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Spin modes in electron Fermi liquid of organic conductors

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The propagation of spin waves in Q2D layered conductors placed in a magnetic field is studied. It is shown, that at certain orientations of the magnetic field with respect to the layers the collisionless absorption is absent and weakly damping spin waves can propagate even under the strong spatial dispersion. We have analyzed the spectrum of spin modes at an arbitrary form of Landau–Silin correlation function.

PACS: 72.15.Nj Collective modes (e.g., in one-dimensional conductors).

Keywords: magnetic field, spin waves, quasi-two-dimensional electron energy spectrum.

A large family of tetrathiafulvalene-based ion-radical salts of the form $(\text{BEDT-TTF})_2\text{X}$ (X stands for a set of various anions) possess layered structure with a pronounced anisotropy of electrical conductivity. Observation of Shubnikov–de Haas magnetoresistance oscillations [1,2] in a magnetic field about 10 T, prove that the free path time τ in these layered conductors can be sufficient for charge carriers to manifest their dynamic properties and their cyclotron frequency ω_B may exceed significantly τ^{-1}. Under that condition, varios type of weakly attenuating collective mode may exist in Fermi liquid of conduction electrons. At present paper we study the collective modes arising from the oscillations of the spin density in layered structures. The paramagnetic spin waves in quasi-isotropic metals were predicted by Silin [3] and observed in alkaline metals by Dunifer and Schultz [4]. The wave processes in layered conductors are characterized by a number of features associated with the quasi-two-dimensional electron energy spectrum. The electrons energy $\varepsilon(\mathbf{p})$ depends weakly on the momentum projection $p_z = \mathbf{pn}$ on the normal $\mathbf{n}$ to the layers and can be represented in the Fourier series with respect to p_z:

$$\varepsilon(\mathbf{p}) = \varepsilon_0(p_x, p_y) + \sum_n^{\infty} \varepsilon_n(p_x, p_y, \eta) \cos\left(\frac{np_z}{p_0}\right). \quad (1)$$

Here $\varepsilon_{n+1}(p_x, p_y, \eta) << \varepsilon_n(p_x, p_y, \eta)$. The coefficient at the first harmonic $\varepsilon_1(p_x, p_y, \eta)$ is of the order of $\eta\varepsilon_F << \varepsilon_F$ where ε_F is the Fermi energy, η is the parameter of the quasi-two-dimensionality of the spectrum, $\hbar$ is the Planck constant. The ratio of the conductivity across the layers to the in-plane conductivity in the absence of a magnetic field, is about η^2. We assume that in an external magnetic field $\mathbf{B}_0 = (B_0 \sin \vartheta, 0, B_0 \cos \vartheta)$ cross-sections $S(\varepsilon_F, p_B)$ of the Fermi surface by the plane $p_B = (\mathbf{pB_0})/B_0 = \text{const}$ are closed for $\pi/2 - \vartheta > \eta$.

Kinetic properties of the system of fermions should be described by means of the kinetic equation for the density matrix $\hat{\rho}$ and the Maxwell equation. In the quasiclassical case when $\hbar\omega_B \lesssim T << \eta\varepsilon_F$ the quantization of the charge carriers energy in the magnetic field does not affect essentially the magnetization $\mathbf{M}$ (T is the temperature). Under these conditions the density matrix can be presented as an operator in the space of spin variables and as a function depending on coordinates and momentum. The Fermi liquid interaction between electrons can be described with the aid of the Landau-Silin correlation function [5,6]

$$\Lambda(\mathbf{p}, \hat{\boldsymbol{\sigma}}, \mathbf{p}', \hat{\boldsymbol{\sigma}}') = L(\mathbf{p}, \mathbf{p}') + S(\mathbf{p}, \mathbf{p}')\hat{\boldsymbol{\sigma}}\hat{\boldsymbol{\sigma}}', \quad (2)$$

where $\boldsymbol{\sigma}$ are Pauli matrices. The second term on the right-hand part of (2) corresponds to the exchange interaction of electrons.

