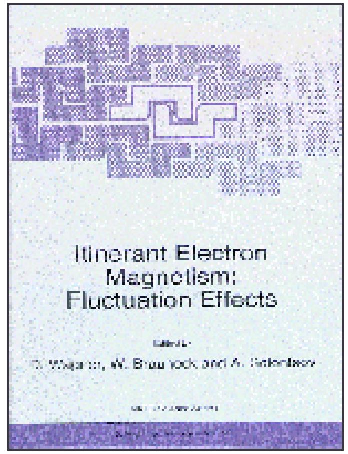
Excitations, Dynamics and Relaxation of the Electron Fermi Liquid.

Alexander Solontsov

State Center for Condensed Matter Physics,

M. Zakharova str., 6/3, 115569, Moscow, Russia,

e-mail: asolontsov@mail.ru



In these lectures I will focus on the properties of collective excitations of electrons (with the emphasis on anharmonic spin fluctuations) which at elevated temperatures have a non-linear nature and may essentially influence radiation damage of metallic materials.

- **D. Wagner, W. Brauneck, and A. Solontsov, Eds., Itinerant electron magnetism: Fluctuation Effects and Critical Phenomena (Kluwer Ac. Publ., 1998);**
- **A. Solontsov, Int. J. Modern Phys. B 19 (2005);**
- **V. Antropov and A. Solontsov, PRB 81 (2010).**

Outline

1. Effect of collective electronic excitations on the radiation damage.

- stopping power;
- energy dissipation in the electron system;
- evidence for spin fluctuations: resistivity and INS.

2. Excitations and dynamics of the electron Fermi liquid.

- individual and collective excitations;
- electron-ion dynamics;
- spectra of transverse spin and charge fluctuations.
- kinetics of electronic Fermi excitations and fluctuations.

3. Thermodynamics of anharmonic SF.

- models: paramagnons, SCR SM;
- magnetic equation of state: criterion of magnetic instability;
- magneto-volume effect and thermal expansion anomalies.

4. Non-linear magnetic dynamics and relaxation.

- Fermi liquid vs Ginzburg-Landau approaches;
- spectrum of longitudinal SF: linear vs non-linear regimes;
- scenarios of temperature dependence of the spectrum of longitudinal SF.

I. Fermi and Bose excitations of the electron Fermi liquid.

1. Effect of collective electronic excitations on the radiation damage.

Stopping power (slow ions $v_{ph} << v \le v_F$),

***J. Lindhard, 1954*):**

$$\frac{dE}{dx} \sim -\left(\frac{Z_i e}{v}\right)^2 \int_o^{|\mathbf{k}|v} \omega d\omega \sum_{\mathbf{k}} \frac{1}{|\mathbf{k}|^3} \operatorname{Im} \varepsilon^{-1}(\mathbf{k}, \omega, T), \qquad \textbf{(1)}$$

Here $-Z_i e$ and e and are the ion and electron charges, v, v_{ph} and v_F are the ion, phonon and Fermi velocities.

Inverse dielectric constant (permittivity):

$$\varepsilon^{-1}(\mathbf{k}, \omega, T) = 1 + \phi_{eff}(\mathbf{k}, \omega)\chi_e(\mathbf{k}, \omega, T), \qquad \textbf{(2)}$$

$\phi_{eff}(\mathbf{k}, \omega)$ **- effective electron-electron interaction, without lattice screening**

$$\phi_{eff}(\mathbf{k}, \omega) = 4\pi e^2 / \mathbf{k}^2.$$

Electron dynamical susceptibility $\chi_e(\mathbf{k},\omega,T)$

(linear response of the electron system to the external electrostatic potential)

defines the spectrum of CF

$$I_{cf}(k,T) = \frac{1}{\omega}\operatorname{Im}\chi_e(k) = \chi_e(\mathbf{k},T)\frac{\omega_{cf}(k)}{\omega_{cf}^2(k)+\omega^2}, \qquad \textbf{(3)}$$

$\omega_{cf}(k) = \Gamma_{e0}|\mathbf{k}|\chi_e^{-1}(\mathbf{k}) \sim |\mathbf{k}|$ **- frequency of CF,**

Γ_{e0} **- Landau relaxation rate of CF.**

STOPPING OF SLOW IONS IS AFFECTED BY CHARGE DENSITY FLUCTUATIONS OF THE ELECTRON SYSTEM

Energy dissipation in the electron system:

$$e_{\mathbf{p}}^{\sigma} \leftrightarrow e_{\mathbf{p}'}^{\sigma} + \xi \quad (\xi = \text{CF, SF, ph, sw})$$

$$\frac{dE_e}{dt} = \int d\omega \hbar\omega \Gamma(\omega)$$

(I. Koponen, PRB 47, 1993),

Boltzmann collision integral $(e \to e' + \xi)$:

$$\Gamma(\omega) \sim \sum_{\mathbf{p},\mathbf{p}',\mathbf{k}} \sum_{\sigma,\sigma',\xi} I_{col}\{n_{\mathbf{p}}^{\sigma}, n_{\mathbf{p}'}^{\sigma'}, N_k\} = \sum_{\mathbf{p},\mathbf{p}',\mathbf{k}} \sum_{\sigma,\sigma',\xi} \left|\hat{W}_{\mathbf{p},\mathbf{p}'}^{\sigma,\sigma'}(\mathbf{k},\omega,\xi)\right|^2 \operatorname{Im}\chi_{\xi}(k)\delta(\varepsilon_{\mathbf{p}}^{\sigma} - \varepsilon_{\mathbf{p}'}^{\sigma} - \hbar\omega)\times$$

$$\delta(\mathbf{p} - \mathbf{p}' - \hbar\mathbf{k})[n(1-n')N_{\omega} - (1-n)n'(N_{\omega}+1)], \qquad \textbf{(4)}$$

$n(n') = n_{\mathbf{p}}^{\sigma}(n_{\mathbf{p}'}^{\sigma'})$ **- electronic distribution function with momentum** $\mathbf{p}(\mathbf{p}')$ **and spin** $\sigma(\sigma') = \pm 1$, N_{ω} **- distribution function of fluctuations (CF, SF, ph, sw),**

$\hat{W}_{\mathbf{p},\mathbf{p}'}^{\sigma,\sigma'}(\mathbf{k},\omega,\xi)$ **- matrix element of electron-fluctuation interaction,**

$\chi_{\xi}(k)$ **- electronic, spin or lattice susceptibilities,** $k = (\mathbf{k},\omega)$.

Electron mean free path $l(T)=\tau(T)\mathrm{v}_F$ and lifetime $\tau(T)$:

$$\tau^{-1}(T)=\left\langle \frac{\delta\Gamma}{\delta n_{\mathbf{p}}^{\sigma}} \right\rangle_{\mathbf{p},\sigma} =\tau_0^{-1}(T)+\tau_{ph}^{-1}(T)+\tau_{sw}^{-1}(T)+\tau_{cf}^{-1}(T)+\tau_{sf}^{-1}(T), \quad \textbf{(5)}$$

$\tau_0^{-1}(T)$ **is due to scattering by defects and electrons.**

Electroresistivity: Matthiesen's rule:

$$\rho(T)\approx\frac{m_e}{e^2 n_e \tau(T)}=\rho_0(T)+\rho_{ph}(T)+\rho_{sw}^{-1}(T)+\rho_{cf}(T)+\rho_{sf}(T), \quad \textbf{(6)}$$

where $\rho_{0,\xi}(T)\sim\tau_{0,\xi}^{-1}(T)$, m_e **and** n_e **are the electron mass and density**

$$\rho_\xi(T)\sim\frac{1}{T}\sum_{\mathbf{k}}|\mathbf{k}|\int_0^\infty \omega d\omega\,\mathrm{Im}\,\chi_\xi(\mathbf{k},\omega)N_\omega(N_\omega+1), (\xi=\textbf{CF, SF, ph, sw}) \quad \textbf{(7)}$$

Experimental evidence for spin fluctuations.

Spin fluctuation resistivity.

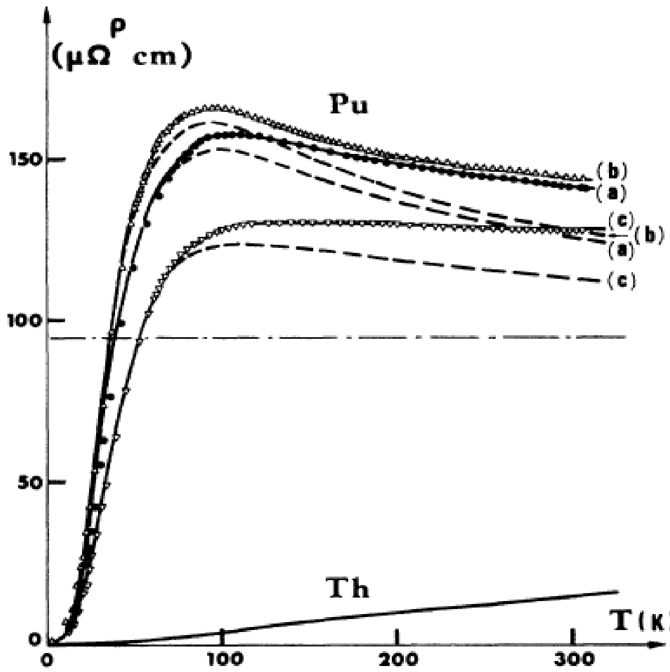


Fig. 1. Temperature dependence of resistivity of $\alpha - Pu$: a) polycrystals , b) and c) monocrystals with current along (010) and (100), dashed lines – calculated SF contributions, solid lines – theoretical curves with addition of the *Th* contribution, triangles and dots – experimental data (R. Jullien et al, PRB 9,(1974)).
Similar results: actinides (Pu,U)Al_2, Np, UN, transition metals.

IN ACTINIDES AND TRANSITION METALS SYSTEMS WITH MAGNETIC INSTABILITIES KINETIC PHENOMENA MAY BE DOMINATED BY ELECTRON SCATTERING ON SPIN FLUCTUATIONS

Inelastic neutron magnetic scattering.

Cross-section:

$$\frac{d^2\sigma}{d\Omega dE} = F_{t,l}(\hat{\mathbf{k}}) I_{t,l}(k,T)(N_\omega + 1) \qquad \textbf{(8)}$$

Intensity of transverse (*t*) and longitudinal (*l*) SF

$$I_{t,l}(k,T) = \frac{1}{\omega} \operatorname{Im} \chi_{t,l}(k) \qquad \textbf{(9)}$$

Quasielastic SF: central Lorenz peak:

$$I_l(k,T) = \frac{1}{\omega} \operatorname{Im} \chi_l(k) = \chi_l(\mathbf{k},T) \frac{\omega_{sf}(k,T)}{\omega_{sf}(k,T) + \omega^2}, \qquad \textbf{(10)}$$

$\omega_{sf}(k) = \Gamma_0 |\mathbf{k}| \chi_l^{-1}(\mathbf{k}) \sim |\mathbf{k}|$ **- characteristic frequency of l-SF,**

$\Gamma_0 \sim \chi_P \nu_F$ **- Landau relaxation rate of SF,** ν_F **- electronic density of states at the Fermi surface.**

Quasielastic l-SF in heavy fermion compound *UPt_3*

(N. Bernhoeft and G. Lonzarich, J. Phys. Cond Mat. 7 (1995).

