

Localized magnetic moments in metals; reservoirs of novel electronic ground states and phenomena

e.g.,

* Formation ("survival") of magnetic moments of T, Ln, Ac ions with partially-filled d- or f-electron shells in metal

* Kondo effect

* Reentrant SC due to Kondo effect

* Coexistence of SC and AFM

* Reentrant SC due to FM

Generation of new oscillatory magnetic state that coexists with SC due to SC-FM interactions

* Heavy fermion compounds ($m^* \sim 10^2 m_e$)

* Unconventional SC in heavy fermion compounds

Pairing with $L > 0$, nodes in energy gap
Magnetic pairing mechanism

* NFL behavior associated with QCPs

* SC in the vicinity of ~~QCP~~ magnetic QCP associated by pressure } AFMs
FM's

* Heavy fermion behavior and SC in $PrOs_4Sb_{12}$

possibly due to fluctuations of electric quadrupole moments, rather than magnetic dipole fluctuations

* Survey these ~~modern~~ electronic ground states and phenomena

* Emphasis on experiment

* Introduce some ideas and review some history -
useful in our discussion of experiments

* Blackboard, V6's

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Outline

1. Moment formation (or "survival") in metal

Magnetic moment

Experimental observations

Friedel-Anderson model

Virtual bound state

Schrieffer-Wolff transformation

Exchange ~~of~~ interaction

Kondo effect

RKKY interaction

Competition between Kondo effect and RKKY interaction

2. Localized magnetic moments in conventional SC's

Superconductivity w/o localized moments

Paramagnetic impurities in SC's

Magnetically ordered sublattices in SC's

Magnetic field induced SC

3. Valence fluctuations in bc compounds

4. Heavy fermion compounds (primarily $f_{d,d}$ ^{-fluctuations} example of d)

Normal state

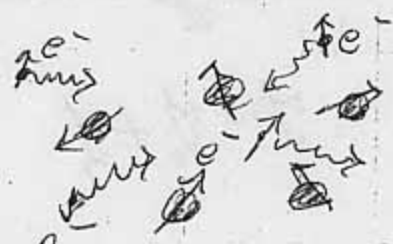
SC'ing state (unconventional)

5. Non Fermi liquid (NFL) behavior near quantum critical points (QCP's)

6. SC near QCP's

7. Heavy fermion behavior and unconventional SC in $\text{PrO}_{7-x}\text{Sb}_x$; driven by electric quadrupole fluctuations? (rather than magnetic dipole fluctuations?)

The system: magnetic moments imbedded in sea of conduction electrons

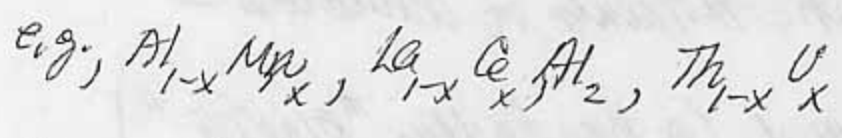


* Magnetic moments, μ derived from e^- -

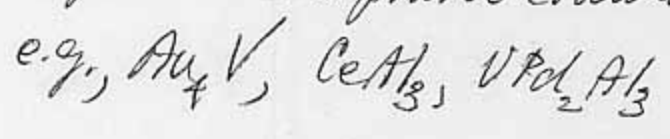
1st row transition metal T (3d), lanthanide Ln (4f), actinide Ac (5f) ions with partially-filled d or f-electron shells

* T, Ln, Ac atoms

substituted for other atoms (low concentration: "impurity")



Component of compound (ordered sublattice)



* Interaction between localized magnetic moments and conduction electron spins

Exchange interaction

$$\mu_{ex} = -2J \sum_{\vec{n}} \vec{S} \cdot \vec{S} \quad T \text{ and } \mu = 0 \text{ "quenched"}$$

$$\mu_{ex} = -2J \frac{\langle \vec{L} \cdot \vec{J} \rangle}{J(J+1)} \sum_{\vec{n}} \vec{J} \cdot \vec{S} = -2J \frac{J(L+1)}{J(J+1)} \sum_{\vec{n}} \vec{J} \cdot \vec{S}$$

Ln, Ac ions ($L \neq 0$, $\vec{J} = \vec{L} + \vec{S}$)

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J-exchange interaction parameter

$J \sim J_0 + J_1$ (FM) $J_0 > 0$ Heisenberg

(AFM) $J < 0$ Hybridization of localized
d-or f states and conduction
electron states

(Very important!)

Look at magnetic moments in insulators -

How are the formed (or how do they "survive")
in metal

Local moment paramagnetism (insulators)

$$\vec{\mu} = -g \mu_B \vec{J}$$

$$\vec{J} = \vec{L} + \vec{S}$$

$$g_J = 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)} \quad \text{Landé } g\text{-factor}$$

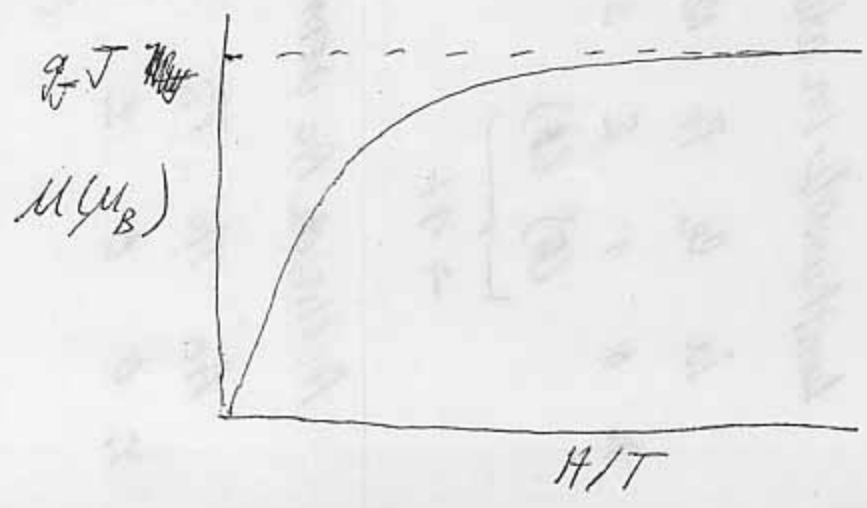
$$\mu_B = \frac{e\hbar}{2mc} = 0.927 \times 10^{-20} \text{ erg/gauss}$$

$$E = m_J g_J \mu_B H; \quad m_J = J, J-1, \dots, -J$$

($2J+1$ equally spaced levels)

$$M = N g_J \mu_B B_J(x), \quad x = g_J \mu_B H / k_B T$$

$$B_J(x) = \frac{2J+1}{2J} \text{ctnh} \left(\frac{(2J+1)x}{2J} \right) - \frac{1}{2J} \text{ctnh} \left(\frac{x}{2J} \right)$$



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$x \ll 1$

~~WAAATTTT~~ $(N g_J^2 J(J+1) \mu_B^2 / 3k_B T) H =$

$M \approx (N \mu_{eff}^2 / 3k_B T) H = \chi A$

$$\chi = \frac{M}{H} = \frac{N \mu_{eff}^2}{3k_B T} = \frac{C}{T}$$

 Curie law

$C = N \mu_{eff}^2 / 3k_B$ Curie constant

$\mu_{eff} = g_J [J(J+1)]^{1/2} \mu_B$ effective moment

~~WAAATTTT~~

$x \gg 1$



$M \approx g_J J \mu_B$ saturation moment

A, L, J derived from Hund's rules

Ionic configuration $4f^n$ (lanthanide)