The closed electron orbits in momentum space almost the same for different values of the momentum projection p_B. So the area $S(\varepsilon_F, p_B)$ of the section of the Fermi surface by the plane $p_B = \text{const}$ and the components v_x and v_y of the velocity of conduction electrons in the plane of the layers, depends weakly on p_B, with the order of smallness $\eta \tan\vartheta$. This results that the electrons energy and the Landau correlation function can be expanded into the asymptotic series about η, the leading term of the expansions being not dependent on p_B. In the main approximation in the small parameter η the functions $L(\mathbf{p},\mathbf{p}')$ and $S(\mathbf{p},\mathbf{p}')$ is independent of p_B and can be presented as

$$L(\mathbf{p},\mathbf{p}') = \sum_{n=-\infty}^{\infty} L_n(\varepsilon_F)\mathrm{e}^{in(\varphi-\varphi')},$$
$$S(\mathbf{p},\mathbf{p}') = \sum_{n=-\infty}^{\infty} S_n(\varepsilon_F)\mathrm{e}^{in(\varphi-\varphi')}. \qquad (3)$$

We have chosen the integrals of motion of an electron in a magnetic field ε and p_B and the phase $\varphi = \omega_B t_1$ at its orbit in the magnetic field as variables in the **p** space. Here t_1 is the time of motion along the trajectory $\varepsilon(\mathbf{p}) = \varepsilon_F$, $p_B = \text{const}$. Because of the symmetry of the function $\Lambda(\mathbf{p},\hat{\boldsymbol\sigma},\mathbf{p}',\hat{\boldsymbol\sigma}')$ with respect to its arguments, the coefficients in (3) satisfy the condition $L_n = L_{-n}$, $S_n = S_{-n}$. Allowing for next-order terms of the expansion for the correlation function about η does not lead to the noticeable correction of the results.

The paramagnetic spin waves represent the high-frequency collective modes for which $\omega >> \tau_1^{-1} + \tau_2^{-1}$, where τ_1 and τ_2 are the relaxation times for the electron momentum and spin density respectively. They result from the oscillations of the electron spin density $\mathbf{g}(\mathbf{r},\mathbf{p},t) = \mathrm{Tr}_\sigma(\hat{\boldsymbol\sigma}\hat\rho)$. The function **g** should be presented as a sum of the equilibrium spin density $\mathbf{g}_0 = -\mu\mathbf{B}_0(\partial f_0/\partial\varepsilon)$ and the nonequilibrium correction $-(\partial f_0/\partial\varepsilon)\boldsymbol\xi(\mathbf{r},\mathbf{p},t)$, where $f_0(\varepsilon)$ is the the Fermi function, $\mu = \mu_0/(1+S_0^\sim)$, μ_0 is the magnetic momentum of an electron, $S_n^\sim = \nu(\varepsilon_F)S_n$, $\nu(\varepsilon_F)$ is the density of states at the Fermi level.

The components $\Phi^{(\pm)} = \Phi_{x_1} \pm i\Phi_y$ of the re-normalized nonequilibrium correction

$$\boldsymbol\Phi = \boldsymbol\xi + \langle S\boldsymbol\xi\rangle \equiv \boldsymbol\xi + \int \frac{2d^3p'}{(2\pi\hbar)^3}\left(-\frac{\partial f_0(\varepsilon')}{\partial\varepsilon'}\right) S(\mathbf{p},\mathbf{p}')\boldsymbol\xi(\mathbf{r},\mathbf{p}',t)$$

to the spin density satisfy the integral equation [7,8]

$$\Phi^{(\pm)} = \int_{-\infty}^{\varphi} d\varphi' \exp\left(\frac{i}{\omega_B}\int_{\varphi'}^{\varphi} d\varphi''(\tilde\omega \mp \Omega - \mathbf{kv}(\varphi'',p_B))\right)\times$$
$$\times\left(i\frac{\mu_0}{\omega_B}(\mathbf{kv}(\varphi',p_B) \pm \Omega)B_\pm^\sim - i\frac{\omega}{\omega_B}\sum_{p=-\infty}^{\infty}\lambda_p \overline{\Phi}_p^{(\pm)}\mathrm{e}^{ip\varphi'}\right), \qquad (4)$$

