MAGNETIC-FIELD-INDUCED SUPERCONDUCTIVITY IN LAYERED MOLECULAR CRYSTALS WITH MAGNETIC ANIONS

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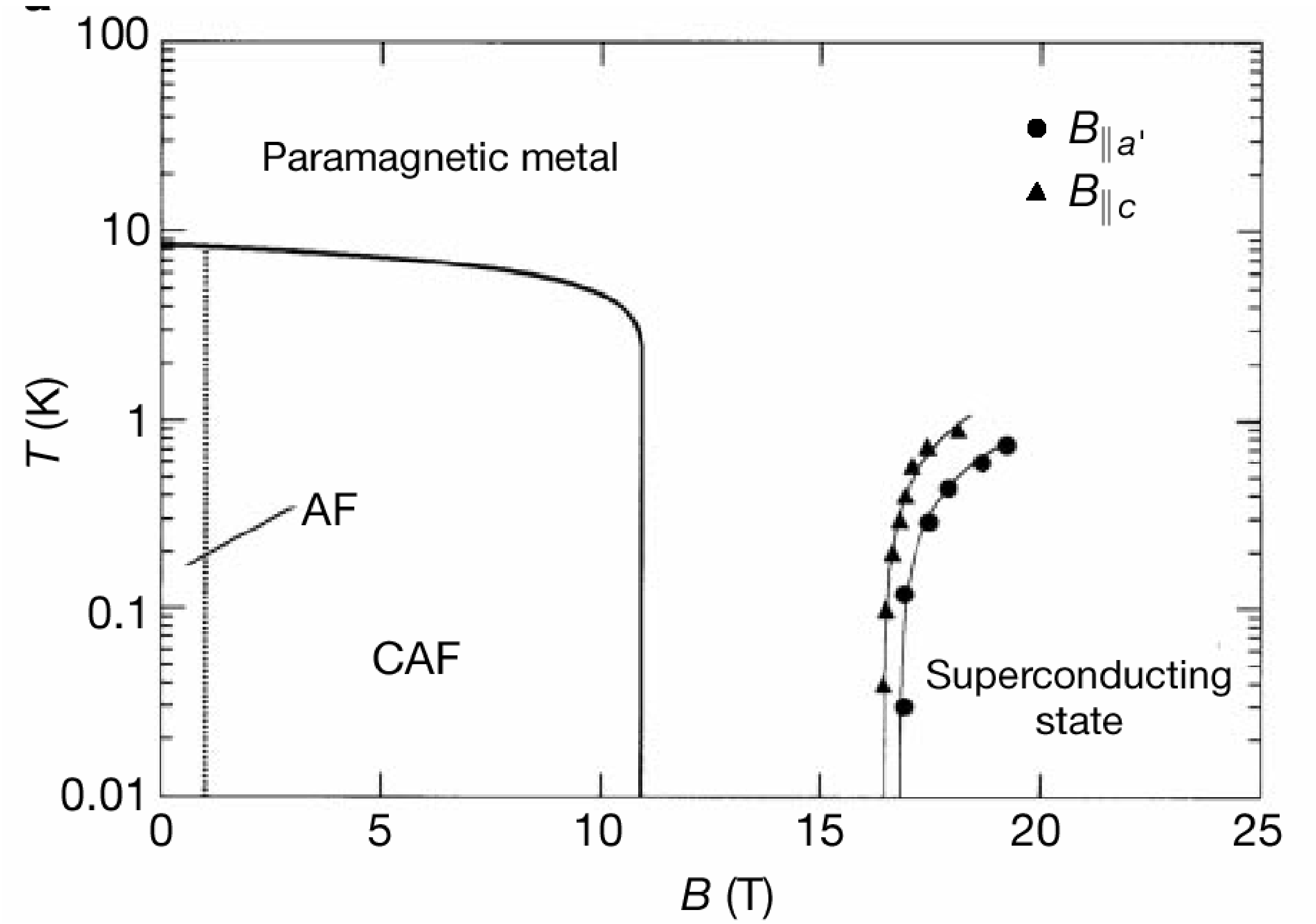
Outline