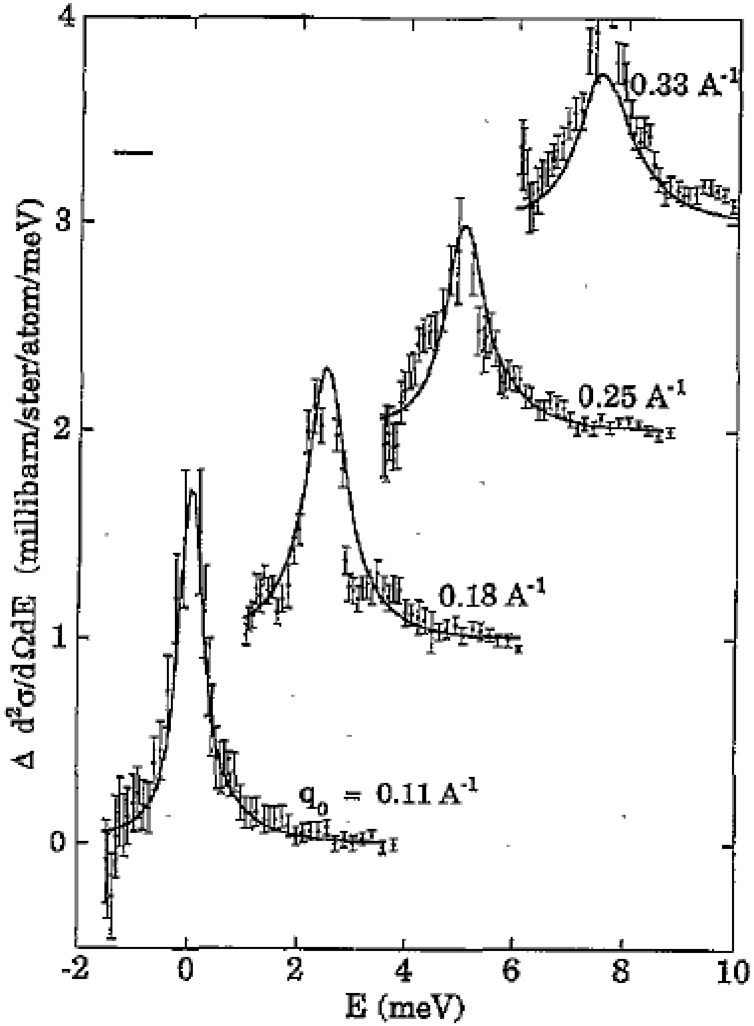


Fig.2. Inelastic neutron scattering cross-section in UPt_3 at 10K as a function of the energy transfer $E = \hbar\omega$ and scattering wave-vectors 0.11, 0.18, 0.25 and 0.33A^{-1}. The origins are shifted by 2.5 mev in the abscissa and 1 unit in ordinate.

Quasielastic l-SF in itinerant antiferromagnets *UN* (perspective nuclear fuel)

(T. Holden et al, PRB 30 (1984)).

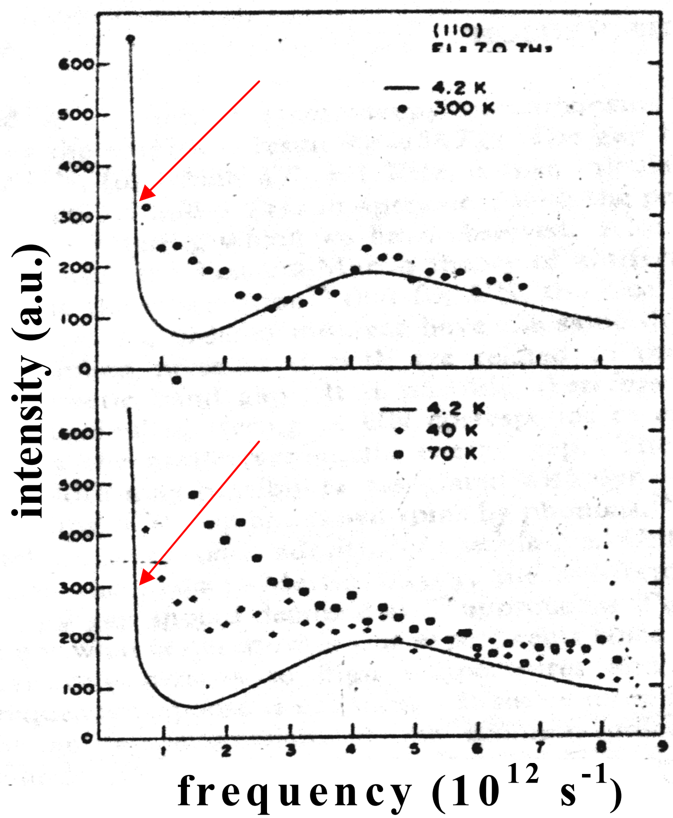


Fig.3. Temperature dependence of the magnetic inelastic neutron scattering in antiferromagnetic *UN* (T_N=53K) near the reciprocal-lattice point (110) at temperatures 4.2, 40, 70 and 300K.

The scattering is dominated by longitudinal quasielastic SF which become giant as the temperature is raised. The temperature dependence of SF cannot be accounted for by the temperature dependence of the static magnetic susceptibility.

Similar features of the inelastic neutron scattering were observed in the antiferromagnetic metallic uranium phosphide *UP* and arsenide *UAs* *(P. Erdos and J. Robinson, 1983)* and transition metal compounds *(T. Moriya, 1985).*

SPIN FLUCTUATIONS IN ACTINIDE AND TRANSITION METAL COMPOUNDS ARE DIRECTLY OBSERVED BY INELASTIC NEUTRON SCATTERING AND SHOULD PLAY AN IMPORTANT ROLE IN THERMODYNAMICS AND NON-LINEAR MAGNETIC DYNAMICS

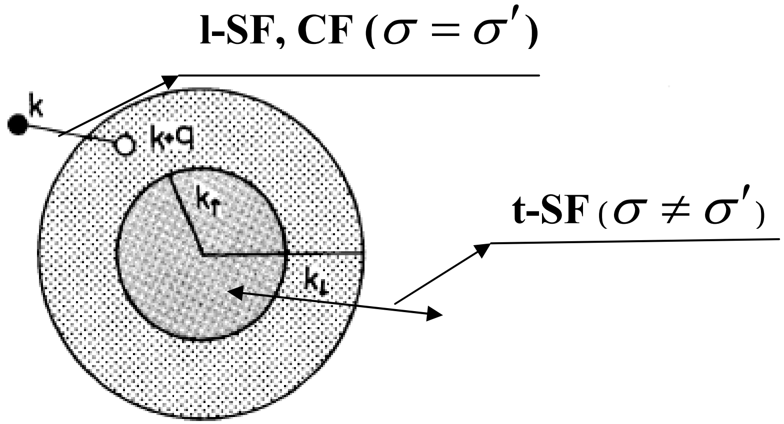
2. Excitations and dynamics of the electron Fermi liquid.

Electronic correlations $\frac{\varepsilon_{int}}{\varepsilon_{kin}} \sim \frac{e^2}{\hbar v_F} \sim 1 \Rightarrow$ **NO ~~HFA~~, ~~RPA~~, etc.**

Electronic correlations: $\Rightarrow$ **electron Fermi liquid.**

Individual and collective excitations.

Fig.4. Fermi spheres and electron-hole pair excitations.

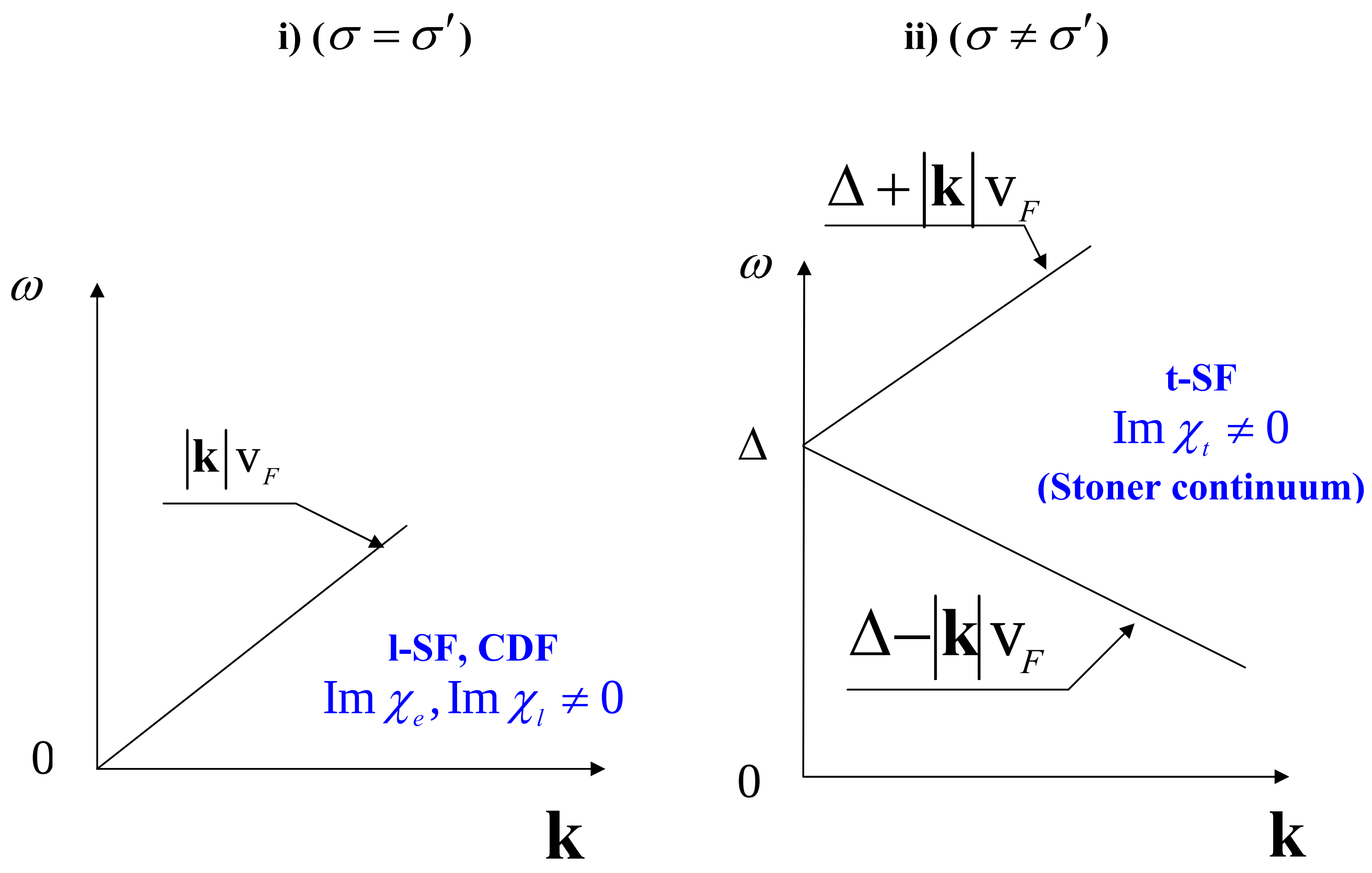


$e^{\sigma}_{\mathbf{p}+\hbar\mathbf{k}} \rightarrow e^{\sigma'}_{\mathbf{p}}$

Individual Fermi excitations - quasiparticles

Collective Bose excitations in the electron-hole

Fig.5. Electron-hole continua



Concept of the electron Fermi liquid

(L.D. Landau 1956, V.P. Silin 1957)