where $\Phi_{x_1} = \Phi_x\cos\vartheta - \Phi_z\sin\vartheta$, the x_1 axis is orthogonal to both the y axis and vector $\mathbf{B}_0$, $\lambda_p = S_p^\sim/(1+S_p^\sim)$, $\overline{\boldsymbol\Phi}_p \langle \mathrm{e}^{-ip\varphi}\boldsymbol\Phi\rangle/\langle 1\rangle$, $\tilde\omega = \omega + i0$, $\Omega = \omega_s/(1+S_0^\sim)$, $\omega_s = -2\mu_0 B_0/\hbar$ is the frequency of spin paramagnetic resonance $\mathbf{v} = \partial\varepsilon(\mathbf{p})/\partial\mathbf{p}$ is the electron velocity. The wave process is supposed to be harmonic, and we present the coordinate and time dependencies of all variable quantities in the form $\exp(i\mathbf{kr} - i\omega t)$.

For the frequencies $\omega << kc$, the alternating magnetic field $B_\pm^\sim = B_{x_1} \pm iB_y$, produced by the spin oscillations is determined from the equation

$$\mathbf{B}^\sim(\omega,\mathbf{k}) = 4\pi[\mathbf{M}^\sim(\omega,\mathbf{k}) - \frac{\mathbf{k}}{k^2}(\mathbf{kM}^\sim(\omega,\mathbf{k}))], \qquad (5)$$

where $\mathbf{M}^\sim(\omega,\mathbf{k}) = \mu_0\langle\boldsymbol\xi(\mathbf{p},\omega,\mathbf{k})\rangle$ is the high-frequency magnetization, c is the velocity of light.

After multiplying the equation (4) by $\mathrm{e}^{-in\varphi}$ and then integrating with respect to $\beta = p_B/p_0\cos\vartheta$ and φ, we obtain the infinitesimal set of linear equations for the coefficients

$$\overline{\Phi}_n^{(\pm)} = \frac{1}{(2\pi)^2}\int_0^{2\pi} d\varphi \int_{-\pi}^{\pi} d\beta\, \Phi^{(\pm)}(\varepsilon_F,\beta,\varphi)$$

$$\sum_{p=-\infty}^{\infty}\left(\delta_{np} - \lambda_p\frac{\omega}{\omega_B}\langle f_{np}(\beta)\rangle_\beta\right)\overline{\Phi}_p^\pm =$$
$$= -\mu_0 B_\pm^\sim \left\langle \frac{1}{2\pi i\omega_B} \frac{\int_0^{2\pi}\int_0^{2\pi} d\varphi d\varphi_1(\mathbf{kv}(\beta,\varphi-\varphi_1) \mp \Omega)\exp\left[-in\varphi + i\frac{\tilde\omega \mp \Omega}{\omega_B}\varphi_1 - iR(\varphi,\varphi_1)\right]}{1 - \exp\left[\frac{2\pi i}{\omega_B}\langle\tilde\omega \mp \Omega - \mathbf{kv}\rangle_\varphi\right]}\right\rangle \equiv F_n, \qquad (6)$$

$$f_{np}(\beta) = \frac{1}{2\pi i}\frac{\int_0^{2\pi}\int_0^{2\pi} d\varphi d\varphi_1 \exp\left[i(p-n)\varphi - ip\varphi_1 + i\frac{\tilde\omega \mp \Omega}{\omega_B}\varphi_1 - iR(\varphi,\varphi_1)\right]}{1 - \exp\left[\frac{2\pi i}{\omega_B}\langle\tilde\omega \mp \Omega - \mathbf{kv}\rangle_\varphi\right]}. \qquad (7)$$

Here

$$R(\varphi,\varphi_1) \equiv \frac{1}{\omega_B} \int_{\varphi-\varphi_1}^{\varphi} d\varphi' \, \mathbf{kv}(\beta,\varphi'),$$

$$\langle \ldots \rangle_\varphi = \frac{1}{2\pi}\int_0^{2\pi} d\varphi \ldots, \quad \langle \ldots \rangle_\beta = \frac{1}{2\pi}\int_{-\pi}^{\pi} d\beta \ldots,$$

δ_{np} is the Kroneker symbol. The dependence of the cyclotron frequency on p_B should be taken into account in the expression $k_x v_x/\omega_B$ in the exponent only provided that $\eta k v_F \gtrsim \omega_B$ where v_F is the Fermi velocity.