$4f$ electron - $l=3, s=1/2$

$S =$ maximum value $\sum_i (s_i)$

$L =$ maximum value $\sum_i (l_i)$ (subject to Pauli principle)

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$J = |L - S|$ shell less than half filled ($n < 7$)

$J = L + S$ shell more than half filled ($n > 7$)

{ Hund's rules }

e.g., Ce^{3+} ($4f^1$) $S = \frac{1}{2}, L = 3, J = L - S = \frac{5}{2}$

Pr^{3+} ($4f^2$) $S = 1, L = 5, J = L + S = 4$

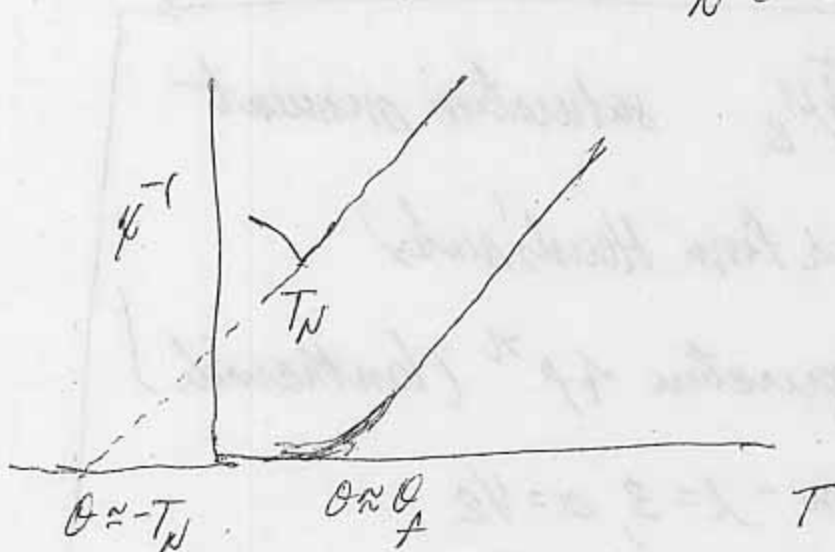
Curie-Weiss law

$$\chi = N\mu_{eff}^2 / 3k_B(T - \Theta)$$

Θ - Curie-Weiss temperature

ferromagnet $\Theta \approx T_c$ (Curie temperature)

antiferromagnet $\Theta \approx -T_N$ (Néel temperature)



NOTE:

Θ is measure of other ~~magnetic~~ interactions
 e.g., Kondo temperature T_K
 valence fluctuation temperature T_0

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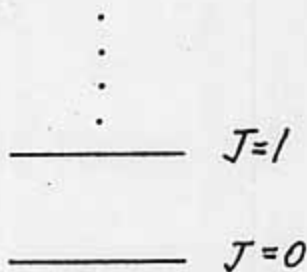
(2) Van Vleck anomalies in χ due to small multiplet splittings of S_m and E_u -

e.g., $E_u^{3+} - 4f^6$

ground state -

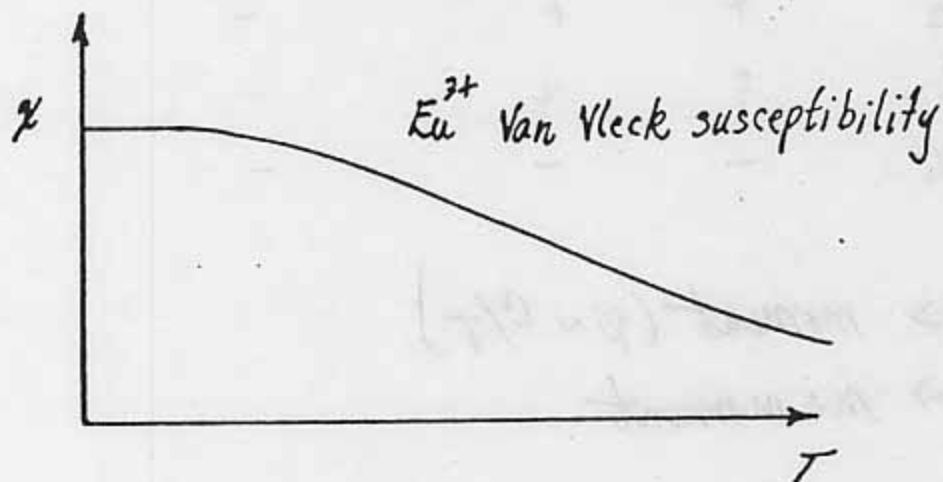
$$S=3, L=3, J=|L-S|=0$$

spectroscopic notation - 7F_0



superscript (7) - $2S+1$

subscript (0) - J

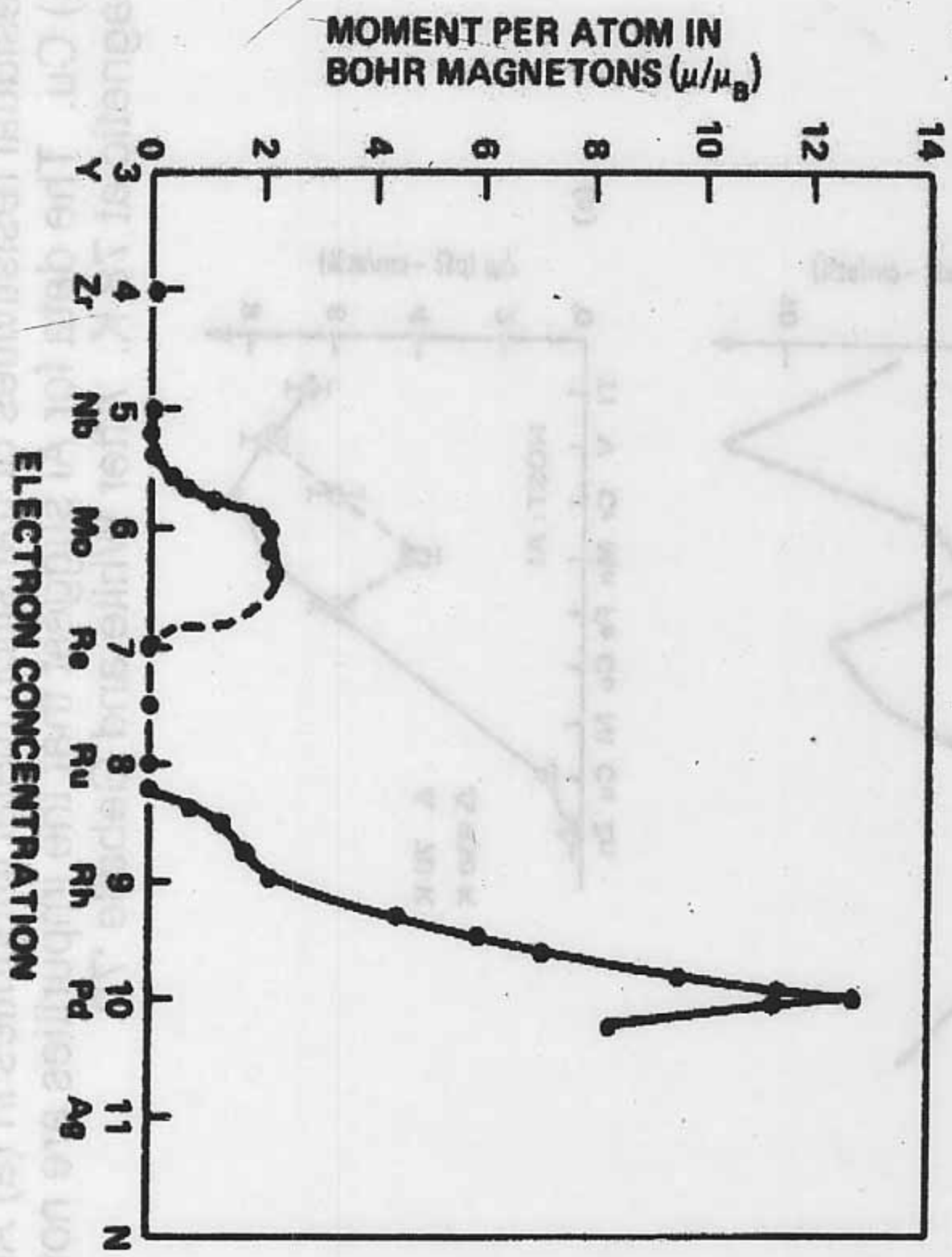


$$k_B T \gg E_1 - E_0 \quad \chi \sim \frac{N |\langle 1 | \mu_z | 0 \rangle|^2}{k_B T}$$