Quasiparticles: Fermi excitations described by the density matrix $\hat{\rho}$:

$$\left\langle \sigma, \mathbf{p} + \hbar\mathbf{k}/2 \middle| \hat{\rho}(\omega) \middle| \sigma', \mathbf{p} - \hbar\mathbf{k}/2 \right\rangle = \delta_{k,0}\delta_{\sigma\sigma'} n^{\sigma}(\mathbf{p}) + \delta\rho_{\mathbf{p}}^{\sigma\sigma'}(k) \qquad \mathbf{(11)}$$

Effective Hamiltonian $\hat{\varepsilon}$:

$$\left\langle \sigma, \mathbf{p} + \frac{\hbar\mathbf{k}}{2} \middle| \hat{\varepsilon}(\omega) \middle| \sigma', \mathbf{p} - \frac{\hbar\mathbf{k}}{2} \right\rangle = \delta_{\sigma\sigma'}\delta_{k,0}\varepsilon_{\mathbf{p}}^{\sigma} + \delta\varepsilon_{\mathbf{p}}^{\sigma\sigma'}(k) \qquad \mathbf{(12)}$$

$$\delta\varepsilon_{\mathbf{p}}^{\sigma\sigma'}(\mathbf{k},t) = \delta_{\sigma\sigma'}\left[\phi(\mathbf{k})n(k) + N_i\Lambda(\mathbf{k})(\mathbf{k},\mathbf{u}(k))\right] + \left(\hat{\boldsymbol{\sigma}}\right)_{\sigma\sigma'}\psi(\mathbf{k})\mathbf{m}(k),$$

$$2\sum_{\mathbf{p}}\delta\rho_{\mathbf{p}}^{\sigma\sigma'}(k) = \delta_{\sigma\sigma'}n(k) + (\hat{\boldsymbol{\sigma}})_{\sigma\sigma'}\mathbf{m}(k), \qquad \mathbf{(13)}$$

$$n(k) = \sum_{\mathbf{p}}\delta\hat{\rho}_{\mathbf{p}}(k) \text{ and } \mathbf{m}(k) = Sp\sum_{\mathbf{p}}\hat{\boldsymbol{\sigma}}\delta\hat{\rho}_{\mathbf{p}}(k)$$

are amplitudes of CF and SF, $\mathbf{u}(k)$ are lattice displacements,

$\psi(\mathbf{k})$**,** $\phi(\mathbf{k}) = 4\pi e^2/\mathbf{k}^2 + \varphi(\mathbf{k})$ **and** $\Lambda(\mathbf{k}) = 4\pi Ze^2/\mathbf{k}^2 + \lambda(\mathbf{k})$

describe the exchange interaction, long-range Coulomb and short-range electron-electron and electron-ion interactions, $-Ze$ is the charge of ions of the lattice,

$\lambda(\mathbf{k}) = const + O(\mathbf{k}^2)$ **- deformation potential.**

Electron-ion dynamics.

Liouville dynamical equation for density matrix:

$$\frac{d\hat{\rho}}{dt} = \frac{i}{\hbar}(\hat{\rho}\hat{\varepsilon} - \hat{\varepsilon}\hat{\rho}), \qquad \textbf{(13)}$$

Dynamics of the crystal lattice (longitudinal oscillations):

$$M[\omega^2 - D_i(\mathbf{k})]\mathbf{u}(k) = -\mathbf{F}_e(k), \qquad \textbf{(14)}$$

$D_i(\mathbf{k}) \approx \omega_p^2 + c_i^2\mathbf{k}^2$ **- dynamical matrix of the crystal without effects of electrons,** $\omega_p \sim \omega_D$ **and** M **- ion plasma and Debye frequencies and mass of ions of the lattice,** $\mathbf{F}_e(k) = 2i\Lambda(\mathbf{k})$ **- amplitude of the electronic force imposed on ions**

Dynamical screening of the electron-electron interaction:

$$\phi(\mathbf{k}) \rightarrow \phi_{eff}(k) = \phi(\mathbf{k}) + \frac{\omega_p^2}{\omega^2 - \omega_p^2 - c_i^2 k^2} \frac{\Lambda(\mathbf{k})}{Z} \approx \qquad \textbf{(15)}$$

$$\begin{cases} (B - B_e)/B_e \nu_F = const(\mathbf{k}), \omega << \omega_p \text{(adiabatic screening by lattice)}, \\ \phi(\mathbf{k}), \omega >> \omega_p \text{ (no screening)}, \end{cases}$$

where $B = B_e(1 + \phi_{eff}(0)\nu_F)$ **- bulk modulus,** $B_e = Z^2 N^2 / \nu_F$ **- electronic contribution,** N **- density of ions of the lattice,** ν_F **- electronic density of states at the Fermi surface.**

Criterion for the isostructural lattice instability
(analogue of the Stoner criterion of itinerant magnetism)
($\gamma \rightarrow \alpha$ transition in Ce compounds)

$$1 + \phi_{eff}(0)\nu_F < 0 \qquad \textbf{(16)}$$

Dynamical susceptibilities.

$$\chi_e(k) = \frac{\chi_e(k)_{\phi_{eff}=0}}{1+\phi_{eff}(k)\chi_e(k)_{\phi_{eff}=0}}, \; \chi_{t,l}(k) = \frac{\chi_{t,l}(k)_{\psi=0}}{1+\psi(\mathbf{k})\chi_{t,l}(k)_{\psi=0}} \qquad \textbf{(17)}$$

i) Relaxational regime inside electron-hole continua, limit $\omega \to 0, \mathbf{k} \to 0$:

$$\chi_\xi^{-1}(k) \approx \chi_\xi^{-1}(\mathbf{k}) - i\frac{\omega}{\Gamma_{\xi 0}(\mathbf{k})} \; (\xi = e,t,l), \qquad \textbf{(18)}$$

$\Gamma_{\xi 0}(\mathbf{k}) \sim |\mathbf{k}|$ **- linear relaxation rate due to Landau damping in the electron-hole continua.**

ii) Spin-wave regime outside the Stoner continuum.

Magnons in ferro- (FM) and antiferromagtets (AFM):

$$\chi_t(k) = \chi_t(\mathbf{k}) \begin{cases} \dfrac{\omega_m(\mathbf{k})}{\omega_m(\mathbf{k}) - \omega - \tau_m^{-1}}, & \text{FM}, \\[2ex] \dfrac{\omega_m^2(\mathbf{k})}{\omega_m^2(\mathbf{k}) - \omega^2 - 2i\omega\tau_m^{-1}}, & \text{AFM}, \end{cases} \qquad \textbf{(19)}$$

$\omega_m(\mathbf{k}) \sim \mathbf{k}^2$ $(\sim|\mathbf{k}|)$ **- magnon frequency in FM (AFM),** $\chi_t(\mathbf{k}) \sim \mathbf{k}^{-2}$.

Spectra of of spin and charge fluctuations.

$$I_\xi(k,T)=\frac{1}{\omega}\operatorname{Im}\chi_\xi(k),\ (\xi=e,t,l) \qquad (20)$$

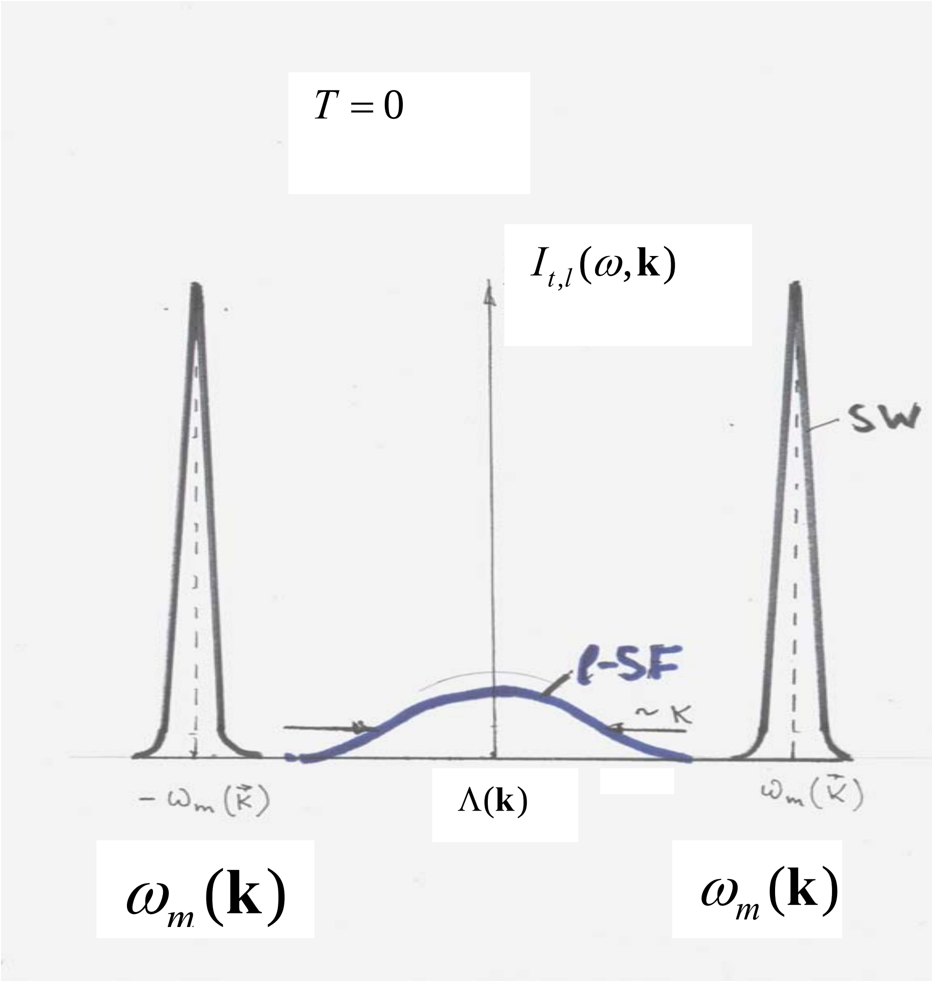


Fig.5. Ground state ($T=0$) intensities of the spin-wave and l-SF spectra $I_{t,l}(\omega,\mathbf{k})$, where $\Lambda(\mathbf{k})=\omega_{sf}(\mathbf{k})=\Gamma_0(k)\chi_l^{-1}(\mathbf{k})$ is the HWHM of the quasielastic central peak defined by the linear Landau relaxation mechanism.

Charge fluctuations: l-SF $\rightarrow$ CDF, magnons $\rightarrow$ phonons.

Local magnetic moments or average amplitude of SF.

$$M_L^2 = \langle \mathbf{m}^2(\mathbf{r},t) \rangle = \sum_{\nu=t,l} \sum_{\mathbf{k},\omega} (m_\nu^2)_k =$$

(21)

$$4\hbar \sum_{\nu=t,l} \sum_{\mathbf{k},\omega} \operatorname{Im} \chi_\nu(\mathbf{k},\omega) \left(N_\omega + \frac{1}{2} \right) \equiv \left(M_L^2 \right)_{Z.P.} + \left(M_L^2 \right)_T$$

(fluctuation-dissipation theorem, *Callen, Welton, 1952*),

factors $N_\omega = \left[\exp\left(\hbar\omega / k_B T \right) - 1 \right]^{-1}$ **and 1/2 are related to thermal** $(M_L^2)_T$**, and zero-point and** $(M_L^2)_{Z.P.}$ **contributions,** $(m_\nu^2)_k$ **- spectral density of SF.**

Kinetics of electronic Fermi excitations and fluctuations.