The coefficients of the Fourier series for the smooth function $S^{\sim}(\mathbf{p},\mathbf{p}')$ decrease significantly with their number increasing, so we will restrict ourselves to account of a finite number of terms. Making use of this equation it is easy to obtain the magnetic susceptibility, taking into account time and spatial dispersion

$$\chi_\pm(\omega,k) = \frac{\partial M_\pm^{\sim}(\omega,\mathbf{k})}{\partial B_\pm^{\sim}} = \mu\nu(\varepsilon_F)\frac{\Phi_0^{(\pm)}(\omega,\mathbf{k})}{B_\pm^{\sim}} =$$

$$= \chi_0 \frac{\det\left[\delta_{0p}F_n + \left(\delta_{np} - \lambda_p \dfrac{\omega}{\omega_B}\langle f_{np}(\beta)\rangle_\beta\right)(1-\delta_{0p})\right]}{\det\left[\delta_{np} - \lambda_p \dfrac{\omega}{\omega_B}\langle f_{np}(\beta)\rangle_\beta\right]}. \quad (8)$$

The properties of the spin waves are determined by the magnetic susceptibility tensor $\chi_{ik}(\omega,k)$. For the frequencies, that do not coincide with the frequency of eigen-oscillations of the spin density, the components $\chi_{ik}(\omega,k)$ is of the order of the static paramagnetic susceptibility $\chi_0 \simeq \mu_0^2\nu(\varepsilon_F) \sim 10^{-6}$. For this reason, in order to find the spectrum of the spin waves, it suffices to use the homogeneous system of equations corresponding to (6). In the expression (6) we can neglect the small non-uniform correction proportional to $\mu_0 B_\pm^{\sim}$ which allows for influence of the self-coordinated field $B_\pm^{\sim}$. The dispersion equation for «free» oscillations of the spin density is of the form

$$D(\omega,\mathbf{k}) \equiv \det\left[\delta_{np} - \lambda_p \frac{\omega}{\omega_B}\langle f_{np}(\beta)\rangle_\beta\right] = 0. \quad (9)$$

The frequency of eigen-oscillations of the magnetization to within the terms proportional to $\chi_0 \sim \mu_0^2\nu(\varepsilon_F)$ coincides with the frequency of the spin density «free» oscillations. At this frequency the magnetic susceptibility has a sharp maximum, and the determinant $D(\omega,\mathbf{k})$ is of the order of χ_0.

The collisionless absorption of the spin waves is absent if the following inequality is satisfied

$$\left|\omega - n\omega_B \mp \Omega\right| > \max\left|\langle \mathbf{kv}\rangle_\varphi\right|. \quad (10)$$

In the opposite case the integrand in the formula (7) has a pole and after integration with respect to p_B the dispersion equation acquires an imaginary part responsible for the strong absorption of the wave. In a layered conductor the velocity $\mathbf{v}_B = \langle \mathbf{v}\rangle_\varphi$ of drift along the magnetic field of conduction electrons is an oscillatory function of the angle ϑ between B_0 and the normal to the layers. For certain values of the angle $\vartheta = \vartheta_i$ the velocity $\mathbf{v}_B$ is close to zero. At $\vartheta = \vartheta_i$ the collisionless absorption is absent and existence of collective modes is possible [7,8] even under the condition $\eta k v_F \gtrsim \omega_B$. For ω and $\mathbf{k}$ such that $kv_F >> (\omega_B, \omega)$, the solution of the dispersion equation (9) takes the form

$$\omega = n_1\omega_B \pm \Omega + \Delta\omega, \quad \Delta\omega << \omega_B, \quad n_1 = 0, 1, 2 \ldots \quad (11)$$