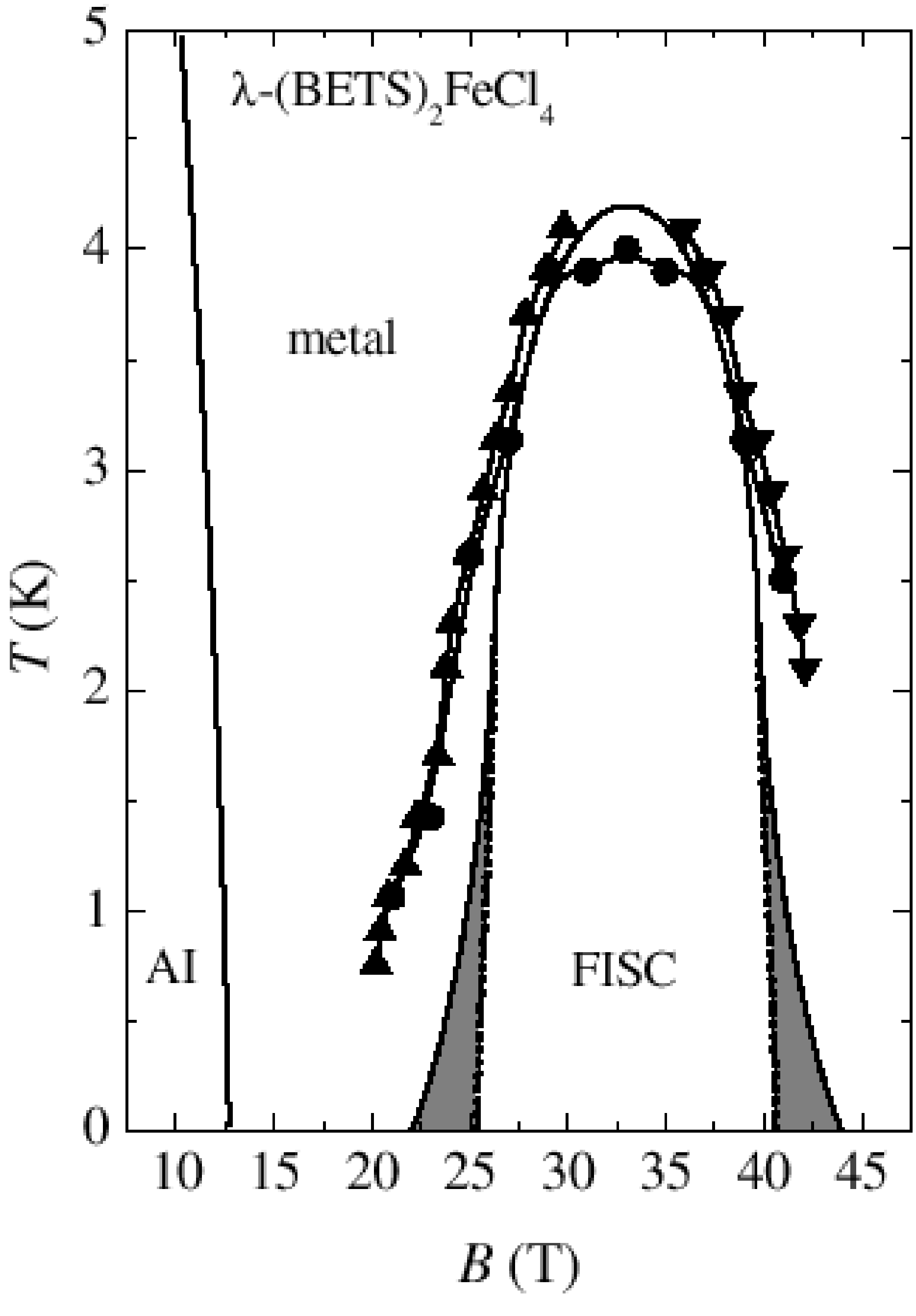
- The simplest strongly correlated electron model that can describe the λ-(BETS)$_2$ X molecular crystals is a Kondo-Hubbard model at half filling.
- The insulating phase in λ-(BETS)$_2$FeCl$_4$ is an antiferromagnetic Mott insulator, similar to that seen in the absence of the magnetic ions.
- The magnetic-field-induced superconductivity is due to the Jaccarino-Peter effect. The field acting on the spins is compensated by an exchange field $H_e = JS/g\mu_B$.
- H_e leads to beating of the magnetic oscillations. Beat frequency is consistent with the optimal field for superconductivity.