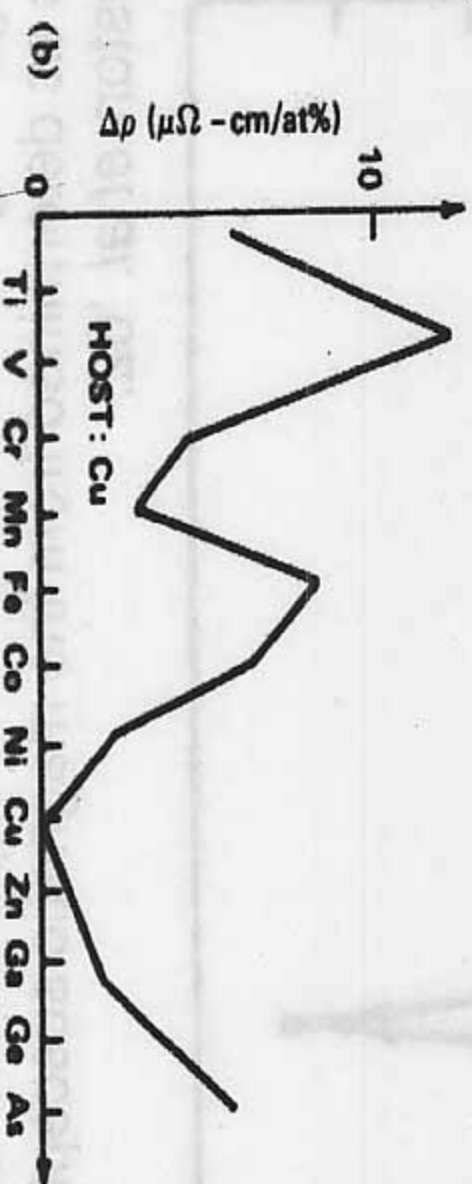
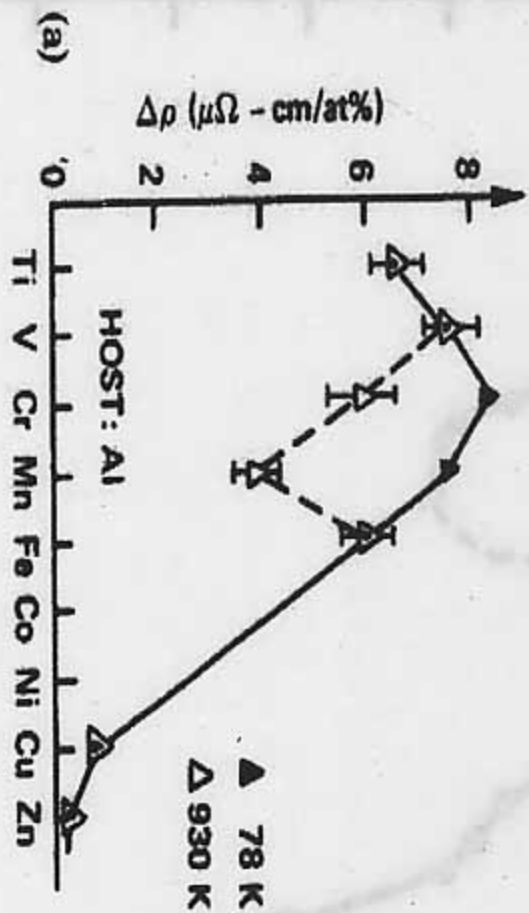
$$k_B T \ll E_1 - E_0 \quad \chi \sim \frac{2N |\langle 1 | \mu_z | 0 \rangle|^2}{E_1 - E_0}$$

relevant for discussing S_m^{2+} -

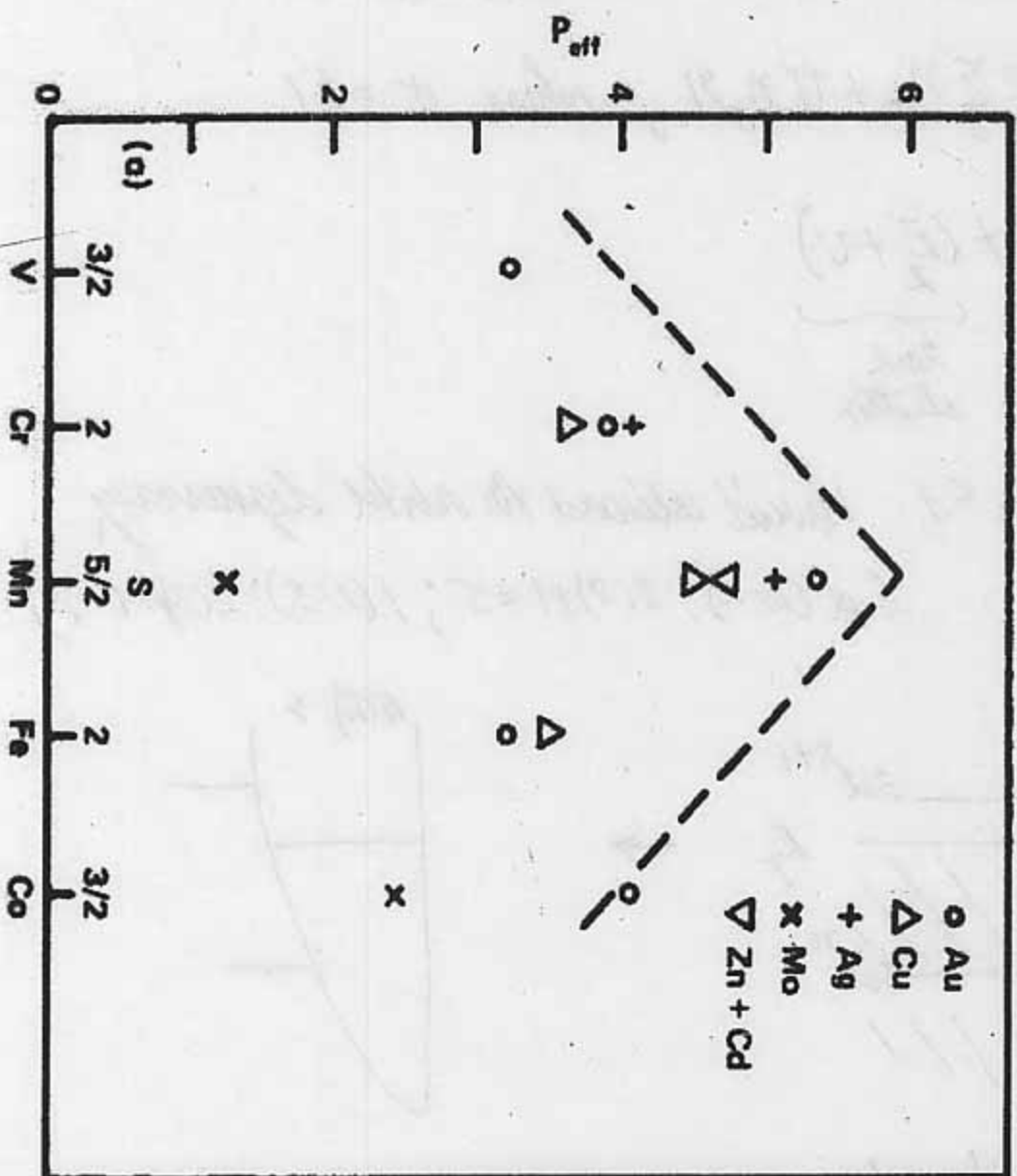
The average magnetic moment of Fe impurities in 4d metals and alloys as determined from the magnetic susceptibility. After Clogston *et al.* '62.