The correction to the resonance frequency may be written as

$$\Delta\omega = \frac{n_1\omega_B \pm \Omega}{\pi k_x r_0}\gamma, \quad (12)$$

where $r_0 \equiv v_F/\omega_B$ and γ_i are roots of the equation

$$\det\left|\delta_{np} - \lambda_p \gamma^{-1}\langle I_{np}(\beta)\rangle_\beta\right| = 0, \quad (13)$$

$$I_{np}(\beta) = \sum_\alpha \kappa(\varphi^\alpha)\exp\frac{\left[-iR(\varphi^{(\alpha)},\varphi_1^{(\alpha)}) - i(n-p)\varphi^{(\alpha)} - ip\varphi_1^{(\alpha)} + i\dfrac{\pi}{4}s\right]}{\sqrt{\left|\det\left(R''_{\varphi\varphi_1}(\varphi^{(\alpha)},\varphi_1^{(\alpha)})\right)\right|}}. \quad (14)$$

The summation in the formula (14) should be carried out over all stationary points $\varphi^{(\alpha)} = (\varphi^{(\alpha)},\varphi_1^{(\alpha)})$ which are determined from the equations $v_x(\varphi) = 0$, $v_x(\varphi - \varphi_1) = 0$. Here $\kappa(\varphi^{(\alpha)}) = 1$ if the stationary point is inside the domain of integration $0 < \varphi^{(\alpha)} < 2\pi$, $0 < \varphi_1^{(\alpha)} < 2\pi$ and $\kappa(\varphi^{(\alpha)}) = 1/2$ if it is located on a boundary of the domain,

$$s = \operatorname{sign} R''_{\varphi\varphi_1}(\varphi^{(\alpha)},\varphi_1^{(\alpha)}) = \nu_+(R''_{\varphi\varphi_1}) - \nu_-(R''_{\varphi\varphi_1}),$$

$$\nu_+(R''_{\varphi\varphi_1}) \text{ and } \nu_-(R''_{\varphi\varphi_1})$$

are the numbers of positive and negative eigenvalues of the matrix

$$R''_{\varphi\varphi_1} \equiv \frac{\partial^2 R(\varphi^{(\alpha)}, \varphi_1^{(\alpha)})}{\partial\varphi\partial\varphi_1},$$

respectively [9].

At $kr_0 >> 1$, $\omega - \Omega > kv_F$, the phase of exponent in (7) has no stationary points. Integrating (7) by parts and substituting the result (9) we obtain follow asymptotic expression for the spectrum of spin mode

$$\omega = \gamma_i k v_F, \qquad (15)$$

where γ_i are the root of equation

$$\det\left[\delta_{np} - \lambda_p \left\langle \frac{e^{i(n-p)\varphi}}{1 - \gamma^{-1}\mathbf{kv}(\beta,\varphi)/(kv_F)} \right\rangle_{\beta,\varphi}\right] = 0,$$

$$\langle \ldots \rangle_{\beta\varphi} = \frac{1}{(2\pi)^2} \int_0^{2\pi} d\varphi \int_{-\pi}^{\pi} d\beta \ldots .$$

Confining ourselves to account of two terms in the formula (1) and neglecting the anisotropy in the layers plane, we have $\varepsilon_0(\mathbf{p}) = (p_x^2 + p_y^2)/2m$, $\varepsilon_1(\mathbf{p}) = \eta v_F p_0$, where $v_F^2 = 2\varepsilon_F/m$, m is the electron effective mass in the layers plane.

Assuming that the wave vector $\mathbf{k} = (k_x, 0, k_z)$ is oriented in the xz plane, one can obtain, at $kr_0 >> 1$, $\omega < kv_F$, following asymptotic expression for the coefficients f_{np}

$$f_{np}(\beta) = \frac{1}{k_x r_0 (1-\rho^2)} \left\{ \cos[(n-p)\delta] \cot\left(\pi \frac{\langle \tilde{\omega} \mp \Omega - \mathbf{kv} \rangle_\varphi}{\omega_B(\beta)}\right) + \right.$$
$$\left. + \sin\left[\frac{1}{\omega_B(\beta)} \int_{-\delta}^{\delta} d\varphi\, \mathbf{kv} + \pi \frac{\langle \tilde{\omega} \mp \Omega - \mathbf{kv} \rangle_\varphi}{\omega_B(\beta)} - 2\delta \frac{\tilde{\omega} \mp \Omega}{\omega_B(\beta)} + (n+p)\delta\right] \sin^{-1}\left(\pi \frac{\langle \tilde{\omega} \mp \Omega - \mathbf{kv} \rangle_\varphi}{\omega_B(\beta)}\right) \right\}. \qquad (16)$$