PHASE DIAGRAM of $\lambda-(BETS)_2FeCl_4$
in a magnetic field parallel to the layers

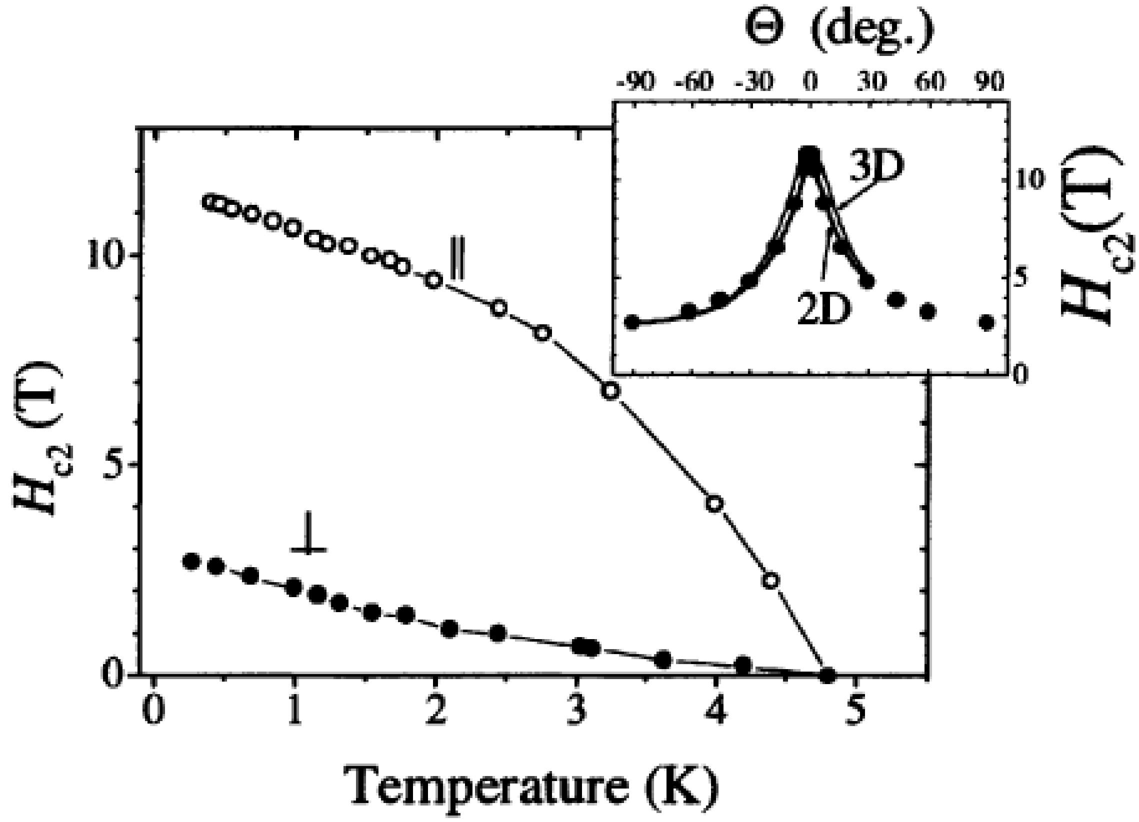
S. Uji *et al.*, Nature **410**, 908 (2001).



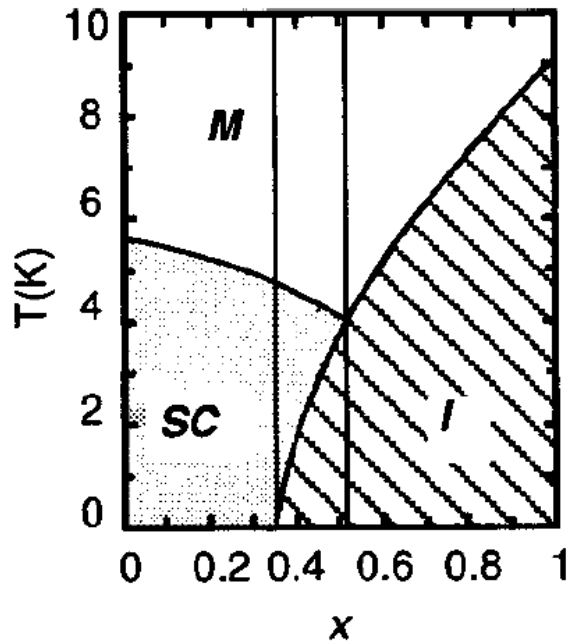
L. Balicas *et al.*, PRL **87**, 067002 (2001).