Residual resistivities of transition metal impurities in (a) Al and (b) Cu. The data for Al suggest that the impurities are not magnetic at 78 K. After White and Geballe '79.



Effective magnetic moments of transition metal ions in various hosts. Dashed line: free ion value $2[S(S+1)]^{1/2}$. After Rizzuto '74.



"Resonant state" or "virtual bound state" (vbs)

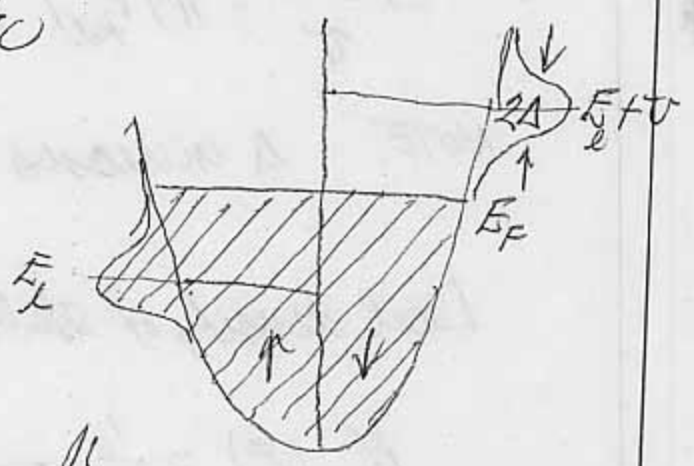
Magnetic moment: $\mu = (\langle n_{\uparrow} \rangle - \langle n_{\downarrow} \rangle) \mu_B = 0$

Magnetic state

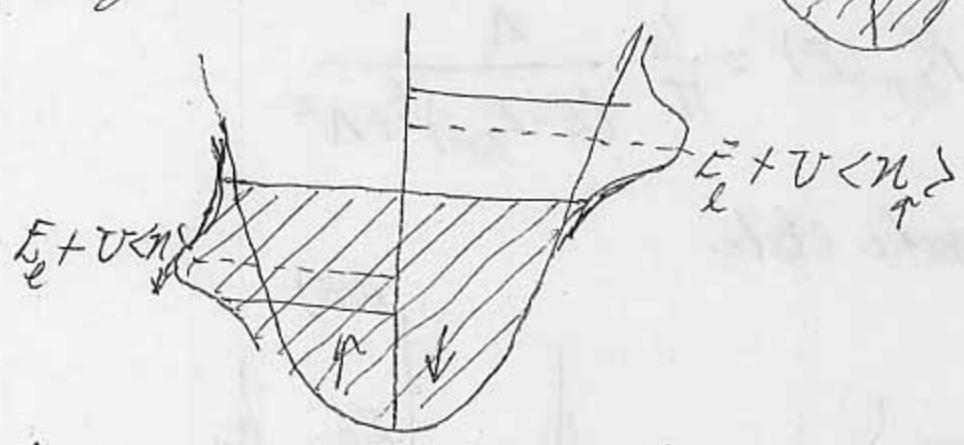
U sufficiently large \Rightarrow magnetic state

Criterion for magnetic moment for symmetric case

$\Pi \Delta / U < 1$ magnetic



Demagnetization



Leads to nonintegral moments

Increase Δ further or decrease $(E_F - E_{LT})$ further

\Rightarrow nonmagnetic solution $\langle n_{\uparrow} \rangle = \langle n_{\downarrow} \rangle$

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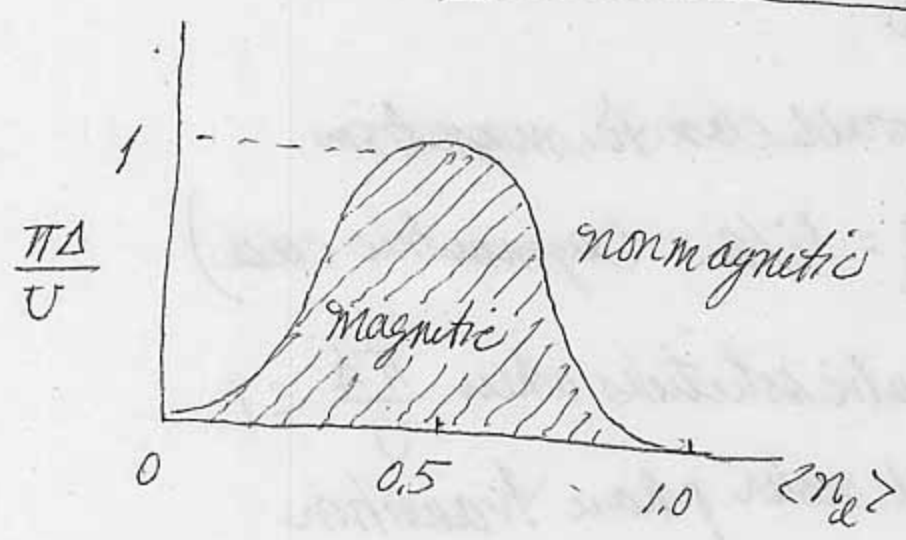
Phase boundary

$\langle n_{d\uparrow} \rangle \neq \langle n_{d\downarrow} \rangle$ magnetic solution

$\langle n_{d\uparrow} \rangle = \langle n_{d\downarrow} \rangle$ nonmagnetic solution

Non magnetic when

$$1 \geq U N_d(E_F) = \frac{U}{\pi \Delta} \sin^2(\pi \langle n_d \rangle)$$



⇒ localization most probable near middle of solute transition metal series

Explains Table of moment formation of 3d solute in Au, Cu, Ag, Al (order of increasing $N(E_F)$)

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"Virtual bound state" of transition metal impurity in metallic host

Friedel sum rule (band or scattering theory)

$$Z = \frac{Z}{\pi} \sum_L (2L+1) \eta_L(k_F)$$

η_L - phase shift of scattering particle

Z charge differential between solute and metallic host

$$\Delta\rho = \frac{4\pi n_i}{ne^2 k_F} \sum_L (2L+1) \sin^2(\eta_L - \eta_{L+1})$$

If only one phase shift is large

$$\eta_L(k_F) = \pi Z / 2(2L+2) = \pi Z / (4L+2)$$

$$\Rightarrow \Delta\rho = \frac{4\pi n_i}{ne^2 k_F} (2L+1) \sin^2\left(\frac{\pi Z}{4L+2}\right)$$

$$\text{For } Z=5, \eta_L = \frac{5\pi}{4(2)+2} = \frac{\pi}{2} \quad \sin(\pi/2) = 1$$

$\Delta\rho$ max.