Separation of individual and collective variables:

$$\delta\hat{\rho} = \delta\hat{\rho}^{(ind)} + \delta\hat{\rho}^{(fl)},$$

$$\delta\hat{\rho}_{\mathbf{p}}^{(fl)}(k) = F_{cf}(\mathbf{p})n(k) + F_{sf}(\mathbf{p})\hat{\boldsymbol{\sigma}}\mathbf{m}(k) \qquad \textbf{(22)}$$

$$\sum_{\mathbf{p}} \delta\hat{\rho}_{\mathbf{p}}^{(ind)}(k) = 0, \ \sum_{\mathbf{p}} F_{cf}(\mathbf{p}) = \sum_{\mathbf{p}} F_{sf}(\mathbf{p}) = 1$$

SF: Slow time and spatial variation of spectral densities of SF

(averaging out fast variations)

$$(m_\nu^2)_k \to (m_\nu^2(\mathbf{r},t))_k = 4\hbar \operatorname{Im}\chi_\nu(\mathbf{k},\omega)\left(N_k(\mathbf{r},t) + \frac{1}{2}\right) \qquad \textbf{(23)}$$

$N_k(\mathbf{r},t)$ **- non-equilibrium distribution of SF.**

Variation of electronic distribution

$$n_{\mathbf{p}}^{\sigma\sigma'}(\mathbf{r},t) = \delta_{\sigma\sigma'} n_{\mathbf{p}}^{\sigma} + (\delta\rho_{\mathbf{p}}^{\sigma\sigma'}(\mathbf{r},t))^{(ind)}$$

(usually assumed $(\delta\rho_{\mathbf{p}}^{\sigma\sigma'})^{(e)} \approx \delta_{\sigma\sigma'}(\delta\rho_{\mathbf{p}}^{\sigma\sigma})^{(e)}$**)**

Kinetic equations

$$\frac{dn_{\mathbf{p}}^{\sigma}}{dt} = \sum_{\mathbf{p}',\sigma',k,\nu=t,l} I_{col}\{n_{\mathbf{p}}^{\sigma}, n_{\mathbf{p}'}^{\sigma'}, N_k, \nu\},$$

$$\frac{d(m_{\nu}^{2})_k}{dt} \sim \sum_{\mathbf{p},\mathbf{p}',\sigma,\sigma',k} I_{col}\{n_{\mathbf{p}}^{\sigma}, n_{\mathbf{p}'}^{\sigma'}, N_k, \nu\} + \text{(mode-mode coupling)}$$

(24)

$$I_{col}\{n_{\mathbf{p}}^{\sigma}, n_{\mathbf{p}'}^{\sigma'}, N_k,\ \nu\} = \left|\hat{W}_{\mathbf{p},\mathbf{p}'}^{\sigma,\sigma'}(k,\nu)\right|^2 \operatorname{Im}\chi_{\nu}(k,T)\,\delta(\varepsilon_{\mathbf{p}}^{\sigma} - \varepsilon_{\mathbf{p}'}^{\sigma} - \hbar\omega)\,\delta(\mathbf{p} - \mathbf{p}' - \hbar\mathbf{k})\times$$
$$[n_{\mathbf{p}}^{\sigma}(1 - n_{\mathbf{p}'}^{\sigma'})N_k - (1 - n_{\mathbf{p}'}^{\sigma'})n_{\mathbf{p}}^{\sigma}(N_k + 1)]$$

SF spectrum $\operatorname{Im}\chi_{\nu}(k,T)$ accounts for mode-mode coupling effects in thermodynamics and magnetic dynamics

Mechanisms of the temperature dependence of the SF spectrum.

$$\chi_\nu(\mathbf{k},\omega) = \chi_\nu(\mathbf{k},\omega,T)$$

Temperature dependence of the SF spectrum

Thermodynamics:

$$\chi_{t,l}(k=0) = \chi_{t,l}(T)$$

Non-linear magnetic dynamics and relaxation:

$$\Gamma_{t,l0}(\mathbf{k}) \rightarrow \Gamma_{t,l}(k,T)$$

3. Thermodynamics of anharmonic SF.

Effective Hamiltonian and free energy of the electron Fermi liquid.

$$\hat{H} = \sum_{\mathbf{p},\sigma} \varepsilon_{\mathbf{p}}^{\sigma} n_{\mathbf{p}}^{\sigma} + \frac{1}{2}\sum_{\mathbf{k}} \psi(\mathbf{k})|\mathbf{m}(\mathbf{k})|^2 + \frac{1}{2}\sum_{\mathbf{k}} \hat{\phi}_{eff}(\mathbf{k})|n(\mathbf{k})|^2 \qquad \textbf{(25)}$$

Free energy (per unit volume)

$$F(M,T) = F_e(M,T) + \Delta F,$$

M **- magnetic order parameter: (sublattice) magnetization,**

$$F_e(M,T) = F_0(T) + \frac{1}{2\chi_0}M^2 + \frac{\gamma_0}{4}M^4,$$

$$\frac{\partial \Delta F}{\partial \psi} = \frac{1}{2} M_L^2\{\hat{\chi}(\psi,k,T)\} \Rightarrow \qquad \textbf{(26)}$$

$$\Delta F = \frac{1}{2}\int d\psi M_L^2 = 2\hbar \sum_{\nu} \sum_{\mathbf{k},\omega>0} \left(N_\omega + \frac{1}{2} \right) \int d\psi \,\mathrm{Im}\, \chi_\nu(\mathbf{k},\omega),$$

$\chi_0 = \nu_F / (1 + \psi \nu_F)$ **- static paramagnetic susceptibility,**

$\gamma_0 = \gamma_0\{\nu_F\}$ **- coupling constant.**

Models: Theory of paramagnons (*1960's, see T. Moriya, 1985*)

$$\chi_\nu(\mathbf{k},\omega) = [\chi_\nu(\mathbf{k},\omega)]_{RPA} = \frac{(\chi_\nu)_{\psi=0}}{1+\psi(\chi_\nu)_{\psi=0}}$$

Landau relaxation:

$$[\chi_\nu^{-1}(\mathbf{k},\omega)]_{RPA} = \chi_\nu^{-1}(\mathbf{k})]_{RPA} - i\frac{\omega}{\Gamma_0(\mathbf{k})}.$$

Free energy of quasielastic paramagnons:

$$\Delta F = 2\sum_\nu \sum_{\mathbf{\kappa},\omega} F_0(\omega) \frac{[\omega_{SF}^{(\nu)}(\mathbf{k})]_{RPA}}{\left[\omega_{sf}^{(\nu)}(\mathbf{k})\right]_{RPA}^2 + \omega^2} \equiv \sum_\nu \Delta F_{RPA}\left\{[\chi_\nu(\mathbf{Q},\omega)]_{RPA}\right\}, \quad \textbf{(27)}$$

$$[\omega_{sf}^{(\nu)}(k)]_{RPA} = \Gamma_0 |\mathbf{k}| [\chi_\nu^{-1}(\mathbf{k})]_{RPA}$$

Drawbacks:

$[\chi_\nu(\mathbf{k},\omega,T)]_{RPA}$: **too weak T - dependence due to Fermi excitations**

(SF contributions neglected).

SCR theory of SF *(T. Moriya 1973-1985, G. Lonzarich 1984-1985)*

(analogue of the theory of anharmonic crystals, see A.D. Bruce and R.A. Cowley 1981)

$$[\chi_\nu(\mathbf{k}=0)]_{RPA} \to \chi_\nu,$$

$$[\omega_{sf}^{(\nu)}(k)]_{RPA} \to \omega_{sf}^{(\nu)}(k) = \Gamma_0(\mathbf{k})\chi_\nu^{-1}(\mathbf{k}) \qquad \textbf{(28)}$$

$$\chi_t^{-1} = (1/M)(\partial F/\partial M),\ \chi_l^{-1} = \partial^2 F/\partial M^2 \ .$$

Magnetic equation of state *(G. Lonzarich 1984)*

$$\frac{B}{M} = \chi_0^{-1} + \gamma_0[M^2 + \frac{5}{3}M_L^2(T)] \qquad \textbf{(29)}$$

SCR approximations *(A. Solontsov 1993):*

Weak spin anharmonicity

$$g_{sf} << 1,$$

$$g_{sf} = \frac{4}{3}\gamma_0 \hbar \sum_\nu \sum_{\mathbf{k},\omega} \mathrm{Im}[\chi_\nu(\mathbf{Q},\omega)\chi_\nu(\mathbf{Q},\omega)]\left(N_\omega + \frac{1}{2}\right) = \frac{\gamma_0}{3}\sum_\nu \left|\frac{\partial M_L^2}{\partial(\chi_\nu^{-1})}\right| = g_{Z.P.} + g_T,$$

$$g_{Z.P.} \sim (M_L^2)_{Z.P.}/\mu_B^2 N_e^2 << 1,\ g_T \sim \sqrt{\tau_G T_C / |T - T_C|} << 1$$

(Ginzburg-Levanyuk criterion, T_c is the critical (Curie or Neel) temperature).

Actinides and transition metals:

$$(M_L^2)_{Z.P.} \sim \mu_B^2 N^2$$ **(GIANT zero-point SF),**

$$g_{Z.P.} \sim 1,$$ **(strong spin anharmonicity)**

Soft-mode theory of SF:

(A. Solontsov, D. Wagner, PRB 51 (1995))

$$\chi_0^{-1} \to \chi^{-1} = (1-5g_{Z.P.})\chi_0^{-1} + \frac{5}{3}(M_L^2)_{Z.P.}, \qquad \textbf{(30)}$$

$$\gamma_0 \to \gamma\{g_{Z.P.}\}$$

Criterion of magnetic instability and local magnetic moments

$$S_0^{-1} = 1 + \psi\nu_F \to S^{-1} = (1-5g_{Z.P.})(1+\psi\nu_F) + \frac{5}{3}(M_L^2)_{Z.P.} < 0$$

(31)

$$(S_0^{-1})_{loc} = 1 + \psi(\nu_F)_{loc} \to (S^{-1})_{loc} = (1-5g_{Z.P.})[1+\psi(\nu_F)_{loc}] + \frac{5}{3}[(M_L^2)_{Z.P.}]_{loc} < 0$$

Criteria for the magnetic order and local magnetic moments in Pu.