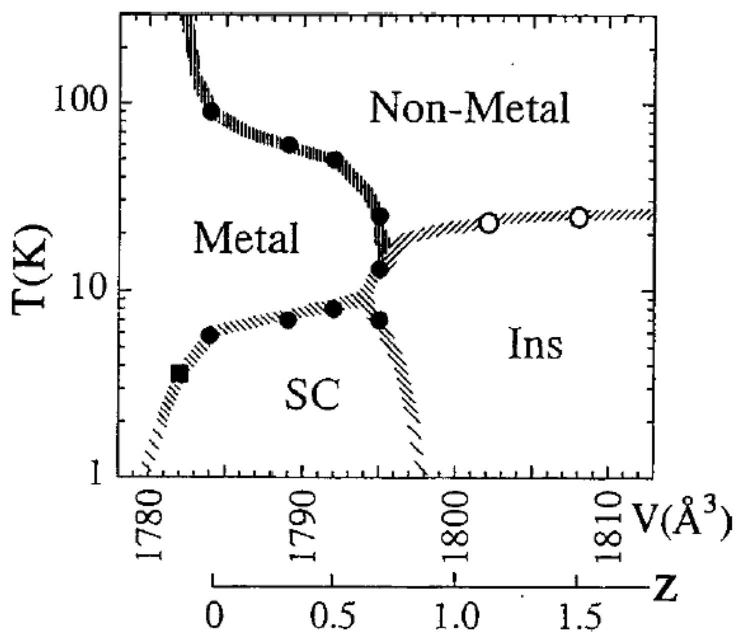
versus that of λ-$(BETS)_2GaCl_4$



PHASE DIAGRAM of λ-$(BETS)_2Fe_xGa_{1-x}Cl_4$
in zero magnetic field



PHASE DIAGRAM of λ-$(BETS)_2GaBr_zCl_{4-z}$
in zero magnetic field



Need a model Hamiltonian that can describe the subtle competition between metallic, superconducting, and antiferromagnetic insulating phases.

HUBBARD-KONDO MODEL

Brossard *et al.*, Eur. Phys. J. B, 1998.

$$\mathcal{H} = \sum_{\mathbf{ij},\sigma} t_{\mathbf{ij}}(c^\dagger_{\mathbf{i},\sigma} c_{\mathbf{j},\sigma} + h.c.) + U \sum_{\mathbf{i}} n_{\mathbf{i},\uparrow} n_{\mathbf{i},\downarrow}$$

$$+ J \sum_{\mathbf{i}} \vec{S}_{\mathbf{i}} \cdot \vec{\sigma}_{\mathbf{i}} + g_a \mu_B H \sum_{\mathbf{i}} S^z_{\mathbf{i}} + g \mu_B H \sum_{\mathbf{i}} \sigma^z_{\mathbf{i}}$$

$\vec{S}_{\mathbf{i}}$ is a spin-$S = 5/2$ operator for the Fe^{3+} local moments.
$\vec{\sigma}_{\mathbf{i}} \equiv \frac{1}{2} \sum_{\alpha,\beta} c^\dagger_{\mathbf{i}\alpha} \vec{\sigma}_{\alpha\beta} c_{\mathbf{i},\beta}$ is the spin-1/2 operator for the hole on site **i**.
- $t_{\mathbf{ij}}$ are tight-binding hopping integrals within the layers (from band-structure calculation). Very similar for Ga and Fe compounds.

- J is the Kondo coupling between the $S = 5/2$ Fe^{3+} local moments and the BETS electron spins. $J = 0$ for Ga^{3+} ($S = 0$).

-Hubbard U is the electron-electron repulsion on BETS molecules.

-One electron for two molecules. Arrangement of BETS molecules in dimers means model is effectively at half filling.

- For $U \gg t_{\mathbf{ij}}$ system is a Mott insulator.

- For small U system is a metal due to poor nesting of the Fermi surface.

- The $J = 0$ model gives a good description of κ-(BEDT-TTF)$_2$X. [R. McKenzie, Comments Cond. Mat. **18**, 309 (1998).].

NATURE OF THE INSULATING STATE

- Small U, spin-density-wave (SDW) picture (Ziman).

The local moments order antiferromagnetically due to RKKY interaction. The associated periodic potential causes Bragg reflection of the electrons, opening gaps on the Fermi-surface and eventually destroys it if J is large enough.

A magnetic field destroys the AF order of the local moments restoring the metallic state.

The charge gap $\Delta \sim JS\mu$, where μ is the SDW moment.
$\Delta \sim k_B T_{MI}$.
Δ will be field-dependent.
Metal-insulator transition will probably be second order?

OR ?

- A Mott insulator

Electronic layers of BETS close to a metal-insulator transition. Small changes drive the system into one or the other phase.

The charge gap Δ should be determined by U/t and weakly dependent on J. Δ should be comparable to that of λ-(BETS)$_2$Ga Br$_z$Cl$_{4-z}$ for $z > 0.8$.
Δ will be weakly field-dependent.
Metal-insulator transition is first order.

Is $\lambda-$(BETS)$_2$FeCl$_4$ a Mott insulator? What happens when Ga is replaced by Fe?

MOTT INSULATING STATE STABILISED BY J

When replacing Ga by Fe, we have to take into account energy of the local moments. $S = 5/2$ spins can be treated classically.