As increase Z , vbs sinks into Fermi sea

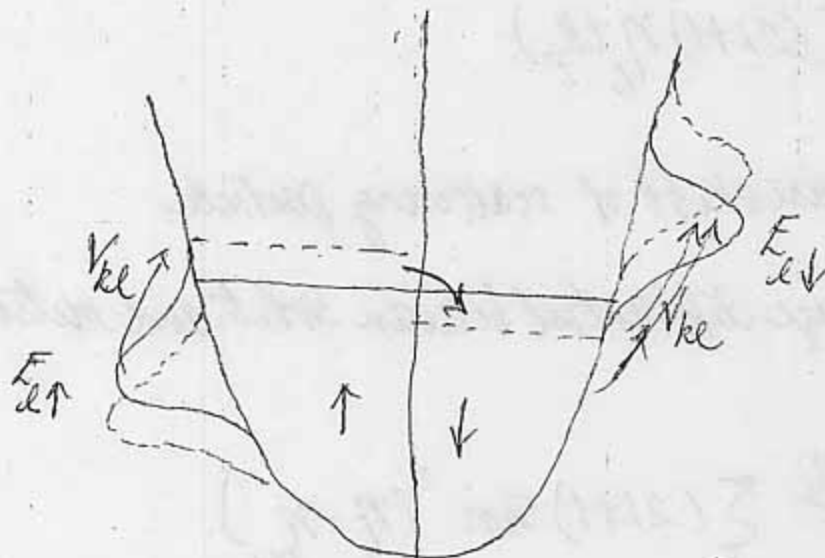
$\Delta\rho$ goes through maximum when centroid of vbs at E_F

One peak for nonmagnetic vbs / two peaks for magnetic vbs (Pauli V)

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Effect of mixing interaction on the magnetic state

- V_{kl} { (1) broadens localized states
- (2) allows electron transfer between localized state and conduction electron states



Matrix element of perturbing Hamiltonian between two levels results in repulsion of the two levels from one another (levels "repel" each other)

Dashed lines in Figure - after repulsion

The correct Fermi level, electrons flow from ↑ states to ↓ states

Results in spin polarization of conduction electrons opposite to impurity moment

Antiferromagnetic interaction

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$$\mu_{ex} = -2D \sum_{\vec{r}} \frac{\rho}{N}$$

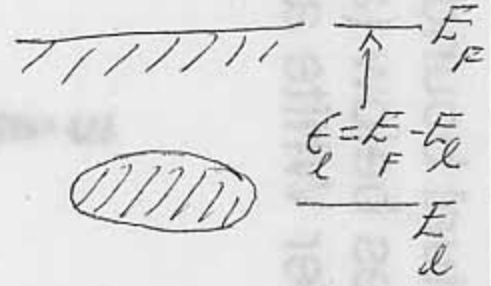
where

$$g = \frac{-|V_{kel}|^2 U}{\epsilon_d (\epsilon_d + U)} < 0$$

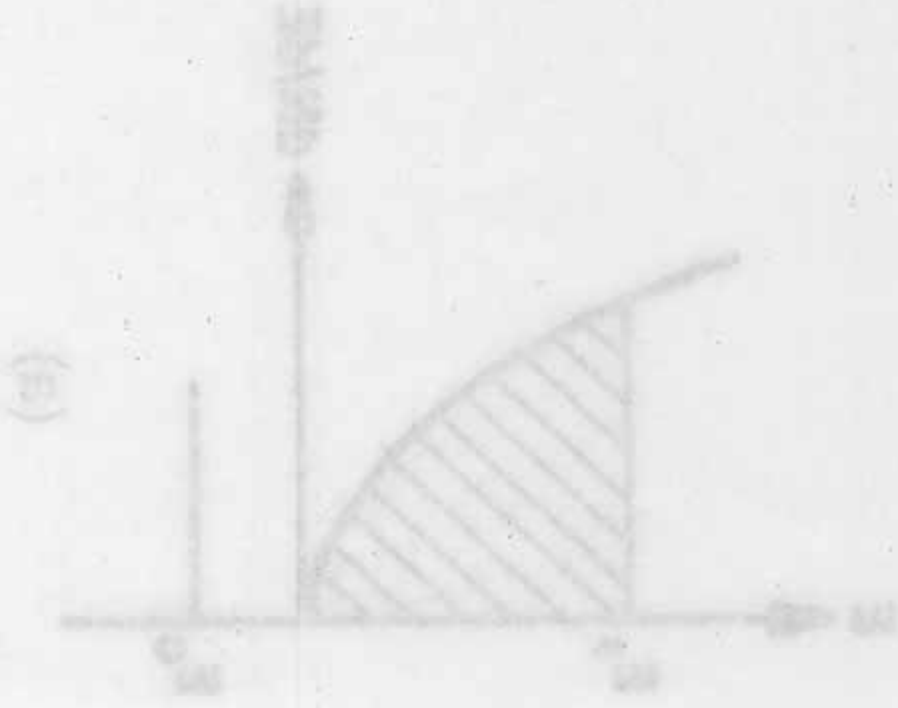
Schrieffer-Wolff transformation

For $U \gg \epsilon_d$,

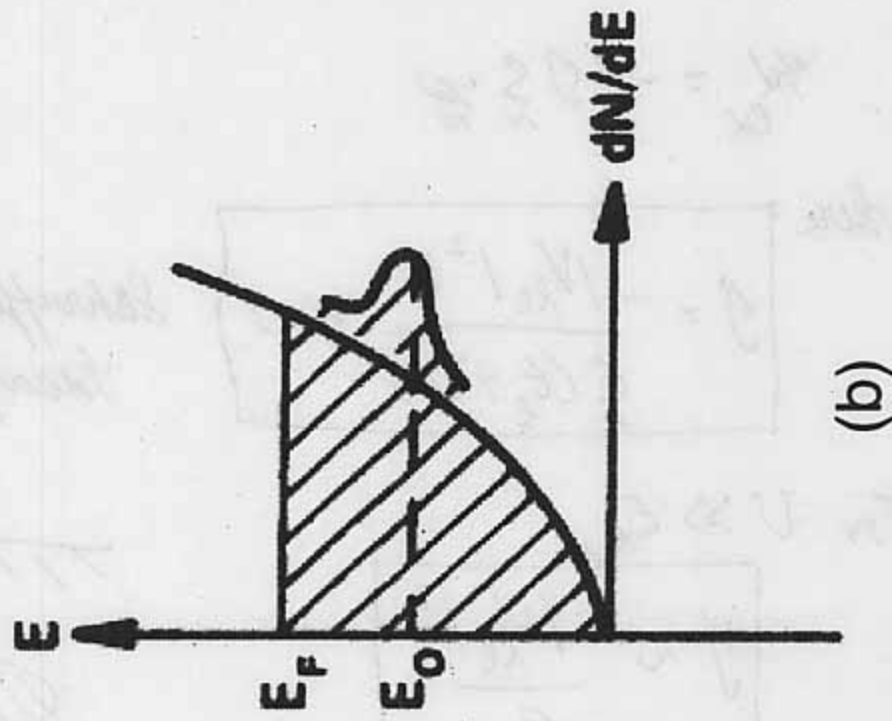
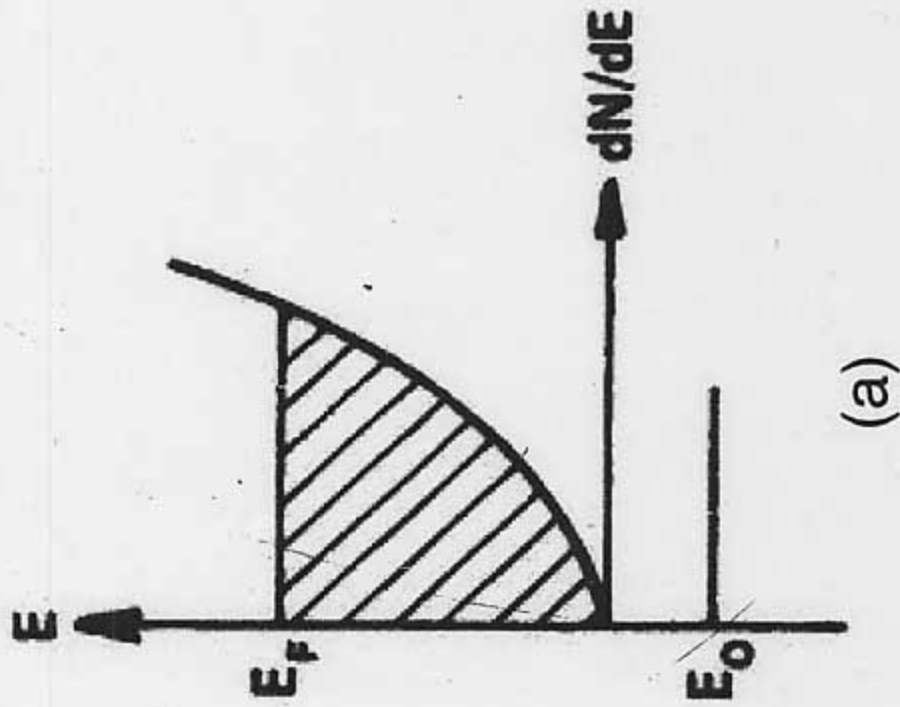
$$g \sim \frac{-|V_{kel}|^2}{\epsilon_d}$$



