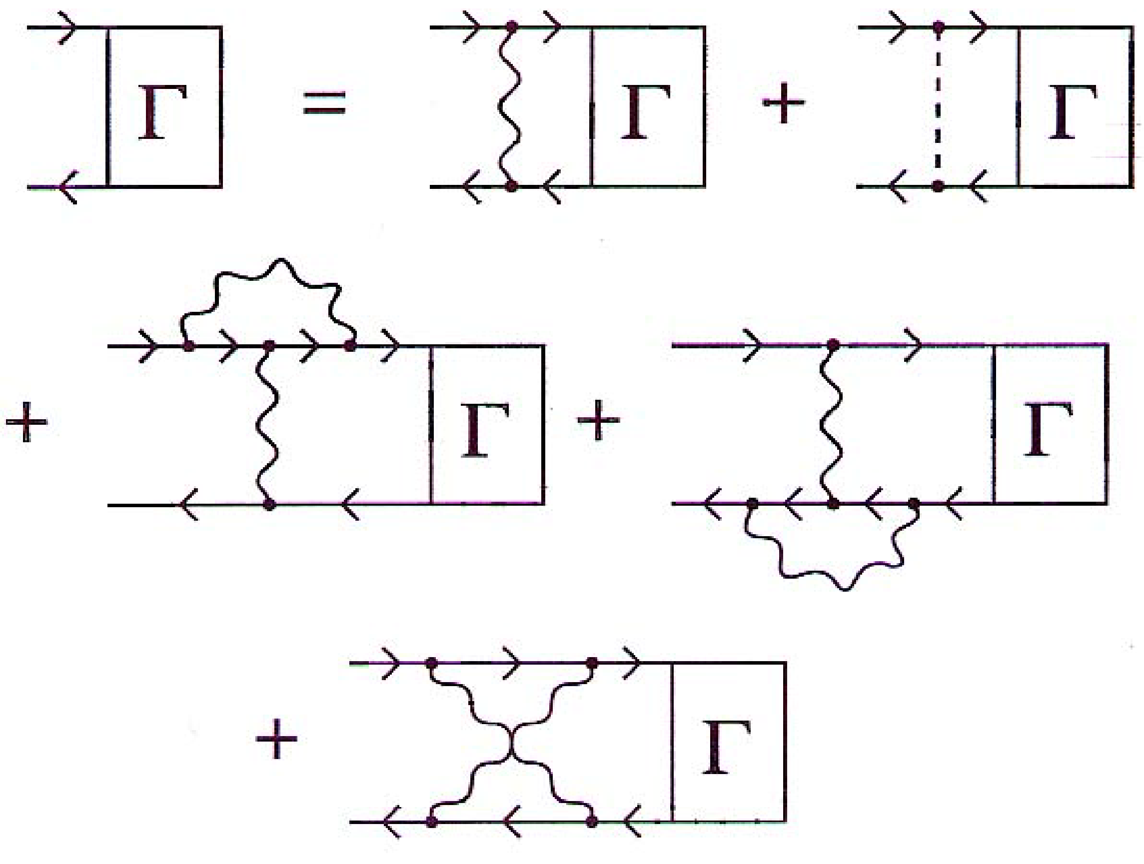
- Migdal-Eliashberg



$\Rightarrow \chi = \mu_B^2 N(E_F) \qquad [\mu_B^2 N(E_F)/(1+I)]$

- no el-ph renormalization of χ
- no isotope effect on spin susceptibility

Nonadiabatic Pauli susceptibility



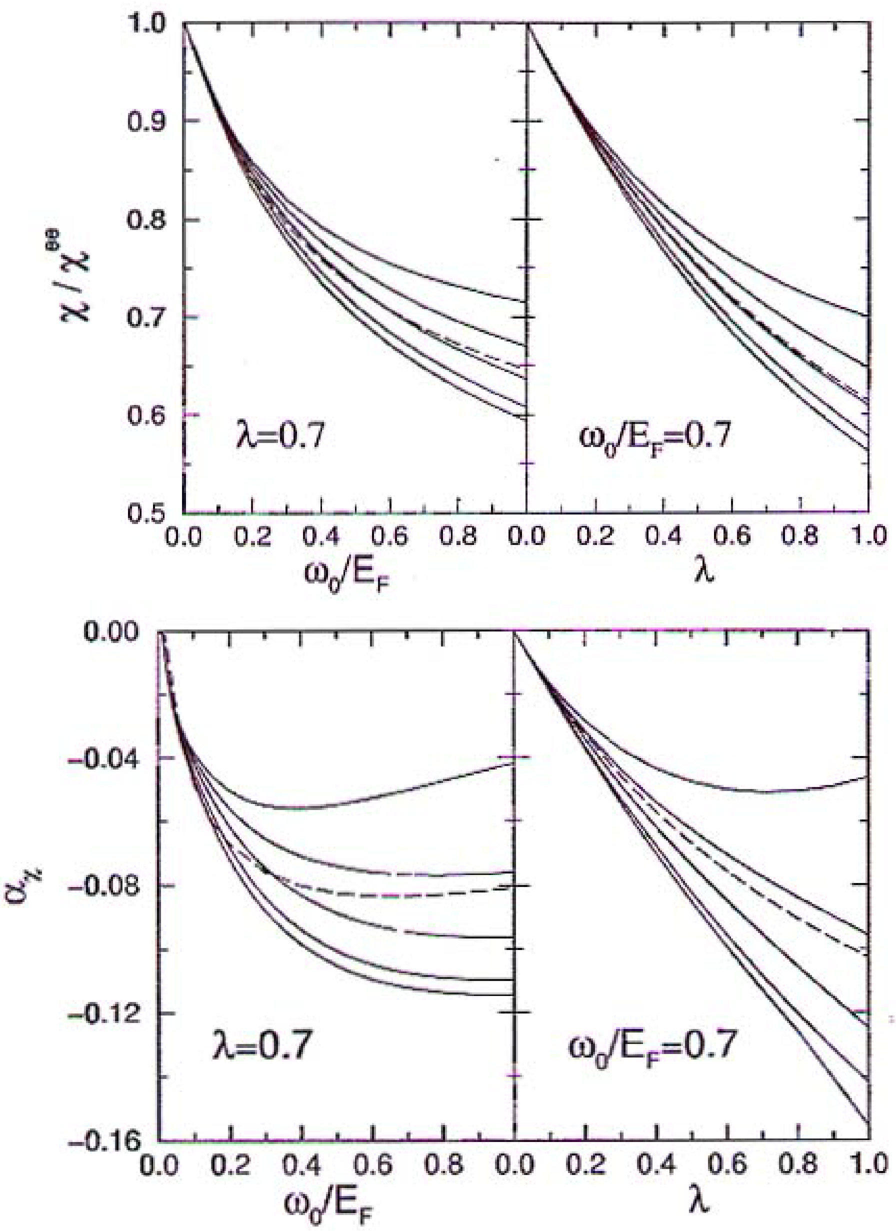
again:

- $\chi \longrightarrow \chi(\lambda, \omega_{\mathrm{ph}}/E_{\mathrm{F}})$

$\Rightarrow$ Electron-phonon renormalization

$\Rightarrow$ Finite isotope effect

Dependence of χ on λ and ω_{ph}