Comparison of the total energies of:

• **Metallic phase**: local moments interacting via RKKY interactions (energy of a classical antiferromagnet):

$$E_{Metal}(J) = E_{Metal}(J = 0) - zJ^2\chi S^2$$

where χ is the electron spin susceptibility at $Q = Q_{AF}$, z the number of magnetic bonds.

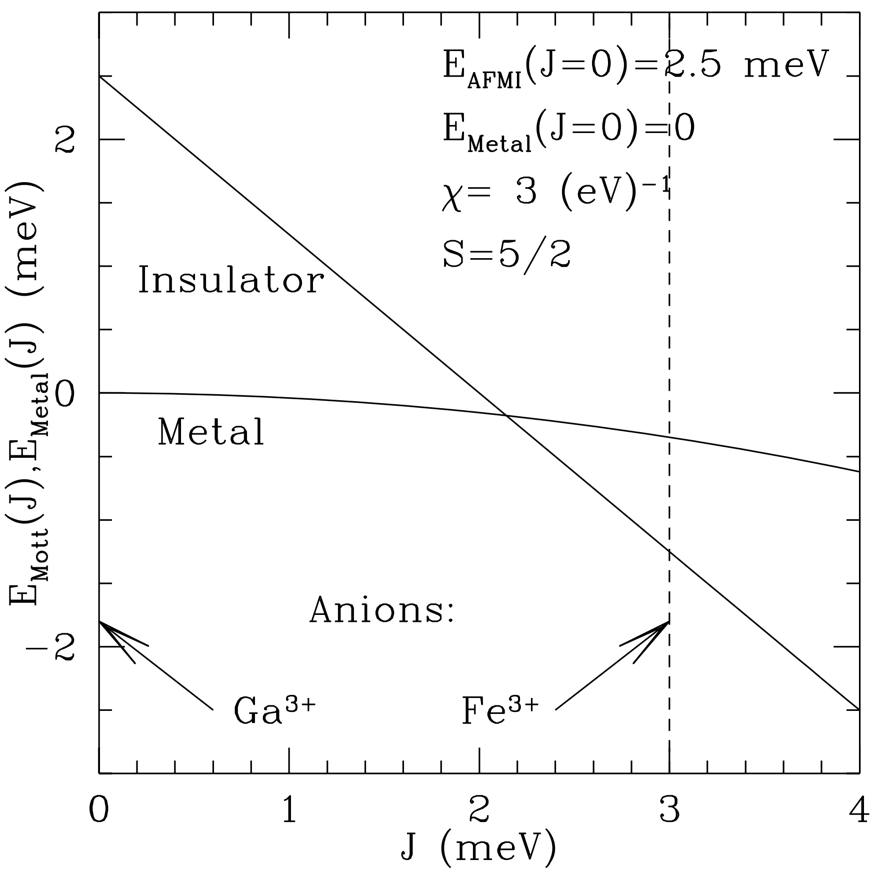
• **Antiferromagnetic Mott phase**: AF local moments + localised electrons interacting with the Kondo coupling. (Large U assumed, energy of a classical magnet with two species of spins).

$$E_{Mott} = E_{AFMI}(J = 0) - JS/2$$

λ-(BETS)$_2$GaCl$_4$ ($J = 0$) is a metal (SC): $E_{Metal}(J = 0) < E_{AFMI}(J = 0)$. The energies are close since λ-(BETS)$_2$GaBr$_z$Cl$_{4-z}$ with $z > 0.8$ is an insulator.