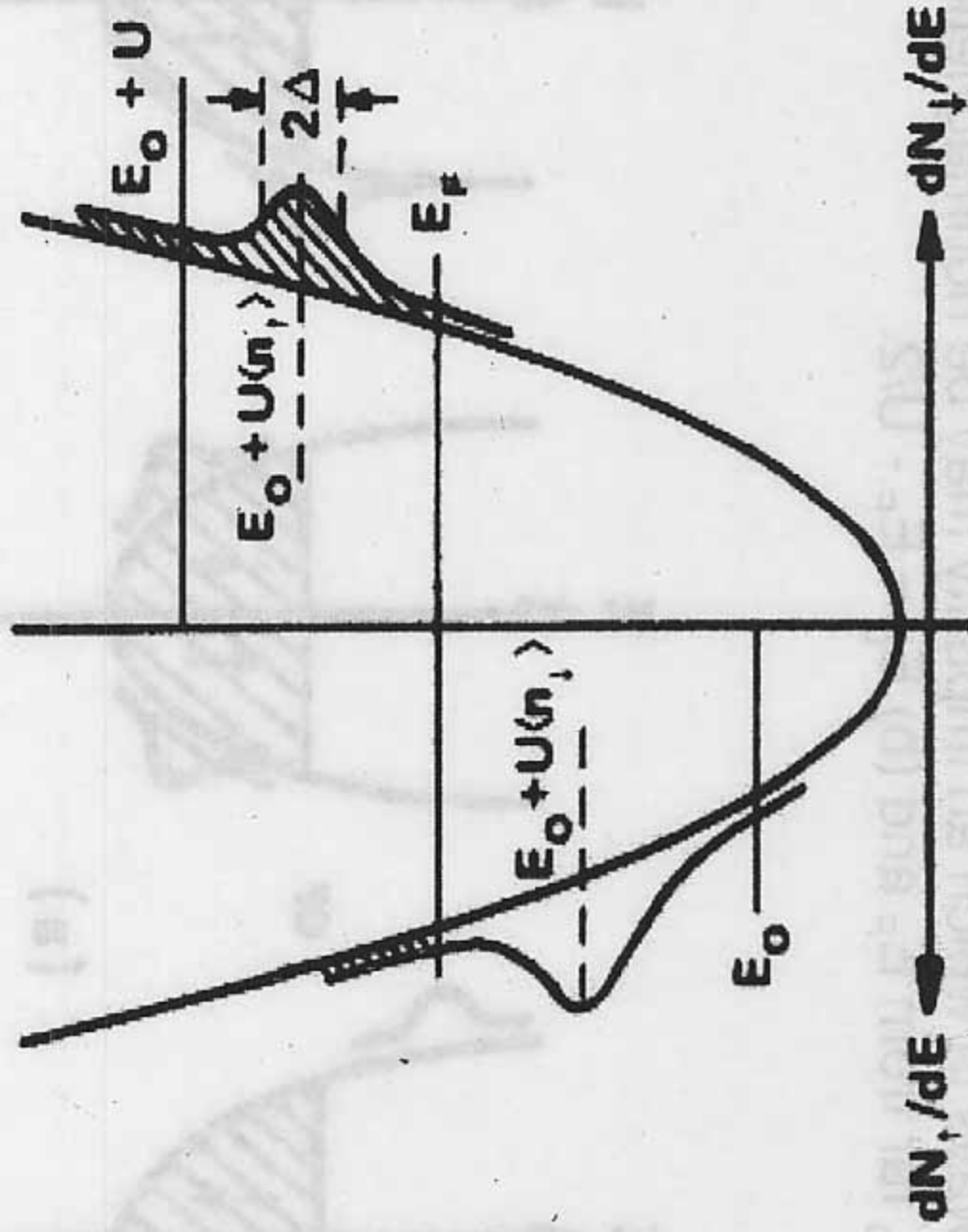
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Bound state (a) and resonance, or virtual bound state, (b) formed when energy E_0 of a localized state lies below (a) or within (b) the continuum of free electron states. After White and Geballe '79.