Here $\delta = \arccos(\tilde{\omega} \mp \Omega)/\omega_B k_x r_0$, $\rho = (\tilde{\omega} \mp \Omega)/\omega_B k_x r_0$, $\omega_B(\beta) = \omega_B(1 + \eta \tan\vartheta\, J_1(\alpha)\cos\beta)$ is the cyclotron frequency of quasi-particles in the first order approximation in η, $J_n(\alpha)$ is the Bessel function.

For the direction of magnetic field in which $\alpha = (mv_F/p_0)\tan\vartheta$ is equal one of the zeros of $J_0(\alpha)$, the average

$$\langle \mathbf{kv} \rangle_\varphi = \eta v_F J_0(\alpha)(k_x \tan\vartheta + k_z)\sin\beta \qquad (17)$$

is of the order of η^2 and the Landau absorption is absent. Under condition $\omega \mp \Omega << k_x v_F$, the spin waves with the frequencies close to the resonance frequencies $\omega_r = n\omega_B \pm \Omega$ can propagate in layered conductors at an arbitrary orientation of the wave vector and selected directions of the external magnetic field. The solution of (9) can be represented in the form (11),(12) where γ should be determinated from equation

$$\det\left|\delta_{np} - \lambda_p \gamma^{-1}\left(\cos\frac{\pi}{2}(n-p) + (-1)^{n_1}\left\langle \sin\left(R(\vartheta) + \frac{\pi}{2}(n+p)\right)\right\rangle_\beta\right)\right| = 0, \quad R(\vartheta) = \frac{1}{\omega_H(\beta)} \int_{-\pi/2}^{\pi/2} \mathbf{kv}(\varphi)d\varphi. \qquad (18)$$

Let us consider the propagation of spin waves along the magnetic field direction in the case when the magnetic field $\mathbf{B}_0 = (0,0,B_0)$ is orthogonal to the conducting layers. The integral equation (4) takes form

$$\Phi^{(+)} = -\frac{\mu_0 \tilde{B}_+ (k_z v_z + \Omega)}{\tilde{\omega} - k_z v_z - \Omega} +$$
$$+ \omega \sum_{p=-\infty}^{\infty} \lambda_p \frac{\overline{\Phi}_p^{(+)} e^{ip\varphi}}{\tilde{\omega} - k_z v_z - p\omega_B - \Omega}, \qquad (19)$$

and we can obtain the simple analytical expression for spin wave spectrum at an arbitrary values of $\eta k_z v_F$. Determine $\langle \overline{\Phi}^{(+)} \rangle_{\beta,\varphi}$ from (19), we find the high-frequency magnetic susceptibility

$$\chi_+(\omega, \mathbf{k}) = \chi_0 \frac{\sqrt{(\omega-\Omega)^2 - (\eta k_z v_F)^2} - \omega\,\mathrm{sign}(\omega-\Omega)}{\sqrt{(\omega-\Omega)^2 - (\eta k_z v_F)^2} - \lambda_0 \omega\,\mathrm{sign}(\omega-\Omega)}. \qquad (20)$$

The frequency of spin magnetization oscillations are given by

$$\omega = \frac{\Omega + \sqrt{\lambda_0^2 \Omega^2 + (\eta k_z v_F)^2 (1-\lambda_0^2)}}{(1-\lambda_0^2)}. \qquad (21)$$

In long-wave-length limit the frequency of spin waves coincides with frequency of spin paramagnetic resonance ω_s. The specifics of Q2D electron energy spectrum of layered conductors lead that in main approximation in the parameter of the quasi-two-dimensionality η, the Landau–Silin correlation function can be presented as Fourier series (3), the coefficients of which are independent of the momentum projection p_B on the magnetic field direction. This circumstance simplifies essentially the integral equation for spin density and makes it possible to obtain the dispersion equation for rather general form of the correlation function.

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