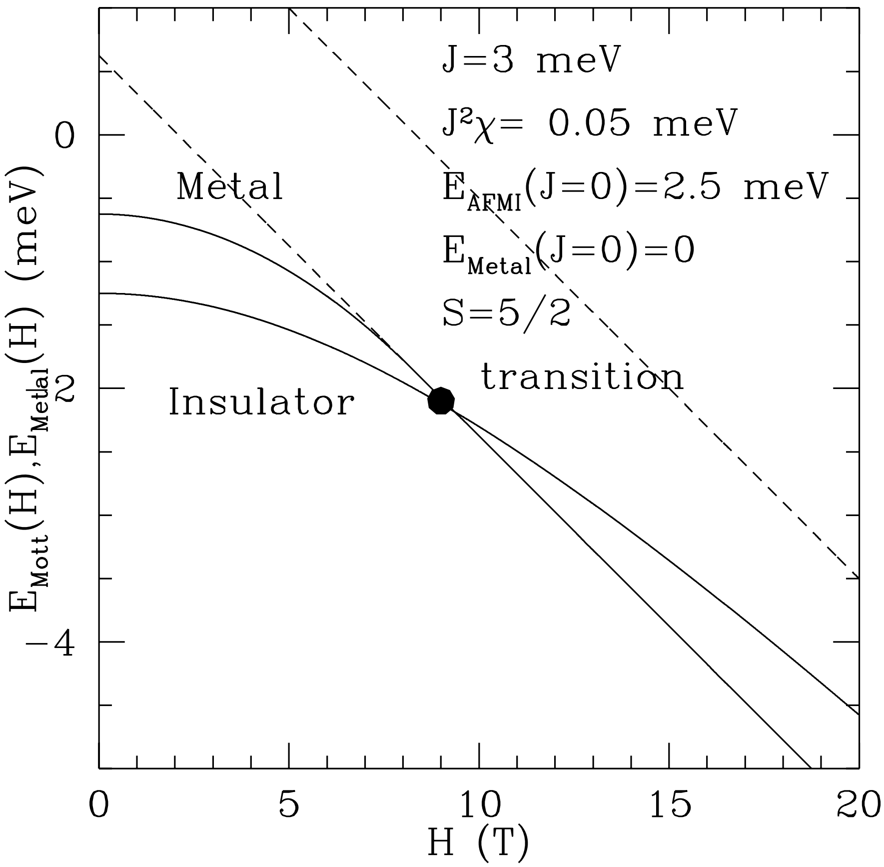
$J\chi \sim J/E_F \ll 1$.

The AF Mott phase is more stable if J is large enough and the difference in energies at $J = 0$ is small enough.



INSULATING STATE IS DESTABILISED BY A MAGNETIC FIELD

Same type of calculation: classical magnetic energies as a function of field.



MAGNETIC-FIELD INDUCED SUPERCONDUCTIVITY:

The Jaccarino-Peter effect

• For high fields all the local moment spins are aligned by the field.

• When $H \parallel$ layers the upper critical field due to orbital effects is large. $H_{c2}^{\parallel}$ is determined by Zeeman splitting.

• Effective exchange magnetic field acting on the electron spins is

$$H_{eff} = H - H_e$$

where the compensating magnetic field is $H_e = J\langle S^z \rangle / g\mu_B$

Superconductivity can occur when

$$|H - H_e| < H_P$$

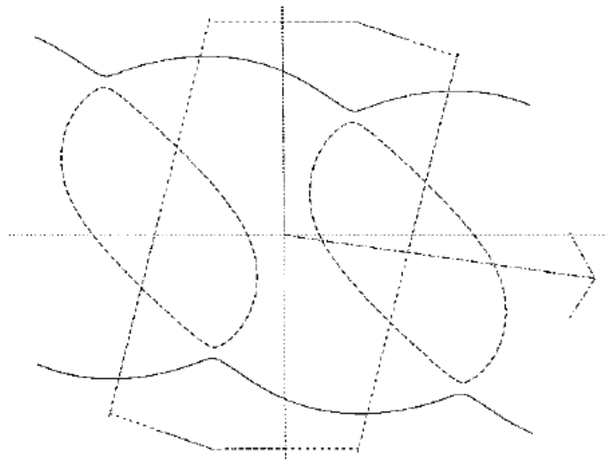
Experiment suggests
$H_e \simeq 30$ T and $H_P \simeq 10$ T.

This value of H_P is consistent with the observed $H_{c2}^{\|,Ga} = 12$ T.

To confirm the JP explaination need
• to check that $J > 0$. See superexchange estimations: (Hotta and Fukuyama).
• an independent measurement of H_e
• to show effect of $S = 5/2$ spin fluctuations is small.

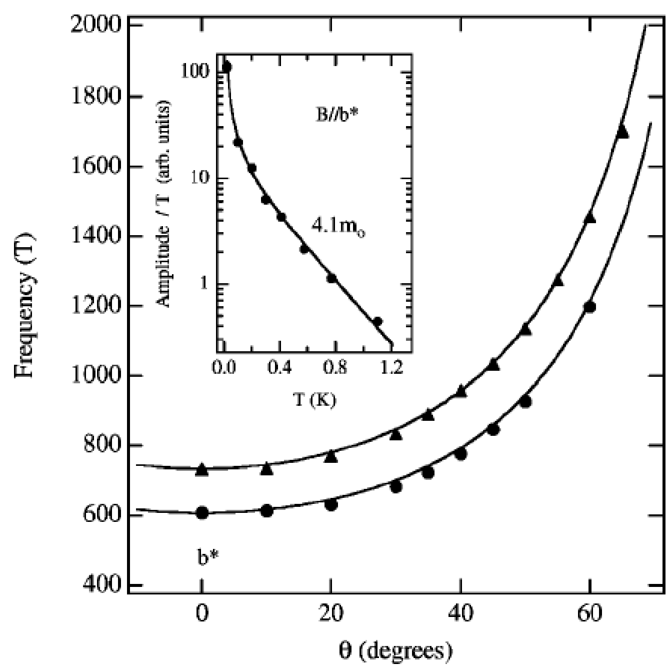
FERMI SURFACE OF λ-$(BETS)_2FeCl_4$

• Band structure calculation for a layer:



→ metallic, not insulating!