(V. Antropov, A. Solontsov, PRB 109 (2011))

	α-Pu	β-Pu
S_0^{-1}	**-0.8**	**-0.9**
S^{-1}	**-0.4**	**-0.5**
$(S_0^{-1})_{loc}$	**-0.5**	**-0.7**
$(S^{-1})_{loc}$	**0.02**	**-0.2**

Magneto-volume effect and thermal expansion anomalies.

$$(\frac{\Delta V}{V})_m = \frac{C_0}{B}(M_L^2)_{tot} = \frac{C}{B}(M_L^2)_T, \quad (M_L^2)_{tot} = M^2 + (M_L^2)_{Z.P.} + (M_L^2)_T, \qquad \textbf{(32)}$$

$C_0 = -(1/2)\partial\chi_0^{-1}/\partial \ln V,$ $C = -(1/2)\partial\chi^{-1}/\partial \ln V = \Gamma_m / 2\chi$ **and** Γ_m **-**

magnetoelastic and Gruneisen constants

Temperature dependence of local magnetic moments

(A. Solontsov et al, Kluwer, 1998)

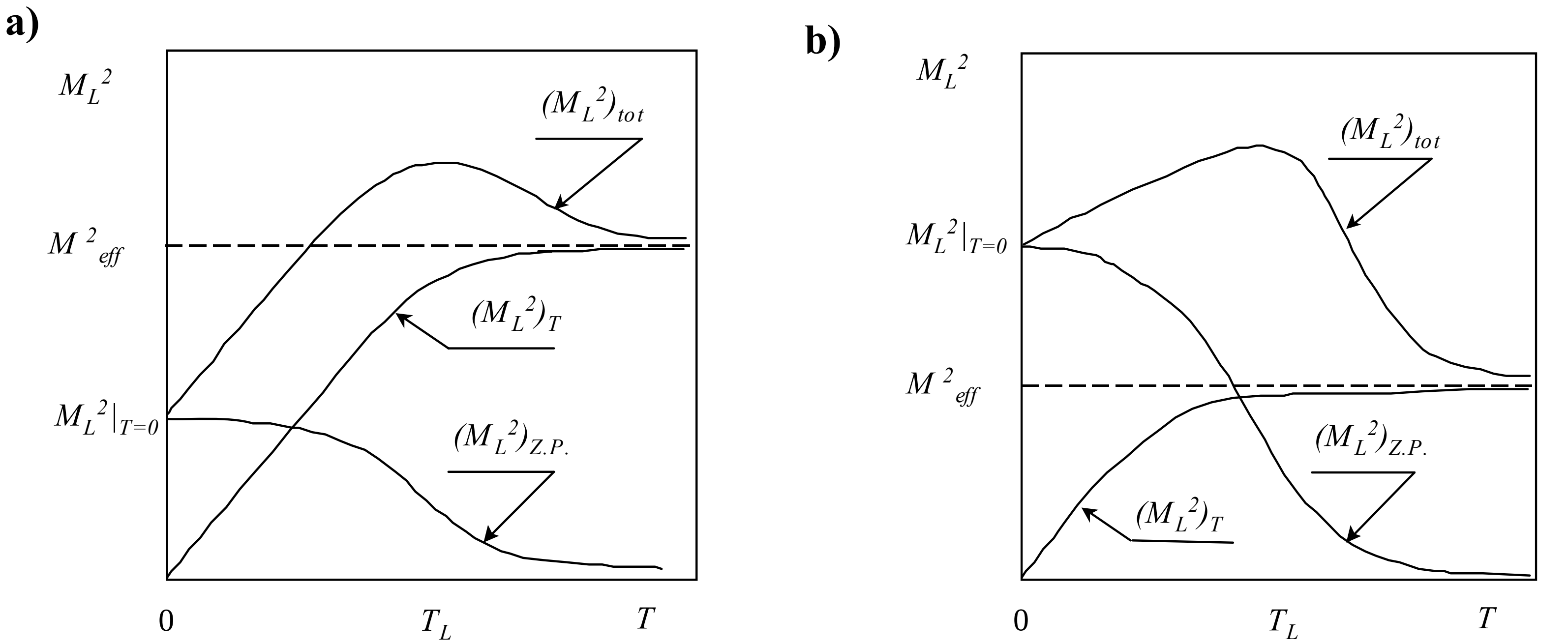


Fig.6. Temperature dependencies of the total squared local magnetic moments $(M_L^2)_{tot}$ and zero-point and thermal contributions to it, $(M_L^2)_{Z.P.}$ and $(M_L^2)_T$ for paramagnets with a) $M_{eff}^2/(M_L^2)_{tot}|_{T=0}>1$ and b) $M_{eff}^2/(M_L^2)_{tot}|_{T=0}<1$. M_{eff}^2 and T_L indicate the Curie-Weiss (CW) saturated moment and the temperature of a crossover to the CW regime.

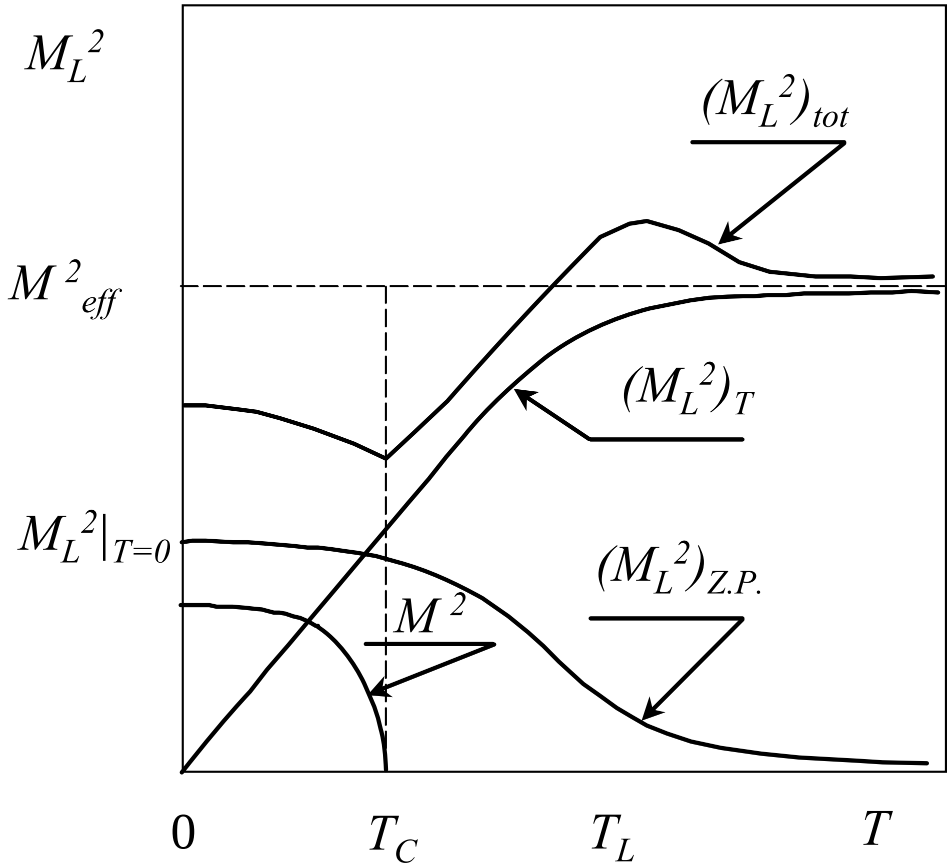


Fig. 7. Temperature dependencies of the total squared local magnetic moments $(M_L^2)_{tot}$, $(M_L^2)_{Z.P.}$ and $(M_L^2)_T$ for ferro- or antiferromagnets with $M_{eff}^2/(M_L^2)_{tot}|_{T=0} > 1$.

Invar phenomenon

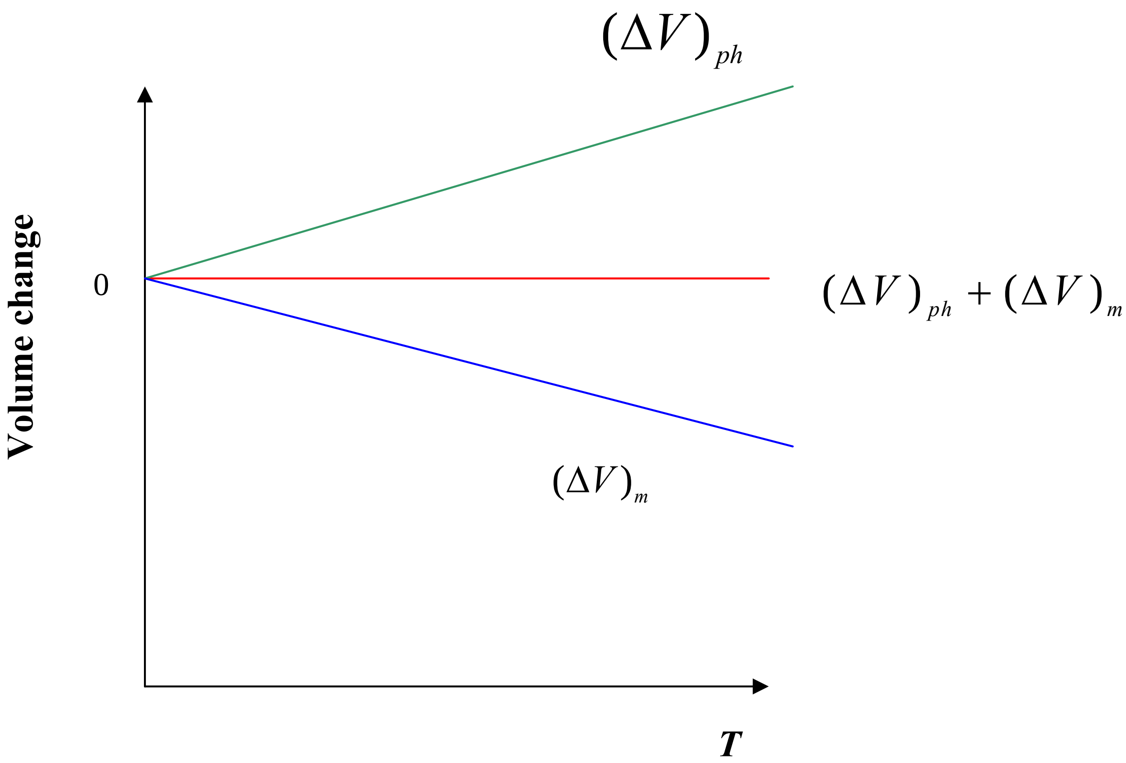


Fig.9. Invar effect in ferro- and antiferro-magnets with positive Gruneisen constants $\Gamma_m > 0$

Invar alloys

Alloy	T_C, K	$\chi^{-1}\cdot 10^3$	$\gamma\cdot 10^{-2}$, Гс	Γ_m	*B*, Mbar
$Fe_{0,70}Ni_{0,30}$	334	-0,986	17	64,8	1,14
$Fe_{0,65}Ni_{0,35}$	495	-3,14	17	19	1,25
$Fe_{0,60}Ni_{0,40}$	570	-6,8	17	6,8	1,30
$Fe_{0,72}Pt_{0,28}$	448	-5,00	0,22	4,4	1,82

Anomalous thermal expansion of *δ-Pu* ($\Gamma_m < 0$).