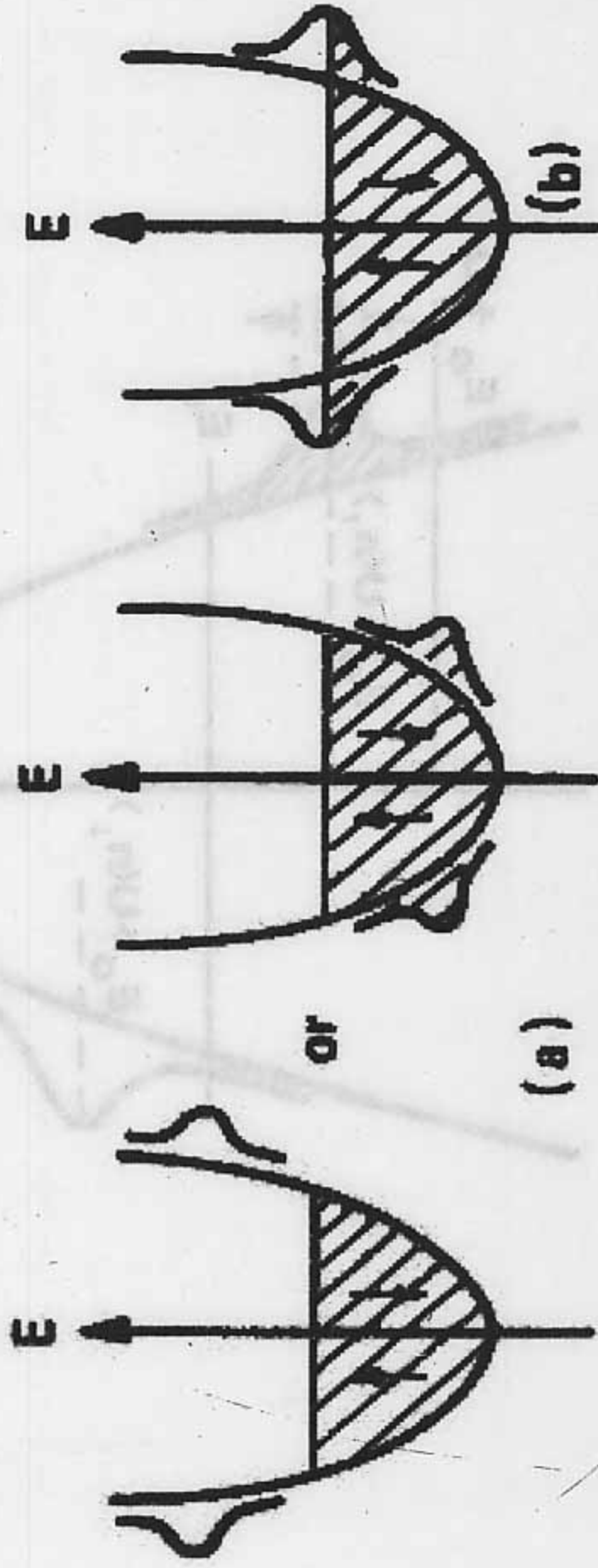


Density of states associated with a magnetic impurity (Anderson model).



Two ways in which an impurity may be nonmagnetic:

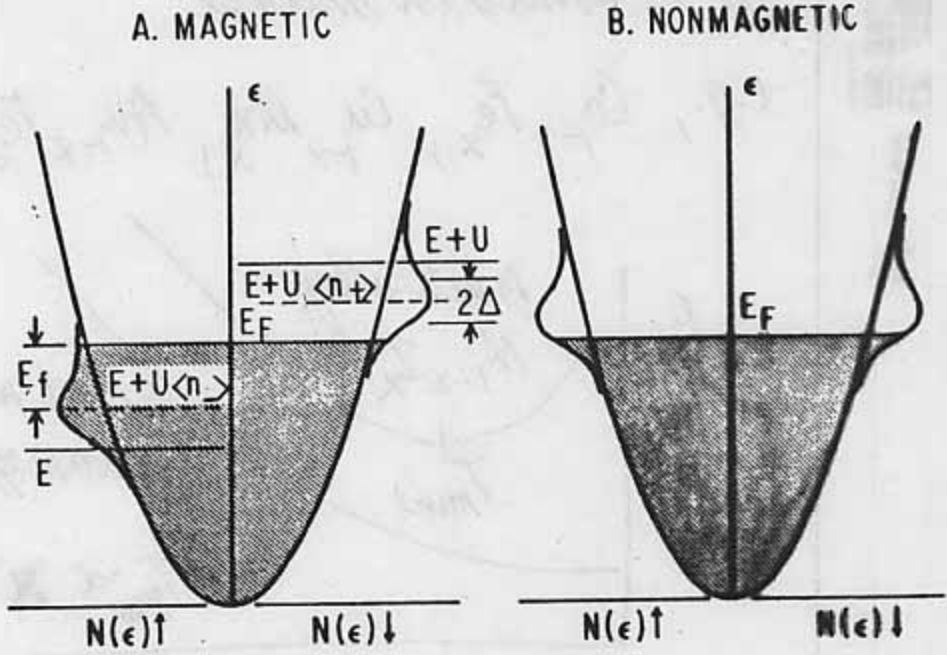
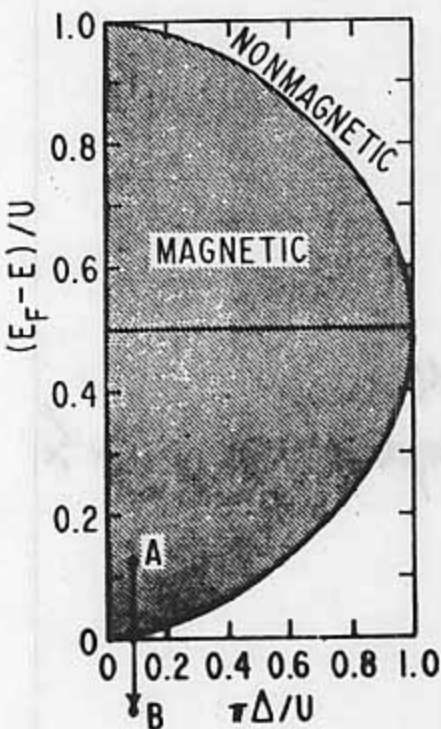
(a) E_0 far from E_F and (b) $E_0 = E_F - U/2$.



$$dN_1/dE \longleftrightarrow \frac{3p_1}{Np} \longleftrightarrow \frac{dN_1}{dE}$$

Anderson model —

Consider nondegenerate orbital state ($S=1/2, L=0$)
 (not appropriate for RE's — return to this later)



$$\Delta = \pi \langle V_{kf}^2 \rangle N(E_F); \quad N_f(E_F) = \frac{1}{\pi} \frac{\Delta}{E_f^2 + \Delta^2}$$

$$\mathcal{H}_k = \sum_{k\sigma} \epsilon_k n_{k\sigma}$$

$$\mathcal{H}_f = \sum_{\sigma} \epsilon_f n_{f\sigma} + U n_{f\uparrow} n_{f\downarrow} \quad n_f = c_{f\sigma}^{\dagger} c_{f\sigma}$$

$$U = \int |\phi_f(r_1)|^2 \frac{e^2}{r_{12}} |\phi_f(r_2)|^2 d\tau_1 d\tau_2$$

$$\mathcal{H}_{sf} = \sum_{k\sigma} V_{kf} (c_{k\sigma}^{\dagger} c_{f\sigma} + c_{f\sigma}^{\dagger} c_{k\sigma})$$

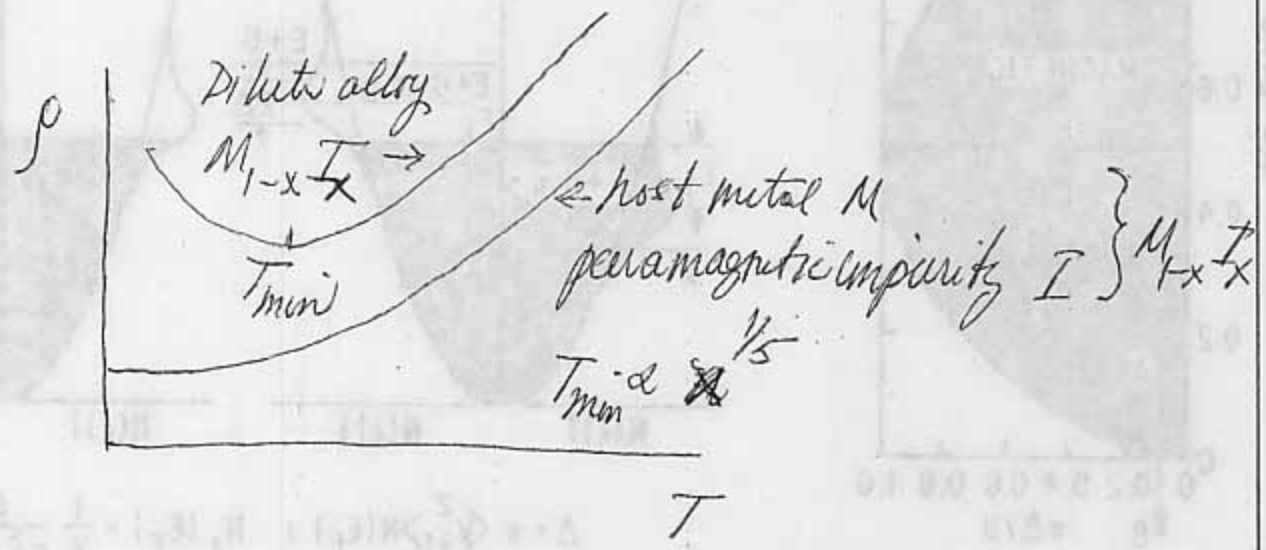
$$V_{kf} = \langle \phi_f(r) | H | \phi_k(r) \rangle$$