• Shubnikov-de Haas quantum oscillations show two frequencies:
S.Uji *et al.*, PRB 2001.

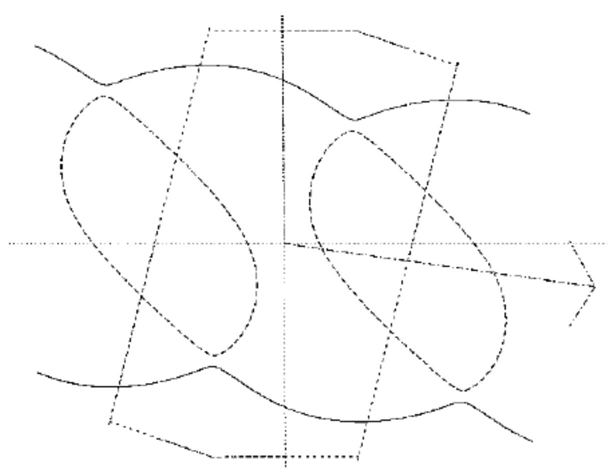


EXTRACTING H_e FROM SHUBNIKOV DE-HAAS OSCILLATIONS

• Without magnetic ions:
Oscillating part of resistance $\cos(2\pi F/B\cos\theta)$ where F is proportional to the area of the Fermi surface and θ is the angle between the field and the normal to the layers.
There is a spin-splitting factor

$$R_s = \cos(\pi S_0/2\cos\theta) \qquad S_0 = g^* m^*/m_e$$

• With magnetic ions:
Effect of H_e at high magnetic field: two Fermi surfaces for spin ↑ and spin ↓.



spin-splitting factor becomes field-dependent: $R_s = \cos[\pi S_0(H_e/H - 1)/2\cos\theta]$. This produces two SdH frequencies:

$$(F \pm S_0 H_e/4)/\cos\theta$$

From $\Delta F = 130\text{T}/\cos\theta$,and $m^* = 4.1 m_e$, $g^* = g$ the compensating field is $H_e = JS/g\mu_B = 32$ T.
This agrees with the optimal field for superconductivity!

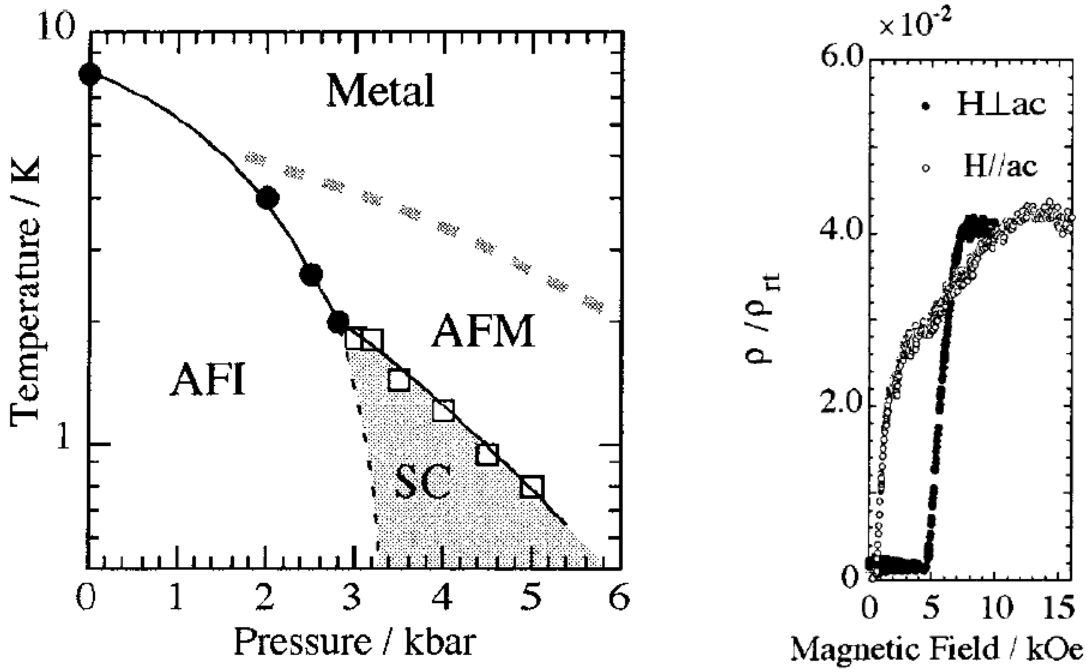
- Other interpretation for the beats?