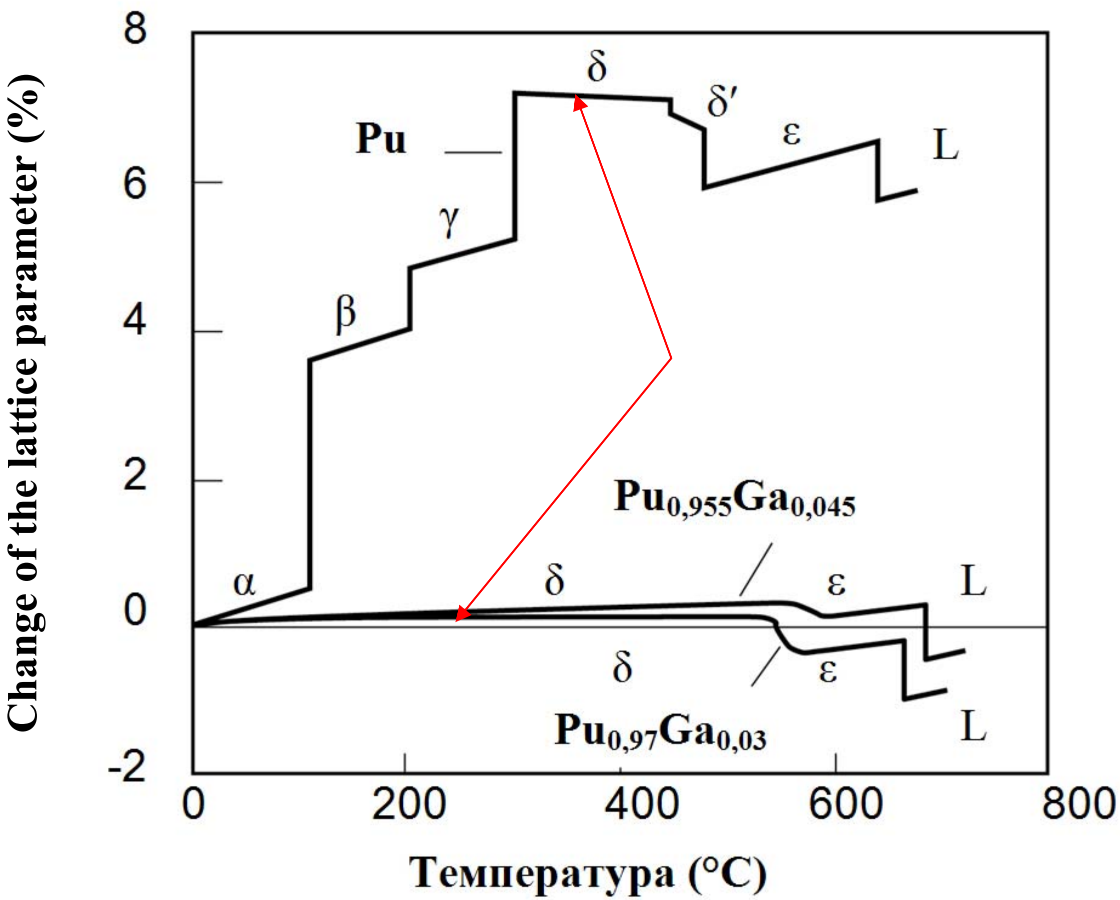


Fig.8. Change of the lattice parameter of *Pu* and *Pu-Ga* alloys at the phase transitions.

a) **b)**

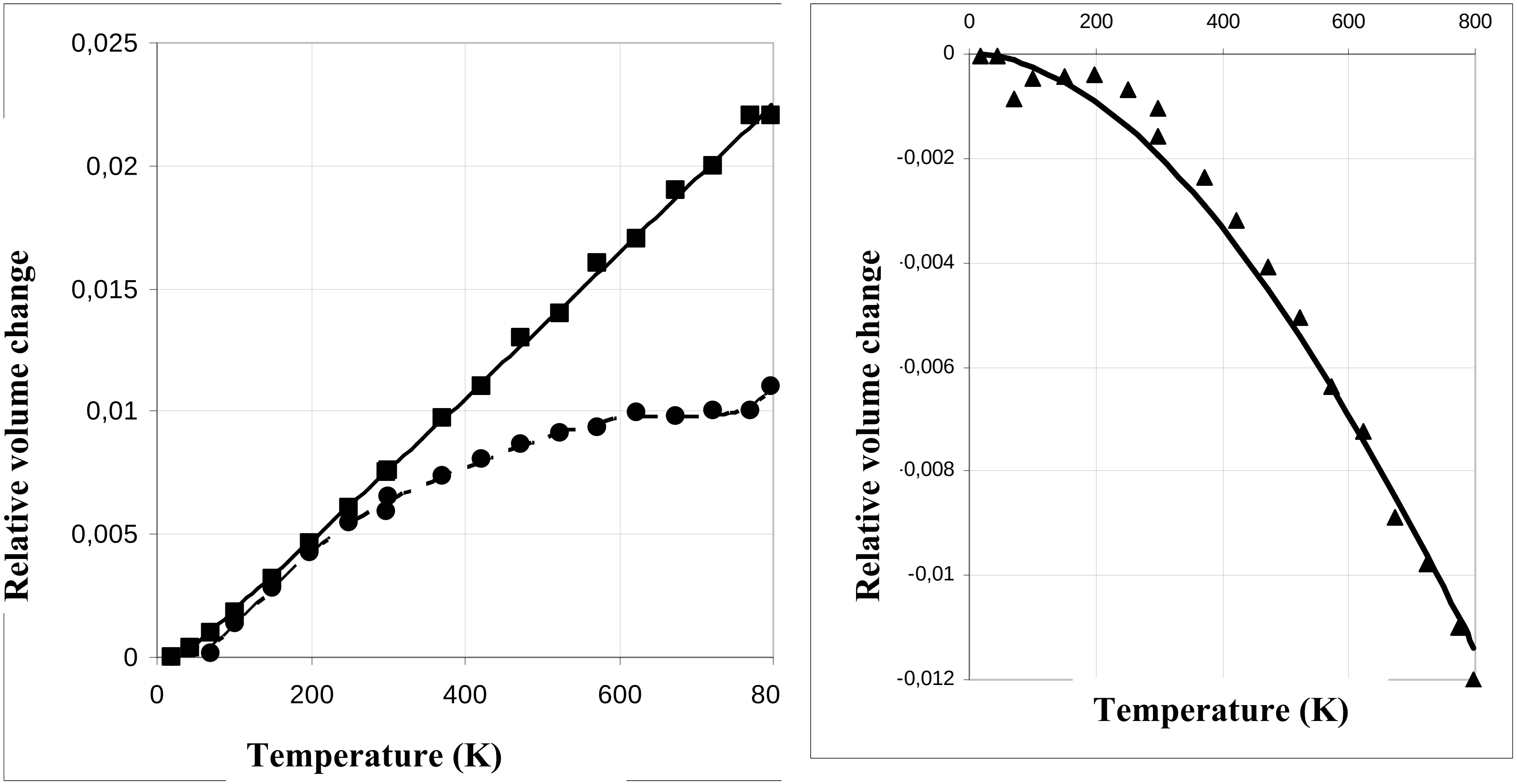


Fig.9. Temperature expansion of *$Pu_{0.96}Ga_{0.04}$*: a) full circles are results of neutron diffraction measurements (A. Lawson et al, Philos. Mag. 86, 2005), full squares are the calculated Debye contribution, b) triangles are non-lattice contribution, solid curve is the calculated SF contribution with $\hbar\omega_{sf} \approx 0.187 meV$ $\Gamma_m \approx -1,75$

(A. Solontsov, V. Antropov, PRB 81 (2010)).

SPIN FLUCTUATIONS VIA MAGNETO-VOLUME EFFECT GIVE RISE TO THE INVAR PHENOMENA AND ANOMALOUS THERMAL EXPANSION IN ACTINIDE AND TRANSITION METAL COMPOUNDS

4. Non-linear magnetic dynamics and relaxation.

Integrating out individual quasiparticle and lattice variables:

$$\frac{d\hat{\rho}}{dt} = \frac{i}{\hbar}(\hat{\rho}\hat{\varepsilon} - \hat{\varepsilon}\hat{\rho}) \Rightarrow$$

Equations of magnetic dynamics: Fermi liquid

$$\chi_t^{-1}(k)m_t(k) = -(\hat{W}_{ltt}m_l m_t)_k + O(m^3),$$

(33)

$$\chi_l^{-1}(k)m_l(k) = -(\hat{W}_{ttl}m_t m_t^*)_k + O(m^3),$$

three-mode coupling **higher order couplings**

Matrix elements

$$\hat{W} = \hat{W}' + i\hat{W}'' = \hat{W}\{\hat{\varepsilon}, \hat{\rho}\}$$

contain all information about dynamics of electronic and lattice systems.

<u>i) Spin wave regime</u> $\Rightarrow$ ~ precession motion, violation of LL equation;

ii) <u>Relaxational regime:</u> non-linear SF dynamics.

<u>Time-dependent Ginzburg-Landau (GL) equations:</u>

$$\frac{1}{\Gamma_0(\mathbf{k})}\frac{\partial \mathbf{M}(\mathbf{k},t)}{\partial t} = -\frac{\delta H_{GL}}{\delta \mathbf{M}(-\mathbf{k},t)} \qquad \textbf{(35)}$$

$$H_{GL} = \frac{1}{2}\sum_{\mathbf{k}} \chi_0^{-1}(\mathbf{k})\left|\mathbf{M}(\mathbf{k})\right|^2 + \frac{\gamma_0}{4}\sum_{\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4=0}\left(\mathbf{M}(\mathbf{k}_1)\mathbf{M}(\mathbf{k}_2)\right)\left(\mathbf{M}(\mathbf{k}_3)\mathbf{M}(\mathbf{k}_4)\right),$$

$\mathbf{M}(\mathbf{k},t) = \delta_{\mathbf{k},0}\mathbf{M} + \mathbf{m}(\mathbf{k},t)$ **- time-dependent order parameter,**

$\chi_0(\mathbf{k})$ **- paramagnetic susceptibility without account of SF.**

Thermodynamics:

$$\Delta F = \frac{1}{2}\int d\psi M_L^2 \rightarrow \frac{1}{2}\int d\chi_0^{-1}(\mathbf{k}=0)M_L^2 \qquad \textbf{(36)}$$

Dynamics: mode-mode coupling:

$$(W_{ltt})_{k=0} = (2W_{ttl})_{k=0} \approx \chi_l^{-1}(\boldsymbol{\kappa}=0)/M \approx 2\gamma M \qquad \textbf{(37)}$$

Non-linear magnetic relaxation of l-SF.

$$\Gamma_0^{-1}(\mathbf{k}) \rightarrow \Gamma^{-1}(k,T) = \Gamma_0^{-1}(\mathbf{k}) + \Gamma_n^{-1}(k,T), \qquad \textbf{(38)}$$

where $\Gamma_n^{-1}(k,T)$ - nonlinear relaxation rate due to mode-mode scattering.