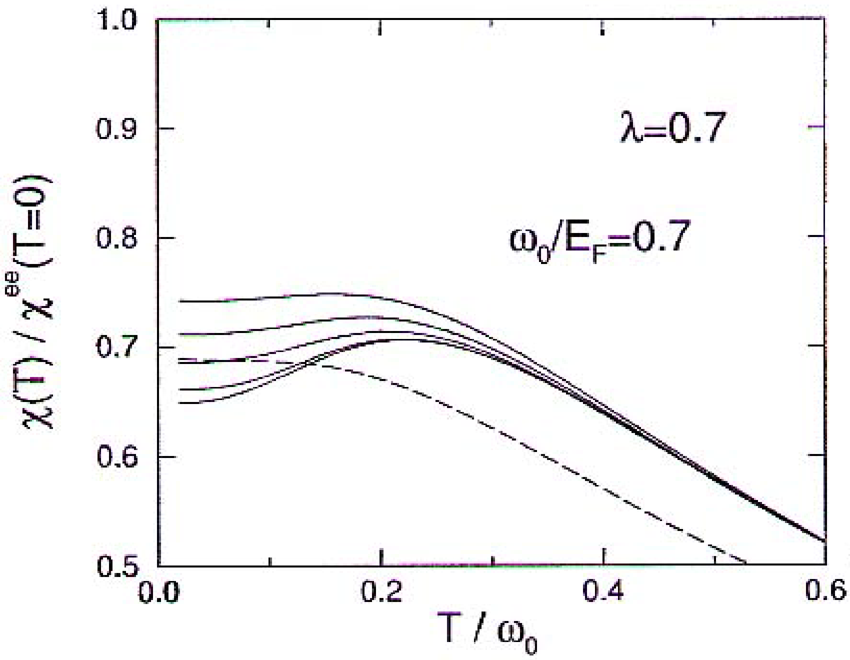


Temperature dependence of χ

In Migdal-Eliashberg: $\chi = \mu_B^2 N(E_F)$ constant in temperature $T \ll E_F$

Finite bandwidth effects: decrease of χ with temperature

Non monotonic temperature dependence in nonadiabatic theory



Conclusions

- Fullerenes: molecular crystal
- Small Fermi energies
- New physics (nonadiabaticity)
- Anomalous metal properties
- Anomalous superconductivity (high-T_c)
- New perspectives (material engineering)