A corrugated 3D Fermi-surface would lead to

$$\Delta F \propto J_0(\gamma \tan\theta)/\cos\theta$$

that vanishes at Yamaji's angles, contrary to what is observed.

“FRAGILE” SUPERCONDUCTIVITY IN λ-$(BETS)_2FeCl_4$ UNDER PRESSURE

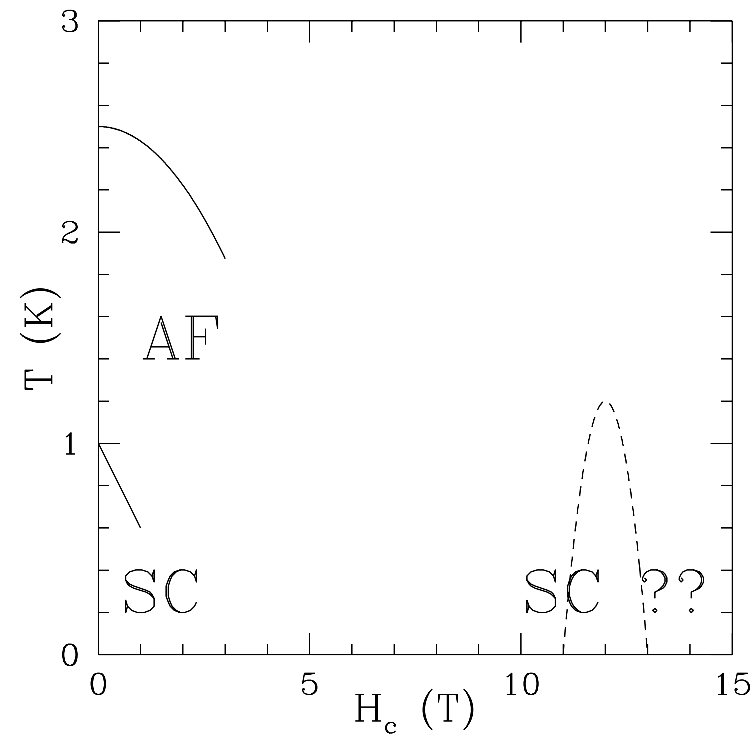


- A very small in-plane magnetic field ($H_{c2}^{\|,Fe}$ $= 0.2$ T $\ll H_{c2}^{\perp,Fe}$) destroys the superconductivity. Compare with $H_{c2}^{\|,\mathrm{Ga}}$=5 T.
- Effect of the magnetic anions?

$$|J\langle S_z\rangle - g\mu_B H_{c2}^{\|,Fe}| \simeq g\mu_B H_{c2}^{\|,Ga}$$

At small field, $\langle S_z\rangle = \chi(S=5/2)H$ with a large factor $1/J^2\chi_e$. Hence, $H_{c2}^{\|,Fe}$ is greatly reduced!

PREDICTION OF FIELD-INDUCED SUPERCONDUCTIVITY IN $\kappa-(\mathrm{BETS})_2\mathrm{FeBr}_4$



Coexistence of antiferromagnetism and superconductivity is possible because the RKKY interaction is not modified at Q_{AF}.

Shubnikov de Haas [Balicas *et al.*, Uji *et al.*] has beats with $\Delta F = 100$ T.

If $J > 0$, we then expect a *compensating* field of 12 T.

$H_{c2}^{\parallel,Ga} \sim 1-2$ T gives superconductivity for 11 T $< H <$13 T.

Conclusions

- The insulating phase in λ-$(BETS)_2X$ with X=$FeCl_4$ is an antiferromagnetic Mott insulator, similar to that seen in X=$GaBr_zCl_{4-z}$ with $z > 0.8$.
- The magnetic-field induced superconductivity is due to the Jaccarino-Peter effect. The field acting on the spins is compensated by an exchange field $H_e = JS/g\mu_B$.
- Superconductivity is the same as that found in the X=$GaCl_4$ material ($J = 0$).
- H_e leads to beating of the magnetic oscillations. Beat frequency is consistent with the optimal field for superconductivity.
- Compensating field H_e should also be measured using electron spin resonance.
- Fluctuations in the spins of the local moments have negligible effect.
- We predict field-induced superconductivity in κ-$(BETS)_2FeBr_4$ near 10 tesla.