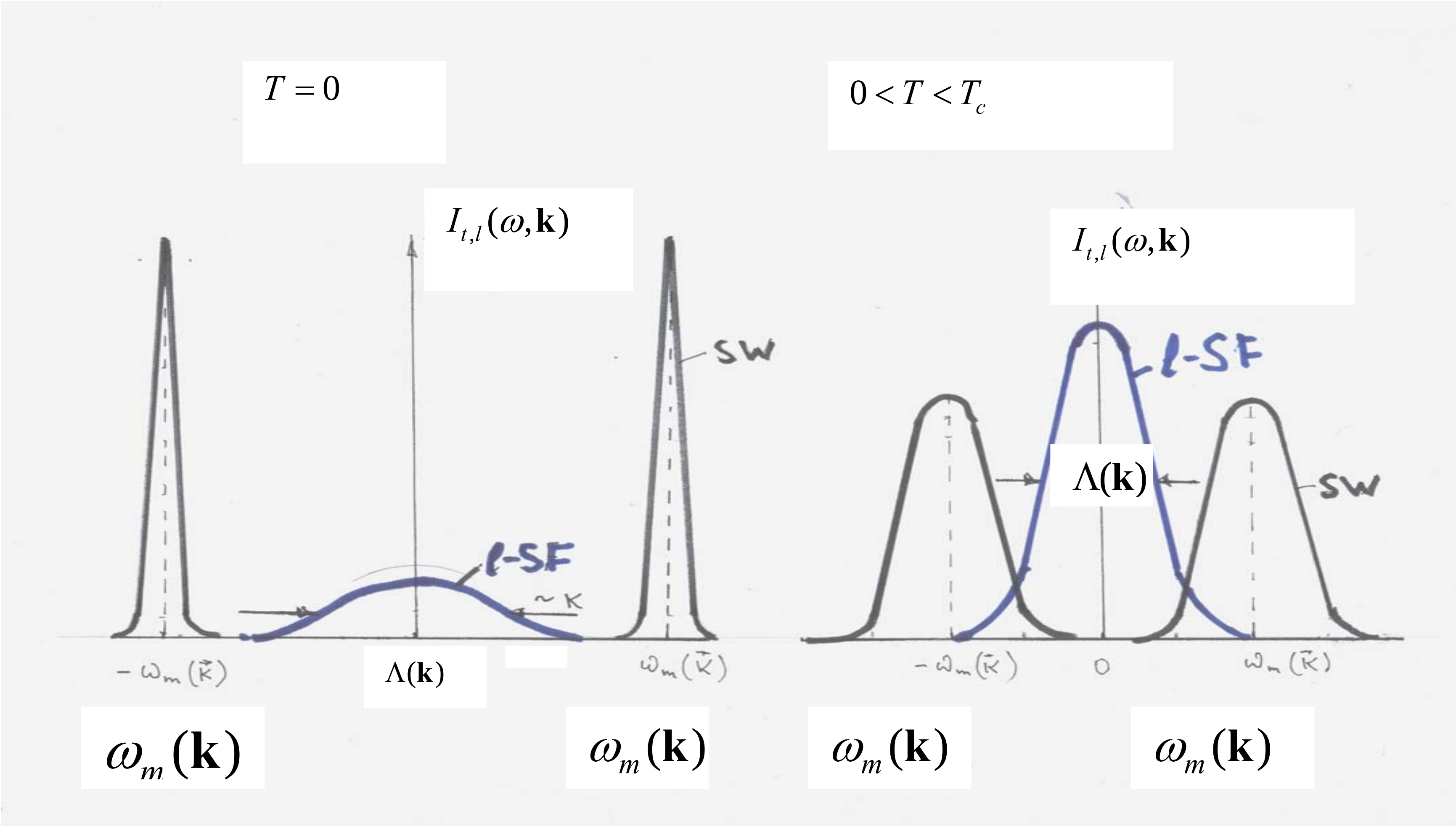


Fig.10. Evolution of the intensity of the SF spectra $I_{t,l}(\omega,\mathbf{k})$ from the linear regime $(T=0)$ to a non-linear one $(T\neq 0)$, $T=0$, where $\Lambda(\mathbf{k},T)=\omega_{sf}(\mathbf{k},T)=\Gamma(k,T)_{\omega=0}\,\chi_l^{-1}(\mathbf{k})$ is the temperature dependent HWHM of the quasielastic central peak.

Three-mode scattering processes ($l \leftrightarrow l' + t$):

(A. Solontsov, JMPB 24 (2005), PRB 81 (2010))

$$\Gamma_n^{-1}(k,T) = \frac{2\hbar}{\omega}\sum_{k'}\left|W_{ltt}\right|^2 \operatorname{Im}\chi_t(k')\operatorname{Im}\chi_t(k+k')\times\left(N_{\omega'} - N_{\omega'+\omega}\right) \quad \textbf{(39)}$$

$$\chi_t(k) \sim \begin{cases} [\omega_m(\mathbf{k}) - \omega - \tau^{-1}]^{-1}, FM, \\ [\omega_m^2(\mathbf{k}) - \omega^2 - 2i\omega\tau^{-1}]^{-1}, AFM, \end{cases}$$

$\omega_m(\mathbf{k}) \sim |\mathbf{k}|^2$ **in FM and** $|\mathbf{k}|$ **in AFM.**

Scattering by magnons:

a) **emission (absorption) of a l-SF by a magnon (FM and AFM):**

$$\omega_m \leftrightarrow \omega'_m + \omega_{SF}$$

b) **annihilation (creation) of magnons resulting in a l-SF (AFM, <u>absent in FM</u>):**

$$\omega_m + \omega'_m \leftrightarrow \omega_{SF}$$

<u>**CDF:**</u> $l - SF \to CDF:\ m \leftrightarrow m' + e,\ m + m' \leftrightarrow e$

or $m \to ph:\ ph \leftrightarrow ph' + e,\ ph + ph' \leftrightarrow e$.

Spectrum of SF: linear vs non-linear regimes

Mode-mode coupling parameter

$$\xi(k,T)=\frac{\Gamma_0\left(\mathbf{k}\right)}{\Gamma_n\left(k,T\right)}, \qquad \textbf{(40)}$$

describes non-linear effects.

Intensity of SF

$$I(k,T)=\frac{1}{\omega}\operatorname{Im}\chi_l(k)=I_0\frac{\omega_m^2(\mathbf{k})/[1+\xi(k,T)]}{\omega_{sf}^2(\mathbf{k})/[1+\xi(k,T)]^2+\omega^2}, \qquad \textbf{(41)}$$

$I_0=\chi_l(\mathbf{k})\omega_{sf}(\mathbf{k})/\omega_m^2(\mathbf{k})$ **is the normalization factor.**

Low-frequency limit $\Rightarrow$ quasielastic SF

$\omega << \omega_m(\mathbf{k}) << k_B T/\hbar$:

$$\xi(k,T) = \xi_0(\mathbf{k},T) = 2\frac{\Gamma_0}{\Gamma_n}\frac{k_B T}{\hbar\omega_m(\mathbf{k})} \sim \frac{T}{\omega_m(\mathbf{k},T)}, \qquad \textbf{(42)}$$

$$\Gamma_n^{-1} \sim W^2 \approx const(\mathbf{k})$$

Weak coupling limit $\xi_0(\mathbf{k},T) << 1$:

weak linear quasielastic peak, HWHM

$$I(k,T) \sim 1/\omega_{sf}(\mathbf{k})$$

$$\Lambda_0(\mathbf{k}) = \omega_{sf}(\mathbf{k}) \qquad \textbf{(43)}$$

Strong coupling limit $\xi_0(\mathbf{k},T) >> 1$:

giant non-linear quasielasic peak, HWHM

$$I(k,T) \sim \xi_0(\mathbf{k},T)/\omega_{sf}(\mathbf{k})$$

$$\Lambda(\mathbf{k}) = \Lambda_0(\mathbf{k})/\xi_0(\mathbf{k},T) >> \Lambda_0(\mathbf{k}) \qquad \textbf{(44)}$$

Fine structure of the SF spectrum:

inelastic l-SF near magnon frequencies $\rightarrow$ **non-propagating l-SF at** $\omega = \pm\omega_m(\mathbf{k})$:

$$\xi(k,T) \sim \xi_0 \ln\left|\frac{1}{[\omega_m(\mathbf{k}) \mp \omega]^2 + \tau^{-2}}\right|, \qquad \textbf{(45)}$$

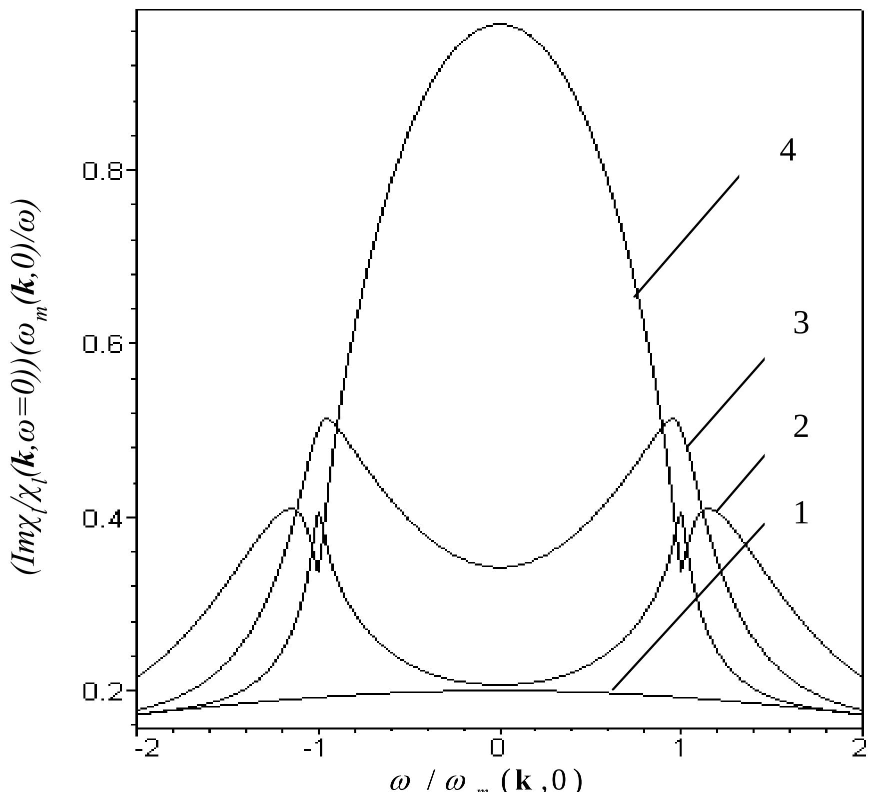


Fig.11. Schematic temperature dependence of the spectrum of l-SF in ferro and antiferro-magnets: 1 – low-temperature weak coupling limit ($\xi_0 << 1$), 2 and 3 – spectra at temperatures $T_2 < T_3 << T_c$, 4 – strong coupling limit ($\xi_0 >> 1$) at higher temperatures.

Scenarios of temperature dependencies of the spectrum of longitudinal SF vs ratio $\eta(\mathbf{k}) = \omega_{sf}(\mathbf{k}) / \omega_m(\mathbf{k})$.

a) No quasielastic dip) $\eta(\mathbf{k}) < 9/2$ (at $T = 0$ quasielastic and magnon peaks are well separated).