↑ one electron Hamiltonian

Rondo effect

Rondo effect developed to explain resistivity minimum phenomenon first observed in nominally pure noble metals such as Au in 30's and 70's and later for 3d solutes in metals

e.g., $Cu_{1-x}Fe_x$, $Cu_{1-x}Mn_x$, $Au_{1-x}Fe_x$



Rondo calculated spin dependent scattering of conduction electrons (σ) by paramagnetic impurity ions (S) via exchange interaction for $g \ll 20$ obtained

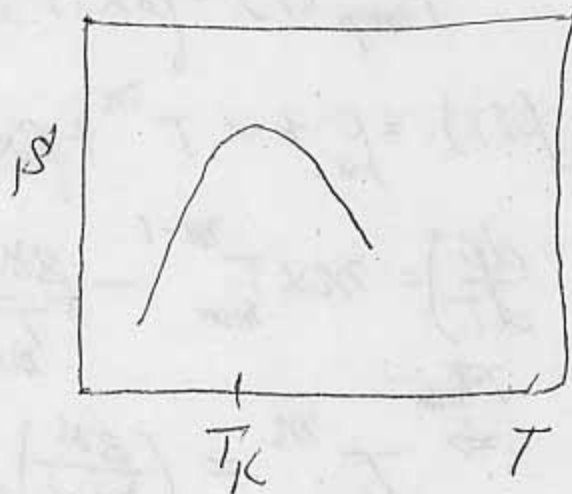
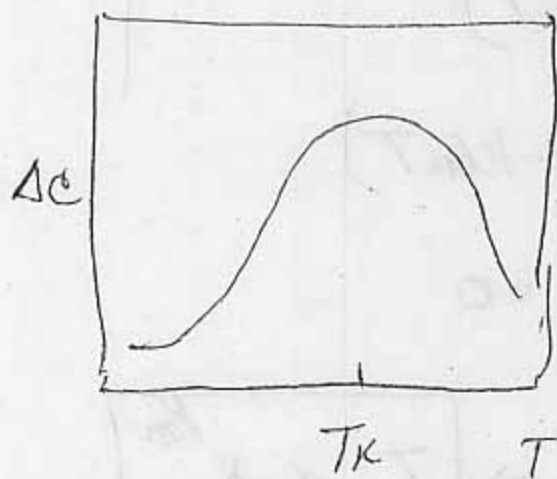
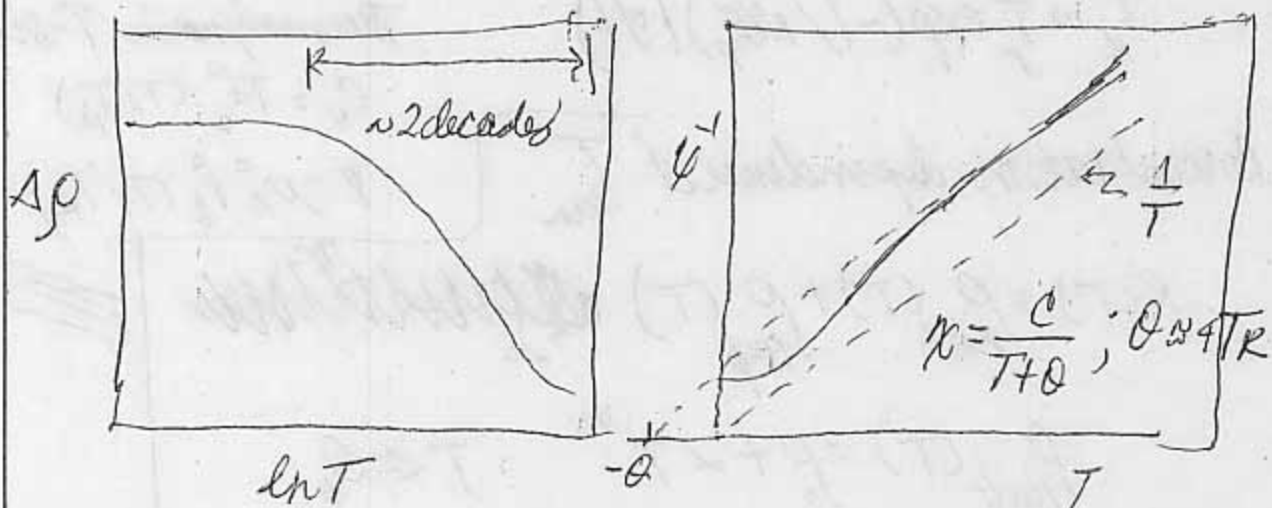
$$\rho = \rho_m^B [1 + 2N(E_F)g \ln(T/T_F) + \dots]$$

$$\rho_m^B = \frac{\pi m N(E_F)}{e^2 k N} n g^2 S(S+1)$$

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Basic properties scale with T_K



Physical picture

Many body singlet forms as T decreases below T_K
 AFM screening of \tilde{S} by \tilde{S} 's of conduction electrons (\tilde{S} 's of \tilde{S} 's),
 systems

3d transition metal (Cr, V, Mn, Fe, Ni); e.g., $\text{Cu}_{1-x}\text{Fe}_x$
 4f lanthanide (Ce, Pr, Yb); e.g., $\text{La}_{1-x}\text{Ce}_x\text{Al}_2$
 5f actinide (U); e.g., $\text{Th}_{1-x}\text{U}_x$

series diverges at "Rondo temperature" T_R

$$T_K \sim T_F \exp(-1/N(E_F) |g|)$$

Thermodynamic T-scale

$$C_V = T_F^2 C(T/T_K)$$

$$\chi \sim \nu_F^2 f_\chi(T/T_K)$$

Concentration dependence of T_{min}

$$\rho(T) = \rho_{host}(T) + \rho_{imp}(T) \approx \rho_0 + \alpha T^m + \beta \chi (1 - \delta \ln T)$$

$$\rho_{host}(T) = \rho_0 + \alpha T^m \quad T \ll \Theta_D$$

$$\rho_{imp}(T) = \beta \chi (1 - \delta \ln T)$$

$$\Rightarrow \rho(T) = \rho_0 + \alpha T^m + \beta \chi (1 - \delta \ln T)$$

$$\left. \frac{d\rho}{dT} \right|_{T_{min}} = m\alpha T_{min}^{m-1} - \frac{\beta \delta \chi}{T_{min}} = 0$$

$$\Rightarrow T_{min}^m = \left(\frac{\beta \delta \chi}{m\alpha} \right) \chi \Rightarrow T_{min} \propto \chi^{1/m}$$

Block = ?
Dimension } $m=5 \Rightarrow T_{min} \propto \chi^{1/5}$

NOTE: T_R is not at T_{min} !

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