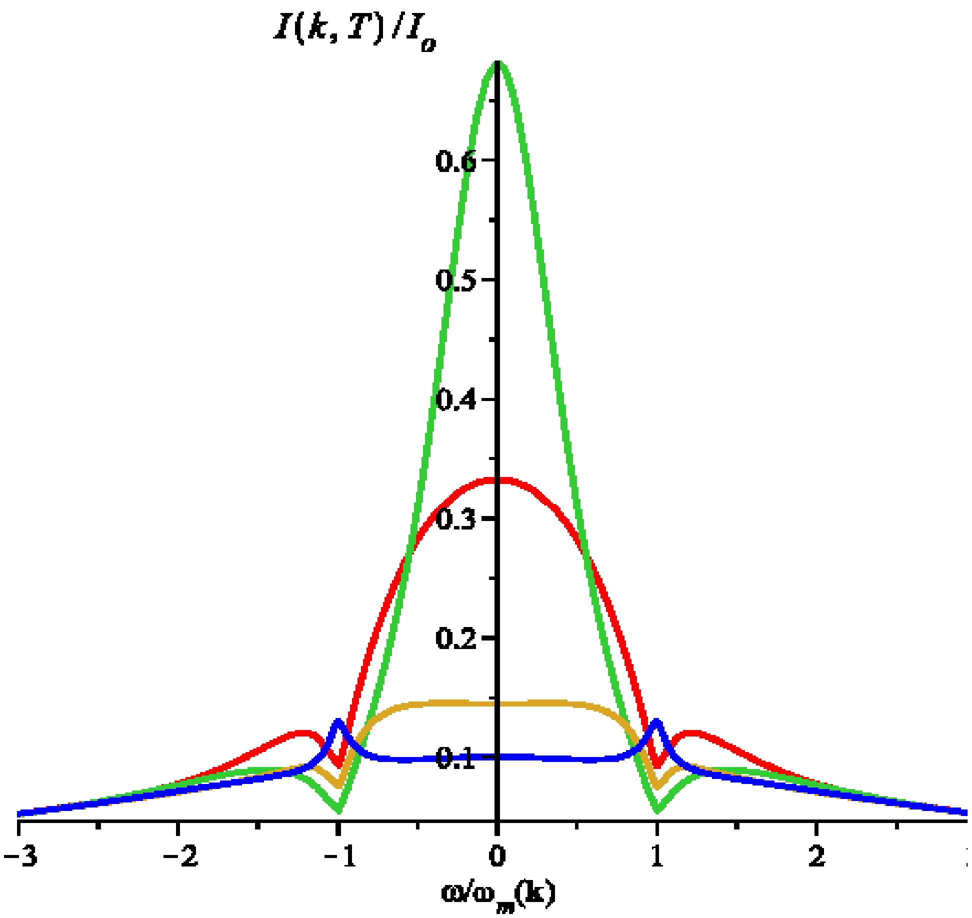
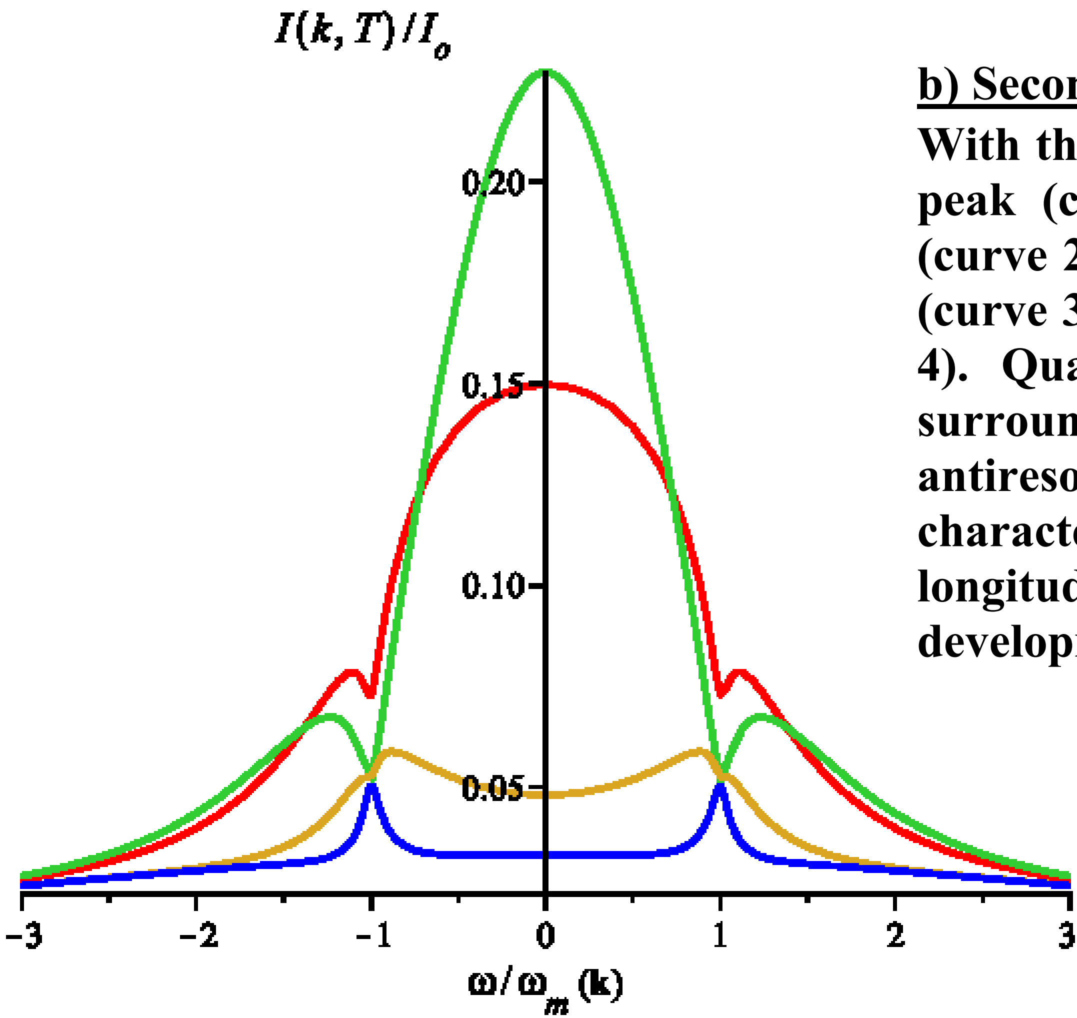


Fig. 9: Spectrum of longitudinal SF in itinerant antiferromagnets as a function of temperature $t = k_B T / \hbar\omega_m(\mathbf{k})$ (for $\omega_m(\mathbf{k})\tau = 0.001$ and $\Gamma_0 / \Gamma_n = 10$). Curves 1 (blue), 2 (dirty yellow), 3 (red), and 4 (green) are calculated for t=0.05, t=0.2, t=0.3 and t=0.5.

(a) First scenario with $\eta^2(\mathbf{k}) = 10$.

The intensity of the central quasi-elastic peak increases with the increase of T and anharmonicity, accompanied by resonances (curve 1), or antiresonances (curves 2, 3, and 4) related to non-propagating l-SF near the magnon frequencies $\omega = \pm\omega_m(\mathbf{k})$.

b) Second scenario $\eta(\mathbf{k}) > 9/2$ (linear quasielastic peak $\rightarrow$ dip $\rightarrow$ non-linear peak) (at $T = 0$ quasielastic and magnon peaks are murged).



<u>b) Second scenario with</u> $\eta(\mathbf{k}) = 30$.

With the increase in T the wide central peak (curve 1) transforms into a dip (curve 2), then back into a central peak (curve 3) and grows in intensity (curves 4). Quasielastic spin fluctuations are surrounded by resonances (curve 1), or antiresonances (curves 2, 3, and 4) characterizing non-propagating longitudinal spin fluctuations developing near $\omega = \pm\omega_m(\mathbf{k})$.

INS in $La_{0.7}Ca_{0.3}Mn_3$: non-linear longitudinal SF

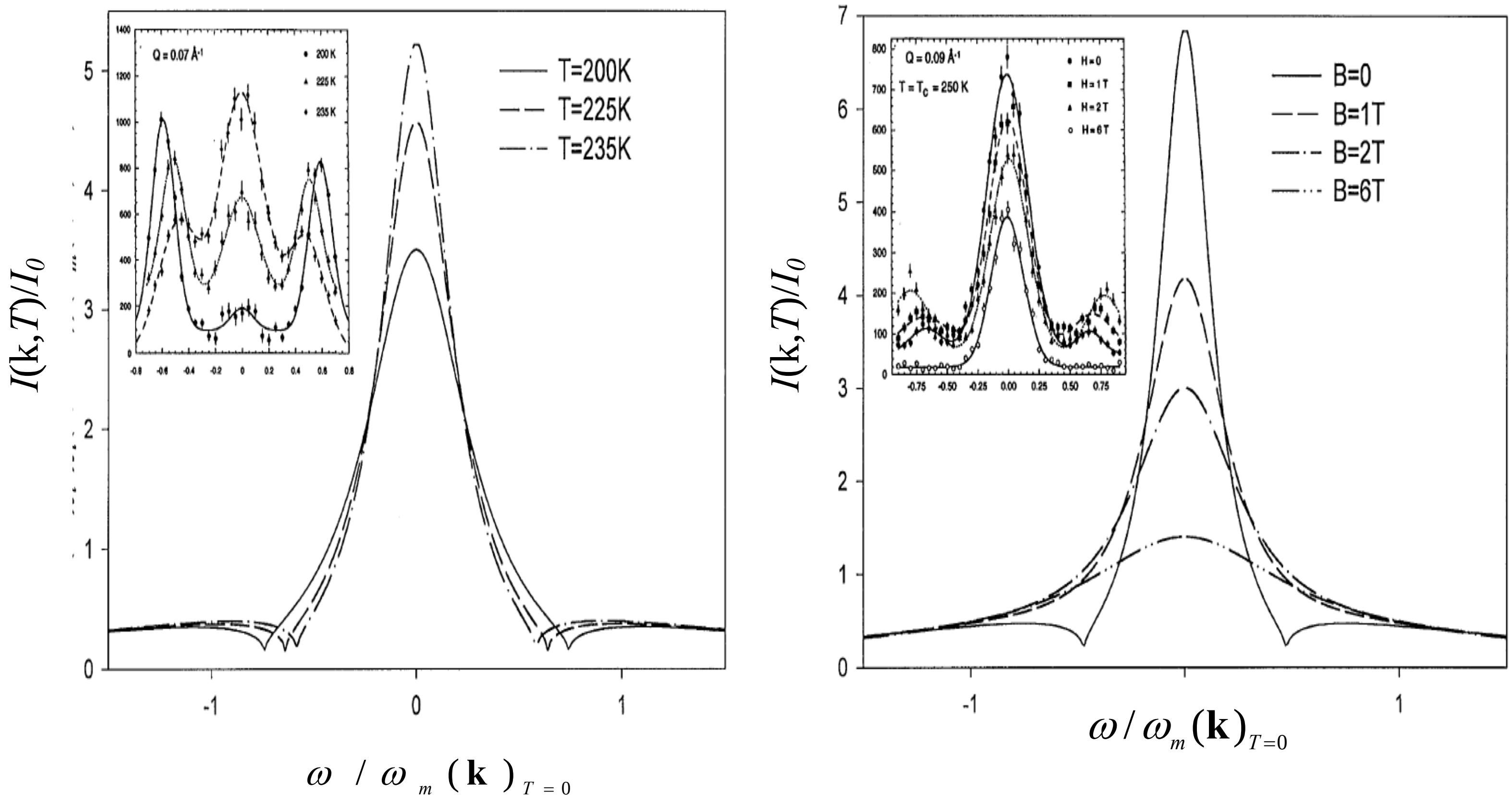


Fig.10. Calculated temperature (a) and field (b) dependencies of the longitudinal SF spectrum in $La_{0.67}Ca_{0.33}MnO_3$.The inserts show the spectrum of the transverse and longitudinal SF in this material measured ***(J.W. Lynn et al, JAP 81 (1997)).***

INELASTIC NEUTRON SCATTERING CONFIRM NON-LINEAR NATURE OF SF IN ITINERANT ELECTRON MAGNETS AT ELEVATED TEMPERATURES

SIMILAR TEMPERATURE DEPENDENCIES EXHIBIT THE SPECTRUM OF CDF WHICH AT ELEVATED TEMPERATURES IS DOMINATED BY NON-LINEAR MECHANISMS OF RELAXATION DUE TO COUPLING WITH MAGNONS AND PHONONS

SUMMARY

- **most of nuclear materials may be regarded as itinerant electron magnets with strong spin fluctuations affecting thermal and kinetic properties;**

- **charge and spin fluctuations essentially contribute to the dissipation of energy by electrons and may strongly influence radiation damage;**

- **giant zero-point spin fluctuations and caused by them strong spin anharmonicity affect the ground state and thermal properties of itinerant magnets;**

- **the excitation spectrum of itinerant magnets at elevated temperatures may be dominated by non-linear spin fluctuations caused by mode-mode couplings (anharmonicity).**

- **traditional concept of linear charge and spin fluctuations in the electron-hole continuum does not hold neither for the ground state nor for finite temperatures.**